Quantum Dot Lasing: From Prehistoric Times until Now

Chemically synthesized quantum dots (QDs) can potentially enable new classes of highly flexible, spectrally tunable lasers processible from solutions [1,2]. Despite a considerable progress over the past years, colloidal-QD lasing, however, is still at the laboratory stage and an important challenge - realization of lasing with electrical injection - is still unresolved. A major complication, which hinders the progress in this field, is fast nonradiative Auger recombination of gain-active multicarrier species such as trions (charged excitons) and biexcitons [3]. Recently, we explored several approaches for mitigating the problem of Auger decay by taking advantage of a new generation of core/multi-shell QDs with a radially graded composition that allow for considerable suppression of Auger recombination by “softening” the electron and hole confinement potentials [4]. Using these specially engineered QDs, we have been able to realize optical gain with direct-current electrical pumping [5], which has been a long-standing goal in the field of colloidal nanostructures. Further, we apply these dots to practically demonstrated the viability of a “zero-threshold-optical-gain” concept using not neutral but negatively charged particles wherein the pre-existing electrons block either partially or completely ground-state absorption [6]. Such charged QDs are optical-gain-ready without excitation and, in principle, can exhibit lasing at vanishingly small pump levels. All of these exciting recent developments demonstrate a considerable promise of colloidal nanomaterials for implementing solution-processible optically and electrically pumped laser devices operating across a wide range of wavelengths and fabricated on virtually any substrate using a variety of optical-cavity designs.


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Semiconductor quantum dots (QDs) obtained by epitaxial growth are regarded as one of the most promising solid-state sources of triggered single- and entangled-photons for applications in emerging quantum technologies.


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On-chip single-photon emission from deterministically positioned and embedded InP-based colloidal quantum dots

Solid-state emitters are attractive as non-classical light sources because of the scalability of their host system\textsuperscript{1}. In particular, zero-dimensional quantum dots are prime candidates due to their atom-like properties and the possibility of on-demand single-photon emission. Colloidal quantum dots (cQDs) are often overlooked for this application due to their alleged blinking behaviour or poor stability. However, blinking of cQDs can now be almost completely suppressed through development of appropriate shelling procedures resulting in bright and photo-stable emitters. Usually, this shelling gives rise to multi-exciton emission at high pump rates, a limiting factor for applications requiring pure single photons with unity quantum yield. Recent room temperature characterization of single InP/ZnSe cQDs showed a nearly blinking free emission and a high pure single photon emission ($g^2(0)<0.03$), even well beyond the saturation intensity\textsuperscript{2}. Cryogenic single cQD spectroscopy revealed line-shapes consisting of fine-structure split zero-phonon lines down to 36 µeV width, phonon-assisted transitions resulting in a broad band, as well as spectral jitter. Though clearly promising, isolated cQDs as single photon sources are only usable when they can be deterministically positioned, both for practical purpose and to couple the cQDs to nanophotonic cavities. The latter will be required to further boost the brightness and directionality of single-photon emission. Here, we show the deterministic positioning of single InP/ZnSe cQDs using electron-beam lithography followed by embedding into a silicon nitride host matrix. After the processing, the dots retain their excellent single photon emission at room temperature. Our results pave the way for reliable embedding of cQDs into nanophotonic cavities which will enable bright InP-cQD based single photon emitters.

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A quantum light emitting diode for the standard telecom window around 1550 nm

Quantum communication networks are expected to enable new applications, such as cryptography secured by physical laws, and distributed quantum computing. An essential building block for these applications is a source of pure single photons and entangled pairs, compatible with the low-loss fibre telecom window around 1550 nm. Previous work based on gallium arsenide quantum dots, colour centres in diamond or single atoms has been largely limited to wavelengths unsuitable for long distance fibre quantum network applications. While efforts have been made to use standard gallium arsenide based quantum dots for these applications by extending their operating wavelength range, electrically driven quantum light emission from quantum dots in this ideal telecommunication window has not been possible yet.

Here, we develop indium phosphide based quantum dot devices to address this gap. Recently, it has been shown that QD growth using metalorganic vapour phase epitaxy (MOVPE), which is the industry favoured growth method, can create droplet QDs with low fine structure splitting (FSS). We now extend this growth scheme to produce the first optoelectronic devices for single and entangled photon emission in the 1550 nm telecom window. We show single photon emission with multi-photon events suppressed to 0.11±0.02. From the biexciton cascade, we further obtain entangled light with a maximum fidelity of 0.87±0.04 sufficient for the application of error correction protocols. We can further extend the device working temperature up to 93 K, allowing operation with liquid nitrogen or simple closed-cycle coolers. Our quantum photon source can be directly integrated with existing long distance quantum communication and cryptography systems, and provides a new material platform for developing quantum information technology such as quantum network hardware.

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Scalable quantum photonics using quantum dots

Quantum photonic devices require methods to efficiently generate photonic qubits and to create strong photon-photon interactions. Quantum dots can provide both of these crucial functionalities. They are ideal single photon sources that exhibit both high efficiency and indistinguishability. Furthermore, coupling them to high Q cavities with small mode volumes enables the strong coupling regime where a nonlinearity can enter the single photon regime.

In this talk I will describe our effort to attain scalable quantum photonic devices using quantum dots. I will first describe a quantum transistor where a single photon can control a quantum dot spin and vice versa [1]. This switch realizes a transistor operating at the fundamental quantum limit, where in picoseconds timescales a single photon flips the orientation of a spin and the spin flips the polarization of the photon. This device provides the key mechanism to achieve photon-photon interactions and generate photonic entanglement [2], which is one of the two central requirements for photonic quantum information. I will discuss how this device can be used to achieve efficient optical readout of a quantum dot spin using a cavity QED system [3, 4]. This approach utilizes the spin-dependent cavity reflectivity to determine the spin state, and is particularly important for qubits such as quantum dot spins that do not possess a good cycling transition for resonance fluorescence detection.

I will also describe our efforts to scale the quantum photonic circuits to larger number of devices. I will show a technique to achieve on-chip tuning of quantum dots in order to create indistinguishable emitters coupled to cavities and waveguides [5–7]. I will also describe new fabrication methods we are pursuing for hybrid integration of quantum dots with silicon photonics [6].

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Quantum Frequency Conversion of a Quantum Dot Single-Photon Source Through Integrated Nanophotonics

Future quantum networks may require a combination of different physical platforms such as atoms, ions and solid-state devices, with single photons as information carriers [1]. Implementation of such a hybrid network would require photons of different colors to interact and transfer information between the nodes [1,2]. Therefore, efficient and low-noise frequency conversion of single photons is required to connect network components. Semiconductor quantum dots (QDs) have shown bright single-photon emission, and have been used in quantum frequency conversion experiments using cm-scale quasi-phase matched waveguides [2,3]. In comparison, nanophotonic devices are attractive for frequency conversion due to their size, potential for scalable integration, and low power operation [4]. Here, we report the first demonstration of frequency conversion of single photons from an InAs/GaAs quantum dot through ultra-compact, low-power nanophotonics, via four-wave-mixing Bragg scattering (FWM-BS) in a Si$_3$N$_4$ microring resonator. We measure an on-chip conversion efficiency >10%, and frequency-converted light that is antibunched with $g^{(2)}(0) < 0.5$. In our experiment, single photons at 917 nm from a QD in a micropillar cavity are shifted into two idlers in the presence of two 1550 nm pumps (<20 mW total power). The frequency-converted idlers are spectrally separated from the signal by an amount equal to the difference in pump frequencies, and subsequent spectral filtering selects one of the two idlers. Prior to frequency conversion, the QD photons are antibunched with an intensity autocorrelation $g^{(2)}(0) = 0.08 \pm 0.003$. The frequency-converted idler remains antibunched with $g^{(2)}(0) = 0.29 \pm 0.032$. This verifies that after frequency conversion, the signal is still dominantly composed of single photons.

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Two-photon interference from remote GaAs quantum dots. A prospect towards deterministic multi-photon applications on-demand

Photonic quantum technologies are on the verge of finding applications in everyday life with quantum cryptography and the quantum internet on the horizon. Single epitaxial quantum dots are emerging as near-optimal sources [1] of bright [2], on-demand, highly indistinguishable single photons [3] and entangled photon pairs [4]. In order to build up quantum networks, it is now essential to interface remote quantum emitters. However, this is still an outstanding challenge, as the states of dissimilar quantum dots have to be prepared on-demand with high fidelity, and the generated photons have to be made indistinguishable [5]. Here, we overcome this major obstacle and show an unprecedented two-photon interference (visibility of 51±5%) from remote strain-tunable GaAs quantum dots emitting on-demand photon-pairs. We achieve this result by exploiting the full potential of the novel phonon-assisted two-photon excitation scheme [6], which allows for the generation of highly indistinguishable (visibility of 71±9%) entangled photon-pairs (fidelity of 98±0.5%), it enables push-to button biexciton state preparation (fidelity of 80±2%) and it outperforms conventional resonant two-photon excitation schemes in terms of robustness against environmental decoherence. It allows us to perform a quantum teleportation experiment [7] from one quantum dot (fidelity of 72±1%), laying the foundation for teleporting with two remote quantum dots. Our results mark an important milestone for the practical realization of quantum repeaters and complex multi-photon entanglement experiments involving dissimilar quantum dots.


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Single quantum dot coherence enhanced with nanophotonics

Efficient harvesting and control of the coherence generated by single emitters in solids is a prerequisite for their use in future optical information technologies. Over the last decade, four-wave mixing microspectroscopy (FWM) \cite{1} has emerged as a successful approach to access coherence dynamics of excitons confined in individual quantum dots (QDs). The major difficulty in this experiment lies in distinguishing a weak response of a QD exciton from the resonant excitation background, dominating the signal typically by ten orders of magnitude in the intensity.

Recently, we have demonstrated that using suitable nanophotonic devices - like planar \cite{1} and pillar \cite{2} microcavities, photonic waveguide antennas \cite{3}, and deterministic microlenses \cite{4} - one can locally amplify the field around the QD, enhancing generation of the excitonic FWM signals, and drastically reducing the resonant background. In this talk, I will show how this winning strategy has helped us to progress in the investigations of optical coherence of individual excitons, by accessing the FWM dynamics with a high signal-to-noise ratio.

I will then focus on our current efforts to achieve the long-range radiative coupling within a pair of QDs, by exploiting propagative coherence in deterministically defined photonic waveguides. As an outlook, I will highlight emerging ideas, which now arise from combining coherent nonlinear nanophotonics with optomechanics and two-dimensional materials. Specifically, I will unveil a novel intriguing concept, in which the single exciton coherence is employed as: source, tuning parameter and detector of the motion of semiconductor mechanical resonators.

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Improving a solid-state qubit through an engineered mesoscopic environment

Quantum dots (QDs) offer the current state of the art in intensity and coherence for single photon generation, and serve as a development testbed for quantum networking. Their high brightness is used to generate high-bandwidth spin-photon entanglement [1]. However, the purity and usefulness of the entangled state is severely limited by the dephasing time $T_2^*$ of the QD spin, typically $\sim 2$ ns for electrons in InGaAs QDs. This limit is imposed by fluctuations of the host nuclear spin ensemble. In this work [2], we demonstrate a tenfold extension of $T_2^*$ enabled by prior preparation of the nuclear spin ensemble in a low-entropy state. Taking advantage of a narrowing mechanism based on coherent population trapping (CPT), we lock the nuclear spins around a dark-state resonance, thereby reducing the nuclear field fluctuations [3]. We directly access the narrowed nuclear ensemble by Ramsey interferometry and monitor its dynamics both during preparation and relaxation. Modeling the evolution of the nuclear probability distribution with a Fokker-Planck formalism, we conclude that the prepared state is optimal given the nuclear spin diffusion rate in our system. The CPT-induced correlations within the nuclear ensemble relax over a characteristic time $T_c = 46.4$ ms which lies between the nuclear $T_1$ and $T_2$ times [4]. Our preparation scheme can be readily implemented in quantum information protocols utilizing spin-photon entanglement. Moreover, further access to such a correlated spin ensemble sets the stage for investigations of quantum many-body physics in QDs, possibly leading to a squeezed nuclear ensemble or quantum memories. [1] K. De Greve et al. Nature 491, 421 (2012); W. B. Gao et al, Nature 491, 426 (2012). [2] G. Éthier-Majcher et al., Physical Review Letters 119, 130503 (2017). [3] X. Xu et al., Nature 459, 1105 (2009). [4] G. Wüst et al., Nature Nanotechnology 11, 886 (2016).

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High indistinguishability single photons generation from an in-plane integrated quantum dot waveguide device

Integrated single photon light sources are key building blocks for realizing scalable devices for the on-chip quantum information processing. For such applications highly coherent and indistinguishable single photon are required fulfilling strict requirements imposed by the quantum optics multi-photon-interferometry. Among different kinds of quantum emitters resonantly driven semiconductor quantum dots coupled to the photonic structures have been shown to be one of the brightest sources fulfilling abovementioned conditions [1,2]. Here we report on a resonance fluorescence investigations of an InAs/GaAs quantum dot coupled to a few millimeter long waveguide device. The light confinement and guiding was achieved by distributed Bragg-reflectors in vertical and ridges defined in a horizontal direction. Optical excitation was carried out from the top of the waveguide and emitted photons collected from the side facet of the ridge after few millimeters travel distance simulating a realistic integrated circuit device. A high degree of linear polarization of the emitted photons above 98% and guiding over 2 mm distances in the waveguide was observed. Based on the three-dimensional finite-difference time-domain calculations the coupling efficiency of the quantum dot emission into the waveguide, as well as an overall efficiency of a whole device was estimated. Under pulsed, resonant excitation conditions a second order correlation function as well as a two-photon interference histograms were recorded. In this case, resonance fluorescence photons have been observed with a single-photon purity of >99% and indistinguishability of >98% opening a route towards the fully integrated and thus scalable quantum information processing. X. Ding, Y. He, Z.-C. Duan, N. Gregersen, M.-C. Chen, S. Unsleber, S. Maier, C. Schneider, M. Kamp, S. Höfling, C.-Y. Lu, and J.-W. Pan, Phys. Rev. Lett. 116, 20401 (2016). Y. He, J. Liu, S. Maier, M. Emmerling, S. Gerhardt, M. Davanço, K. Srinivasan, C. Schneider, and S. Höfling, Optica 4, 802 (2017).

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Coherent Coupling of a Single Spin to a Single Photon

Electron spins are excellent candidates for solid state quantum computing due to their exceptionally long quantum coherence times, which is a result of weak coupling to environmental degrees of freedom. However, this isolation comes with a cost, as it is difficult to coherently couple two spins in the solid state, especially when they are separated by a large distance. Here we combine a large electric-dipole interaction with spin-orbit coupling to achieve spin-photon coupling [1]. Vacuum Rabi splitting is observed in the cavity transmission as the Zeeman splitting of a single spin is tuned into resonance with the cavity photon. We achieve a spin-photon coupling rate as large as $g_s/2\pi = 10$ MHz, which exceeds both the cavity decay rate $\kappa/2\pi = 1.8$ MHz and spin dephasing rate $\gamma/2\pi = 2.4$ MHz, firmly anchoring our system in the strong-coupling regime [2]. Moreover, the spin-photon coupling mechanism can be turned off by localizing the spin in one side of the double quantum dot. These developments in quantum dot cQED, combined with recent demonstrations of high-fidelity two-qubit gates in Si, firmly anchor Si as a leading material system in the worldwide race to develop a scalable quantum computer [3].


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High Purcell factor generation of indistinguishable on-chip single photons

On-chip single-photon sources are key components for integrated photonic quantum technologies. Semiconductor quantum dots can exhibit near-ideal single-photon emission but this can be significantly degraded in on-chip geometries owing to nearby etched surfaces. A long-proposed solution to improve the indistinguishability is by using the Purcell effect to reduce the radiative lifetime. However, until now only modest Purcell enhancements have been observed. Here we use pulsed resonant excitation to eliminate slow relaxation paths, revealing a highly Purcell-shortened radiative lifetime (22.7 ps) in a waveguide-coupled quantum dot-photonic crystal cavity system [1]. The quantum dot exhibits near-lifetime-limited single-photon emission which retains high indistinguishability (93.9%) on a timescale in which 20 photons may be emitted. On longer timescales, a modest drop in indistinguishability is observed, indicating the presence of spectral wandering on a timescale of tens of nanoseconds. These results indicate that a large Purcell enhancement significantly improves the measured coherence of quantum dots in on-chip geometries by suppressing both pure dephasing and spectral wandering. When exciting the cavity and collecting from the waveguide, nearly background-free pulsed resonance fluorescence is achieved under π-pulse excitation. This enables the demonstration of an on-chip, on-demand single-photon source with very high potential repetition rates (~10 GHz). Furthermore, we note that the demonstration of a short radiative lifetime has important consequences for other important schemes in quantum information processing such as fast single photon switching and photonic cluster state generation.


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Resonance fluorescence revival in a voltage-controlled semiconductor quantum dot

Solid-state systems like semiconductor quantum dots are very attractive as building blocks for quantum information processes. A lot of effort has been devoted to minimizing dephasing due to coupling of the quantum dots with their surrounding solid-state matrix. Interaction with phonons[1] reduces the degree of indistinguishability and charge or spin noise lead to inhomogeneous broadening of the emission line. Moreover, charge noise can be detrimental as it strongly limits or even suppresses the QD resonance fluorescence (RF). The RF quench has been attributed to the structure residual doping and defects which create a fluctuating electrostatic environment. This can lead to a Coulomb blockade effect preventing the photocreation of an electron-hole pair in the QD. We demonstrate[2] how a revival of the RF can be achieved by using a suitably designed voltage-controlled device that stabilizes the resonantly photogenerated electron-hole pair in the dot. The resonant excitation is realized in a one-dimensional waveguide geometry, while the single photons are collected from the top. By controlling the QD electric field environment by the gate voltage, the charging/discharging mechanisms from the nearby trap states to the QD are disabled. The resonantly photogenerated electron-hole pairs give rise to a very intense RF line and an increase in the coherence time. However, the radiative limit is not reached, suggesting that charge and/or spin noise are still present, leading to residual inhomogeneous broadening. A lower bound for the photons indistinguishability of 0.5 was found. Still, the gate control allows the conversion of one laser pulse into one emitted photon and optimization of the collection efficiency remains the last step to achieve for using such a device in quantum technology applications.


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Optimized quantum photonics

The fundamental building block for implementing numerous quantum technologies is a high quality single photon source, exhibiting high efficiency, high purity (and strong multiphoton probability suppression), high indistinguishability of emitted photons, and high repetition rate. Such a source, with addition of classical optical devices, and possibly quantum memories, could be used to efficiently generate large entangled states of light (such as NOON and cluster states), as well as to implement building blocks of quantum repeaters and quantum simulators. However, in addition to a high quality single photon source, necessary ingredients for implementation of such technologies are high quality photonic components and circuits that are scalable, robust to errors, and exhibit minimal losses.

We have demonstrated high quality quantum light sources based on the effects of photon blockade and tunneling in a strongly coupled InAs/GaAs quantum dot-cavity system [1], and resonant excitation of a quantum dot in a cavity [2]. Despite the very high purity, indistinguishability, and efficiency of such sources, scaling and integration of multiple systems is a challenge because of quantum dot inhomogeneous broadening. On the other hand, color centers embedded in wide bandgap materials, such as silicon vacancy (SiV) color centers in diamond, exhibit far less inhomogeneous broadening. With this platform, we have demonstrated single photon sources with cooperativity C=1.5 and Purcell enhancement > 10, approaching the strong coupling regime of cavity quantum electrodynamics [3], and cavity enhanced Raman scattering from a single SiV in a diamond cavity for detunings of up to 100GHz [4], exceeding the 30GHz inhomogeneous broadening of SiVs in optical structures.

In addition to developments in platforms for light-matter interaction, improvements in classical photonics are also critical to implementing practical quantum technologies. In particular, losses in in/out coupling to chips and increased circuit complexities resulting from post-fabrication tuners are particularly detrimental to quantum circuits. Our recent work on inverse design in photonics offers a powerful tool to design and implement quantum photonic components and circuits with superior properties, including robustness to errors in fabrication and temperature, compact footprints, novel functionalities, and greater than 97% coupling efficiencies for very simple designs [5]. We will show our results on applying inverse design approach to implement optimal quantum photonic circuits.

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A strain-tunable deterministically fabricated single-photon source with high efficiency

Innovative concepts of quantum communication and computation require bright single-photon sources emitting at a precisely determined wavelength. Quantum repeater networks, for instance, need quantum-light sources at coinciding wavelength to enable entanglement distribution [1] and an overlap with atomic resonances is crucial for the storage of photonic quantum states in quantum memories [2,3].

Quantum dots (QDs) can be exploited for the generation of single photons and entangled photon pairs using the biexciton-exciton radiative cascade. However, due to the random nature of the self-assembled growth process, their emission wavelength varies over a wide range. Deterministic nanoprocessing needs to be applied to realize QD-devices with matching target wavelength and an additional fine-tuning is required to meet resonances of other emitters or quantum memories.

Here, we deterministically fabricate wavelength-tunable QD-microlenses by in-situ electron-beam lithography (EBL), which leads to single-photon sources with photon-extraction efficiencies beyond 40% for an NA of 0.4 and low $g^{(2)}(0)$-values.

Precise wavelength adjustment is achieved by employing strain-tuning of the deterministic QD-microlenses. The sample is transferred onto a piezoelectric actuator using a thermocompression goldbonding step, before the in-situ EBL process is conducted on the remaining 570 nm thin QD membrane.

By applying an electric field between -20 and +20 kV/cm to the piezoelectric crystal, the emission of an X transition is tuned repeatably over a range of 0.6 meV which perfectly meets our requirements since the spectral pre-selection accuracy of the in-situ EBL technique is about 0.25 meV. We demonstrate an adjustment tuning accuracy on the order of 1 µeV and strong suppression of multi-photon emission with $g^{(2)}(0) < 0.08$. Together with readily available extensions to the microlenses for fiber-coupling [4] or electrical excitation [5], wavelength-tunable QD microlenses are promising candidates for future plug&play single-photon sources for applications in quantum communication and computation.

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Integration of multiple quantum-dot single-photon sources on a photonic waveguide by transfer printing

Two-photon interference lies at the heart of photonic quantum information processing based on Fock states. Toward scalable quantum information processing, it is vital to implement on-chip two-photon interference using dissimilar single-photon sources (SPSs) integrated on a photonic circuit. In this regard, self-assembled III-V semiconductor quantum dots (QDs) are highly promising because of their deterministic and indistinguishable single photon emission. However, the QDs are inherently random in their emission wavelengths and spatial locations, hindering the integration of multiple identical QD SPSs on a single photonic chip. Here, we report a novel approach for the on-chip integration of multiple QD SPSs. We employ transfer printing, which utilizes an adhesive rubber stamp for picking up a proper QD SPS from a source substrate and subsequently placing it on a photonic chip prepared independently. In this way, it is possible to place any suitable QD SPSs at will in desired locations in the photonic circuit. Experimentally, we first implement an InAs-QD SPS with a GaAs-based photonic crystal nanobeam cavity, which supports a near unity coupling of the QD radiation into a glass-clad waveguide placed underneath. Simply by repeating the transfer process, we succeeded in the integration of two dissimilar SPSs on a photonic waveguide. Both the integrated sources show Purcell-enhanced single-photon emission, confirmed through an exit port attached to the waveguide. It is noteworthy that the demonstrated three-dimensional integration with proper QDs will require highly daunting processes if using conventional hybrid integration approaches, such as wafer bonding. In addition, QD SPSs are bonded to the photonic chip solely by van der Waals force, allowing for the integration with various photonic platforms including those based on silicon photonics. We believe that the transfer-printing-based approach will solve the existing major issues when using QD SPSs and opens the way for highly-functional photonic quantum integrated circuits.

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Colloidal quantum dots for optical gain and lasing
enhanced gain characteristics by QD design

Colloidal quantum dots (QDs) are highly attractive as the active material for optical amplifiers and lasers. Traditionally, optical amplification by QDs is due to stimulated emission from bi-excitons; a transition that provides large gain coefficients yet competes with the efficient non-radiative loss of biexcitons through Auger recombination.

Here, we first address the question how biexciton-related gain in CdSe/CdS core/shell QDs can be optimized by adjusting the core/shell structure. Based on an initial gain model, we predict that reducing the thickness of the CdS shell grown around a given CdSe core will increase the maximal material gain, while increasing the shell thickness will lower the gain threshold. We assess this trade-off by means of transient absorption spectroscopy. Our results confirm that thin-shell QDs exhibit the highest material gain. On the other hand, the significant threshold reduction expected for thick-shell QDs is absent. We relate this discrepancy between model and experiment to a transition from attractive to repulsive exciton-exciton interactions with increasing shell thickness. The spectral blueshift that comes with exciton-exciton repulsion leads to competition between stimulated emission and higher energy absorbing transition, which raises the gain threshold. This makes large-core/thin-shell QDs that feature attractive exciton-exciton interactions the overall champion core/shell configuration in view of highest material gain, lowest threshold exciton occupation and longest gain lifetime.

In a second part, we discuss an approach to circumvent the limitations of stimulated emission by multi-excitons. We start from the experimental observation that HgTe QDs exhibit size-tunable stimulated emission throughout the near-infrared telecom window at thresholds unmatched by any QD studied before. We attribute this unique behaviour to surface-localized states in the bandgap that turn HgTe QDs into 4-level systems. The resulting long-lived population inversion induces amplified spontaneous emission under continuous-wave optical pumping at power levels compatible with solar irradiation and direct current electrical pumping. These results introduce an alternative approach for low-threshold QD-based gain media based on intentional trap states that paves the way for solution-processed infrared QD lasers and amplifiers.

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**Heterogeneous integration for on-chip quantum photonics with single InAs quantum dots**

Single quantum emitters are an important resource for quantum photonics, constituting building blocks for single-photon sources, qubits, and deterministic quantum gates. Robust implementation of such functions, however, can only be achieved through systems that provide both strong light–matter interactions and a low-loss interface between emitter and probing optical fields. We developed a heterogeneous photonic integration platform that provides such features in a scalable on-chip implementation, allowing direct integration of GaAs nanophotonic devices containing self-assembled InAs/GaAs quantum dots within passive photonic circuits composed of low-loss Si$_3$N$_4$ waveguides. We demonstrate a highly efficient optical interface between Si$_3$N$_4$ waveguides and single-quantum dots in various types of GaAs geometries (waveguides, ring and disk resonators, photonic crystals), with photonic performance approaching that of devices optimized for each material individually. With this class of devices, we demonstrate efficient single-photon emission into passive, on-chip Si$_3$N$_4$ waveguides, quantum dot radiative rate enhancement in hybrid microcavities, and outline a path for reaching the non-perturbative strong-coupling regime within our hybrid photonic integration platform. The introduction of single quantum dot based devices as functional elements in quantum photonic circuits enables significant scaling of on-chip photonic quantum information systems, in two complementary ways. First, by acting as chip-integrated on-demand, bright single-photon sources, these devices can significantly boost the photonic flux available for non-deterministic, linear-optics based quantum computation. Furthermore, single-emitters strongly coupled to on-chip cavities provide a path towards single-photon nonlinearities, which would enable deterministic quantum operations through cavity quantum electrodynamics within a quantum network formed by a photonic integrated circuit. While several other hybrid/heterogeneous integration technologies are currently being explored our work is unique in allowing independent, flexible, and high-resolution tailoring of both active and passive photonic circuit elements with precise and repeatable, sub-50 nm alignment defined strictly by lithography.

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Quantum dot rapid adiabatic passage by ultrafast Stark tuning

For the case of pulsed laser fields and in the absence of decoherence, an exciton in a single QD represents a quantum bit, which can be tuned by electric fields. A selective population transfer from the ground state into a desired target state can be performed by Rabi rotations [1]. A robust state preparation can be achieved by using polarization tailored pulses [2] or frequency chirped pulses for the realization of a rapid adiabatic passage (RAP) [3,4]. More flexible and scalable as chirped optical pulses is a hybrid approach using the combination of transform-limited optical pulses and a fast electrically induced chirp by Stark shift. This allows performing RAP with an optical system of reduced complexity in comparison to chirped optical pulses. Our system is based on self-assembled InGaAs QDs embedded in a low capacitance Schottky-photodiode. The photodiode is closely connected to a SiGe:C BiCMOS chip, which delivers transients of the exciton energy as fast as 3.6 µeV/ps at low temperature. The ps transient is synchronized to the optical pulse. The resulting occupancy of the QD can be read out quantitatively by photocurrent detection. We are able to observe the transition from an un-chirped Rabi scenario to a clear RAP signature when the electric chirp is applied. Our experimental findings are in good agreement with a theoretical model based on the optical Bloch equations. References


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Optical properties of armchair and zigzag graphene quantum dots

Semiconductor quantum dots confine Schrödinger Fermions while Dirac Fermions are confined in graphene quantum dots (GQD). The lateral size, shape, edge, number of layers and carrier density allows for engineering of the electronic, optical and magnetic properties of graphene, including continuous tuning of the bandgap from THz to UV [1]. In this work, we present new results illuminating the evolution and oscillation of the energy gap of armchair and zigzag GQDs with size. The GQDs energy spectra are computed using the tight binding and ab-initio density functional methods. The tight binding energy spectra are analyzed by dividing zigzag graphene quantum dots into concentric carbon rings, for which the analytical solutions for the wavefunctions and energy levels, including energy gap, are known. The effect of the tunneling of electron into inner rings is determined and the energy gap is shown to scale as 1/R where R is the radius of the GQD. The growth of a zigzag terminated GQD into armchair terminated GQD is shown to be associated with addition of a one-dimensional Lieb lattice of carbon atoms with a shell of energy levels in the middle of the energy gap of a zigzag terminated GQD. This results in a different dependence energy gap on GQD size and oscillation of the gap energy with size. These results are compared with predictions from confined Dirac Fermion model and tested against ab-initio calculations, including td-DFT calculation of optical gap [2].[1] D.Guclu, P.Potasz, M. Korkusinski and P.Hawrylak, “Graphene Quantum Dots”, Springer-Verlag (2014); P Hawrylak, F Peeters, K Ensslin, Editors, Carbononics—integrating electronics, photonics and spintronics with graphene quantum dots, Focus issue, Physica status solidi (RRL)-Rapid Research Letters 10 (1), 11(2016).[2] Y. Saleem, L. Najera Baldo, A. Delgado Gran, L. Szulakowska and P. Hawrylak, to be published (2018).

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Progress in quantum dot photonics: From science to practical realization

Since the first proposal of semiconductor quantum dots in 1982, the quantum dots have been intensively studied for both fundamental solid-state physics and advanced device applications. Fully quantum mechanical confinement of electrons has enabled the realization of high performance quantum lasers, high-sensitivity quantum dot infrared detectors, and quantum information devices such as single photon sources. Quantum dots can be also applied to solar cells with a forecasted conversion efficiency over 75% toward the future sustainable renewable energy system. Moreover, embedding a single quantum dot inside a photonic nanocavity provides a new platform for studying solid-state cavity quantum electronics (cavity-QED).

In this presentation, we overview recent progress in quantum dot photonics, including practical realization of quantum dot lasers and single photon sources operating above room temperature. Moreover, advances in quantum dot cavity-QED as well as future prospects of the quantum dot photonics are also discussed.

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Colloidal Photocatalysis

Colloidal quantum dots (QDs) have many advantages of heterogeneous and homogeneous photocatalysts for reactions relevant to energy conversion and organic synthesis. This talk focuses on how the surface chemistry of the particle can be tuned to promote selectivity for certain reaction pathways, enantioselectivity of products, and the formation of colloidal assemblies for energy and charge funneling, for reactions such as proton and CO2 reduction, and carbon-carbon coupling.

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Formation of MoS2 Quantum Dots by Thermal Annealing and Memory Device Application

Recently, a few-layer MoS$_2$ was extracted by using a liquid exfoliation technique in organic solvents, and the MoS$_2$ quantum dots (QDs) were generated from bulk MoS$_2$. It is a challenge for making high quality and large-scale MoS$_2$ QDs through extraction of materials from such liquid dispersions without layer aggregation. In this work, we propose a new approach to the synthesis of molybdenum disulfide (MoS$_2$) QDs that uses a single layer MoS$_2$ grown by chemical vapor deposition (CVD) method as a precursor. The MoS$_2$ QDs were formed by a thermal annealing process at 500 °C for 70 hrs to reduce the particle size. Then, CVD-MoS$_2$ covered almost area of the substrate, which showed a typical single layer thickness and 2H-MoS$_2$ structure. A blue shift from 682 nm to 548 nm wavelength was observed in photoluminescence spectra. It shows a successful formation of QDs with 4-7 nm small size from the large sized layers. This process is relatively easy for defining MoS$_2$ QDs with a density of $10^{11}$ cm$^{-2}$ and shows potential advantages for the fabrication of a wide range of electrical optical devices using QDs. We demonstrate also the fabrication of a memory device by employing poly-methyl methacrylate (PMMA) as insulating layer. The fabricated devices have a PMMA-MoS$_2$/metal configuration and exhibit a bistable resistance switching behavior with high/low-current ratio around $10^3$.

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Advances and Challenges in Quantum-Dot Luminescent Solar Concentration for Building Integrated Photovoltaics

The transition to fully energetically sustainable architecture, through the realization of so-called ‘net zero-energy’ buildings, is currently in progress in low density populated areas. This is however, not yet true in cities, where the cost-of-land for ground PV installation is prohibitively high and the rooftop space is too scarce to accommodate the PV modules necessary for sustaining the electrical requirements of tall buildings. For this reason, new technologies are being investigated to integrate solar into building façades in the form of PV windows or envelope elements. Luminescent solar concentrators (LSCs) are the most promising technology capable of realizing semi-transparent, electrodeless PV glazing systems that can be integrated ‘invisibly’ into the built environment without detrimental effects to the aesthetics or the quality of life of the inhabitants. After forty years of research, recent breakthroughs in the realization of broadband absorbing, reabsorption free colloidal semiconductor nanocrystals, have boosted the LSC technology to such a degree that they could be commercialized for building integrated applications in the near future. In this 'tutorial' talk, I will review the successful strategies that have allowed this historical change of pace and discuss the issues that remain to be investigated for further progress. Since a growing number of QD scientists are becoming active in this field, I will dedicate particular attention to provide guidelines for LSC design, lifting common ambiguities on key processes, suitable device architectures and materials.


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Reaching for Commercial Viability of Quantum Dot Photovoltaics

Photovoltaic (PV) modules based on colloidal quantum dots (QDs) thin films could be lightweight and flexible opening up new applications and rapid modes of fabrication and installation. However, as with any new solar PV technology QD PVs must reach low production costs to compete with today’s market-leading crystalline silicon and commercial thin-film PV technologies. We analyzed the cost of synthesizing QD solutions at the large scale needed for PV module production. We used our experience with commercial QD scale-up and developed a Monte Carlo model to analyze the cost of synthesizing lead sulfide and metal halide perovskite QDs with 8 different reported synthetic methods. We considered the cost of solution-phase ligand exchange for preparing deposition-ready PbS QD inks, as well as the manufacturing cost for roll-to-roll solution-processed PV modules using these materials. Results show that present QD synthesis costs are prohibitively high for cost-competitive PV applications. Development of new low-cost synthetic methods is, therefore, critically important for the commercial relevance of QD PVs. Using our cost model, we identify strategies for reducing synthetic cost and propose a cost target of $5/g to move QD solar cells closer to commercial viability.

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Towards perfect photon entanglement with a quantum dot

The on-demand generation of bright entangled photon pairs is an essential resource in quantum optics, quantum communication and quantum sensing. However, a quantum light source combining both high entanglement fidelity and on-demand bright emission has proven elusive with current leading photon technologies. In this work we present a new bright nanoscale source of strongly entangled photon pairs generated with a quantum dot in a nanowire waveguide that satisfies both of these criteria [1, 2]. The quantum dot guarantees highly entangled photon pairs are produced on-demand, while the shape of the nanowire waveguide with a unique taper towards the tip allows the entangled photon pairs to be efficiently collected. These results will be put in perspective with respect to state-of-the-art entangled photon sources with the viewpoint of going beyond in the future towards near-unity fidelity and efficiency. Finally, we show that is possible to reach perfect fidelity with current technology by considering also the detection process in addition to the generation process, even in an indium rich quantum dot with a large nuclear spin [3].


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Unconventional Lasing from Quantum Dots in the Cavity-QED Regime

In the realization of ultra-small semiconductor nanolasers, cavity-QED effects can be leveraged to enhance spontaneous emission and enable lasing with gain from only a few solid-state emitters. Operation in this regime fosters correlation effects that leave their fingerprint especially in the emission dynamics. Through time-resolved photon-correlation spectroscopy of a quantum-dot (QD) photonic-crystal nanolaser, we find that coherence associated with lasing is established on a different timescale than the emission itself [1]. Using a microscopic semiconductor laser model [2], we show that this breakdown of the traditional lasing picture is due to non-Markovian dynamics that are not captured by laser rate-equation theories.

We performed two-photon correlation spectroscopy using a Hanbury Brown and Twiss interferometer to access the correlation function $g^{(2)}(t_1,t_2)$. The device studied contains a QD ensemble that can be temperature-tuned into resonance with a high-quality-factor cavity. Precise control of the emitter number in the cavity allows tuning between LED and lasing regimes. The two-time maps of $g^{(2)}$ show the dynamical transition from thermal ($g^{(2)} = 2$) to coherent ($g^{(2)} = 1$) and back; surprisingly, the formation of coherent emission is delayed by ~250 ps, resulting in the intense portion of the pulse being largely thermal despite the device operating in the regime of stimulated emission.

This is in contrast with the emission dynamics of conventional macroscopic and mesoscale semiconductor lasers in which the onset of coherent emission coincides with the pulse maximum. Using a semiconductor laser model, we find that the delay arises from the non-Markovian time evolution of carrier-photon correlations. By expressing these quantities in a Markovian way, the delay disappears. These findings have direct implications for the modulation response, repetition rate, noise characteristics, and coherence properties of future devices based on high-quality-factor nanolasers.


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On-Chip Quantum Optics in III-V Nano-Photonic Systems

In this talk some of the new physics which may be accessed by confining light on the nanoscale in III-V semiconductor systems will be described. It will be shown that such confinement enables on-chip quantum optics from self-assembled quantum dots with very little interference from environmental effects. Attention will be focussed on three topics: chiral phenomena in non-chiral structures, very large modulations of transmission by single quantum emitters, and the production of very high indistinguishability single photons on-chip. In the latter two cases very close to the transform limit for emission and scattering from single quantum dots is achieved. This is realised by the highly beneficial contribution of Purcell enhancement and field-stabilisation, rendering any residual dephasing and spectral wandering insignificant, opening up a viable route towards scalable quantum optical circuits.

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**Whispering gallery modes of electrons in emission spectra of semiconductor quantum dots.**

Propagation of waves along a closed circular path is quantized and the resulting resonant modes, called whispering gallery modes (WGMs), have wavelengths equal to integer fractions of a path length. Such modes are well known for sound and for photons, and for photons they are widely used, for example, in lasers. Recently WGM cavities for electrons were claimed to be realized in molecular nano-ring [1] and graphene sub-micron-disk [2] and they are expected to be key elements of future solid-state integrated electron optics. In any quantum system electrons behave as localized waves, and one can expect that quantum dots (QDs) can act as the electron WGM cavity. Here we present high-spatial-resolution near-field scanning optical microscopy (NSOM) study of the photoluminescence (PL) spectra of InP/GaInP self-organized QDs containing few electrons, in which we observed WGM. We measured quantum confinement (QC) and electron number (N), using PL spectrum, and size of the maxima (D1) of charge density distribution (CDD), using NSOM imaging, in a series of QDs having QC=0.5-5 meV and N=1-9. We also performed calculations of CDD using configuration interaction approach. The e-WGMs were observed for few N=1 and N=5 QDs, denoted as S and P, and they appear as emission lines, which D1 size measured, is few times larger than the calculated one. In S- and D-dots, e-WGMs have size 120 and 80 nm and were observed for QC~0.7 and 4 meV, respectively. The azimuthal number estimated is 5 and 10. We also observed suppression of WGM in magnetic field (MF) and built up MF induced by WGM. For S-dot we observed decrease of QC in MF, which can indicate fractional charge. [1]G. Reecht et al, Physical Review Letters, 110, 056802, 2013. [2]Y. Zhao et al, Science, 348, 672-675, 2015.

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Tuning individual InAs quantum dots into resonance in photonic network structures with micro-laser processing

Qubit scalability remains one of the most significant challenges to create quantum networks for information processing and communication. Photonic networks of solid state single photon emitters (SPEs) integrated in on-chip structures is a promising approach to this challenge, but variation in solid state SPE emission frequencies is a major impediment to scalability. Semiconductor InAs quantum dots (QDs) are the most advanced solid-state SPEs, with high photon purity, indistinguishability, and generation rate. We present a significant step toward producing many identical QDs with in situ micro-laser processing that locally changes the strain and alloy profile. We demonstrate that individual QD emission frequencies can be deterministically tuned into resonance with other QDs embedded within the same suspended GaAs bridge waveguide and photonic crystal membrane diodes. These structures enable precise control over many of the QD optoelectronic properties, including charging with a controllable number of electrons and holes that can serve as spin qubits.

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GaN/AlN dots-in- a-wire photodetectors

The nanowire geometry offers interesting features for the fabrication of quantum dot devices. In a wire, dots can be stacked keeping a similar diameter, with a height that is controlled by the growth rate (i.e. reduced randomness in comparison with the Stranski-Krastanov approach), and they can be almost free of the influence of structural defects.

Nanowire photodetectors attract broad interest due to their low dimensionality, small electrical cross-section, and ultrahigh photocurrent gain. III nitride nanowires are particularly suitable for spectrally-selective UV photodetection, thanks to their band gap energy and their stability against chemical, mechanical or electrical stress. 1 In a single-GaN- nanowire UV photodetector, the efficiency can be enhanced by the insertion of GaN/AlN quantum dots, which leads to an increase of the responsivity by about two orders of magnitude and improved linearity, while maintaining a UV/visible contrast larger than six orders of magnitude. 2

On the other hand, the insertion of GaN/AlN quantum dots in GaN nanowires is also interesting for infrared photodetection using intraband transitions. We systematically investigated intraband transitions in GaN/AlN nanowire heterostructures, varying their geometry and doping level, targeting absorption at the 1.55 µm telecom band. 3 Based on this study, we present the first single-nanowire quantum well infrared photodetector (NW-QWIP), observing photocurrent at 1.55 µm. 4 Finally, we introduce an extension of the study to cover the mid-infrared spectral range, up to around 6 µm, using intraband transitions in GaN/AlGaN dots-in- a-wire.

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Thermal enhancement of quadrature squeezing in resonance fluorescence

We show that thermal interactions can be harnessed within resonance fluorescence to generate steady-states that emit single photons with a higher degree of quadrature squeezing than in isolated atomic systems. Thermal environments arise naturally, for instance, in solid-state and molecular emitters. Considering the example of a driven quantum dot coupled to phonons, we demonstrate that it is feasible to surpass the maximum level of squeezing theoretically obtainable in an atomic system that lacks such thermal transitions, and indeed come close to saturating the fundamental upper bound on squeezing from a two-level quantum emitter. We thus identify a scenario in which thermal processes in solid-state emitters can be used to access regimes unavailable to their atomic counterparts.

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A control of magneto-optical properties of magnetically doped semiconductor nanocrystals by hyperfine interactions

The control of individual spins in semiconductor nanocrystals is an emerging scientific field which undoubtedly play an important role in the development of new spin-based technologies. Generation of individual spins via incorporation of magnetic ions into colloidal nanocrystals became a hot topic in recent years. These diluted magnetic semiconductor (DMS) nanocrystals are subject to a size confinement on photo-generated carriers, which markedly enhances the mutual carrier-dopant spin-exchange interaction, consequently leading to a pronounced modification of the host's optical and magneto-optical properties. Here we described the synthesis and characterization of DMS nanocrystals based on colloidal nanoplatelets as the host semiconductor matrices, embedded with extremely low concentration of Mn$^{+2}$ ions. This work includes host nanocrystals with a core-shell design (one semiconductor covered by another semiconductor) with dopant ions positioned either in the core or in the shell, using the layer-by-layer deposition. Thus, the work here discusses the influence of internal design on the magneto-optical properties of DMS colloidal nanocrystals. The research involved the use of magneto-photoluminescence (MPL) and optically detected magnetic resonance (ODMR) spectroscopy. The MPL spectra recorded at various temperatures and strength of magnetic field revealed the generation of giant magnetization (~30 Tesla) and g-factor (-4) of the exciton upon doping. The ODMR spectra of those samples designated a resonance line with a distinguished sextet related to the hyperfine interaction between the photo-generated electron and the surrounding nuclear spins of the dopants. The ODMR explored, for the first time, a control of nuclear spins on the magneto-optical properties of DMS nanocrystals. Furthermore, a time resolved ODMR experiment also revealed a shortening of the spin coherence time with respect to estimations, due to the hyperfine interaction. The carrier-nuclear spin may occur either directly, or mediated via the carrier-dopant electron spin (the sp-d interaction).

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Surface science of semiconductor nanocrystals.

Semiconductor nanocrystals are defined by their surface by virtue of their size. Yet their surface remains one of the most poorly understood aspects of their basic science. Here we exploit photoluminescence spectroscopy, resonance Raman spectroscopy, and electron transfer theory to provide insight into the nature of their surface. Temperature dependent and size dependent PL spectroscopy reveals that surface PL is ubiquitous and can be rationalized in terms of semi-classical electron transfer theory in which charges move between the core and the surface states. The theory successfully reproduces the phenomenology of PL from these nanocrystals under the assumption of exciton-ligand coupling at the surface. Our most recent resonance Raman experiments are the first to show direct ligand-NC coupling via resonance enhancement of the ligand modes upon resonance excitation of the core states of the NC. These experiments suggest that the ligands are electronically coupled to the excitonic states of the NC and thus form a composite system.

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Intermixing of direct and indirect states in Si quantum dots: size and shape effect

Quantum dots lack the translational symmetry of the underlying bulk-periodic solids. Therefore electron and hole wave functions represent a mix of bulk bands over different wave vectors and band indices. Here, using single-dot spectroscopy and atomistic pseudopotential calculations, we show how bands of direct and indirect character intermix in silicon nanocrystals as a function of particle size and shape. While bulk Si is an indirect bandgap semiconductor, a strong energy-dependent intermixing is found in quantum dots, affecting their emission and absorption properties. Emission and absorption spectroscopy on individual silicon nanodots [1] and nanorods [2] was performed as a function of temperature. Narrow luminescence linewidth at low temperature and several steps in the absorption curve were found for quantum-confined silicon nanostructures [1]. Dielectric confinement effect was decoupled for nanorods using polarization-dependent measurements [2]. Semiempirical pseudopotentials were used to calculate energy level structure of silicon nanostructures in a broad energy range [1-3]. Since direct and indirect band edges in Si are far in energy, we found that the emission state is only slightly affected by quantum confinement, while higher-lying absorption states get strong intermixing of the direct character [1]. This leads to stronger absorption in nanostructures than in Si bulk per atom. Shape effect is found to change non-linearly with aspect ratio, reflecting giant molecule-like nature of these nanoparticles [2]. The results reveal deficiencies of the commonly used effective mass approximation in describing fundamental properties of quantum dots [3].


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Spins of surface dangling bonds in colloidal CdSe nanocrystals: magnetic polaron formation and giant Zeeman splitting

Colloidal nanocrystals, due to incomplete passivation of surface ions have dangling bonds (DBs), the spins of which can act as the localized spins of magnetic ions. Therefore, non-magnetic nanocrystals demonstrate magnetic properties typical for diluted magnetic semiconductors. We present two examples of effects induced by the DB spins in CdSe nanocrystals at cryogenic temperatures.

First, magnetic polaron formation is observed due to spontaneous orientation of DB spins in the absence of external magnetic field in spherical 2.8 nm diameter CdSe quantum dots [1]. It occurs on time scales of hours and results in (i) spectral shift of the emission lines under selective laser excitation and (ii) decrease of the intensity ratio of zero-phonon line and LO-phonon replica. DB magnetic polaron parameters evaluated from the experiment are: binding energy $\sim 7$ meV, number of DBs per particle $\sim 60$.

Second, in applied magnetic field, the DB spins are oriented resulting in large Zeeman splitting for the exciton interacting with these spins, similar to the known effect of the giant Zeeman splitting in diluted magnetic semiconductors, like (Cd,Mn)Se. The circular polarization degree about 50% in magnetic field of 3 T is observed in bare 1.2 and 1.5 nm thick CdSe nanoplatelets. Further increase of magnetic field leads to unusual behavior: circular polarization decreases to about 20% in 15 T due to increasing of the quantum efficiency from vertically oriented nanoplatelets with unpolarized photoluminescence.

Our findings show that the dangling bond spins are inherent for colloidal nanocrystals. The magnetism that originates from them should affect all types of colloidal nanostructures – with various shapes, and synthesized not only from semiconductors, but also from metals and oxides. [1] L.Biadala, E.V.Shornikova, A.V.Rodina et al., Nature Nanotechnol., 12, 569 (2017).

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Ultrase sensitive, high spectral resolution photocurrent detection of QDs excitons

In optical experiments on single quantum dot photodiodes the exciton ground state transition appears as a two-level system with a lifetime limited linewidth of a few μeV. Photocurrent spectroscopy is a quantitative method to read out the occupancy of the QD. In the past photocurrent detection was limited to a regime with high tunneling rates and elevated excitation powers, which results in photocurrents in pA-range. Therefore the linewidth of QD excitons, as observed in the photocurrent measurements, was broadened due to the lifetime and power broadening. Refinements of the photocurrent detection enabled us to improve its sensitivity down to the fA-range. Using resonant cw laser spectroscopy with high spectral resolution allows us therefore to investigate the linewidth of QDs at exceptionally low tunneling rates and at very low excitation powers. For this work we have fabricated Schottky photodiodes with embedded high quality MBE grown QDs. In resonant excitation experiments we can read out the occupancy of the QD via ultrase sensitive photocurrent spectroscopy. We have been able to demonstrate the linewidth of the excitonic ground state as low as 1.62 μeV in the low tunneling regime. Our result is close to the Fourier transform limit of such systems, as shown by Kuhlmann et al. using resonance fluorescence [1]. References[1] A.V. Kuhlmann et al. Nature Physics 9, 570-575 (2013).

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A Rational Control of the Surface Structure and Electronic Passivation of Lead Halide Perovskite Nanocrystals

Colloidal lead halide perovskite nanocrystals (NCs) recently emerged as promising candidate materials for many optoelectronic applications. Building efficient and long-lasting devices from perovskite NCs however remains a challenge: the unstable composition, related to its ionic core, combined with the labile surface ligand binding lead to structural degradation, it complicates the post-synthesis purification routines, limits the options for ligand exchange, and reduces the colloidal stability. In this study, we introduce a structural framework for classification of the perovskite NC core and surface, deduce the most commonly found aging mechanism using density functional theory, and compute its effect on the electronic structure. Based on our detailed insight into the aging process at the perovskite NC surface, we propose a general strategy for increasing colloidal stability and eliminating traps. By performing tailored experiments, we demonstrate that such an approach is useful to obtain purified CsPbBr$_3$ NCs samples, washed up to three times in several solvents, with near unity photoluminescence quantum yields.

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Light-matter interaction in quantum dot and graphene hybrids

The aim of this talk is to study light matter interaction in hybrids made of quantum dots (QDs) and gapless nanostructures (GNSs) deposited on polaritonic materials. Here we consider graphene, germanene, silicene and narrow band gap semiconductors as GNSs [1, 2] which have surface plasmons. Examples for polar materials are SiO2, SiC, 2D-BN and they have optical phonons [3]. Light scattering in GPNs is controlled by surface plasmons and hence, this research is also known as plasmonics [3]. QDs have bound electron-hole pairs which are called excitons. Electronic and optical properties in QDs can be manipulated by excitons. By using various combinations of GPNs and QDs one can create enormous numbers of hybrid nanostructures. These hybrids are an ideal platform for fundamental and applied research for light-matter interactions [4]. In this talk we mainly investigate the interaction of electromagnetic light (photons) with surface plasmon of GNSs using Maxwell equations and density matrix method. Due to this interaction, new types of particles are created called surface plasmon polaritons and surface phonon polaritons. Energy transfer from external light to hybrids will be studied for energy storage. The photon-plasmon interaction and the energy transfer can be controlled by changing the shape, size and relative positions of the constituents. These are very interesting properties of these nanostructures and they can be used to make nanodevice for applications in nanotechnology and nanomedicine.

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Single Molecule Spectroscopy of Short-Wave Infrared Emitting Quantum Dots

Access to efficient and bright short-wave infrared (SWIR) quantum dot emitters opens up a host of potential applications ranging from high-contrast biological imaging to quantum computation with entangled photon sources. Our lab has made much progress in improving the quantum yield and photostability of SWIR emitting quantum dots by synthesizing core-shell structures of III-V semiconductors using a continuous precursor injection technique. Among these, InAs/CdSe core-shell quantum dots can be spectrally tuned from 1000-1500 nm while maintaining a relatively high quantum yield in the excess of 20%. However, the physical origin of broad emission lineshapes in these systems is poorly understood due to the difficulties associated with performing single-molecule spectroscopic measurements in the SWIR region of the electromagnetic spectrum. Here, we present shell-size dependent single nanoparticle emission linewidth measurements of InAs/CdSe quantum dots obtained by Solution-Photon Correlation Spectroscopy (S-PCFS) performed in the SWIR. Our findings show that the single particle linewidth is significantly narrower than the observed ensemble emission spectrum and moreover increases with shell-growth up to a certain maximum shell thickness. This trend is consistent with the model of an increased exciton-phonon coupling for quantum dots with larger shell diameters. The results suggest that unlike the visible CdSe-based quantum dots whose emission approaches that of the homogeneous linewidth, SWIR emitting quantum dots hold much potential for improvement in their spectral characteristics. Further temperature-dependent studies of their spectral properties will yield detailed information on physical processes that mediate the emission linewidth broadening, paving the way to a better understanding of their fundamental photophysics and opening routes to more optimized synthetic strategies.

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Band Edge Exciton in CdSe Nanocrystals – Revisited

We summarize major corrections to the dark/bright exciton theory [1] which should be used for quantitative description of the band edge exciton in II-VI and III-V compound quantum dot nanocrystals (NCs). The theory which was developed initially for description of CdSe NCs did not take into account the long-range exchange interaction, resulting in the underestimation of the splitting between the upper bright and lower dark or quasi-dark exciton. Several experimental groups have measured this splitting in photoluminescence excitation experiments and resonance photoluminescence excitation and reported the underestimation of this splitting by the old theory. Another type of correction originates from the energetic overlap of the $1P_{3/2}$ and $1S_{3/2}$ hole manifolds in small size NCs. The thermal occupation of the optically forbidden $1S_{3/2}1P_{3/2}$ exciton levels changes the radiative decay time both at helium and at room temperatures. The effect becomes even more pronounced in NCs comprised of semiconductors with spin-orbit coupling, $\Delta_{SO}$, smaller than that of CdSe, where $\Delta_{SO}=420$ meV. Indeed, calculations within the 6-band model demonstrated that the $1S_{3/2}$ and $1P_{3/2}$ hole levels can switch order in small size CdS NC[2] with $\Delta_{SO}=62$ meV and in InP NC $\Delta_{SO}=108$ meV.[3] We demonstrate the role of both effects on the optical properties of CdSe NCs, and compare our predictions with available experimental data. [1] Al.L. Efros, M. Rosen, M. Kuno, M. Nirmal, D.J. Norris, and M.G. Bawendi, Phys. Rev. B 54, 4843 (1996). [2] G.B. Grigoryan, E.M. Kazarayn, Al.L. Efros, and T.V. Yazeva, G.B. Grigoryan, E.M. Kazarayn, Al.L. Efros, and T.V. Yazeva, Soviet Solid State 32, 1031 (1990). [3] T. Richard, P. Lefebvre, H. Mathieu, and J. Alle`gre, Phys. Rev. B 53, 7287 (1996).

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Challenges and opportunities for colloidal quantum dots: a chemist’s perspective

Colloidal chemistry has revolutionized the synthesis of inorganic nanomaterials. The field has evolved tremendously, both in the fundamental understanding of nucleation, growth and surface chemistry of nanocrystals, and in the ability to provide a toolset for preparation of functional materials for various applications. However, current methodology has several limitations that need to be carefully explored and addressed.

The lack of atomic precision in nanomaterial synthesis restricts our ability to harness all the power of this broad and diverse class of materials. Heterogeneity introduces broadening of the absorption and emission spectra, reduces charge carrier mobility in nanocrystal solids, and generally restricts our ability to engineer nanomaterials. I will discuss new approach for colloidal synthesis of nanomaterials with minimal, ideally no, size distribution. The concept is inspired by gas-phase atomic layer deposition (ALD) widely used in microelectronics. Our studies show that the ALD concept can be successfully implemented in solution and, when applied to nanomaterials, enables layer-by-layer growth of crystalline lattices with close-to-atomic precision.

The other general limitation of traditional colloidal chemistry is related to thermal stability of organic solvents at high temperatures required for some hard-to-crystallize materials. Very few traditional solvents remain liquid above 400°C, and solvent or ligands decomposition becomes a serious problem at even higher temperatures. The use of inorganic salts as solvents eliminates this issue and offers new opportunities. As an example, we found that III-V quantum dots synthesized in organic solvents generally have high concentration of vacancies and antisite defects. Annealing or ion-exchanging in a molten salt can eliminate these structural defects without sintering. We envision multiple exciting opportunities for colloidal chemistry in molten salts.

I will also discuss the advances in the surface chemistry of semiconductor nanostructures. Molecular inorganic species can be designed to electronically couple individual nanostructures into nanocomposite materials with high electron mobility. By making surface ligands photochemically active, we introduce a general approach for photoresist-free, direct optical lithography of functional inorganic nanomaterials (DOLFIN). Examples of patterned materials include quantum dots, metals, oxides, magnetic and rare earth compositions. No organic impurities are present in the patterned layers, which helps achieve good electronic and optical properties. The ability to directly pattern all-inorganic layers using a light exposure dose comparable to that of organic photoresists opens up a host of new opportunities for additive nanomanufacturing.

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Photon-spin conversion using gate-defined GaAs quantum dots

Quantum state conversion from a photon polarization state described by a Poincaré sphere to an electron spin state described by a Bloch sphere provides an elemental technology which would contribute to realize a long distance quantum communication. We pursue a quantum interface ”Poincaré interface” based on the quantum state conversion using electron spins in gate-defined GaAs quantum dots (QDs) [1]. Trapping and detecting the single electrons generated by single photons have been developed using a GaAs double QDs [2,3]. Subsequently angular momentum from single photons to single electron spins has been demonstrated in a double QD [4]. Recently, we show paired generation of a single electron in a GaAs quantum dot and a single photon from a single polarization-entangled photon pairs [5]. In this presentation, we also discuss the recent progresses on the improvement of the photon-electron spin conversion efficiency and prospects toward quantum interfaces.


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Electrical initialization of electron and nuclear spins in a single quantum dot at zero magnetic field

The emission of circularly polarized light from a single quantum dot relies on the injection of carriers with well-defined spin polarization. Here we demonstrate single dot electroluminescence (EL) with a circular polarization degree up to 35% at zero applied magnetic field. This highly efficient electrical injection of spin polarized electrons is achieved by combining ultrathin CoFeB electrodes on top of a spin-LED device with p-type InGaAs quantum dots in the active region [1]. The CoFeB ultrathin injector (with a thickness of about 6 atomic planes [2]) presents a Perpendicular Magnetic Anisotropy (PMA) thanks to Fe(Co)–O bondings, a key property in order to inject electron spin along the growth axis of the structure. In addition, we measure an Overhauser shift of several micro-eV at zero magnetic field for the positively charged exciton (trion X+) EL emission, which changes sign as we reverse the injected electron spin orientation. This is a signature of dynamic polarization of the nuclear spins in the quantum dot induced by the hyperfine interaction [3] with the electrically injected electron spin [4]. Both EL circular polarization and Overhauser shift follow the hysteresis cycle of the magnetic electrode. This study paves the way for electrical initialization of electron and nuclear spins in a single quantum dot without any external magnetic field.


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Coherent Raman manipulation of a single quantum dot-confined spin.

Self-assembled InGaAs quantum dots (QDs) can function as high-bandwidth quantum network nodes, providing an ultrafast interface between a single spin and well-defined optical modes [1]. Thus far, full coherent spin manipulation in optically active QDs has made use of far-detuned ps-length laser pulses [2]. Whilst the rotations realised by these pulses have been shown to be highly coherent, they are limited to a single rotation axis and require a mode-locked laser source.

Here, using ns-length laser pulses derived from the coherent sidebands of a sinusoidally modulated laser, we perform full coherent control of a single spin confined to a QD by driving a two-photon transition between its two ground states. This all-optical coherent control technique is fully equivalent to, and as versatile as, microwave spin control. The direction and magnitude of the effective drive linking the two ground states on the Bloch sphere is set by the laser modulation. Controlling the phase of this modulation allows us to choose the angle of our rotation axis in the equatorial plane of the Bloch sphere, giving us fast access to any rotation and allowing us to construct arbitrary composite pulses [3].

Using Rabi oscillations and Ramsey interferometry, we demonstrate complete control of both an electron and a hole spin qubit. We construct a composite pulse sequence which allows us to achieve high fidelity qubit gates even when operating on timescales comparable to the inhomogeneous dephasing time of the spin qubit. Our investigation demonstrates versatile and programmable spin control, and provides a route towards on-chip all-optical spin manipulation.


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Photonic quantum technologies: Metrology, Spins and Photons

Photons are well known as carriers of information in quantum secured key distribution, as gatherers of information in quantum metrology and processors of information in quantum computing. We have made great strides in linear optics quantum information processing in Bristol but come up against scaling limitations due to probabilistic gates. In theory, 1D cavity systems coupled with ground state spins could deterministically generate spin-photon entanglement and thus show great promise to address this scaling challenge. Here I will discuss our ongoing work to realise spin-photon entanglement using quantum dots and colour centres in diamond. They exhibit ground state spins that lead to spin dependent transitions and the potential to entangle ground state spin with emitted or reflected photons[1]. Spin-photon entanglers are effectively universal and deterministic quantum gates enabling quantum memories, quantum repeaters [2] and eventually large scale quantum computation. In our latest work we show that low Q-factor pillar microcavities efficiently couple a dot to a single mode (ie have high β-factor) this enables us to detect large spin dependent phase shift (Faraday rotation) limited by drift in the dot line centre [3]. When we measure the rotation over timescales shorter than the spectral diffusion time (~100 μs) and herald times when the the dot is on resonance we see 80% of photons receive a pi phase shift with 20% of scattered photons not interacting with the dot due to mode mismatch [4]. Our present measurements were performed in a Faraday geometry magnetic field separating spin up and down transitions. Recent measurements in low field and Voigt geometry and on uncharged dots showing single photon switching will be discussed. Quantum dot spins are hard to control with decoherence times measured in microseconds. In contrast the ground state spin in NV-centres in diamond has spin coherence out to milliseconds at room temperature and the polarisation of spin is simply done by reading out the spin in fluorescence measurements. Fabrication of cavity structures coupled to NV-centres is still not perfected so single shot measurements of spin are still limited by light collection efficiency. A review of our progress in NV-centre spin-photon interfaces will be presented.

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Unveiling Kondo physics in a hybrid dot-wire system

Previous experimental studies on interactions between separate bound spins have largely been focused on the system in which a quantum dot is coupled by another one. Here we present a quantum dot-wire system in a GaAs/AlGaAs heterostructure, and employ the quantum dot as a tunable spin impurity to control and investigate spin physics in the nearby quantum wire. In such a hybrid structure, competition between the Kondo effect and the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction, the effects related to single and multiple spins surrounded by itinerant electrons, has been observed and investigated with respect to temperature and magnetic fields. It is worth mentioning that the Kondo-like signatures have been widely studied in quantum wires but there is still no consensus about its origin. In other words, we provide an approach to unveil its mechanism and the results support the formation of emergent localized spins, essential for both Kondo and RKKY effects.

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Fine structure of an exciton coupled to a single Fe2+ ion in a CdSe/ZnSe quantum dot

Here we present a polarization-resolved photoluminescence study of the exchange interaction effects in a prototype system consisting of an individual Fe2+ ion and a single neutral exciton confined in a CdSe/ZnSe quantum dot [1]. A maximal possible number of eight fully linearly polarized lines in the bright exciton emission spectrum is observed, evidencing complete degeneracy lifting in the investigated system. We discuss the conditions required for such a scenario to take place: anisotropy of the electron-hole interaction and the zero-field splitting of the Fe2+ ion spin states. Neglecting either of these components is shown to restore partial degeneracy of the transitions, making the excitonic spectrum similar to those previously reported for all other systems of quantum dots with singlemagnetic dopants [2].


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Environmentally Friendly Quantum Dots for Display Applications

Ever since the physics of quantum dot (QD) materials was discovered, much research effort has been carried out for more than 30 years, and lots of applications adopting QDs have been proposed. Especially, wide color gamut displays using QDs as active light emitting materials have drawn much attention from the academia as well as the industry. And, the QD-based consumer displays such as LED TVs, tablets, and special monitors are now on the market. They provide best color gamut, reasonable power efficiency, and affordable price showing superior competitive edge to OLED technology.

However, up to this point, still there are issues and argues using Cadmium containing materials in practical consumer devices. In spite of the European RoHS Exemptions, we need to be aware the environmental risk of producing large quantity of Cd-containing materials and using them in the consumer electronics. And, this growing apprehension for environmental issues formed great limitation for QD’s applications. Therefore, we have dedicated to develop more environmentally friendly InP based QDs that showed considerably high efficiency and saturated color spectrum compared to the Cd-containing materials. The structure of Cd-free QD was specially tailored for display applications and the synthetic process was optimized to produce reliable materials in commercial scales. In order to improve the efficiency and stability of the QDs in the devices operating under severe atmosphere, specific composite materials were designed and the fabrication process was optimized. From 2015, Samsung has released Cd-free QD adopted UHD TV for major product line-up which show the best color gamut among the current displays. Now we are trying to make additional breakthroughs in displays by using established QD technology and broaden the technology to wider optoelectronic applications.

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Nanoscale Ion-Exchange: a Versatile Route to Tailored Colloidal Quantum Dots

Colloidal quantum dots (QDs) are versatile nanomaterials, whose properties are determined by their size, shape, composition, and compositional profile (i.e., single component, gradient/homogeneous alloy, heterostructured). Heterostructured QDs, quantum rods (QRs) and nanosheets (NSs) are particularly attractive, since the spatial localization of photogenerated charge carriers in these nanocrystals can be manipulated by controlling the energy offsets between the materials that are combined at the heterointerface, which has a dramatic impact on their properties. Moreover, the colloidal nature of these nanomaterials allows one to take full advantage of nanoscale effects to combine size-, shape- and composition-dependent properties with easy surface manipulation and solution processing. These features have turned them into promising materials for many applications, motivating extensive research into synthesis strategies. One particularly promising approach is Nanoscale Ion Exchange, which has emerged in recent years as a resourceful route to size-, shape- and composition-tailored QDs, QRs and NSs that would otherwise not be attainable. The reaction is often topotactic, allowing the product nanocrystals to inherit the size, shape, and heteroarchitecture of the template nanocrystals, thereby circumventing thermodynamic and kinetic barriers, and making it possible to post-synthetically tailor the material properties under constant size and shape. In this contribution, we will discuss recent work by our group in which post-synthetic ion exchange is exploited to achieve the synthesis of colloidal nanocrystals that cannot be obtained by direct synthesis (e.g., PbSe/CdSe core/shell QDs; ultranarrow (Zn,Cd)Te/CdSe heteronanowires; (Zn,Cd)Se core/shell and alloy QDs, wurtzite CuInS₂ QDs, QRs and nanosheets, CuInSe₂/CuInS₂ dot core/rod shell nanorods and concentric core/shell QDs, doped CsPbBr₃:M perovskite QDs). The novel materials and insights provided by our work constitute a significant contribution to the development of targeted synthesis strategies for tailored QDs, QRs and nanosheets.

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Control of Surface Ligands Density on Blue Emitting Core/Shell-Type CdZnS/ZnS Quantum Dots

Core/shell type quantum dots (QDs) have attracted much attention for the application to thin film display and solid-state lighting, owing to their narrow band emission with high photoluminescence quantum yields (PLQY), color tunability, and solution processability. We synthesized deep blue emitting CdZnS/ZnS QDs by conventional hot-injection method. Generally, QDs are purified by precipitation/dispersion method. In this study, we purified the QDs by gel permeation chromatography (GPC) with toluene eluent. PLQY of the QDs purified by GPC was higher (73%) than that of the QDs purified by typical precipitation/dispersion method (64%), due to the thorough removal of free ligands and high boiling point solvent such as 1-octadecene. The GPC purification also enabled to selectively separate the QDs with different ligand density, which was confirmed by NMR and thermogravimetric analysis. Difference of ligand density between the QDs with fewer ligands and the QDs with more ligands was 1.53 times. The both QDs showed almost same photoluminescent properties with 71-73% of high PLQY, 444 nm of deep blue emission maximum, and 22 nm of narrow FWHM. Usually, deligand or ligand exchange process largely deteriorate photoluminescent properties because of the formation of quenching defects at surface of QDs. In this case, we controlled surface ligand density on the blue emitting QDs without changing the photoluminescent properties.

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Halide Perovskites Nanocrystals: Synthesis, Transformations and their Application in Devices

Halide perovskite semiconductors can merge the highly efficient operational principles of conventional inorganic semiconductors with the low-temperature solution processability of emerging organic and hybrid materials, offering a promising route towards cheaply generating electricity as well as light. Perovskites not only show exceptional primary optoelectronic properties such as a direct bandgap, small exciton binding energy, low carrier recombination rates, ambipolar transport, and tunability of the bandgap covering a wavelength range from the near-infrared to the ultraviolet, but they are also very attractive for their ease of processability for mass production (e.g. printing from solution) and for the large availability of their chemical components. Following a surge of interest in this class of materials, research on halide perovskite nanocrystals as well has gathered momentum in the last three years. In such a narrow time span, several properties/features of halide perovskite nanocrystals were investigated, among them electroluminescence, lasing, anion-exchange, as well as control of size and shape such that nanocrystals in the quantum confinement regime were recently reported. The present talk will highlight the research activities of our group on halide perovskite and perovskite-related nanocrystals and films, with emphasis on synthesis, as well as structural, chemical, and surface transformations, and their applications in various types of devices.\(^1\)\(^3\)


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Synthesis of III-V colloidal quantum dots

To facilitate the use of colloidal quantum dots (QDs) in the industry, interest is shifting from the well-characterized cadmium-based QDs to cadmium-free alternatives such as indium phosphide. We recently proposed protocols based on aminophosphine-type precursors that allow for a cost efficient, up-scaled syntheses of indium phosphide (InP) QDs of different sizes.\(^1\) A detailed understanding of the reaction chemistry is a key in the development of colloidal QDs synthesis. We present a complete investigation of chemical reactions leading to the formation of InP starting from aminophosphine-type precursors.\(^2\) This mechanism is innovative in the sense that it points out a double role of the phosphorus precursor in the reaction as both a reducing agent and the source of the phosphorus needed to form InP. Its understanding furthers the general use of aminopnictogens for the synthesis of III-V QDs such as InAs.\(^3\) Finally, We show that InP QDs can be processed in polymer layer and that their structure can be optimized in order to obtain more efficient and cheaper lighting devices.\(^4\)


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Fabrication and optical characterization of a site-controlled three-fold symmetric III-Nitride quantum dot grown by MOCVD

The solid-state single photon emitters based on III-Nitride material have been interested due to the properties such as high degree of linear polarization and room temperature operation. However, anisotropic shapes of QDs from Stranski-Krastanov growth method make difficult to generate polarization entangled photon pairs. The way to control symmetric properties of III-Nitride QD has been significantly issued in order to use them in the quantum information technology. In this work, we demonstrate the fabrication of a site- and dimension-controlled three-fold symmetric InGaN QD using three-fold symmetric nano-pyramidal structure. Nano-pyramids were grown by metal-organic chemical vapor deposition with nano-scale openings which are fabricated by electron beam lithography. The formation of three-fold symmetric GaN pyramidal structure has been established with nitrogen dominant ambient in carrier gases. We controlled the top width of pyramids as scale as QD lateral size (~20 nm) following self-limited growth model with optimizing the growth temperature. The symmetricity of structures was systematically investigated varying growth conditions by scanning electron microscopy. Degradation of the symmetricity has only occurred on the hexagonal structure after complete pyramidal shape even though in the self-limited growth regime, not on the three-fold symmetric structure. We regrew InGaN/GaN single quantum well on the three-fold symmetric structure to form site- and dimension-controlled QD at the apex of the pyramid. We characterize the optical properties (e.g. degree of linear polarization) with three- and six-fold InGaN QD with low temperature micro-photoluminescence measurement. As a result, we confirm that symmetry-controlled III-Nitride QDs were successfully fabricated with the assistance of the nano-structure. These results suggest that III-Nitride QDs have also possibility to demonstrate polarization entangled photon pairs.

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Engineering CQD solids at the suprananocrystalline level and their synergism with 2D materials for optoelectronic applications and solar harvesting

CQD solids lie at the heart of current CQD optoelectronics and solar cells. Albeit, until now huge efforts have been focused on reaching exquisite control over the surface chemistry of CQDs to improve their optoelectronic quality, little attention has been paid in engineering these complex solids at the suprananocrystalline level leveraging electronic interactions within heterogeneous QD composites. In the first part of my talk I will present recent progress in my group on exploiting such remote charge transfer processes in reaching high quality, optoelectronic grade CQD thin films demonstrated in solar cells with record high open circuit voltage and light emitting diodes with very high quantum efficiency. In the second part of the talk the focus will turn to integration of CQD thin films with the a new material platform of 2D materials towards high performance photodetectors and their integration in modern CMOS electronics for broadband image sensor

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Improving quantum dot synthesis with machine learning

Colloidal quantum dots have just reached 12% certified photovoltaic performance by using solution-based ligand exchange that reduces defect densities and improves carrier extraction. Recently, we have uncovered size polydispersity as the major contributor to Stokes shift and open circuit voltage deficit in QD solar cells. Using the data of 2000 batches of PbS QD synthesis we trained the machine learning algorithm to explore the parameter space further and demonstrated improved monodispersity and linewidth. Using similar techniques, we now screen for new Cd- and Pb-free materials in a QD form that can be used to fabricate wider-bandgap solution-processed films and create tandem cells complementing and improving silicon.

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The Colloidal Synthesis of Luminescent Nitride-Based Quantum Dots

III-V nitride semiconductors offer an exciting possibility to tune the intrinsic band gap from the infra-red through the visible into the ultraviolet spectrum by alloying and quantum confinement effects. Whilst nitride based systems have been mainly grown via CVD and MBE routes, they have been used to great effect in light emitting devices and photovoltaic devices [1,2]. At present, the routes to colloidal-based nitride nanoparticles are limited and have mainly utilised nitrification of oxide nanoparticles [3]. More recently, the direct synthesis of nitrides has focused on the thermal decomposition of metal precursors with ammonia, nitrates, azides or sodium amide, for which the resulting particles have demonstrated interesting catalytic and plasmonic features [1,4,5,6,7]. Whilst indium nitride (InN) nanoparticles have been synthesised via these methods, no photoluminescence has been reported thus far [5,6]. However, bright Zn$_3$N$_2$ has been synthesised via the thermal decomposition of highly pyrophoric diethyl zinc whilst bubbling highly volatile ammonia gas through the solution [7]. Alternatives to these highly volatile reagents for large scale manufacture are highly desirable. Here we will present the synthesis and characterisation of bright zinc, indium based nitride nanocrystals, using sodium amide as the nitrogen precursor with tunable emission from the visible into the near infrared and quantum yields as high as 30%.


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Colloidal nanocrystals of APbX$_3$ perovskites [A=Cs$^+$, CH(NH$_2$)$_2^+$, X=Cl$^-$, Br$^-$, I$^-$]: synthesis, spectroscopy and applications

We review the differences that exist in the chemistry and physics of colloidal lead halide perovskite nanocrystals (APbX$_3$ NCs, A=Cs$^+$, FA$^+$, FA=formamidinium; X=Cl, Br, I) as compared with more conventional colloidal QDs. We survey the synthesis methods and prospects of these NCs for optoelectronic applications such as in displays, LEDs, and solar cells, emphasizing the practical hurdles that remain to be overcome. The spontaneous and stimulated emission spectra of these NCs are readily tunable over the entire visible spectral region of 410-700 nm [1-2]. The photoluminescence of these NCs is characterized by narrow emission line-widths of 12-42 nm, wide color gamut covering up to 140% of the NTSC color standard, and high quantum yields of up to 100%. Cs$_{1-x}$FA$_x$PbI$_3$ and FAPbI$_3$ reach the near-infrared wavelengths of 800 nm [3]. These Cs- and FA-based perovskite NCs are highly promising for luminescence downconversion (bright and narrow emission at 530 and 640 nm needed in backlighting of LCD displays or for LEDs). Their practical deployment is, however, hampered by the loss of colloidal and structural integrity due to the facile desorption of surface capping molecules during isolation and purification. To address this issue, we have developed a new ligand capping strategy utilizing common and inexpensive long-chain zwitterionic molecules, resulting in much improved chemical durability [4]. In particular, this class of ligands allows for the isolation of clean NCs with high photoluminescence quantum yields of above 90% in after 4 rounds of precipitation/redispersion. Densely packed films of these NCs exhibit high photoconductivity, high photoluminescence quantum yields and low thresholds for amplified spontaneous emission.1. L. Protesescu et al. Nano Letters 2015, 15, 3692–3696 2. M. V. Kovalenko et al. Science 2017, 358, 745-750 3. L. Protesescu et al. ACS Nano 2017, 11, 3119–3134 4. F. Krieg et al. ACS Energy Letters 2018, DOI: 10.1021/acsenergylett.8b00035

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Quantum of Optical Absorption in 2D PbSe & CdSe Nanocrystal Superlattices

2D semiconductors have been extensively investigated during the last few decades in view of their optoelectronic properties and applications as transistors, light-emitting diodes, photovoltaic cells and lasers. Fang et al. demonstrated that the magnitude of the light absorption on quantum membrane occurs in steps of 1.6%\(^1\). They specifically showed that each interband between the 2D subband results in a quantum unit of \(\pi \alpha / n_c\) (1.6%), where \(\alpha\) is fine structure constant and \(n_c\) is optical local field correction factor. Recently, 2D square and honeycomb superstructure of lead- and cadmium-chalcogenide quantum dots (QDs) have been prepared. These superstructures are formed by assembling PbSe nanocrystals in a monolayer at the toluene suspension air/interface after which the nanocrystals attach via their four vertical \{100\} facets\(^1,2\). CdSe superlattices are then obtained by cation exchange. Theoretical studies show that these 2-D systems have profoundly distinct band structures compared to other continuous nanosheets with the appearance of Dirac cones in the case of the honeycomb\(^3,4\). A tight-binding calculation of the light absorptance shows that square and honeycomb systems have a different absorption spectrum. For CdSe square lattice, the step-wise behavior between each interband transition is observed. Our latest measurements on (Pb-Cd)Se square superstructure show an absorptivity of 1.5 % which is in a good agreement with recent works\(^1,6\). On the other hand, for honeycomb CdSe superlattice, the calculated absorption shows more astounding futures, very different from a quantum well. Since the spectral response of photoconductivity measurement generally follows the absorption spectrum, we performed photoconductivity measurement on lithographically defined electrodes with a layer of honeycomb superstructure on top of them. In addition, detectivity and frequency response of the superlattices were also investigated.


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Kinetic Growth Instabilities in Isotropic Materials Lead to Nanoplatelets

Colloidal nanoplatelets—quasi-two-dimensional sheets of semiconductor exhibiting efficient, spectrally pure fluorescence—form when liquid-phase syntheses of spherical quantum dots are modified. Despite intense interest in their properties, the mechanism behind their anisotropic shape and precise atomic-scale thickness remained unclear, and even counterintuitive when the underlying crystal structure is isotropic. Proposed explanations include templated growth within molecular templates exhibiting layered structures. We experimentally disprove this mechanism for CdSe nanoplatelets by synthesizing them in isotropic melts containing only cadmium carboxylate and chalcogen, a result incompatible with previous mechanisms. Based on our experimental results we developed a theoretical model in which intrinsic kinetic instabilities in growth kinetics cause enhanced growth rates on narrow surface facets. This ultimately leads to the formation of nanoplatelets\(^1\). Additionally, our model predicts lateral Ostwald ripening within each thickness population and complete absence of growth in thickness on nanoplatelets with large surface areas. We show that thicker populations evolve from thinner nanoplatelets with small surface areas, provided through lateral size distribution broadening induced by lateral Ostwald ripening\(^2\). By mixing and heating CdS and CdSe nanoplatelets, we were able to synthesize CdSe/CdS core/crown nanoplatelets. This unequivocally demonstrates material transfer between nanoplatelets and confirms this previously unknown form of Ostwald ripening. We anticipate that our experimental results and the theoretical understanding for controlling the shape at the nanoscale will lead to broader libraries of quasi-two-dimensional materials.\(^1\) Riedinger, A.; Ott, F. D.; Mule, A.; Mazzotti, S.; Knusel, P. N.; Kress, S. J. P.; Prins, F.; Erwin, S. C.; Norris, D. J., Nature Materials 2017, 16 (7), 743-748. 2. Ott, F. D.; Riedinger, A.; Ochsenbein, D. R.; Knüsel, P. N.; Erwin, S. C.; Mazzotti, M.; Norris, D. J., Nano Letters 2017, 17 (11), 6870-6877.

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Directional emission and two photon absorption in colloidal CdSe quantum wells

Intrinsically directional light emitters are potentially important for applications in photonics including lasing and energy efficient display technology. We propose a new route to overcome intrinsic efficiency limitations in light-emitting devices by studying a CdSe nanoplatelets monolayer that exhibits strongly anisotropic and directed photoluminescence. We briefly introduce the electronic and exciton dynamic properties of these nanoplatelets. In the following our analysis of the two-dimensional k-space distribution of the nanoplatelet absorption and emission reveals the underlying internal transition dipole distribution. The observed directed emission is related to the anisotropy of the electronic Bloch states governing the exciton transition dipole moment and forming a bright plane. The strongly directed emission perpendicular to the platelet is further enhanced by the optical local density of states and local fields. In contrast to the emission directionality, the off-resonant absorption into the energetically higher 2D-continuum of states is isotropic. These contrasting optical properties make the oriented CdSe nanoplatelets, or superstructures of parallel-oriented platelets, an interesting and potentially useful class of semiconductor-based emitters for photonic applications. We also demonstrate by 2D k-space spectroscopy that two-photon absorption (TPA) is highly anisotropic in CdSe nanoplatelets, thus promoting them as a new class of directional two-photon absorbers with extremely large cross sections.


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Spin physics in colloidal nanocrystals

While spin physics is a well-developed field for epitaxially fabricated quantum dots, much less knowledge has been worked out for the spin excitations in colloidal crystals. We will give here an overview of the tools that can be used to assess and characterize the spins in these structures. With this knowledge we will give three examples for studying the spin dynamics in colloidal nanostructures: (1) We will first consider the spin relaxation dynamics of charged excitons in quantum dots and identify the underlying mechanisms; (2) then we will show how the spin dynamics can be used to assess the charging state of the nanocrystals after pulsed excitation that is often used for photo-charging; (3) finally we will study the impact of dangling bond spins at the surface of the nanocrystals on excitons in the quantum dots. Their interaction can lead to the formation of magnetic polarons, facilitating also dark exciton recombination.

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Tailor Made Nanocrystals for Materials and Life Science Applications

We will present in situ studies of the nucleation and growth of nanocrystals by optical spectroscopy, x-ray synchroton investigations and mass spectrometry. Magic sized clusters were observed during the growth of cadmium selenide quantum dots. We will show time and temperature dependent data on the growth mechanisms and discuss the special role of magic sized clusters during size focusing and Ostwald ripening conditions. To fully exploit and utilize the unique properties of nanocrystals in nanocomposite materials and life science applications their chemical interaction with their environment must be adjusted. This may lead to either homogeneously dispersed or controlled aggregated nanocrystals in the surrounding matrix or biological environment and determines the finally obtained properties. We will show examples for ligand exchange and encapsulation of quantum dots, plasmonic and magnetic nanocrystals resulting in nanocomposites with tailor made mechanic and electronic properties or adjustable bioresponse.

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CQD: Silicon heterostructures for fast, ultrasensitive infrared detection

Silicon technology currently dominates electronics, photonics, and photovoltaics. Colloidal quantum dots (CQDs) can introduce capabilities to integrated circuits that are beyond silicon capacity; for example, infrared absorption enables detection of light at wavelengths of interest for night vision, optical communications, and photonics circuits. Importantly, IR absorption can be used to extend the spectrum of energies silicon solar cells are sensitive to, increasing the efficiency of photovoltaic modules. Other attractive functionalities which CQDs could add to silicon comprise multiple exciton generation and photon upconversion. However, the integration of conventional silicon technology with novel nanomaterials has thus far been limited and delayed the uptake of technologies such as CQDs.

Here we present - for the first time - the silicon:CQD heterojunction. The fabrication process was designed to overcome fundamental band alignment limitations that would otherwise impede collection of infrared photocharge from the heterojunction; only by developing a novel halide-based surface treatment we were able to make the heterojunction functional. We perform an in-depth investigation of the interface electrostatics (UPS, XPS, Kelvin probe, surface carrier-lifetime) and characterize in full the performance of the junction as an IR photodiode.

The Si:CQD heterostructure is then used to develop a novel infrared-sensitive phototransistor that we term photovoltage field-effect transistor. We use a CQD film as a photo active gate to modulate the conductivity of a silicon channel by controlling the extension of the depletion region arising at the sides of the Si:CQD junction. We demonstrate high sensitivity of ~1e12 Jones, high gain in excess of 1e6, and a fast response time of ~10us, realizing the highest gain*bandwidth CQD based photodetector so far developed. In this work, we break a fundamental limitation that has, until now, exclusively produced high-amplification-slow-response or unity-gain-fast-response CQD light detectors. Our results show that CQDs can be an efficient, cost-effective platform for silicon-based photodetection that is competitive with epitaxial semiconductors. This work was recently presented in the journal “Nature”.

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High Performance Mid-Wave Photodetection with Colloidal HgTe

Thermal imaging in the mid-wave infrared (MWIR) has applications in areas such as autonomous vehicles, environmental monitoring, and diagnostics. However, semiconductor thermal cameras, based on epitaxial materials such as mercury cadmium telluride (MCT), indium antimonide (InSb) or superlattices, are very costly (>\$50,000) and this severely restricts their use. The challenge is to realize a cheap material with a fast response and good sensitivity with operation near ambient temperature. Colloidal quantum dots (CQDs) inks are cheap to manufacture. They are easily deposited and widely compatible with different substrates. In contrast to the flip-bonding technology which is increasingly difficult and costly for high resolution chips, ink coatings work just as well down to any desirable pixel size. Focusing on single element detectors and HgTe CQDs, simple dropcasting methods were used in the fabrication of photovoltaic MWIR photodetectors. Electrical and optical characterizations were performed over a temperature range of 50K to 295K under vacuum, using a calibrated blackbody source. The 5 microns detectors achieved background-limited photodetection (BLIP) below 140K with faster than microsecond response time. Specific detectivities of $10^{11}$ Jones at 80K, $10^{10}$ Jones at 200K and $10^9$ Jones at 250K were achieved with an external quantum efficiency up to 40%. Although these performances already approach commercial HgCdTe and InSb detectors, there should be much room for improvement. With further efforts on synthesis and device structure, mid-IR CQDs have the potential to transform thermal infrared technologies.

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An emerging field of nanocrystal optoelectronics: all-colloidal nanocrystal lasers of quantum dots to wells

Solution-processed semiconductor nanocrystals have attracted great interest in photonics including color conversion and enrichment in quality lighting and display backlighting. Optical properties of these colloidal nanocrystals can be conveniently controlled by tailoring their shape, composition, and size in an effort to realize high-performance light generation and lasing. These nanocrystals span different types and heterostructures of colloidal semiconductors in the forms of quantum dots and rods to more recently emerging quantum wells. Based on the rational design and control of excitonic processes in these nanocrystals, it is possible to achieve highly efficient light-emitting diodes and optically pumped lasers. In this talk, we will present all-colloidal lasers developed by incorporating nanocrystal emitters as the optical gain media in fully colloidal cavities for the first time. As an extreme case of solution-processed tightly-confined quasi-2D colloids, we will also discuss that the atomically flat heteronanoplatelets uniquely offer record high optical gain coefficients and ultra-low threshold stimulated emission. In addition, we will show that controlled stacking of these nanoplatelets provides us with the ability to further tune their excitonic properties. Given the recent accelerating progress in nanocrystal optoelectronics, solution-processed quantum materials hold great promise to challenge their epitaxial counterparts in the near future.

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Carbon dot photoluminescence lifetime thermal sensing

Temperature is a fundamental parameter that affects the behavior of physical, chemical, and biological systems in a wide variety of ways. Thermal sensing and imaging are important in many fields, including integrated photonic devices, nano/microelectronics, biology, and medical diagnostics. Temperature also affects many cellular processes and varies between cells because of biochemical reactions associated with normal cellular metabolism or responses to external stimuli. For example, in hyperthermia therapy, where cancer cells are killed by heating, it is essential to precisely control the temperatures of both the cancer cells and the surrounding healthy tissues in order to make the treatment efficient and safe. Consequently, there is a need for technologies that enable reliable and accurate monitoring of the temperatures of biological systems. In our work, we report the application of carbon dots (CDs) for in vitro intracellular temperature sensing, using the photoluminescence (PL) lifetime of CDs as the temperature-dependent variable. PL lifetime measurements enable accurate temperature determination because they are independent of fluctuations in diverse experimental parameters including the concentration of the CDs and the intensity of the excitation source. The results presented in this work show that CD-based nanothermometers perform extremely well in vitro in human cervical cancer (HeLa) cells. Because CDs have many qualities that are advantageous in biological compounds (good water dispersibility, low cytotoxicity, high biocompatibility, superior photo- and thermostability, extraordinary functional and concentration independency) and not found in other nanothermometers, we believe that these findings open up a wide range of promising applications in biological, biochemical, and medical disciplines where precise monitoring of intracellular temperatures is needed. For example, CD-based nanothermometers could be used to monitor thermal stress in plants or to monitor and increase the effectiveness of temperature-assisted drug delivery/release.

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Flexible colloidal quantum dot triboelectric field-effect transistor with application of touch sensing

In the process of seeking low-cost, printing-compatible materials for flexible systems, semiconductor colloidal quantum dots (CQDs) are gradually appearing for being able to be solution-processed. Other attractive features of CQDs include tunable absorption spectrum and narrow emission linewidth brought by sizes. Besides the above-mentioned properties, the amorphous nature of CQD thin-films enables them to be mass-produced on lightweight, flexible plastic substrates using reel-to-reel printing, spray painting and ink-jet printing. Till now, CQDs have been widely used in solar energy harvesting, logic device and displays. However, CQDs haven’t been widely used on strain and pressure sensing systems since CQDs lack proper transduction mechanisms (i.e. piezoelectric, piezoresistive) to respond to strain or pressure. To expand the territory of CQD to mechanical sensing, we introduced triboelectric phenomenon into CQD device architecture to create a CQD triboelectric field-effect transistor (TFET) combining triboelectric effect and CQD FET to create high-performance touch sensors for flexible systems. In our touch sensor, we use a simple PDMS film to generate triboelectric voltage in response to touching and a floating-gate CQD FET to produce a source-drain current driven by the triboelectric voltage. This design makes it possible to generate a much larger current change compared to the free-standing PDMS touch sensor. The fabrication process of CQD TFET is free of expensive techniques (reactive ion etching etc.) which enables the potential of our devices to be mass produced by printing technologies at low cost in industry. We also enhance the performance of our TFET touch sensors by surface modification strategy. This strategy will largely enhance the mobility of CQD solid, thereby increase the current response, which will pave the way of CQD TFET touch sensors from lab to a huge market.

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Beyond Displays – How Quantum Dots Will Help Power and Feed Future Cities

In recent years, quantum dots (QDs) have gained interest as narrowband emitters for displays, but have traditionally suffered from reliability issues, high manufacturing cost, and toxicity concerns, which prevent them from being incorporated into products beyond displays. At UbiQD, we envision making QDs ubiquitous across many industries with a new kind of QD that is intrinsically more stable, lower-cost, and avoids toxic compounds. We are currently focusing on bringing to market QD-tinted luminescent solar concentrators (LSCs) that enable electricity-generating windows and optimize greenhouse agriculture. In my talk I will highlight the unique optical properties of our QDs and recent LSC developments.

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Quantum optical upconversion photodetector

Night vision, automatic driving, virtual reality, long distance diagnosis all require fast response and high sensitivity infrared detector. Infrared-to-visible upconversion devices offer a simple route for pixel-free infrared detecting and imaging. In this work, quantum dots (QDs) based light harvesting layer and light emitting layer were integrated in a monolithic structure to develop a quantum optical upconversion photodetector. By inserting permanent carriers trapping centers, an electrical field can be formed in the carriers transporting layer under illumination, which allows the formation of carriers tunneling and realization of high external quantum efficiency up to 8000%. The photodetector only device generates overall detectivity over $10^{13}$ jones and response time below 1.5 ms, while the optical upconversion photodetector exhibit remarkably low turn on voltage of 2 V and maximum luminescence intensity over 100 cd/cm$^2$. In the end, we fabricated a flexible optical upconversion photodetector to showcase its potential for wearable devices.

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CuInS2 Nanocrystals: Ideal Optical Properties via Synthetic Insights

CuInS2 nanocrystals have emerged as a practical stand-out within the ternary nanocrystal material class for optoelectronic applications, ranging from photovoltaics to luminescent solar concentrators (LSCs). Owing to unique defect-related photoluminescence physics, CuInS2 nanocrystals show high photoluminescence quantum yields and tunable emission wavelength via size and composition control. However, synthetic methods have lagged behind the application of CuInS2 nanocrystals, which has thereby limited the number of spectroscopic insights available for this material. Here, we present a generalized method for the preparation of CuInS2 nanocrystals with ZnS shells, which show the first evidence of a "true" shell formation, as opposed to a "cation-exchanged" shell. By understanding the kinetic dynamics of shell growth, we present a number of novel nanostructures which ultimately allow us to showcase optimal CuInS2 nanocrystals, with high quantum yield, narrow photoluminescence linewidth, and high photostability.

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Surface Chemistry of III-V Semiconductor Nanocrystals for Photovoltaics

Wet chemistry synthesis and shape/size control of III-V (InP, InAs) colloidal quantum dots (CQDs) has been challenging, compared to well-established synthetic solution chemistry for II-VI (CdSe, CdS) and IV-VI (PbS, PbSe) CQDs. The difficulty stems from the limited choices of precursors suitable for solution chemistry of the III-V materials. At more fundamental level, the difficulty lies in the inherent covalency of the III-V materials, which does not allow an easy solution chemistry to control their surfaces. Therefore, the key is how to control the surface chemistry of rather covalent III-V materials. Based on our previous experience on the atomic control of surface chemistry for IV-VI rock-salt materials, a novel surface chemistry of tetrahedrally-coordinated III-V colloidal quantum dots will be presented. Further control over the surface energy of specific facet allow morphology evolution from tetrapod to tetrahedral shape. Finally, defect-controlled conductive CQD solids as an electron transport component of photovoltaics will be discussed.


S. Tamang, S. Lee, H, Choi, and S. Jeong, Tuning Size and Size Distribution of Colloidal InAs Nanocrystals via Continuous Supply of Prenucleation Clusters on Nanocrystal Seeds, Chem. Mat., 2017, 28, 8119

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Tuning color and transparency in colloidal quantum dot solar cells via multi-objective optimization

Controlling the reflection and transmission spectra of solar cells, resulting in tinted and semi-transparent devices, is of particular interest for applications in building-integrated photovoltaics and vehicular heat and power management. Solution-processed solar cells are suitable for these applications due to their ease and flexibility of fabrication, thin film and lightweight nature, associated low costs, and high efficiency potential. Two advantages of colloidal quantum dots (CQDs) for color-tuned photovoltaics are their tunable band gap and their large spectral absorption range that can extend into and beyond the near infrared (NIR) portion of the spectrum. Color-tuning and semi-transparency are traditionally achieved at the expense of material absorption in the visible portion of the spectrum. However, if suitable spectral engineering techniques could be employed, these losses in the visible portion of the spectrum could be overcome via strong NIR absorption. Using thin film interference engineering and multi-objective optimization algorithms, we developed a technique for generating arbitrary spectral profiles in multilayered solar cell stacks. To achieve an objective apparent color or transparency level, we employ population-based multi-objective optimization algorithms, combined with electric field profile calculations within multilayer stacks, to maximize reflection or transmission over an applicable wavelength range while maximizing photocarrier generation. Experimentally, we fabricate color-tuned devices with photocurrents of 10-15 mA/cm² and semitransparent devices with ~ 30% visible transparency. Our results indicate that designs with minimum transparency do not necessarily translate to the highest attainable photocurrents, thus providing a route to highly-efficient color-tuned devices. We fabricated proof-of-principle blue, green, yellow, red and semi-transparent devices with measured reflectance and transmittance spectra that match well with predicted color and transparency levels. Our optimization technique provides a sustainable foundation for designing spectrally-selective optoelectronic devices.

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Mixed Quantum Dot Solar Cells

Colloidal quantum dots are emerging solution-processed materials for large-scale and low-cost photovoltaics. The recent advent of quantum dot inks has overcome the prior need for solid-state exchanges that previously added cost, complexity, and morphological disruption to the quantum dot solid. Unfortunately, these inks remain limited by the photocarrier diffusion length. Here we devise a strategy based on n- and p-type ligands that judiciously shifts the quantum dot band alignment. It leads to ink-based materials that retain the independent surface functionalization of quantum dots, and it creates distinguishable donor and acceptor domains for bulk heterojunctions. Interdot carrier transfer and exciton dissociation studies confirm efficient charge separation at the nanoscale interfaces between the two classes of quantum dots. We fabricate the first mixed-quantum-dot solar cells and achieve a power conversion of 10.4%, which surpasses the performance of previously reported bulk heterojunction quantum dot devices fully two-fold, indicating the potential of the mixed-quantum-dot approach.

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Enhancing Perovskite Nanocrystal Stability and Luminescence by Self-Assembly and Molecular Passivation for LED Applications

Metal halide perovskite, APbX$_3$ (A is a cation and X is a halide), nanocrystals are an emerging class of nanomaterials with high photoluminescence quantum yield and readily tunable emission wavelengths. However, they suffer severely from chemical and phase instabilities, and are commonly capped by ligands that insulate the nanocrystals, thus precluding their utility in optoelectronic devices. We present two strategies to stabilize these nanocrystals in charge transport-compatible configurations. In the first strategy, we developed a bottom-up anisotropic self-assembly approach for constructing large free-standing uniform sheets of CsPbBr$_3$ perovskite (μm$^2$ in area; >100 nm in thickness) from CsPbBr$_3$ nanocrystals (<10 nm). As ionic materials, perovskites have fast growth kinetics and labile building-blocks, which makes it challenging to implement such assembly strategies. Through atomic-scale investigation of the assembly intermediates we unravel the self-assembly mechanism and, moreover, reveal a “staggered” adhesion mode between aligned building-blocks that is free of long carbon-chain ligands. Uniquely, the large (much larger than quantum-confinement scales) single-crystal sheets with domains isolated by atomic-staggers retain the desirable luminescence properties of the original nanocrystals. Solid-state films fabricated from the assembled CsPbBr$_3$ sheets, in contrast to the ligand-protect CsPbBr$_3$ nanocrystal building-blocks, are photoluminescently stable (in both intensity and wavelength) and have higher quantum yield. In a second strategy, we developed a post-synthesis passivation process for CsPbX$_3$ nanocrystals by using short bidentate ligands. The passivated nanocrystals exhibit narrow photoluminescence with exceptional quantum yield (in some cases close to unity) and substantially improved stability. The passivation strategy enabled us to realize nanocrystal-based LEDs with much higher external quantum efficiency and luminance than LEDs made from the non-passivated nanocrystals. Our work paves the way to further the exploitation of perovskite nanocrystal materials in optoelectronics through self-assembly and surface-passivation.

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Low-Threshold Up-Converted Lasing in Perovskite Quantum Dots

Perovskite quantum dots (PQDs) have emerged as a promising candidate for application in lighting technologies, including LEDs and Lasers. Here we investigate the nonlinear optical response of these materials and how those properties can be applied to up-converted lasing. We show, for the first time, the size dependence of two-photon excited optical gain in CsPbBr$_3$ PQDs, demonstrating that this is directly linked to the large biexciton binding energy present in this material. By performing two-photon absorption (2PA) spectroscopy in a series of CsPbBr$_3$ PQDs with size varying from 7.4 to 12.5 nm, we have verified that PQDs possess large 2PA cross section, which can reach over $10^6$ GM (1 GM = $10^{-50}$ cm$^4$s), and increases linearly with the nanoparticle volume. Taking advantage of the large 2PA cross section, we demonstrate extremely low up-converted lasing threshold, which is as low as 1 mJ/cm$^2$, under two-photon excitation at 1.55 eV. Considering the 2PA cross section, we calculate the lasing threshold in terms of the average number of excitons per PQD. We have found that this threshold goes from about 0.8 for the smallest PQDs to about 1.4 for the largest one, indicating a clear size dependence. In order to understand the physical mechanism responsible for this size dependence, we investigated the biexciton binding energy and observed that it goes from 60 meV to 40 meV as the size of the PQDs increases, indicating that the large red-shift caused by the biexciton binding energy drastically reduces the re-absorption losses favoring amplified spontaneous emission at low population regime.

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Colloidal Quantum Dot Heterostructures with Near-Unity Quantum Yield and Suppressed Blinking

We demonstrate novel heterostructured quantum dots (QDs) with CdS/CdSe/CdS seed/spherical quantum well/shell (SQW) geometry that show near-unity photoluminescence quantum yield (PL QY) at room temperature and suppressed blinking simultaneously, which have never been exhibited with conventional core/shell heterostructures. The comparative studies based on spectroscopic analysis and XRD patterns with the conventional CdSe/CdS core/shell QDs reveals that the misfit defects are origins of the low PL QY in conventional thick-shell QDs. In addition, the reduced lattice mismatch between CdSe quantum well and CdS seeds suppresses the formation of misfit defects as a result of the coherently strained heterostructure and the thick CdS shell can eliminate the effect of surface defect on the electron and hole wavefunction in CdSe emissive layer¹. Consequently, SQW heterostructure with a thick CdS shell (≥ 5 nm) allows the near-unity PL QY and substantially suppressed blinking.

Comprehensive studies with various geometries of SQW QDs suggest the effect of SQW heterostructure on the optical properties of QDs. The outstanding optical properties of SQW QDs are maintained even in the cases of concentrated solutions and films under thermal stress. To the best of my knowledge, this study is the first propose of the coherently strained heterostructure in colloidal QDs. This study will boost the role of thick-shell heterostructured QDs for applications.


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Colloidal quantum dot lasers

Colloidal quantum dots are appealing gain mediums for compact, substrate independent and wavelength tunable lasers. They can be processed in solution and their emission wavelength can cover a broad range including ultraviolet, visible and near-infrared. However, due to the loosen restriction in momentum conservation, Auger recombination is accelerated, continuous wave lasing is therefore very challenging. In this talk, I will introduce our efforts towards the continuous wave lasing and perspective about future directions of the colloidal quantum dot lasers.

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The Future of Quantum Dots in Displays

Quantum dots have seen tremendous commercial success recently due to the advent of displays utilizing QD color-converting technology to improve the viewer experience and increase efficiency. We are only exposing the tip of the iceberg, however, as there will be numerous advancements in the coming years. Polymer films containing QDs have become the dominate form factor in QD-enabled displays, but other technologies such as QD color filter replacements, QDs on LEDs, and electroluminescent QD devices are making tremendous progress. Current state-of-the-art technology will be reviewed, but the focus of the talk will be future implementation strategies and technical hurdles where further research is required. The demanding requirements of the display industry are driving QD development, and those who can solve the technical challenges will see tremendous reward.

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Superfluorescence from Superlattices of Lead Halide Perovskite Nanocrystals

Superfluorescence is the cooperative emission of individual emitters that arises from coherent collective coupling to a common light field, leading to strongly enhanced radiative rates. First reported in atomic gases, it has been studied in several solid state quantum materials. However, for the important material class of colloidal nanocrystals, which have applications ranging from displays to photovoltaics, the generation of superfluorescent light was precluded by large inhomogeneous emission broadening, low oscillator strength, and fast exciton dephasing. Recently, CsPbX$_3$ (X = Cl, Br, I) perovskite nanocrystals$^1$ have emerged as a new class of quantum emitters with narrow-band, blinking-free emission and unprecedented high oscillator strength$^{2,3}$. Here we show superfluorescent emission from perovskite quantum dots assembled in highly ordered three-dimensional superlattices. Superfluorescence is characterized by a red-shifted emission with more than ten-fold reduction of the radiative lifetime, extension of first-order coherence time by more than a factor of four, photon bunching and Burnham-Chiao ringing behavior at high excitation density. Our results demonstrate a straightforward solid state approach for the generation of superfluorescent Dicke states in self-assembled superlattices with the potential for on-chip optical and electrical control. Furthermore, these mesoscopically extended coherent states might enable the generation of entangled N-photon bundles for quantum information processing and quantum sensing applications.

References:

2) Rainò et al., ACS Nano, 10, 2485–2490 (2016)

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Design of cadmium-free and II-VI semiconductor quantum dot phosphors exhibiting green and red emissions

CdSe-based QDs are currently used as QD-phosphors in ultra-high definition television compliant displays. In order to avoid using toxic cadmium, alternative materials that do not contain toxic elements are strongly desired. Although InP QDs are particularly promising as alternative, their emission band width is still larger than that of CdSe-based QDs. In 2006, our group reported visible emission of CuInS$_2$-based QDs; however, they exhibited significantly broadened emission and cannot achieve wide gamut of colors. Thus, suitable alternative materials to CdSe have not yet been established. In the present study, we focused on the ZnTe-based alloy QDs as green and red QD-phosphors, because ZnTe alloyed with ZnSe and ZnS exhibits a considerable negative deviation of the energy band gap, termed as large band gap bowing, although ZnTe having 2.25 eV of energy band gap is not suitable as a green and red phosphor. Based on the size and composition dependence of the optical gap of colloidal alloyed QDs of Zn(Te,Se) and Zn(Te,S) calculated using the finite-depth well effective mass approximation method, it was demonstrated that Zn(Te$_{1-x}$Se$_x$) QDs with 0.02≤x≤0.68 are expected to exhibit green emission, and Zn(Te$_{1-x}$S$_x$) QDs with 0.26≤x≤0.37 should give red emission. The results indicates that Zn(Se,Te) and Zn(Te,S) QDs are very promising cadmium-free II-VI semiconductor green and red QD-phosphors, respectively.

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Stabilized self-assembled J-aggregate antennas to enhance NIR QDs emission

We report that the photoluminescent emission of near infrared emitting lead sulfide quantum dots (PbS QDs) can be enhanced more than 8-folds either in liquid or in solid media by self-assembled molecular dye J-aggregates (Light Harvesting Nanotubes or LHNs) thanks to a long-range antenna effect and an efficient Förster resonance energy transfer (FRET). As donor, we applied an amphiphilic cyanine dye (C8S3) which is soluble in methanol and aggregates in water into cylindrical double walled nanotubes (LHNs), showing narrow spectral features in the visible part of the solar spectrum with high oscillator strengths and fast radiative rates. To use QDs acceptors compatible with the aqueous environment of the LHNs and to preserve their high optical properties, we synthesized water-soluble PbS QDs through a new method. We performed a ligand substitution directly during the QDs synthesis, followed by a PEG-functionalization reaction using click-chemistry. Thanks to this method, we were able to maintain for months high optical and morphological properties of the water soluble QDs. In our system, the low concentrated QDs acceptors are weakly coupling with the LHNs. Our experimental measurements and theoretical studies show that not only the quantum yield of the donor but also the long range diffusive exciton (antenna effect) can promote the energy transfer to the acceptor in non-linear way. This opens opportunities for creating host–guest systems with large Stokes’ shift from the visible to the NIR-IR, effectively separating the absorption and the emission.


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Polarons and Polaritons in Lead Halide Perovskites

Lead halide perovskites have been demonstrated as high performance materials in solar cells and light-emitting devices. These materials are characterized by coherent band transport expected from crystalline semiconductors, but dielectric responses and phonon dynamics typical of liquids. Here I will show that the “crystal-liquid” duality and the soft phonon modes are responsible for large polaron formation and screening of charge carriers, leading to defect tolerance, moderate charge carrier mobility, and low radiative recombination rates (Sci. Adv. 2017, 3, e1701217; Sci. Adv. 2017, 3, e1701469; Nat. Mat. 2018, 17). Large polaron formation, along with the phonon-glass character, explains the dramatic reduction in hot carrier cooling rates (Science, 2016, 353, 1409; JACS 2016, 138, 15717). Besides excellent carrier properties, lead halide perovskite have been demonstrated in high efficiency light emission, including nanowire lasing (Nat. Mat. 2015, 14, 636). I will show that both pulsed and CW lasing in lead halide perovskite nanowires originate from polaritons in the bottleneck region (Adv. Opt. Mat. 2018, 6, 1700982). Time resolved measurement provides a direct view of the kinetic condensation process and reveals intriguing Bose-scattering dynamics in low dimensions.

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Bright Triplet Excitons in Cesium Lead Halide Perovskites

According to Hund’s rules, the lowest energy exciton in organic materials is a poorly emitting triplet state. For inorganic semiconductors, similar rules predict an analog of this triplet known as the dark exciton. Because this state releases photons slowly, materials that disobey these rules have been sought. However, despite considerable experimental and theoretical efforts, no inorganic semiconductors have been identified in which the lowest exciton is bright. Here, we will discuss the situation in cesium lead halide perovskites \([\text{CsPbX}_3 \ (X = \text{Cl, Br, and I})]\). Theory shows that this material can exhibit a highly emissive triplet state when the strong spin–orbit coupling in the perovskite conduction band is combined with the Rashba effect.\(^1\) We then apply such a model to \(\text{CsPbX}_3\) nanocrystals,\(^2\) for which we measure size- and composition-dependent fluorescence at the single-nanocrystal level. The bright-triplet character of the lowest exciton immediately explains the anomalous photon-emission rates of these materials, which emit \(\sim 20\) and \(\sim 1,000\) times faster than any other semiconductor nanocrystal at room and cryogenic temperatures, respectively.\(^3\) The bright-triplet exciton is further confirmed by detailed analysis of the fine structure in low-temperature fluorescence spectra. Our results provide criteria for identifying other semiconductors that exhibit bright excitons, with potential implications for optoelectronic devices. Becker, M. A.; Vaxenburg, R.; Nedelcu, G.; Sercel, P. C.; Shabaev, A.; Mehl, M. J.; Michopoulos, J. G.; Lambrakos, S. G.; Bernstein, N.; Lyons, J. L.; Stöferle, T.; Mahrt, R. F.; Kovalenko, M. V.; Norris, D. J.; Rainò, G.; Efros, Al. L. Nature 553, 189 (2018). Protesescu, L.; Yakunin, S.; Bodnarchuk, M. I.; Krieg, F.; Caputo, R.; Hendon, C. H.; Yang, R. X.; Walsh, A.; Kovalenko, M. V. Nano Lett. 15, 3692 (2015). Rainò, G.; Nedelcu, G.; Protesescu, L.; Bodnarchuk, M. I.; Kovalenko, M. V.; Mahrt, R. F.; Stöferle, T. ACS Nano 10, 2485 (2016).

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Long Exciton Dephasing and Bright Emission from Lead Halide Perovskite Nanocrystals

The emission of fully inorganic cesium lead halide perovskite-type nanocrystals is tunable over a wide energy range with photoluminescence quantum yields of up to 90%[1]. Due to their facile solution processability and their potential for high-efficiency photovoltaics and light sources they have gained enormous interest. Experiments on single quantum dots reveal a bright triplet state[2] with photon emission rates ~20 and ~1000 times higher compared to any other conventional semiconductor nanocrystals at room and cryogenic temperatures, respectively. We discuss the radiative nature of this exceptionally fast photon emission by temperature dependent quantum yield measurements. Furthermore we discriminate it from composition dependent “A-type” blinking behaviour in intensity-decay time correlation measurements and demonstrate stable, narrowband emission, with suppressed blinking and small spectral diffusion[3] for single CsPbBr\textsubscript{2}Cl nanocrystals. In order to access the coherence properties of the excitons, we perform degenerate four-wave mixing on CsPbBr\textsubscript{2}Cl nanocrystal films. We obtain a dephasing time of about 27 ps at low temperature and additionally observe signatures of a coherent exciton-phonon coupling. Our experimental findings of extraordinary high oscillator strength together with an exceptionally long dephasing time suggest that lead halide perovskite nanocrystals are a prime candidate for the observation of coherent excitonic phenomena and strong light-matter coupling.

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Excitonic Pathway to Photoinduced Magnetism in Colloidal Nanocrystals with Nonmagnetic Electronic Dopants

‘Electronic doping’ of colloidal semiconductor nanostructures through controlled incorporation of metal impurities, aliovalent with respect to the cation sub-lattice, holds promise for future design of functional nanomaterials and advanced device concepts in optoelectronics, photonics and spin-based technologies. An emerging electronic dopant in III-V and II-VI nanostructures is silver, whose incorporation as Ag⁺ impurity leads to the introduction of intragap electronic states and the development of a largely Stokes-shifted emission resulting from an optical transition coupling the host conduction band to the dopant 4d states. Having a full d electronic shell, Ag⁺ is nominally nonmagnetic and luminescence is often ascribed to transfer of the photoexcited hole from the host valence band to the Ag⁺ site, which then behaves as a radiative acceptor centre for a conduction band electron. The mechanism of this optical-activation process and, specifically, the associated modification of the electronic configuration of the Ag ion remain unclear. In this talk, I will trace a comprehensive picture of the dopant-mediated excitonic process in Ag-doped CdSe nanocrystals by combining site-selective photoluminescence and transient absorption experiments with spectro-electrochemical methods and magnetic circular dichroism spectroscopy. Our results demonstrate that, in contrast to expectations based on silver ionization energy and Jan Teller crystal effects, capture of the photohole by Ag⁺ effectively leads to its transient conversion to paramagnetic Ag²⁺. Thus in Ag doped NCs the process of exciton recombination and photoluminescence is inextricably tied to photoinduced magnetism. Accordingly, we observe strong optically-activated magnetism and diluted magnetic semiconductor behaviour, demonstrating that optically switchable magnetic nanomaterials can be obtained by exploiting the excitonic processes involving nominally nonmagnetic impurities. These discovery of off-equilibrium magnetism in individual electronic-doped NCs could lay the foundation for future (opto)spintronic applications operated by light.


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N- and P-Doping of Colloidal Nanocrystal and Nanowire Assemblies

Doping is used to control free carrier type and concentration in semiconductor materials. In bulk semiconductors, doping is conventionally achieved through impurity substitution, and less well-known, by controlling a material’s stoichiometry. However, doping in low-dimensional, colloidal semiconductors presents both new opportunities and challenges. The electron and hole concentrations in colloidal nanocrystal (NC) and nanowire (NW) assemblies have often been manipulated by “remote doping,” exploiting the large surface-to-volume ratio of nanostructures to add atoms, ions, or ligands to the surface that serve as dopants. For example, we describe methods of 1) thermal evaporation and diffusion and 2) wet-chemical techniques to introduce extrinsic impurities to and non-stoichiometry through the nanostructure surface to passivate surface traps and dope NC and NW assemblies. However, the doping efficiency of colloidal nanostructures is often extremely low, such that only 1% of dopants yield carriers. These low doping efficiencies are consistent with increased ionization energies from quantum and dielectric confinement effects. We show for a given size nanostructure, where quantum confinement effects are fixed, the doping efficiency can be enhanced by >10-fold by encapsulating the NCs and NWs in high dielectric constant materials that reduce the dielectric mismatch between the nanostructure and its surroundings. We give examples where n- and p-type semiconductor NC and NW assemblies are used to construct flexible, electronic transistors and integrated circuits and optoelectronic solar photovoltaics and photodetectors.

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Direct observation of harmonic dipole oscillations in PbS/CdS core/shell nanocrystals with phase-locked interference detection

Semiconductor nanocrystals (NCs) are fascinating systems for fundamental science and optoelectronic devices. Especially, multiple excitons in NCs are being intensively investigated because of their unique quantum properties such as quantized Auger recombination and efficient carrier multiplication [1]. However, the generation process of multiple excitons still remains unclear, despite intensive investigations on the recombination processes of multiple excitons. As predicted in theoretical investigations of the coherent superposition of single and multiple excitons [2], a detailed understanding of the multiple exciton coherence would help to clarify the generation process. In this study, we report on the direct observation of coherent dipole oscillations of multiple excitons in PbS/CdS core/shell NCs. We developed a new high-sensitive spectroscopic technique that employs phase-locked interference detection of transient absorption signals in order to measure the multiple exciton coherences in the ultrafast time regime. Our results suggest that the transient absorption signals show the existence of dipole oscillations with higher harmonic orders even though the photon energy of the excitation pulse is set to the exciton resonance frequency [3]. The excitation fluence dependence of the oscillation amplitudes exhibited a clear correlation with the generation probabilities of multiple excitons. Moreover, we clarified that the decay profiles of the oscillations can be expressed as a function of the number of excitons. These results prove that the dipole oscillations are generated by the harmonic quantum coherences of multiple excitons. The coherent dynamics of multiple excitons provide fundamental insights into the generation mechanisms of multiple excitons.

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A Quantitative Study of Optical Gain Mechanisms in Quasi-2D Nanoscale Materials

Solution processable nanomaterials for photonic applications, in particular light emission and lasing, have received much attention in the past decade(s). A demonstration of ultralow continuous wave optical gain and lasing using CdSe nanoplatelets, quasi-2D materials in a colloidal dispersion, was the most recent milestone in this field. In last few years, colloidal pervoskite nanoplatelets have also emerged as promising nanomaterial having strongly confined exitonic states. Until now, the optical gain in these quasi-2D systems was thought to originate from the biexciton to exciton transition, much as is the case for 0D colloidal QDs. The net gain should therefore occur at biexciton carrier densities and moreover only develop in a narrow region below the lowest energy exciton feature. In this work, we present a comprehensive ultrafast spectroscopy study of the optical gain mechanisms in CdSe and CsPbBr$_3$ nanoplatelets. In particular for CdSe nanoplatelets, a unique combination of quantitative pump-probe, fluorescence and THz spectroscopy is used to reveal particular photo-physics. Large and broadband optical gain is observed in these nanoplatelet which is caused by the occurrence of a carrier cooling bottleneck at high density leading to a hot electron-hole plasma. Furthermore, the excitons are found to co-exist with the plasma and even counter-act the development of net optical gain in certain spectral regions. Our results shed a new light on the role of excitons in highly excited two-dimensional materials and pave the way for the development of more efficient broadband optical gain media and excitonic devices.

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Tale of two excitons: the effect of spectator excitons on nanocrystal photophysics.

Interpretation of transient transmission spectra of hot excitons in nanocrystals hinges on bi-exciton shifting while carriers occupy excited states in the valence and conduction bands, with a transition to state filling once they relax to the discreet band-edge states. In this talk we will describe a novel approach for testing this assignment based on a comparison of pump-probe data starting with a pristine QD sample, to data collected from a sample saturated with cold single excitons. The opportunity for such a comparison rests on the separation of timescales between Auger recombination of multi-excitons, and radiative recombination of singles. Exciting with a fluence which produces significantly above 1, and waiting until Auger recombination is complete, a saturation with cold single excitons is obtained. Results from PbSe, PbS, and CdSe samples demonstrate 1) that band edge bleaching is not linear in the number of absorbed excitons. In the case of Lead salt QDs the band edge bleach introduced is significantly smaller than for the first, while in CdSe it is only one half as strong. Accordingly full bleaching of the band edge absorption band requires more than the expected minimum number of excitons. 2) That the below band edge induced absorption is not the positive half of a band shift as expected. These findings call for a significant revision of our understanding of transient spectral data from excited states of QDs. This includes data which has been pivotal in understanding carrier cooling dynamics as well as assigning degeneracies of low lying exciton states. It therefore is directly relevant to our current understanding of exciton dynamics in nanocrystals and should impact every aspect of the theme of this track.

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Highly Efficient Inverted Structural Quantum Dot Solar Cells

Highly efficient PbS colloidal quantum dot (QD) solar cells based on an inverted structure have been missing for a long time. The bottlenecks are the construction of an effective p–n heterojunction at the illumination side with smooth band alignment and the absence of serious interface carrier recombination. Here, solution-processed nickel oxide (NiO) as the p-type layer and lead sulfide (PbS) QDs with iodide ligand as the n-type layer are explored to build a p–n heterojunction at the illumination side. The large depletion region in the QD layer at the illumination side leads to high photocurrent. Interface carrier recombination at the interface is effectively prohibited by inserting a layer of slightly doped p-type QDs with 1,2-ethanedithiol as ligands, leading to improved voltage of the device. Based on this graded device structure design, the efficiency of inverted structural heterojunction PbS QD solar cells is improved to 9.7%, one time higher than the highest efficiency achieved before.

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**Strain-controlled InGaN quantum dots-based monolithic white light-emitting diodes**

Recently, InGaN-based blue LEDs were commonly used by adding yttrium aluminum garnet phosphor which is converted from blue light to longer wavelengths such as green, yellow and red emissions. However, the drawback of dichromatic white LED needs additional deposition of the light converter. Therefore, many research groups have studied to develop the phosphor-free white LEDs using multi-colored quantum wells (QWs), which have exhibited a low internal quantum efficiency and a significant quantum confinement Stark effect. In this study, we focused on the In phase separated-InGaN quantum dots structure in InGaN-based LEDs using solid phase immiscibility in InGaN-GaN material system. In\textsubscript{0.14}Ga\textsubscript{0.86}N/GaN five QWs were grown on n-GaN for blue emission. For longer wavelength emissions, additional In\textsubscript{0.2}Ga\textsubscript{0.8}N/GaN SQW was grown on blue InGaN/GaN five QW structure. To achieve the In-phase-separated InGaN SQW structure for amber/green emissions, the thickness of green In\textsubscript{0.2}Ga\textsubscript{0.8}N SQW was systematically increased from 2.0 to 3.5 nm using the variation of the growth time. By increasing the well thickness to 2.7 nm for InGaN/GaN SQW, the blue emission from n-side InGaN five QWs was remained at 440 nm, while the green emission peak was redshifted from 505 to 560 nm. Surprisingly, we found the discrete three peaks with triple wavelengths of 440, 500, and 610 nm for 3.5-nm-thick In\textsubscript{0.2}Ga\textsubscript{0.8}N SQW. These three wavelengths would represent white light emissions. From high-resolution TEM measurements, we found that 3.5-nm-thick InGaN SQW has the inhomogeneous well structure which was consisted of dark spot and thin bright regions. The bright region showed InGaN phases with low In content, whereas the dark spot regions represented InGaN phases with high In content. One can see that the amber and green peak should be emitted from phase-separated InGaN quantum dots with bright and dark spot phases formed in the 3.5-nm-thick InGaN SQW.

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SAW Modulation of Nanowire-Embedded Quantum Dots

Quantum dots are an excellent source of single photons because of their atom-like emission. They have the potential to emit triggered single photons, and they can also be embedded into solid state systems that are capable of channeling the single photon emission. Once produced, single photons can be reliably manipulated and travel long distances making it attractive in the field of quantum information.

In this study we investigate the effect of surface acoustic waves (SAWs) on the optical emissions of InAsP quantum dots in an InP nanowire fabricated at NRC. By generating SAWs on the device, a strain field dynamically modulates the quantum dot properties and causes an oscillation in the time integrated transition energy of the quantum dots. In addition, a piezoelectric field can influence the generated electrons and holes. Combining the acoustically modulated quantum dot emission lines with a spectral filtering of detected photon can be used to produce a high frequency single photon source.

Photoluminescence measurements were performed on nanowire-embedded quantum dots cooled to cryogenic temperatures and optically pumped by a 532 nm laser source. Incorporating a layer of ZnO, which is deposited by sputtering, on the InP substrate in order to generate SAWs is also discussed.

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Interfacing single quantum dot spin with photon using a topological photonic crystal.

Topological phonics has given a new direction to designing photonic devices along with opening plethora of applications [1]. In recent years there has been many interesting studies in topological photonics in classical domain [2] but substantial attention is needed towards realizing light matter interaction with topological photonic states in quantum domain. Towards this goal, we developed a topological photonic crystal structure for interfacing single quantum dot spin with photon [3]. We fabricated a distinct photonic crystal on a thin piece of Gallium Arsenide which are embedded with Indium Arsenide quantum dots as in-plane single photon source. This device can support two counter propagating edge states at the interface of two types of photonic crystals with different band topologies. We showed chiral coupling of circularly polarized single photons from the quantum dots under strong magnetic field to the two counter propagating waveguide modes. Owing to the topological nature of these guided modes, we demonstrated these edge states to be robust against sharp corners. This new technique can pave path towards exploring exotic physics like many-body interaction [4,5], unconventional quantum states of light and chiral spin networks [6].


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High-efficiency multiphoton boson sampling

Boson sampling is considered as a strong candidate to demonstrate ‘quantum computational supremacy’ over classical computers. However, previous proof-of-principle experiments suffered from small photon number and low sampling rates owing to the inefficiencies of the single-photon sources and multiport optical interferometers. Here, we develop two central components for high-performance boson sampling: robust multiphoton interferometers with 99% transmission rate and actively demultiplexed single-photon sources based on a quantum dot–micropillar with simultaneously high efficiency, purity and indistinguishability. We implement and validate three-, four- and five-photon boson sampling, and achieve sampling rates of 4.96 kHz, 151 Hz and 4 Hz, respectively, which are over 24,000 times faster than previous experiments. Our architecture can be scaled up for a larger number of photons and with higher sampling rates to compete with classical computers, and might provide experimental evidence against the extended Church–Turing thesis. Another problem for large scale experimental boson sampling is the scalability. We report an experiment on boson sampling with photon loss, and demonstrate that boson sampling with a few photons lost can increase the sampling rate. Our experiment uses a quantum-dot-micropillar single-photon source demultiplexed into up to seven input ports of a 16×16 mode ultra-low-loss photonic circuit, and we detect three-, four- and five-fold coincidence counts. We implement and validate lossy boson sampling with one and two photons lost, and obtain sampling rates of 187 kHz, 13.6 kHz, and 0.78 kHz for five-, six- and seven-photon boson sampling with two photons lost, which is 9.4, 13.9, and 18.0 times faster than the standard boson sampling, respectively. Our experiment shows an approach to significantly enhance the sampling rate of multiphoton boson sampling.

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Integration of PbS/CdS QDs with superconducting nanowire detectors on an amorphous silicon photonic platform

There is potential for a fast-emitting and reliable single-photon source with emission in a telecom band to replace attenuated lasers and decoy state protocols for the application of quantum cryptography. Colloidal PbS/CdS quantum dots (QDs) can be synthesized to emit in the telecom O-band, but are a challenging material to work with because of temperature sensitivity during processing, long exciton lifetimes in the range of a microsecond and a broad emission spectrum. Using a room-temperature lift-off process, we integrated PbS/CdS QDs in the gap of plasmonic antennas on top of amorphous silicon (a-Si) waveguides. The emission was then guided to Superconducting Nanowire Single-Photon Detectors (SNSPDs) located on the same photonic chip, which was cooled to a temperature of 4K in a helium cryostat. Evanescent coupling of the waveguide mode to the SNSPDs located below the a-Si waveguides enabled an efficient detection of the QD emission. Our measurements show that we are able to increase the radiative rate, while not exceeding the thermal budget of the QDs. At the same time, coupling of the QD emission to the a-Si waveguide mode was high - because of the high refractive index contrast - and we were able to achieve a reasonably low waveguide loss of 6-7 dB/cm at a wavelength of 1300 nm. While it should be possible to scale our method of patterning the QDs down to single emitters, the long intrinsic radiative lifetime and broad emission spectrum of PbS/CdS QDs remain a hindrance for any application in Quantum Information Technologies. However, with our approach we are also able to incorporate faster emitters e.g. recently developed InAs-based QDs in the future, making it a generic platform that can be used with different kinds of quantum emitters in the IR.

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Towards quantum control of quantum dots

There are numerous potential applications for quantum dots. Among them, the use of electronic states for quantum computing is of particular interest to the future of computer technology. In this case, the intervention of an external electromagnetic field is required. Our goal is then to understand and manipulate the electronic dynamics of silicon QDs through the use of quantum control, i.e. the use of complex electromagnetic pulses to control phenomena at the pico- or even femtosecond time scale. This will for example allow us to populate a dark electronic state like we did for dark vibrational levels in acetylene in one of our previous works. The first step involves getting knowledge of the structural variables of interest for our system: electronic levels, transition dipole moments, coupling matrix elements, etc. In order to do so, we use DFT and TD-DFT methods. This way, we were able to compute the electronic gap of different sizes and shapes of silicon quantum dots. The second step will make good use of those values to model our system into our control algorithms. Those algorithms implement the Optimal Control Theory by an iterative process in order to control the populations of the electronic levels of our quantum dots.

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Hybrid integration of single photon source from quantum dot in silicon-based photonic circuits

A single photon source is an essential component for the photonic quantum information applications including the quantum communication, the quantum simulation, and the quantum computation [1, 2]. Solid state quantum dots (QDs) have a lot of potential as a single photon source platform due to their various advantages of favorable manipulation and integration with various photonic elements. In particular, the GaAs- and InP-based QDs were appropriate for photonic platforms in telecommunication wavelength. There have been numerous effort to integrate the GaAs- and InP-based QDs with silicon-based photonic circuits, which opening the possibility for various applications using on-chip quantum photonic device [3-5]. However, it was still challenge to fabricate the hybrid integrated device of single QD due to difficult hybrid integration and irregular size and distribution of self-assembled QDs. In this study, we realized hybrid integration of deterministic single photon source from InP-based QDs and the silicon-based waveguide through the pick-and-place method and the selective-area growth technique. We designed the waveguide-integrated QD structures as InGaAs QDs on SiNx waveguides. The QDs embedded in InGaAsP layers were fabricated with one-dimensional cavity by electron beam lithography. The waveguides were fabricated by conventional photo lithography using the SiNx/SiO2/Si layers. The QD-embedded one-dimensional cavity was transferred to the silicon-based waveguides through the pick-and-place technique. The physical dimensions of QD-cavity structures and SiNx waveguides were determined by the finite-difference time-domain simulation. The hybrid integrated device was designed that a single dipole source of the QD was efficiently coupled to the waveguide by adopting a SiOx interlayer between the QD and the waveguide. For optical characterization, the waveguide-integrated QD was excited vertically in the micro-photoluminescence experiment and measured laterally, which confirming that the hybrid integrated device was successfully fabricated. Finally, the waveguide-integrated QD structure could provide a solution for on-chip single photon source in photonic integrated circuits.


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QD2018 – Poster Session 1 Abstracts

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Atomic vapor based memories for quantum dot single photons – A variable EIT delay in Cesium for hybrid quantum networks

Quantum memories will play a central part in synchronizing operational events in quantum networks, e.g. joint measurements between photons from different sources in order to implement entanglement swapping in quantum repeater protocols [1]. A promising platform relies on electromagnetically induced transparency (EIT) in atomic vapor. There, transitions in Rubidium or Cesium between 800 nm – 900 nm provide a convenient wavelength to link the memories to other photon emitters like molecules or quantum dots [2]. In this way the most promising system for pure and fast few-photon sources, i.e., semiconductor quantum dots, can be linked to efficient storage devices in a hybrid approach. In our contribution we introduce our recent experimental results in this direction. First, we demonstrate precise strain-tuning of InGaAs quantum dot emission between two 1.2 GHz hyperfine-split D1 transitions in Cesium vapor at room temperature. Then we show how single photons from the quantum dots can interact with a Cesium cell [3] providing a novel approach to access strong delays even close to room temperature. These recent experiments lay the ground for atomic vapor-based quantum memories in hybrid quantum networks [2] with storage times up to hundreds of microseconds and efficiencies of about 35 %. In a second part of the contribution we also introduce our recent results concerning the conversion of single photons at Cesium wavelengths from quantum dots to the telecom band at 1550 nm [4]. Both experiments lay the ground for hybrid quantum networks involving semiconductor quantum dots, atomic memories, and long-distance fiber transfer. [1] H. J. Kimble, Nature 453, 1023 (2008)[2] J. Wolters et al., Phys. Rev. Lett. 119, 060502 (2017)[3] D. Höckel et al., Phys. Rev. Lett. 105, 153605 (2010)[4] T. Kroh et al., Quantum Sci. Technol. 2, 034007 (2017)

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Photoluminescence imaging of self-assembled quantum dots for deterministic photonic structure fabrication

Bright sources of indistinguishable single photons are key components for advanced quantum optics applications such as quantum communication and linear optical quantum computing. Among different kinds of quantum emitters epitaxially grown self-assembled quantum dots (QDs) have been shown to be one of the prime candidates for efficient single photon generation. In order to maximize number of photons extracted from the device, QDs are frequently embedded into photonic structures such as miropillar or photonic crystal cavities, nanowires, waveguides, ring resonators, gratings and micro-lenses. In order to fully utilize such architectures positions as well as spectral characteristics of the emitters have to be known prior the photonic structure fabrication. In this contribution we demonstrate results of low-temperature microphotoluminescence of InAs/GaAs QDs utilizing wide field illumination for large area imaging. Investigated QD sample with planar cavity structure have been patterned with metal grid by utilizing e-beam lithography and lift-off process. Based on performed experiments positions of QDs in respect to the alignment marks have been determined with around 20 nm precision. For identified QDs spectral characteristics of the emission have been also recorded. Performed measurements allowed to gather information necessary for design of photonic structures matching characteristics of identified QD emitters and make it is feasible to fabricate photonic structures on the top of the bright emitters by utilizing the high-yield e-beam lithography. Based on this method a single-photon device with an extraction efficiency of $\eta = (49 \pm 4)\%$ and the emission of photons with $(94 \pm 2.7)\%$ indistinguishability was fabricated [1]. We believe that these investigations are an important step towards the high-yield fabrication of scalable single photon devices based on the self-assembled QDs.


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Towards Microwave-to-Optical Quantum State Transduction on a Hybrid Quantum Dot/Superconducting Qubit Platform

Hybrid quantum systems, in which two or more quantum technologies are integrated onto the same platform, offer the promise of exploiting the best properties of each individual system [1]. A pressing need for superconducting Josephson junction (JJ) qubits, which are current state-of-the-art in quantum information processing, is a complementary quantum system that can serve as both a memory unit and a microwave-to-optical quantum state transducer. To this end, we will present our efforts towards integrating the long spin coherence and storage times and strong optical coupling of a self-assembled InAs/GaAs quantum dot (QD) with the robust and fast operations of JJ qubits. We propose to couple these two disparate quantum systems through their mutual strong interaction with surface acoustic waves (SAWs). SAWs as a coupling medium offer advantages compared to other approaches, such as their ability to propagate coherently over millimeter distances and at gigahertz frequencies. The former can allow sensitive superconducting electronics to be isolated from the optical excitation, while the latter ensures operation in the resolved-sideband regime and zero occupation of thermal phonons at dilution refrigerator temperatures. Indeed, progress is being made towards achieving strong coupling between a JJ qubit and SAWs on GaAs [2]. The key attribute that makes this platform compelling is the nonlinear response of the QD in both the optical and microwave domains. The coupling strength between the optical excitations (excitons) in a QD to optical photons is larger than any other solid-state system, providing low-loss and high-speed access to optical qubits that can be transported long distances. Furthermore, our calculations reveal that strong QD-SAW cavity coupling is possible. We will present a promising path towards achieving strong coupling between a QD and a JJ qubit and thus transfer of quantum information between the microwave and optical domains with high fidelity.


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Quantum diffusive analysis of two electron spins in double quantum dots

Quantum dot is one of the most dominant candidates to realize multi qubits system. Coherent control and preserve of the quantum information of the electrons are the central issue[1]. It is known that the time evolution of the electrons in the quantum dot monitored by Quantum Point Contact (QPC), which measures the charge state of the electrons, can be discussed with the method called Quantum Diffusive Analysis (QDA)[2].

In our research, we consider two electrons in the double quantum dots with zero total $S_z$ component, and focus on the singlet (1,1) state $|S_{11}\rangle$ and the triplet (1,1) state $|T_{11}\rangle$, as a qubit[3]. The states $|S_{11}\rangle$ and $|T_{11}\rangle$ are hybridized because of the fluctuations of the magnetic field. Then, we study how to stabilize $|S_{11}\rangle$ or $|T_{11}\rangle$. Since the states $|S_{11}\rangle$ and $|T_{11}\rangle$ have the same charge distribution, we cannot distinguish the two states by near-by QPC. If there is an energy detuning however, $|S_{11}\rangle$ is hybridized with $|S_{02}\rangle$, which realizes the different charge distributions. This charge difference enables us to distinguish singlet state and triplet state by detecting the charge. Hence, it is expected that we can achieve the stabilization of singlet (triplet) states by feeding back the information about the charge distribution.

In the poster, aiming the feedback control[4] mentioned above, I discuss the dynamics and decoherence of the following system: two electrons in double quantum dots, and one of the dots is monitored by QPC, by using QDA.


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Path to perfect entanglement

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Unique quadrupole field to generate high efficiency entangled photon pairs

Entangled photon sources are crucial for quantum optics, quantum sensing and quantum communication. Semiconductor quantum dots in nanowires have recently emerged as leading candidates to generate entangled photons due to their high brightness and directional Gaussian emission profile for near-unity fiber coupling. However, the structural asymmetry of the quantum dot leads to a fine-structure splitting (FSS) and severely limits the use of quantum dots as a source of entangled photon pairs with high fidelity. Here, we propose a novel approach for generating a pair of entangled photons from the quantum dot with high fidelity by correcting the spatial asymmetry of the excitonic wave function via application of a quadrupole electrostatic potential. The proposed device architecture and the far field emission profile is presented in Figure 1: Proposed device.

We have performed numerical simulations for the proposed device by Nextnano3 in 2D, which solves the Schrödinger-poisson equation self-consistently. In Figure 2, FSS and electron-hole (e-h) overlap is plotted as a function of quadrupole potential, V applied on the proposed device. Our results demonstrate that the spatial asymmetry of the excitonic wave function can be tuned without compromising the spatial overlap between electron and holes. Importantly, the FSS can be tuned to zero, meaning that the excitonic wavefunction is symmetric, even when the electrical gates are misaligned with respect to the quantum dot asymmetry. Finally, we will present the nanofabrication of the first generation of devices and initial results (Figure 3: fabricated device). This work paves the way toward a deterministic source of entangled photons with high fidelity and unprecedented collection efficiency.

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Noiseless and flexible high-speed quantum dot IR photodetectors

A high-speed infrared photodetector (IRPD), converting light (photon) into current (electron), has various applications, ranging from military uses and imaging sensors to medicine. However, current high speed IRPDs technology are heavy and bulky, because they mainly use InSb (1-5.5 µm) and HgCdTe (1-10 µm) traditional semiconductors, while cryogenics or thermal-electrical cooling systems must be used to reduce thermal background noise. In the report, we demonstrated a flexible quantum dot high speed IRPD (~2 µm) fabricated by spin-coating approach. We characterized the device by combining high speed electronics with ultrafast laser excitation in a photoconductor with sub-40 "ps" response time. We observed a peak photocurrent density of more than $10^6 \text{mA/cm}^2$. The ratio between peak photocurrent and dark current is ten orders of magnitude, indicating the thermal signal reach theoretical noise baseline. The effective carrier mobility is in the order of $10 \text{cm}^2/\text{Vs}$. The ultrahigh photocurrent is attributed to multiple photo-generated excitons undergoing Auger recombination, followed by resonant, rapid tunneling at high energies. On the other hand, the ultralow thermal noise current is due to carrier transport at the band edge.

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Exciton-Ligand Vibronic Coupling in Colloidal CdSe Nanocrystals

Resonance Raman (RR) spectroscopy is an effective tool for probing the extent the exciton-phonon coupling in colloidal semiconductor nanocrystals (NCs); however, there are conflicting reports in the literature of whether exciton coupling to covalently bound surface ligand vibrational modes can be observed. Recent theoretical and computational studies have posited that such couplings can occur through resonance with ligand-nanoparticle charge-transfer states, particularly in the case of asymmetric vibrational modes of ligands with states that lie within the semiconductor bandgap. Such couplings are predicted to lead to large enhancements of the Raman scattering of certain ligand modes. Here we show Raman spectra of ligands covalently binds to the surface of 2-5 nm colloidal CdSe NCs through resonant excitation of the NC. The strong observed Raman scattering of the ligand modes appears to be consistent with theoretical prediction. To conduct these measurements, we use the cheap and commercially available capping ligand thiophenol (PhS), which is a widely employed surface-enhanced Raman scattering molecular tag. PhS conveniently mitigates the strong background photoluminescence which generally impedes RR measurements of semiconductor nanocrystals. It is also effective at displacing commonly used native phosphonic acid or amine ligands at room temperature, thus avoiding the use of higher temperatures which may lead to unwanted CdS shell growth. The approach is simple and can be employed on CdSe nanocrystals synthesized within our group, as well as commercially available samples. Furthermore, this strategy may be made generally applicable to study exciton-ligand interactions in a variety of semiconductor NC materials.


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Investigation of the Broad Emission Feature of Monodisperse Ultrasmall CdSe Quantum Dots via Modification of the Surface Environment

The highly tunable nature of the electronic band structure of quantum dots (QDs) is promising for many reasons. For a given composition, alteration of the size of the QD alone will change the light absorbed and emitted by the QD. Upon entering the ultrasmall regime, sub-2 nm radii, CdSe QDs have been shown to emit white light, i.e. all wavelengths in the visible regime, upon exposure to UV light despite having only two narrow absorption features, corresponding to the first and second excitonic transitions. Two main theories have arisen to explain this phenomenon: 1) a variety of surface traps yielding various energies of emitted wavelengths, or 2) a constantly fluctuating band gap due to a high degree of disorder on the surface. This second theory is known as fluxionality. Here the applicability of the two theories is compared based upon alteration of the ultrasmall CdSe band gap via modification of the surface environment. At this size, approximately 90% of the atoms in the QD are on the surface, thereby allowing for significant control over the QD behavior via design of different surface conditions. A number of techniques have been employed to probe the nature of the light-matter interactions of these systems including time correlated single photon counting (TCSPC), ultraviolet-visible (UV-vis) and photoluminescence (PL) spectroscopy, transmission electron microscopy (TEM), x-ray photoelectron spectroscopy (XPS) and ultraviolet photoelectron spectroscopy (UPS). In this work, QDs are placed in media with a range of dielectric constants to probe the role that the surface plays in the structure of the band gap. In particular, UV-vis, PL, and TEM are used to characterize the QD-solvent interaction. Additionally, simulations of the QDs in the various environments have also been conducted to garner a better understanding of the alterations to the electronic band gap based on the conditions imposed.

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Ultrafast X-ray Absorption Reveals Charge Carrier Modulated Dopant Bonding in Quantum Dots

Semiconductor Quantum Dots have great potential applications in renewable energy generation due to the ability to tune their redox potentials with size. One may also dope quantum dots to alter their electronic structure. Our group recently invented a method to dope each quantum dot with an exact number of guest ions by nucleating the QD around an organometallic seed cluster that contains the guest ions. As a result, each QD has the same number of dopants, which eliminates problems due to inhomogeneity of the dot stoichiometry. These materials were studied using time-resolved X-ray absorption spectroscopy, which allows us to characterize the electronic and coordination structure in both the ground and excited states. It was found that, when dopants interact with charge carriers, they may alter their bonding to the underlying matrix. This phenomenon of charge carrier modulation of dopant bonding has a strong effect on the conductivity properties of doped semiconductors.

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Fully Realized Time Resolved Multi Harmonic Dual Comb Spectroscopy

We propose multi harmonic time resolved dual comb spectroscopy as the ideal tool to study ultra-fast processes in quantum dots and other low dimensional materials. Since the initial realization of dual-comb spectroscopy, its combination with the pump-probe technique has been suggested as a means to achieve high resolution measurements of time-resolved complex transmittance. As the technology has matured, only one group [1] has very recently realized the fabrication of such a machine.

Not only do we present a significantly more refined version of this technology, we introduce the production of multiple harmonics which allows for the probing of various non-linear optical processes. We present an analysis of static rubidium cells in free-space, observing hyperfine splitting of the Rb85 Rb87 isotopes, as well as oxygen overtones in the atmosphere. We follow this by demonstrating saturable absorption in standard SAM semiconducting mirrors at 1550 nm. We show a spectral resolution of 0.002 nm and temporal resolution of less than 100 fs. Moreover, we simultaneously present phase data alongside transmission to characterize the complex permittivity of the material, and subsequently use this data to study dispersion changes between the saturated and non-saturated states of the device.

The successful demonstration of such a device, capable of measuring non-linear transient complex transmittance with great ease, paves the way for new advances in the study of many optical phenomena. Applications include the study of MEG and its potential to increase solar cell efficiency, as well as the population inversion dynamics of lasing media, and various non-linear processes such as two photon absorption, etc.


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Ultrafast Charge Transfer in Quinone Functionalised CdTe/CdS quantum dots for Biosensing Applications

Colloidal quantum dots (QDs) have found numerous applications in biomedical research, ranging from in vitro biosensing to intracellular and in vivo imaging, due to their bright, size-tuneable, spectrally-narrow and photo-stable emission. The sensitivity of QDs to organic ligands and to the chemical environment allows them to be functionalised for tracking specific biochemical processes and the diagnosis of diseases [1,2]. In particular, QD conjugates can be used for intracellular redox-sensing, the effectiveness of which depends on the efficiency of the switchable electron transfer event that leads to luminescence quenching.

In this work, the excited state dynamics in CdTe/CdS core/shell QDs with electron accepting quinone (Q2NS) ligands attached to the surface were studied under oxidised and reduced conditions, using ultrafast laser spectroscopy techniques. These results, along with atomistic modelling of the system, enabled us to determine that the electron transfer pathway is a multistep process incorporating ultrafast (2-8 ps) trapping of hot electrons. The efficiency of this hot-trapping step enables the QD-quinone system to achieve redox detection with a small number of sensor molecules per QD, which preserves their colloidal properties and allows for further functionalization to expand its application. The new insight into the excited state properties and the possibility of ultra-fast charge transfer will have important implications for the study of charge extraction from quantum dots.


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Morphology Impact on the Dephasing Dynamics in CdSe Colloidal Quantum Dots

Colloidal quantum dots (CQDs), due to their tunable opto-electronic properties, have found their place in a host of applications. A comprehensive understanding of the phenomena involved in their carrier dynamics is thus required for optimal design of devices involving CQDs. The strong quantum confinement that carriers are subjected to in CQDs, gives rise to an intricate intraband structure, where energy splittings can vary orders of magnitudes depending on the electron-hole exchange interaction (i.e. QD’s size and shape). In addition, the carrier-phonon couplings greatly influence the dephasing dynamics of coherences involving these states. Ensembles of CQDs have an inherent large inhomogeneous spectral broadening due to the nanoparticles’ size distribution. Resolving the material’s fine structure is thus challenging and restricts the number of techniques capable of studying it. Multi-dimensional coherent spectroscopy (MDCS) is an ideal tool for this purpose. In addition to possessing temporal resolution, MDCS is able to probe the homogeneous response even in the presence of large size inhomogeneity. By spectrally resolving the signal, one is able to measure the resonance linewidths of present states and their coupling strengths. In this work, we perform MDCS on different samples of CdSe CQDs at cryogenic temperatures. Through proper choice of slices on the 2D plots, the response of CQDs with specific resonance energies were isolated, revealing the zero-phonon line (ZPL), bright-dark exciton splitting, and acoustic phonon confinement modes. Multiple CQD samples of different core sizes and shell configurations are further studied to probe the influence of CQD size and shell on the exciton dephasing dynamics. In summary, we show direct measurements of three key components involved in the exciton dephasing process in CQDs. The variation of these peaks are related to changes in CQD structure, of fundamental importance to practical applications of these nanomaterials.

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Formation of colloidal copper indium sulfide nanosheets by two-dimensional self-organization

Ultrathin 2-dimensional nanomaterials (nanosheets, NSs) are attracting increasing attention due to their unique physical, electronic, and structural properties. Ultrathin colloidal semiconductor NSs with thickness in the strong quantum confinement regime are of particular interest, since they combine the extraordinary properties of 2D nanomaterials with versatility in terms of composition, size, shape, and surface control, and the prospects of solution processability. However, synthesis of NSs is challenging and synthesis procedures for materials other than the well-known Pb- and Cd-chalcogenides are still underdeveloped. Copper indium sulfide (CIS) is a direct semiconductor with a bulk bandgap of 1.5 eV and large absorption coefficients, which yields nanocrystals (NCs) with photoluminescence tunable in the vis−NIR (600−1100 nm) spectral range. Here, we present a new approach to CIS NSs and study their structural and optical properties. The CIS NSs form via self-organization and oriented attachment of 2.5 nm sized chalcopyrite CuInS$_2$ NCs, yielding triangular and hexagonal shaped NSs with a thickness of ~3 nm and lateral dimensions ranging from 20 to 1000 nm. The Cu:In ratio of ~3:1, different from the 1:1 stoichiometry of the parent CIS NCs, gives rise to a plasmon resonance in the NIR spectral window. The self-organization of the CIS NCs is induced by a sudden change in their composition due to preferential extraction of In$^{3+}$ by in-situ generated H$_2$S. This fast cation extraction leads to attractive dipolar interactions between the NCs. Primary amines play a crucial role in the formation of the CIS NSs, both by forming in-situ the cation extracting agent, and by preventing the attachment of NCs to the top and bottom facets of the NSs. Moreover, DFT calculations reveal that the amines are essential to stabilize the covellite crystal structure of the product CIS NSs.

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Radiative and non-radiative recombination in CuInS2 nanocrystals and CuInS2-based core/shell nanocrystals

Luminescent copper indium sulfide (CIS) nanocrystals are a potential solution to the toxicity issues associated with Cd- and Pb-based nanocrystals, as they show high absorption coefficients over a broad spectral range and size-dependent optical spectra, which are tunable in the visible to near-IR spectral window. However, the development of high-quality CIS nanocrystals has been hindered by insufficient knowledge of the electronic structure of these materials, and of the factors that lead to luminescence quenching. The photoluminescence (PL) of CIS and CIS-based core/shell NCs is characterized by broad bandwidths (FWHM 300 meV), large global Stokes shifts (~400 meV), and multi-exponential PL decays with long lifetimes (~200 ns). The origin of these characteristics is still under intense debate. A commonly invoked mechanism to explain the PL of CIS NCs is donor-acceptor pair recombination involving native point defects, based on the assumption that the PL mechanisms in nanoscale and bulk CIS are the same. Other models involve one localized charge carrier, that recombines radiatively with the remaining delocalized charge carrier. Yet another model precludes the involvement of localized carriers, ascribing the PL to the recombination of conduction band electron states with dark and bright valence band hole states. Here, we investigate the exciton decay pathways in CIS nanocrystals using time-resolved PL and transient absorption spectroscopy. Core-only CIS nanocrystals with low quantum yield are compared to core/shell nanocrystals (CIS/ZnS and CIS/CdS) with high quantum yields. Our results support the model of radiative recombination of a conduction band electron with a localized hole. Moreover, we find that PL quenching in low quantum yield nanocrystals involves initially uncoupled decay pathways for electron and hole. The electron decay pathway determines whether the exciton recombines radiatively or nonradiatively. The development of high-quality CIS NCs should thus focus on the elimination of electron traps.

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Coherence of a dynamically decoupled quantum-dot hole spin

Single spins in self-assembled InGaAs quantum dots are promising candidates for the implementation of distributed quantum information processing. For the electron the strong hyperfine coupling to the surrounding nuclei dephases the spin states in a few ns, inhibiting the quality of the spin-photon interface. In contrast, a hole spin is expected to experience significantly weaker coupling to the nuclear spin bath, providing a promising alternative. While it has been shown that holes can offer a longer dephasing time [1], future progress hinges on what coherence times can be achieved through decoupling.

In this work [2], we establish the regimes that allow for a highly coherent hole spin in this system. We observe a pick-up and decay with external magnetic field for both the inhomogeneous dephasing time, $T_2^*$, as well as the coherence time, $T_2$. We show that the decoherence of the hole is still dictated by the hyperfine coupling to the nuclear spin environment for fields up to a few Tesla, whereas electrical noise dominates at higher fields. In the latter regime, we implement dynamic decoupling to actively protect the hole spin against high-frequency electrical noise. Our work presents the first demonstration of such decoupling to prolong the coherence of an optically active spin qubit in a semiconductor, allowing us to achieve $T_2$ of 4 μs, the longest ground-state coherence time for any spin in this system. Further, we independently determine the local electrical environment which quantitatively supports the improvement of coherence we achieve with dynamic decoupling. We present the first report of direct transient measurements confirming that the hole spin outperforms the electron in both $T_2^*$ and $T_2$, solidifying the importance of the hole spin for the realisation of solid-state based quantum networks.


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Monitoring single-exciton Rabi oscillations via four-wave mixing

The rich energetic structure of individual quantum dots (QDs) can in some cases be restricted to the lowest excitonic states, forming a two-level system. Its coherent dynamics can be measured and controlled via heterodyne four-wave mixing micro-spectroscopy (FWM): By using two and three exciting pulses, we have access to the dynamics of the microscopic exciton coherences and populations, respectively.

In this combined experimental-theoretical investigation we use single self-assembled QDs embedded in a micropillar cavity, exhibiting a moderate optical Q-factor around 15000, operating in the weak coupling regime. The field cycling within the cavity amplifies the exciton coherent response and sets the pulse duration to around 10ps.

By varying the temperature, we have access to the detuning between the cavity mode and the exciton transition. This way we can tune the coupling strength between the two-level system and the optical field precisely. The Purcell effect is demonstrated and evaluated by measuring the exciton lifetime as a function of detuning in a three pulse FWM experiment.

In a two-pulse FWM configuration, we have recently inferred the coherence dynamics on a picosecond time-scale and we have shown that the phonon induced dephasing strongly depends on the temperature. By systematically varying the pulse areas of the driving laser pulses we here reveal and model two novel effects: (i) The phonon induced dephasing also depends on the exciton occupation after the first pulse. (ii) For large pulse areas additional oscillations of the FWM signal emerge due to the optically driven Rabi oscillations during the action of the first laser pulse.

By performing the analogous measurement with three pulses also the population dynamics during the Rabi oscillations has been experimentally resolved and theoretically described. As such, via the FWM delay dependence, we monitor the optically driven dynamics of the Bloch vector of a single QD exciton.

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Probing vibrational and excitonic coherences in CdSe QDs using absorptive 2D spectroscopy.

Colloidal semiconductor quantum dots (QDs) are an ideal model to probe the impact of many-body effects on structure and dynamics. Over the past decades, incoherent relaxation processes such as Auger and MEG have been studied extensively. Attention is now turning towards coherent dynamics. Once more, QDs provide an ideal model system to study the effects of many-body interactions on coherent dynamics: their structure shields the excitons from solvent perturbation and spatial confinement enhances charge-charge and charge-phonon interactions. Furthermore, different exciton states can share an electron or hole state, reducing the potential sources of decoherence between excitons. Both vibrational and excitonic coherences can be revealed by oscillations of the time-resolved absorption spectrum. Previous transient-absorption (TA) work on CdSe QDs allowed the observation of modulations due to acoustic and LO phonons. Furthermore, there is a growing evidence for the existence of interexcitonic coherences coming from 2D and TA work. Here we study the oscillations in the real absorptive 2DE signals from CdSe QDs at room temperature. The experimental results are compared to model calculations. We show the real absorptive spectrum can be used to discriminate between vibrational and excitonic coherences in this case, in the experimentally simple pump-probe geometry. The experimental data shows a strong signal from the LO phonon, but no electronic coherences can be observed. We outline factors that can reduce the duration and amplitude of interexcitonic coherence in QDs. This informs the design of nanostructures to attain longer lasting coherences.

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Entering the reappearance regime using chirped excitation of semiconductor quantum dots

Semiconductor quantum dots are promising candidates for single photon sources. The electron-phonon interaction is a major obstacle to overcome for an efficient optical state control. Depending on the excitation conditions phonons lead to a damping and hinder the optical state preparation. For extremely high pulse areas a decoupling of the phonons has been predicted for Rabi rotations, where the so-called reappearance regime is entered. In the reappearance regime an unhindered state preparation takes place, even at elevated temperatures. However, due to the high pulse areas the reappearance regime for Rabi rotations is very challenging to reach experimentally. A different excitation scheme is the adiabatic rapid passage occurring for chirped excitation. The adiabatic passage leads to a population inversion being stable against fluctuations in pulse area or chirp. Also here, phonons in general lead to a damping, which occurs only for one sign of the chirp at low temperatures [1]. At elevated temperatures the occupation probability is limited to 0.5 due to phonons. For excitation with strongly chirped pulses it is possible to enter the reappearance regime and, in contrast to Rabi rotations, it can be reached already for moderate pulse areas [2]. In this contribution we analyze the influence of electron-phonon interaction on the optical state preparation and discuss the excitation conditions for entering the reappearance regime. By introducing the dressed state picture we show why the reappearance regime can be entered with adiabatic rapid passage, while for Rabi rotations the phonon influence is always present. We compare our theoretical calculations with recent experimental data demonstrating the decoupling of the electron-phonon interaction for chirped excitation [2]. Our results open the way for a coherent and efficient state preparation.[1] S. Lüker et al., Phys. Rev. B 85, 121302(R) (2012)[2] T. Kaldewey et al. Phys. Rev. B 95, 241306(R) (2017)

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Ultrafast electric phase control of a quantum dot exciton

The application of an optical $\pi/2$-laser pulse to quantum dot (QD) ground state generates an exciton superposition state, with a quantum phase, which is defined by the electric phase of the laser pulse. Within the coherence time of the system, exciton phase is stored in the QD and evolves freely, if no disturbance appears. A measurement of the phase can be performed by a Ramsey experiment, i.e. by applying second optical $\pi/2$-pulse within the coherence time. Here the relative phase between optical pulses is important, which defines the quantum interference to be constructive or destructive. For a case where optical pulses are phaselocked, the occupancy of the excitonic system is extremely sensitive to external manipulations of exciton phase. In this work we take advantage of this sensitivity and we demonstrate a method, which controls only the exciton phase but not its occupation.

In our experiment, we control the coherent phase of exciton deterministically, by ultrafast Stark tuning of exciton energy. The resulting phase shift is measured by quantum interference using second $\pi/2$-pulse. We have already shown, that the coherent phase of a QD exciton can be manipulated electrically by phase-locked RF signals[1]. Here we have designed SiGe:C BiCMOS chips for the generation of ultrafast electric pulses ($t_{\text{rise}}<20\text{ps}$, 4.2K) and ultrafast photodiodes with embedded high quality InGaAs QDs. This hybrid approach enables us to perform electric phase control synchronous to double pulse ps laser excitation. We are able to demonstrate electrically controlled phase manipulations with magnitudes up to $3\pi$. We further introduce a promising protocol for the coherent optoelectronic preparation of an arbitrary quantum state of an exciton qubit by exclusive electric manipulation under condition of time invariant optical clocking[2].

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White-light 2D Coherent Spectroscopy Reveals Coherent Coupling and Crossed Excitons in InAs/InGaAs Nanostructures

Modern active media of semiconductor opto-electronic devices are based on tailored nanostructures of different dimensionality. For excitons in such complex potential landscapes, simplified level schemes often do not capture the essential coupling and scattering pathways. Coherent ultrafast multidimensional spectroscopy is an excellent tool to map new excitonic states and their interaction. The use of spectrally broad probe fields allows us to investigate coherently coupled exciton states with large energy separation up to ~200meV. We report about coherent coupling phenomena in In(Ga)As quantum dot/quantum well systems of a semiconductor optical amplifier (SOA). Coulomb interaction of carriers confined in the QD and those in the surrounding continuum leads to the formation of Crossed Excitons (CEs) that have crucial impact on both the ultrafast and diffusion dynamics in the QW and the ultrafast gain recovery within the QD. To map this multitude of states and couplings, we present a two-dimensional Fourier-transform (FT) experiment based on frequency heterodyning and broadband fs-pulses from a fiber laser. Owing to the white spectra of the pulses, coherent coupling of excitonic states spread over a wide spectral range can be visualized. For an unbiased SOA, the direct QD GS excitons dominate the spectrum along the diagonal, however, there is also a strong coupling (off-diagonal area) to higher energy states that confirms the existence of CEs and allow for a quantitative determination of CE energies that is not possible in simple pump-probe experiments. Under high injection currents, signatures of the QD GS and ES as well as their coupling terms are clearly visible at energies of 0.97eV and 1.04eV. We find identical homogeneous linewidths for the QD GS and ES indicating coherent coupling to the surrounding carrier reservoir, an essential aspect for future device engineering based on control of Coulomb couplings between confined and delocalized charge carriers.

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Current antiresonance in spin-orbit coupled double quantum dots

The coherent control of spin states with ac-driving fields is important for spin-based qubits. At the same time, the spin-orbit-interaction enables electrical control of spin qubits [1], therefore understanding the role of the spin-orbit-interaction is useful. In semiconductor quantum dotsspin resonance effects can be detected via electrical transport measurements [1]. In this work, we consider a double dot in the spin blockade regime and focus on the coupled two-spin states. The resulting states in a static magnetic field are mixed (hybridized) because of the spin-orbit-interaction. Under the application of an oscillating electric field the electrical current flowing through the double dot displays spin resonance effects. By employing a Floquet-Markov open-system approach we calculate the current as a function of the frequency and power of the driving field. For a specific frequency range we predict an interference pattern (antiresonance) in the current as a result of a three-level dynamics. We compute the current in the weak and strong driving regimes and find a rather high sensitivity. We clarify the role of the spin-orbit strength, energy detuning, and g-factor difference. Our findings can be tested with standard electrical transport measurements [1, 2, 3].[1] R. Hanson et al, Rev. Mod. Phys. 79, 1217 (2007).[2] S. Nadj-Perge et al, Phys. Rev. Lett. 108, 166801 (2012).[3] K. Ono et al, Phys. Rev. Lett. 119, 156802 (2017).

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Direct determination of the zero-field splitting for a single cobalt ion embedded in a CdTe/ZnTe quantum dot

When a Co\(^{2+}\) impurity is embedded in a quantum dot [1], crystal strain strongly influences the zero-field splitting between Co\(^{2+}\) states with spin projection \(S_{z} = \pm 3/2\) and \(S_{z} = \pm 1/2\). Experimental evidence of this effect has been given in previous studies [1]; however, direct measurement of the strain-induced zero-field splitting has been inaccessible so far. Here this splitting is determined thanks to magneto-optical studies of an individual Co\(^{2+}\) ion in an epitaxial CdTe quantum dot in a ZnTe barrier. Using partially allowed optical transitions, we measure the strain-induced zero-field splitting of the Co\(^{2+}\) ion directly in the excitonic photoluminescence spectrum [2]. Moreover, by observation of anticrossing of \(S_{z} = +3/2\) and \(S_{z} = -1/2\) Co\(^{2+}\) spin states in a magnetic field, we determine the axial and in-plane components of the crystal field acting on the Co\(^{2+}\). The proposed technique can be applied to optical determination of the zero-field splitting of other transition-metal ions in quantum dots.


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Structural analysis of InGaAs/GaAs Quantum Dots using Inverse NMR Spectra

Nuclear magnetic resonance (NMR) is a very powerful technique in the study of materials at an atomic level. NMR spectra can be measured optically from single self-assembled InGaAs/GaAs quantum dots (QDs) from which important structural information can be determined. Here we present a direct comparison of the structure of 2 different samples – a sample capped with low temperature GaAs and one grown at higher temperature with In-flush. A novel 3-band inverse NMR method was developed to accurately measure the satellite transitions, which reveal structural information that could not be previously derived.

The NMR spectrum of a QD can be divided into distinct peaks formed from the central transition (CT, \( I_z = -1/2 \leftrightarrow 1/2 \)) and the satellite transitions (ST, \( I_z = \pm 3/2 \leftrightarrow \pm 1/2 \), for spin \( I = 3/2 \)). The CT spectra of the 4 most abundant nuclear isotopes in the samples (\(^{75}\)As, \(^{115}\)In, \(^{69}\)Ga, \(^{71}\)Ga) were measured using the inverse NMR technique (see Chekhovich et al., 2012). By comparing the amplitudes of the CT peaks, the relative concentrations of gallium and indium nuclei within the dot region could be estimated - for the In-flush sample \( p_{\text{In}} = 29\% \) and \( p_{\text{Ga}} = 71\% \), and for the low temp. cap sample \( p_{\text{In}} = 13\% \) and \( p_{\text{Ga}} = 87\% \), despite QDs in both structures emitting at about the same wavelength.

The ST spectra are much broader than the CTs due to strong quadrupolar shifts induced by strain in self-assembled dots. Using the newly developed 3-band method an additional 3\(^{rd}\) band was applied in the Inverse NMR waveform that saturates the CT. Surprisingly, a non-zero ST signal was observed (for all isotopes) close to the central frequency corresponding to nuclei in low strain regions which have thus far not been experimentally observed. This 3-band method allows us to extract new, minute structural details of QDs consisting of only a few tens of thousands of nuclear spins.

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Electrically tunable dynamic nuclear spin polarization in GaAs quantum dots at zero magnetic field

In semiconductor nanostructures the electron and nuclear spin dynamics are efficiently coupled via the hyperfine interaction, especially in III-V materials with Ga, Al and In where 100 % of nuclei have non-zero nuclear spin¹. Both electron and nuclear spins can be controlled optically. Stable nuclear spins can potentially be used as a resource for quantum information storage². The nuclear spin dynamic is widely studied, but little is known about the initialization mechanisms. Their control is often achieved in experiments in applied magnetic fields, which is not practical for applications. Here we investigate optical pumping of carrier and nuclear spins in charge tunable GaAs dots grown on 111A substrates using a laser energy one longitudinal optical phonon above the positively charged exciton X⁺ state transition. We demonstrate dynamic nuclear polarization (DNP) at zero magnetic field in a single quantum dot. We show that DNP can be tuned in both amplitude and sign by variation of an applied bias voltage V<sub>g</sub>. Application of ΔV<sub>g</sub> of the order of 100 mV changes the Overhauser splitting (nuclear spin polarization) from -30μeV (-22%) to +10μeV (+7%), although the X⁺ photoluminescence polarization does not change sign over this voltage range³. This indicates that absorption in the structure and energy relaxation towards the X⁺ ground state might provide favorable scenarios for efficient electron-nuclear spin flip-flops, generating nuclear spin polarization during the first tens of ps of the X⁺ lifetime, which is of the order of hundreds of ps. Finally, voltage control of DNP is further confirmed in Hanle experiments.


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Electron-electron interaction in the double quantum dots system defined within the finite flake of silicene.

Since the discovery of graphene [1], the scientists have been looking for other two-dimensional systems with extraordinary properties. One of these material is silicene [2] with the crystal and electronic structure similar to graphene, yet with large spin-orbit coupling [2] and controllable band gap [3].

In this work, we consider a system that consists of the finite flake of silicene with double quantum dots defined electrostatically within. We determine the confined states with the energy levels that lie in the band gap using the tight-binding method occupying the pz orbitals of silicon atoms. The considered flake is large enough to ensure the vanishing of the wave functions on the edges. Moreover, the perpendicular electric field shifts the possible edges states higher/lower in the energy, out of our range of interest, making the quantum dots completely separated from the influence of the edge.

We analyze the quantum dots states as functions of the size/depth of the dots as well as the distance between them, and describe the creation of the bonding/antibonding orbitals extended over the double dot system. Next, we use the one-electron states as a basis for the two-electron calculations with the configuration-interaction approach. For the electron pair we present the results as the functions of the external magnetic field. We consider both, weak and strong interdot coupling limits to investigate the effects of the electron-electron interaction.


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Activation of band-edge excitons in nanocrystals by symmetry breaking: 
Modeling the effect of charged impurities in CdSe Nanocrystals

We analyse the effect of symmetry breaking on the exciton fine structure and optical selection rules in semiconductor nanocrystals and show that the descent of the nanocrystal symmetry from spherical to point group C<sub>s</sub>, which is characterized by just one mirror plane symmetry element, leads to activation of all five \( F=2, F_\perp=\pm 2, \pm 1, 0 \) excitons in CdSe NCs. As an example, we model the effect of symmetry reduction by introduction of charged impurities in quasi-spherical CdSe nanocrystals [1]. The off-center charge breaks the nanocrystal symmetry and affects the optical properties not only by level shifting, but through mixing of the hole sublevels associated with different angular momentum projections and parity. The result of the symmetry breaking is a shortening of the radiative lifetime, even at room temperature, in qualitative agreement with the increase in PL efficiency observed in CdSe nanocrystals doped with positive Ag charge centers [2]. This acceleration persists even with surface compensating charges, whose presence also stabilizes the band edge peak position in PL and absorption spectra. Calculation also shows that absence of spectral shifts with doping in the reported experiments [1] can be explained only if charged centers are at or near the NC surface, not at the NC center, where an interstitial Ag\(^+\) donor would be expected to migrate to due to image forces [3] in the absence of surface compensating charges. [1] P. C. Sercel, A. Shabaev, Al. L. Efros, Nano Lett., 2017, 17, 4820 (2017). [2] A. Sahu, M. S. Kang, A. Kompch, C. Notthoff, A. W. Wills, D. Deng, M. Winterer, C. D. Frisbie and D. J. Norris, Nano Lett. 12, 2587, (2012). [3] F.D. Ott, L.L. Spiegel, D.J. Norris, and S.C. Erwin, Phys. Rev. Lett. 11, 156803 (2014).

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Surface Chemistry Impact on the Electronic Structure of Colloidal Quantum Dots

Surface chemistry modification of as-synthesized colloidal quantum dots (CQDs), commonly referred to as ligand exchange, is mandatory towards effective CQD-based optoelectronic and photocatalytic applications. The widespread recourse to ligand exchange procedures is leading to uncover the marked impact exerted by chemical species at nanoscopic surfaces on the ground state optical properties of CQDs: indeed, CQD surface modification has been shown to induce, among others, optical band gap reduction[1] and broadband optical absorption enhancement[2]. However, the most common descriptions of ligands at the QD surface, which are based on the analogy to core/shell heterostructures that consider CQDs as superposition of core and ligand components and interfaces as abrupt, fail in accounting for the observed ligand-induced CQD ground state spectral changes. Rationally designed thiol-based ligands at the surface of metal chalcogenide CQDs are here exploited to suggest a comprehensive description of such phenomena. Surface chemistry does drastically modify the CQD electronic structure to an extent that scales with surface-to-volume ratio as a result of inherent ligand/core orbital mixing; some heuristic guidelines to assess surface chemistry impact on the CQD electronic structure are suggested. [1] a) R. Koole et al., J. Phys. Chem. 2007, 111, 11208; b) A. Wolcott et al., J. Phys. Chem. Lett. 2011, 2, 795; c) M. T. Frederick et al., Nano Lett. 2011, 11, 5455; d) M. T. Frederick et al., Nano Lett. 2013, 13, 287. [2] a) C. Giansante et al., J. Am. Chem. Soc. 2015, 137, 1875; b) D. Kroupa et al., Chem. Commun. 2016, 52, 13893; D. Debellis et al., Nano Lett. 2017, 17, 1248.

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**Ab initio-based model of electronic coupling between colloidal QDs**

Colloidal semiconductor quantum dots (QDs) can form periodic supercrystals, which can be used for various types of electronic and optical applications[1]. The quantum mechanical atomistic calculation of the electronic properties of experimental-size nanostructures remains a challenging task. We present a method within the framework of the atomic effective pseudopotential (AEP) approach[2]. AEPs are constructed by extracting the local part of the self-consistent effective pseudopotentials using simple DFT calculations within local density approximation (LDA). We show that the surface dipole effects need to be taken into account to model electronic properties of confined structures accurately. The analytical connection to the DFT, good level of transferability and capability to model systems with 10 000 atoms and more is demonstrated. The method opens opportunities to go beyond LDA by introducing correction to the band gap. AEP-based wave functions and eigenvalues of experimental-sized QDs were investigated within Configuration-Interaction (CI) theory[3] to calculate exciton properties. In the current report we demonstrate a model system of two coupled spherical QDs and study the dependence of the electronic states coupling with the QD diameter and separation as well as screening effects of the solvent on the exciton binding energy and dark-bright splitting.[1] Kramer et al., Chemical reviews, 114(1), 863-882 (2013) [2] Karpulevich A. et al., PRB, 94, 205417 (2016) [3] A. Franceschetti et al., PRB, 60, 1819-1829 (1999)

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On the Origins of the Stokes Shift in PbS Quantum Dots: Impact of Polydispersity, Ligands and Defects

Understanding the origins of the excessive Stokes shift in the lead chalcogenides family of colloidal quantum dots (CQDs) is of great importance at both the fundamental and applied levels; however, our current understanding is far from satisfactory. Here, utilizing a combination of ab initio computations, and UV/vis and photoluminescence measurements, we investigate the contributions to the Stokes shift from polydispersity, ligands, and defects in PbS CQDs. The key results are as follows: (1) The size and energetic disorder of a polydisperse CQD film increase the Stokes shift by 20 to 50 meV compared to that of an isolated CQD; (2) Franck-Condon (FC) shifts increase as the electronegativities of the ligands increase, but the variations are small (<15 meV); unlike the aforementioned two minor factors, (3) the presence of certain intrinsic defects such as V\textsubscript{Pb}\textsuperscript{2-} and V\textsubscript{Cl}\textsuperscript{2+} (in Cl-passivated CQDs) can cause substantial electron density localization of the band edge states, and consequent large FC shifts (100’s of meV). This effect arising from defects could explain the excessive Stokes shifts in PbS CQDs and improve our understanding of the optical properties of PbS CQDs.

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Strain Induced Polarization in CdSe(x)S(1-x)/Cd(x)Zn(1-x)S Seeded Nanorods with Graded Composition

Seeded semiconductor nanorods represent a unique family of quantum confined materials that manifest characteristics of mixed dimensionality. They show polarized emission with high quantum yield and fluorescence switching under an electric field, features that are desirable for use in display technologies and other optical applications. So far, their robust synthesis has been limited mainly to CdSe/CdS heterostructures, thereby constraining the spectral tunability to the red region of the visible spectrum. Lately, a novel synthesis of CdSe<sub>x</sub>S<sub>1-x</sub>/Cd<sub>x</sub>Zn<sub>1-x</sub>S seeded nanorods with a radially graded composition was presented. These seeded nanorods show bright and highly polarized green emission with minimal intermittency, as confirmed by ensemble and single nanorods optical measurements. Using Finite Element simulation and multiband k·p theory, we analyze the potential influence of several factors affecting excitonic emission and show that strain induced by the ZnS in the shell is the mechanism promoting linear polarization. It favors energetically light hole excitons over heavy hole ones, via deformation potential, and makes their radiative recombination faster via piezoelectricity. Thus, the controlled addition of Zn influences and improves the nanorods’ optoelectronic performance by providing an additional handle to manipulate the degree of linear polarization and the quantum confinement beyond the common size control approach. These nanorods may be utilized in applications that require the generation of a full, rich spectrum such as energy-efficient displays and lighting.

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Ultra efficient numerical study of non-Markovian phenomena in quantum dots

Modelling realistic quantum devices requires an understanding of quantum systems strongly coupled to their environment. Describing strong coupling, and the non-Markovianity that accompanies it, analytically is extremely challenging and for consistently reliable results one must use numerical methods. Available methods are either restricted to special cases or limited by huge computational requirements. Here we present a revolutionary numerical approach to describing the time evolution of a quantum system coupled to a general non-Markovian environment [1], and show it represents a substantial improvement over other techniques. We demonstrate the applicability of our method to quantum dot systems by calculating exciton dynamics and optical properties, showing that exciton-phonon interactions can lead to observable non-Markovian phenomena. Within the path integral description of open quantum systems, environmental degrees of freedom are integrated out to leave an influence functional of system trajectories only. This influence functional describes all dissipative and non-Markovian physics in the system due to the environment, though performing the remaining integration over system trajectories is intractable in general. We show that discretising the system trajectories allows the path integration to be performed numerically by contracting a tensor network. We efficiently contract this network using a matrix product state representation of the augmented density tensor, a large-dimensional tensor which stores the system state and its previous history [2]. Our method, called "time-evolving matrix product operators" (TEMPO), can be used to calculate both state dynamics and arbitrary multi-time correlation functions of generic open systems, making it widely applicable. Thus, we first use TEMPO to investigate the role of dimensionality on exciton hopping between two spatially separated sites embedded within a common phonon environment. We then use TEMPO to study the impact of exciton-phonon interactions on spectra and single-photon emission properties of a quantum dot [3].


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Electronic properties of magnetic diluted semiconductor quantum dots

In this work, we are interested on the diluted magnetic III-V quantum dot compounds. Using matrix method, we report on the calculation of the electron-hole ground state transition energies in a self-organized quantum dot with a single magnetic impurity. We examine how the impurity position in the quantum dot affect the electron-hole ground state transition energies. Then we compare our results with the undoped III-V single quantum dot.


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Numerical simulations of the Pauli blockade microscopy of the planar quantum dots

We consider the influence of varying electric field introduced by the tip of the atomic force microscope on the electron states in the two-dimensional quantum dots in which the current flow is blocked by the Pauli exclusion principle\(^1,2\). The spin transitions induced by the AC potential of the tip in the presence of the spin-orbit interaction are studied: for two-electron case the AC voltage applied to the tip acting as a scanning gate electrode\(^3\) drives the spin-polarized triplet \(T_{++}\) state to the singlet \(S\) or triplet \(T_{0}\) state in which the Pauli blockade is lifted.

Our study covers a single electron confined in a rectangular elongated quantum dot and two electrons in the tunnel-coupled double quantum dots.

We simulate the proposed electric-dipole spin resonance experiment\(^4-6\) using the stationary Hamiltonian eigenstates that form the basis for solving the time-dependent Schrödinger equation with the AC perturbation introduced by the tip. We observe the direct and higher order transitions and present the transition probability maps as functions of the tip position for \(S\) and \(T_{0}\) in the final states. The rates of the spin-flip transitions depend strongly on the probe position. The similarity between the maps obtained for the fixed resonant frequencies and the form of the transition matrix elements is also discussed. The numerical simulations provide the information about the symmetry of the confined wave functions of the states involved in the transitions. We demonstrate the possibility of manipulating the electron spins in quantum dots, identification of the final transition state and probing the spin densities.


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Photoluminescence of Silver and Alloy Colloidal Quantum Dot from Intraband Transition

The first steady state intraband transition was reported by Jeong et al. in 2014 and suggested new direction of colloidal quantum dot study by investigating the electronic transition occurring in higher quantum states. In this poster, I will present the mid-IR photoluminescence of the intraband transition for the Ag$_2$Se and CdHgSe alloy nanocrystals. This intraband PL of the silver chalcogenide has significantly removed the concerns about the toxicity of the mercury chalcogenide nanocrystals and renders a new possibility of non-toxic CQDs showing the intraband transitions. The Ag$_2$Se nanocrystals was regarded as a nanomaterial having a bandgap transition in the mid-IR region. However, the spectro-electrochemistry and mid-IR emission result of Ag$_2$Se CQDs proved the mid-IR bandgap transition of Ag$_2$Se is the mid-IR intraband transition rather than the bandgap transition. The mid-infrared emission was measured by using the home-made mid-infrared emission spectrometer. Surprisingly, the intraband transition can be generated by cation exchange method. The CdHgSe alloy synthesis has shown the interesting optical property that the major electronic transition shifts from the visible to mid-IR LSPRs by increasing the mercury portion of the nanocrystals. The photosensitivity and the PL results were obtained to determine the stoichiometric effect of sample characteristics.


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Spin Properties in Magnetically Doped Semiconductor Nanocrystals

Recently, research interest has shifted toward semiconductors NCs embedded with magnetic ions, joining a group of materials known as diluted magnetic semiconductors (DMS)[1]. The magnetic ions provide spin-exchange interaction with carrier spins, resulting in giant enhancement of magneto-optical and magnetic properties of semiconductors. Low-dimensional structures (quantum wells, wires, dots) offer advantage for DMS, as the spatial carrier confinement strongly enhances their exchange interactions with magnetic ions[2].

In the present work, we compare the generated magnetism in colloidal nanocrystals of different morphologies, the quantum dots (QDs), seeded nanorods (NRs) and nanoplatelets (NPLs), all with core/shell configuration based on CdSe/CdS composition with a quasi-type-II core-shell band alignment. Those nanocrystals were embedded with a diluted concentration of Mn<sup>+2</sup> impurities, prepared by colloidal layer-by-layer deposition procedures, controlling the position of the magnetic ions in the shell regime.

The magneto-optical properties were investigated by the temperature dependence photoluminescence (PL) study. The results showed a pronounced Stokes shift of the emission in the doped nanocrystals with respect to that of the undoped structures, stemming from the so-called sp-d interaction. Fitting the experimental data to a Brillouin function revealed a generation of internal magnetization varying in the various structures. Further, characterization of carrier-Mn interaction was done utilizing optically detected magnetic resonance (ODMR) spectroscopy. The unique experimental approach permits achievement of high spectral resolution <100 µeV, obligatory for detection of the anticipated interactions (100–800 µeV), revealing spin exchange interactions and manipulation that cannot be otherwise discerned.

The findings of this work showed a control of doping level and positioning as a tool to manipulate spin properties in nanoscaled materials, a topic of main concern in the currently developing spin-based technologies.


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High Contrast Differential Reflection Measurements on a Single Quantum Dot

Excitons in semiconductor quantum dots (QDs) are promising candidates for the realization of quantum information and communication technologies. Resonance fluorescence is a widely used possibility to address resonantly the QD excitons and generate single, indistinguishable photons. Commonly this requires that the laser background is suppressed by seven orders of magnitude, e.g., using cross-polarization [1]. Another possibility is differential reflection, where the ratio between the signal of a single QD and the reflected laser light is determined by lock-in technique. However, this technique was limited to low contrasts between QD photons and back-scattered laser light in the order of 10% [2].

In this contribution we present an optimized sample structure which significantly increases the collection efficiency of the QD photons. A distributed Bragg reflector and an epitaxially grown gate allows us to measure contrasts up to more than 80%, in confocal rejection even exceeding 800%. It enables us to perform measurements on a single dot without the modulation of the lock-in technique. These findings open up the possibility to perform resonant optical spectroscopy on single QDs without the need of suppressing the backscattered laser light by cross-polarization. [1] Kuhlmann, Rev Sci. Instrum. 84, 073905 (2013) [2] Vamivakas, et al., Nano Lett. 7, 2892 (2007)

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Stabilization of quantum dot fluorescence with a built-in feedback loop

The vision of a quantum photonic networks relies on transform-limited single photons on-demand. Resonance fluorescence on a quantum dot provides access to a solid-state single photon source, where unfortunately the environment is a source for spin and charge noise [1], causing fluctuations of the emission frequency and destroying the photon indistinguishability. We demonstrate a built-in stabilization approach for the photon stream, which relies solely on charge carrier dynamics of a two-dimensional hole gas inside a micropillar structure [2]. The hole gas is populated by hole tunneling from field-ionized excitons and influences the energetic position of the excitonic transition by changing the local electric field at the position of the quantum dot. The photon noise is suppressed by 50 percent with an operating frequency range in the used micropillar structure up to 1 kHz. This built-in feedback loop represents an easy way for stabilization of the quantum dot emission in large arrays of single photon emitters and promises to reach higher bandwidth by device optimization.[1] A. V. Kuhlmann, et al., Nat. Phys. 9, 570 (2013).[2] B. Merkel, et al., Phys. Rev. B 95, 115305 (2017).

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A charge-tunable quantum dot deep in the strong coupling regime of cavity QED

The strong coupling regime of cavity quantum electrodynamics represents a fully quantum light-matter interaction. It stands for a coherent exchange between an atomic excitation and a single photon. Achieving strong coupling is key to creating coherent atom-atom couplings and single-photon transistors. Three parameters are crucial: the atom-photon coupling rate $g$, the atom decay rate $\gamma$, and photon loss rate $\kappa$. True strong coupling is achieved only when $g \gg \gamma$ and $g \gg \kappa$. A quantum dot is an ideal “atom” on account of its large optical dipole moment. The conundrum is that a large coupling $g$ is achieved only by nanofabrication of the cavity, a process which usually leads to a large $\kappa$ via scattering-induced losses: $g \gg \gamma$ is readily achieved; $g \gg \kappa$ is more challenging[1–5].

We report here an experiment in which we achieve both $g \gg \gamma$ and $g \gg \kappa$. We use a quantum dot embedded in a highly miniaturized, fully tunable Fabry-Pérot microcavity[6–7]. This gives reasonably large values of $g$, and, crucially, a way to miniaturize without increasing $\kappa$. The quantum dot is embedded in a charge-tunable heterostructure which gives close-to-transform limited optical linewidths, in situ tuning via the Stark effect, and control of the quantum dot charge via Coulomb blockade. We achieve $g/2\pi=3.31$ GHz, $\gamma/2\pi=0.34$ GHz, $\kappa/2\pi=0.63$ GHz (Q-factor=510,000).

Resonant laser spectroscopy shows a very clear avoided crossing at the quantum dot exciton-cavity resonance. The splitting between the two peaks, the coupled exciton-photon modes, is a factor of 14 larger than the individual polariton linewidths. The intensity correlation function $g^{(2)}$ exhibits perfect antibunching when the laser is tuned to one of the polariton resonances. On detuning the laser slightly, we observe pronounced oscillations in $g^{(2)}$, unambiguous evidence of a coherent exciton-photon interaction.

We present this geometry as an ideal platform to explore and exploit both the strong and weak coupling regimes of cavity QED.


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High beta factors and directional coupling of InAs/GaAs quantum dots to photonic crystal waveguides determined by direct spectral imaging

A quantum dot (QD) embedded in a photonic crystal waveguide (PCWG) is a promising system to implement quantum circuits. A fundamental requisite of any network based on a PCWG is that all the spontaneous emission (SE) from the emitter couples to a single waveguide mode. The probability that a spontaneously emitted photon couples to the waveguide mode is given by the beta factor \( \beta \). Previous works have used the lifetime to estimate \( \beta^{1,2} \). This approach approximates the free space emission as constant, which we show creates significant errors. In this work, we image the SE of the QD along the WG onto the input slit of a high resolution imaging spectrometer, at T=5K. The SE is measured both spatially resolved along the waveguide and spectrally resolved. The SE guided by the PCWG and coupled by the bottom and top grating couplers to free space is measured together with the direct QD free space emission. The \( \beta \) factor can be, for the first time, directly determined, using the relative emission intensities. By introducing an external magnetic field, directional emission is demonstrated, as previously observed\(^1\). Using the collected emission intensities, we can directly quantify the directionality of the emission, and consequently the mode circularity at the QD site. Evaluating the mode circularity versus beta factor, we find that high beta factors are only found for small mode circularities, and vice versa. We find beta factors > 97.5% outside of the slow light regime, in deviation from what was reported in \cite{1}. Finally, by measuring the propagation wave vector for different energies, simulations are accurately calibrated to the experimental data. This allows us to determine the position of the QDs within the lattice unit cell from their beta factors and mode circularities.


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Pyridine-Induced Dimensionality Change in Hybrid Perovskite Nanocrystals

Engineering the surface energy through careful manipulation of the surface chemistry is a convenient approach to control quantum confinement and structure dimensionality during nanocrystal growth. Here, we demonstrate that the introduction of pyridine during the synthesis of methylammonium lead bromide (MAPbBr$_3$) perovskite nanocrystals can transform three-dimensional (3D) cubes into two-dimensional (2D) nanostructures. Density functional theory (DFT) calculations show that pyridine preferentially binds to Pb atoms terminating the surface, driving the selective 2D growth of the nanostructures. These 2D nanostructures exhibit strong quantum confinement effects, high photoluminescence quantum yields in the visible spectral range, and efficient charge transfer to molecular acceptors. These qualities indicate the suitability of the synthesized 2D nanostructures for a wide range of optoelectronic applications.

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Synthesis of Bio-compatible Type-II InP/ZnO Quantum Dots

Indium phosphide based quantum dots can be a promising alternative for cadmium and lead-based QDs due to less toxicity and tunable effective band gap photoluminescence covering the blue to near infrared. Although indium-based colloidal quantum dots with Type-I band alignment have attracted significant interest to suppress non-radiative losses in surface trap states and obtain narrower PL line widths and increased quantum yield, little attention has been paid to Type-II InP-based quantum dots, in which the charge carriers are delocalized away from each other. Zinc oxide has the potential to form a Type-II band alignment by forming a shell on an InP core. Herein, we demonstrated the first heavy metal-free Type-II quantum dot. We synthesized the InP core by the hot-injection method, and ZnO shell was grown by thermal decomposition of zinc acetylacetonate. The presence of the InP/ZnO core/shell structure was confirmed by XRD, TEM and XPS measurements. Additionally, the steady-state and time-resolved optical studies revealed the characteristic transition of a Type-II core/shell system and are in agreement with our quantum mechanical calculations. Finally, the biocompatibility of QDs was confirmed by investigating the membrane integrity, morphology and mitochondrial activity of biological cells seeded on QDs.

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Optimization of Near-Infrared I-III-VI Quantum Dots for Scale-Up

Near-Infrared (NIR) emitting fluorophores attract significant attention due to their potential use in a wide variety of industries. Currently available NIR fluorophores lack wavelength tunability, cover only a small portion of the NIR spectrum, suffer from low brightness, have poor stability, low quantum yield (QY), small extinction coefficients, high self-absorption, and/or are composed of Restriction of Hazardous Substances Directive prohibited elements such as Cd, Hg, or Pb. UbiQD solves the problems of existing NIR fluorophores with I-III-VI semiconductor-based quantum dots (QDs). Our QDs are synthesized through a combination of air-free (Schlenk line) and ambient techniques. This presentation focuses on CuInSe$_{x}$S$_{2-x}$/ZnS (CISeS) QDs with emission longer than 1000 nm and the achievements UbiQD reached with these materials, such as, consistently high QY (up to 100%), inherently large Stokes shifts, variable lifetimes, as well as work with surface modification. These reproducible qualities lead to many potential applications for CISeS QDs, including lighting, displays, security, biotechnology, and luminescent solar concentrators. Our development efforts will contribute to a successful synthesis scale-up of UbiQD’s patented technology.


High-Performance CuInS$_2$ Quantum Dot Laminated Glass Luminescent Solar Concentrators for WindowsMatthew R. Bergren, Nikolay S. Makarov, Karthik Ramasamy, Aaron Jackson, Rob Guglielmetti, and Hunter McDanielACS Energy Letters 2018 3 DOI: 10.1021/acsenergylett.7b01346

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Spontaneous Formation of Noble- and Heavy-Metal-Free Alloyed Semiconductor Quantum Rods for Efficient Photocatalysis

Quasi-one-dimensional cadmium chalcogenide nanorods are benchmark semiconductor materials that have been combined with noble metals to constitute quantum rod heterostructures for efficient photocatalysis. However, the high toxicity of cadmium and cost of noble metals are the main obstacles to their widespread use. Herein we present a facile colloidal synthetic approach that leads to the spontaneous formation of cadmium-free alloyed quantum rods (ZnS<sub>x</sub>Se<sub>1-x</sub>) from polydisperse ZnSe nanowires by the etching of alkylthiol. The obtained non-noble metal ZnS<sub>x</sub>Se<sub>1-x</sub> quantum rods can be directly adopted as efficient photocatalysts in water splitting, showing a striking oxygen evolution capability of 3000 μmol g<sup>-1</sup> h<sup>-1</sup>. The alloyed quantum rods based photoanode presents an excellent photocurrent density of 0.75 mA cm<sup>-2</sup> at 1.23 V. The outstanding performances of alloyed quantum rods can be attributed to their increased surface area and active sites induced by etching of alkylthiol.

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Mixed Halide QD Passivation Strategies Enabling High Performance Infrared QD Photovoltaics

Colloidal quantum dots are promising materials for tandem solar cells complementary to silicon and perovskites. These devices are fabricated from solution phase; however, existing methods for making infrared-bandgap CQD inks suffer agglomeration and fusion during solution exchange. Here we develop a new ligand exchange that provides robust surface protection and thereby avoids aggregation. First, we exchanged long oleic acid ligands to a system comprised of chloride ligands; then, we re-shelled the surface using larger halides and pseudohalide ligands that enabled transfer to a polar solvent. Further optimization of this process led to an increased amount halide content on the surface of the QDs, resulting in the fabrication of an infrared QD solar cell capable of adding an additional 1.20% power points to a standard cSi solar cell. This is over a 1.5x enhancement over the current IR QD PV record.

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Interplay between charge and photoluminescence of individual doped semiconductor nanocrystals

Aliovalent doping of CdSe nanocrystals (NCs) can potentially enhance conductivity, making the material better for applications that depend on electrical transport, such as solar cells, transistors, and light emitting devices. While many optical and electronic properties of these doped materials have been reported, including those of Ag⁺ doped CdSe, aspects of the doping process, including the location of the dopant atoms within the NC lattice, and the underlying physics of the optical properties are still not well understood. Electrostatic force microscopy (EFM) was used to determine the charges of individual doped NCs in order to investigate the dopant type with respect to the dopant concentration. Cation doped, including Ag⁺ doped, CdSe NCs were synthesized and were shown to have an enhanced fluorescence over their undoped counterparts. The doped NCs were then studied with EFM to measure their charge as a function of added cations. While there was no direct trend relating the charge to the dopant type, there was a remarkable and unexpected correlation between the charge and PL intensity, regardless of the dopant cation introduced. We conclude that the enhancement in PL intensity, as tracked also by increased charge, is actually a consequence of a radiative rate acceleration caused by symmetry breaking and consequent brightening of a dark state by the introduced charge, rather than a result of the direct Ag⁺ doping within the CdSe NCs.

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Vanishing confinement in semimetal nanocrystals: a path to reach the THz range

Wide and moderate band gap materials have attracted most of interests for synthesis of nanocrystals and lead to visible and near IR optical properties. To push further the absorption of nanocrystal toward the infrared, confined semimetal materials have been used and among them mercury chalcogenides were the first to reach the 3-5µm and later the 8-12µm range. Here we discuss how we can expand even further the available range of wavelength for colloidal nanocrystals and push their optical feature up to the THz range. A first strategy is to explore intraband transition in self-doped nanocrystals made of HgSe.[1,2] In mercury chalcogenides, doping can be obtained without introducing extrinsic impurities and actually result from a combination of large work function, narrow band gap and material non stoichiometry. As a consequence of this doping, intraband air stable transition in the mid IR appears, which pave the way for new design of infrared devices.[1] Using a combination of infrared spectroscopy, photoemission and transport measurement, we elucidate the electronic structure of these nanocrystals and bring evidence for a semiconductor to metal transition when nanoparticle size is increased.[2] In the second part of my talk, I will present a new synthetic procedure for mercury chalcogenides which allows to expand the range of size (from 5nm up to 200nm). This procedure allows for the first time to reach transition in the THz range and close to phonon absorption. Narrow energy transition around 60µm peak (and up to 200µm for cut off) will be demonstrated. 1. Lhuillier et al. Nano Lett. 2016, 16, 1282–1286 2. Martinez et al. ACS Appl. Mater. Interfaces 2017, 9, 36173–36180

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**Novel Fabrication Technique for Functionalized Two-Dimensional Semiconducting Nanoparticles Using Ultrafast Laser Pulses**

Chemical modification and functionalization of 2-dimensional (2D) materials can adversely affect the optoelectronic and electrical properties of these materials, consequently broadening their potential for a wide plethora of applications including imaging, sensing, cancer therapy, optoelectronics, display, catalysis, and energy storage and conversion [1-4]. Research discussing functionalization of 2D semiconductor materials is still in its infancy and experimental reports of successful covalent functionalization of 2D nanoparticles is scarce.

In this study, femtosecond laser pulses (λ=800 nm, τ=30 fs, rep.rate=1kHz and Ep= 2W) were used to irradiate 2D materials (i.e. MoS2, WS2, and BN) dissolved in a 1:1 water:ethanol solvent for 70 mins. The highly energetic laser pulses are sufficient to dissociate the solvent molecules into radical carbon and oxygen functional groups and hydroxyl groups. Simultaneously, the 2D nanosheets were also cleaved into nanoparticles with particle sizes of ~3nm also as a result of the laser treatment. The edges of the freshly cleaved 2D nanoparticles consequently bond with C and O functional groups (dissociated from the solvent molecules) to satisfy the periphery of the edges of the 2D material and hence successfully functionalize the 2D nanoparticle (Figure 1).

This was confirmed by analyzing high resolution TEM images (lattice spacing analysis) and XPS spectra of the fabricated 2D nanoparticles, where the characterization conclusively revealed the attachment of carbon and oxygen functional groups to the periphery of the 2D nanoparticle. The carbon atoms bonding to the 2D nanoparticle was also evident in the Raman spectra, where a characteristic carbon vibrational D+G peaks emerged after laser treatment at 1300-1600 cm⁻¹[5]. Remarkably, it was found that absorption due to carbon, which is typically present at ~260 nm [6], as well as the intrinsic absorption due to the 2D nanoparticle typically located at 600-700 nm [7], were both retained in the functionalized 2D semiconductor materials after the laser treatment (Figure 2). Consequently, the resultant nanoparticles have exhibited absorbance and photoluminescence at the intrinsic 2D, carbon, and oxygen wavelengths, making these promising materials for modern sensing and emitting applications.

This innovative and facile technique can be further developed and extended to potentially functionalize and produce different combinational materials of interest, not only for atomically thin and 2D materials, but also for a broader array of materials from other families.

It is still a massive challenge to synthesize high quality colloidal core shell quantum dots (QDs) especially in the group IV-VI semiconductor. Inconsistent particle size and sensitive towards environment changes that lead to agglomeration are the main challenges to produce good colloidal QDs. Colloidal QDs have attracted great interest due to their unique properties related to the quantum confinement effect. Here, we present our work for the ongoing study to synthesize colloidal PbS/MnS core shell QDs in aqueous solution. The stability of the colloidal PbS/MnS core shell QDs was studied over a period of time, when the samples are kept at low temperature (4o C). Basically, the colloidal PbS/MnS core shell QDs with various thicknesses are synthesized by cation exchange. By introducing Mn2+ precursor, the exchange between Pb2+ and Mn2+ on QDs surface will form MnS shell. Two different methods were used to grow the MnS shell on PbS QDs. First, the MnS shell was growth after the PbS QDs was kept for several days.
This technique allowed enough time for PbS QDs to form. The latter approach, the Mn2+ precursor was added immediately after PbS core is produced. The samples will be further studied by PL Spectroscopy and HRTEM. We expect the presence of MnS shell will improve the photoluminescence efficiency and provide photochemical stability that can avoid agglomeration, thus produce good and stable colloidal PbS/MnS core shell QDs.

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**Optical and Magnetic Properties of Self-doped Colloidal Quantum Dots with Excess Electrons**

Colloidal quantum dots have been of great interest as materials for IR light source and memory in view of their high quantum yield in photoluminescence and their semiconductor property. Steady state intraband transitions arise in nanocrystals with excess electrons in the conduction band, resulting in size-tunable absorption and emission in the mid-wavelength infrared. The steady state intraband transition is observed only when the electrons stay in the discrete electronic states of the conduction band in the absence of possible oxidation reactions that can occur in the presence of water or other oxidants. Therefore, controlling the number of electrons in the lowest energy state of the conduction band of CQDs has been extremely challenging and rarely observed. Here, we present the optical and magnetic properties of singly charged and doubly charged colloidal quantum dots. First, the mercury chalcogenide nanocrystals show the mid-IR intraband photoluminescence at 3-10 μm with the corresponding magnetic properties depending on the number of electrons in the conduction band. Second, we demonstrate that the colloidal silver chalcogenide quantum dot can be an alternative to the mercury chalcogenide quantum dot, removing concerns about the toxicity of the mercury-containing nanocrystals. Thus, the silver chalcogenide nanocrystals will be the next generation of the colloidal quantum dot exhibiting the mid-IR intraband photoluminescence.

2. Mid-Infrared Intraband Emission of Metal Excess Colloidal Ag$_2$Se Nanocrystals Mihyeon Park, Dongsun Choi, Yoonchang Choi, Hang-beum Shin and Kwang Seob Jeong Submitted

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Organometal halides are an emerging class of solution-processed semiconductors that are employed in optoelectronic devices as the active layer. In addition to their now widespread use in light harvesting applications, perovskites have shown great promise as efficient, spectrally tunable light emitting materials. Perovskite quantum dots (PQDs) have excellent charge transport properties and low non-radiative recombination rates, making them attractive alternatives to existing light emission solutions. PQDs with exceptionally high photoluminescent quantum yields (PLQY) of over 90% have been reported in literature; however, these PQDs ubiquitously contain lead, limiting their commercial potential. Replacing lead while maintaining a high performance has thus far proved challenging, as many of the desired properties of PQDs stem from the electronic structure of the lead cation. Aided by first-principle calculations from density functional theory (DFT) several direct-bandgap, energetically stable perovskites, including BaZrS$_3$ and Rb$_3$Sb$_2$I$_9$, have been synthesized in bulk; the bulk materials exhibit poor emission due to high non-radiative recombination rates and exciton dissociation. Here we present the first synthesis and development of the lead-free PQDs of BaZrS$_3$ and Rb$_3$Sb$_2$I$_9$ through a bottom-up approach. In addition, we discuss the challenges associated with these syntheses, and the potential of these materials as candidates as next-generation light-emitting materials for commercial application.

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Synthesis and Electrochemistry of Colloidal Copper and Iron Oxide Nanocrystals

Metal oxide nanoparticles based on first row transition metals are particularly attractive for photocatalytic applications because (i) they absorb visible light, (ii) they are thermodynamically capable of performing desired photoredox chemistry (e.g. proton reduction or water oxidation), and (iii) they are composed of inexpensive, earth-abundant, nontoxic elements. Compared to prototypical colloidal semiconductor materials, such as CdSe and PbS, the synthesis and chemistry of colloidal transition metal oxide nanocrystals are complicated by the fact that transition metals, such as copper and iron, have multiple accessible oxidation states. Complete synthetic control of size, shape, and crystal structure of transition metal oxide nanoparticles combined with a thorough understanding of their redox behavior is required to optimize their function in photocatalytic applications. This talk discusses recent results elucidating the role of organic ligands and solvent in controlling the size and crystal phase of iron oxide nanocrystals synthesized by a solvothermal method. Preliminary investigations into the electrochemical and spectroelectrochemical behavior of copper and iron oxide nanocrystals of various sizes will also be presented.

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Room temperature ferromagnetic behaviors of Co-doped MoS2 film

Two-dimensional (2D) layered materials have been actively studied because of their unique electronic, optical, mechanical properties that make them possible in much of potential applications since the discovery of graphene. In addition, Transition metal doped semiconductor, so called, diluted magnetic semiconductor (DMS) has been attracting much of interests for spintronics application. Recently, two dimensional DMS has been studied in graphene actively. Likewise, if transition metal can be successfully doped in layered MoS2, a two-dimensional diluted magnetic semiconductor will be realized. Then, to realize spintronic application and manipulate ferromagnetic property in MoS2 film becomes a hot issue to be resolved in two-dimensional spintronic research. In this study, we report the room temperature ferromagnetic properties of MoS2 thin film doped with cobalt by using chemical vapour deposition and sulfurization process. The Co-doped MoS2 has a ferromagnetic hysteresis at room temperature and showed two Curie temperatures. The remnant magnetization and coercive field at room temperature are 167 emu/cm$^3$ and 68 Oe.

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CdSe/CdS quantum dot doped nanocomposite polymer fiber

We report our results characterizing the optical behaviour of CdSe/CdS colloidal quantum dots embedded in the core of a polymer optical fiber, pulled from a novel nanocomposite. More specifically, a quantum dot doped polystyrene core with either PMMA or polycarbonate cladding fiber was fabricated following a dry method, where both the core and cladding are polymerized independently to ensure a step-index profile. Quantum dots from μM stock solutions or less are added to polystyrene following a dilution by a factor 102. Preforms are then drawn after core and cladding are bound together by heating in vacuum, removing any residual air gap. Characterizing the resulting fiber shows that less than 100μm diameter was achieved for the core, achieving close to single mode regime for short sections of fiber. Also, we observed that waveguiding in the dielectric fiber modifies the spontaneous emission characteristics of the embedded quantum dots compared to the bulk nanocomposite. Via time resolved photon counting, the Purcell enhancement factor from the fiber-embedded quantum dots was measured. The blinking statistics of the CdSe/CdS quantum dots, both in bulk and fiber, are recovered from the noise power spectrum to assess the effect of engineering the local density of states on blinking.

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**Phonon Interaction in Single FAPbBr₃ Quantum Dots**

Lead halide perovskites are promising materials for light emitting applications in the visible spectral range. Especially formamidinium lead halide (FAPbX₃) quantum dots provide a high photoluminescence quantum yield and an enhanced chemical stability compared to other lead halide perovskites. To fully exploit this application potential, the exciton recombination process – in particular the exciton-phonon interaction - has to be understood in detail.

In order to avoid inhomogeneous broadening effects on spectroscopic details of exciton recombination, we performed single quantum dot experiments on a statistically relevant amount of individual FAPbBr₃ quantum dots. The single quantum dots were prepared by diluting and embedding them into polymethylmethacrylate, and measured with spatially resolved micro-photoluminescence spectroscopy at various temperatures. While analyzing 32 individual quantum dots, 17 show a fine structure splitting in the meV range caused by the Rashba effect. Polarization depended measurements reveal that this splitting is comparable to the recent findings in CsPbX₃. Additionally, distinct phonon replica have been observed within energetic distances of 4.3 ± 0.5 meV, 8.6 ± 0.9 meV and 13.2 ± 1.1 meV from the zero phonon line, attributed to TO₁, TO₂ and TO₃/LO₁ vibrational modes of the lead bromide lattice, respectively. Additional replica have been found at 18.6 ± 0.3 meV and 38.8 ± 1.1 meV and can be related to the liberation modes of the formamidinium cation. Due to a marker positioning system on the sample substrate, we have been able to track single quantum dot luminescence over a large temperature range going from 4 K up to 170 K. The observed broadening of the emission linewidth with temperature can be fitted with a phonon energy of 13 meV, and thus attributed to exciton-LO-phonon interaction.

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Transforming Chalcogenide Nanoparticles to Nanoplates via Partial Cation Exchange Reactions

Cation exchange reactions (CE) emerged as a post-synthetic strategy for the fabrication of ionic nanocrystals that allows the total or partial replacement of host cations in the lattice of a pre-synthesized parent system. Even though CE has been widely investigated, it hasn’t been until recent years that comprehensive studies about the role of the main variables involved in the reactions such as the cation valency, lattice strain and ligands have been carried out. Among these variables, the volume change (ΔV) in the crystal structure between the parent NC and the final composition has been reported as an important parameter since it is related with the morphology preservation after CE. So far, the ΔV reported has not been enough to produce substantial morphological changes, so the question whether it is possible to induce a significant morphological change by stressing the anion framework with CE reactions remains unanswered. Here we investigate the effect of ΔV on the morphology of cubic berzelianite Cu$_{2-x}$Se nanoparticles (NPs) by inducing partial CE with Ge$^{4+}$ and Sb$^{3+}$. We found that in the case of Ge$^{4+}$ (ΔV 11.3%), the NPs size and shape remained basically the same, but when CE with Sb$^{3+}$ was performed, monoclinic nanoplates of around 200 nm were obtained. We observed that when Sb$^{3+}$ goes inside the lattice of the Cu$_{2-x}$Se NPs, it triggers a reorganization of the anion framework because of the high stress induced in the lattice. As this metastable NPs gain energy as Sb$^{3+}$ continues to substitute Cu$^+$ ions, the NPs start to orient and attach as a way to reduce the surface stress and energy, thus forming the nanoplates. This new approach for growing larger crystals can help to overcome the limitations of small NPs and to broaden even further the sea of possibilities to synthesize more and more complex materials.

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**Growth of InGaN dots-in-nanowire by plasma MBE**

InGaN/Al(Ga)N quantum dots (QDs) are in demand for a broad range of electronic and optoelectronic applications including visible/ultraviolet light-emitting diodes (LEDs), laser diodes (LDs), multi junction solar cells, and single photon sources for quantum computing. We have studied molecular beam epitaxy growth of self-organized InGaN/Al(Ga)N quantum dots on Si substrate. The growth parameters including the In/Ga beam flux, growth temperature/duration, nitrogen flow and plasma power were carefully optimized to ensure better radiative efficiency from the dots. In addition, structural and optical properties of the dots-in-nanowire were thoroughly studied by scanning electron microscopy (SEM), high-resolution transmission electron microscopy (HRTEM) and photoluminescence (PL) spectroscopy. It was found that nearly defect-free, self-organized InGaN/GaN QDs (diameter 70-80, thickness 5-15 nm) can be embedded in GaN nanowire. A strong, broad PL emission spectrum centered at ~555 nm was recorded from the dots-in-nanowire sample. Excitation power dependent PL measurements confirm that the emission is from quantum dots rather than defect or impurity related states.

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Synthesize InPZnS alloy quantum dot with THTP(tris(hexylthio)phosphine) as a binary anionic precursor

In recent years, Indium phosphide based quantum dots (QDs) have received a lot of attention for use many application, especially display, and the search for good optical properties has led to various reports on the testing of reaction variables. However, most reports using tris(trimethylsilyl)phosphine(TMS$_3$P)) as a anionic precursors but its easy to disrupted by moisture and air, so very unstable and difficult to handle. Then we use a new anionic precursor, tris(hexylthio)phosphine(THTP), is introduced for synthesis InP-based QDs. On account of its activated phosphine and sulfur atoms, the THTP molecule is a binary reacting anionic precursor for both InP and ZnS QDs. When THTP is reacted with indium and zinc precursors, InPZnS alloy QDs can be made. To observe the synthesis mechanism and probe the intermediate, FAB-mass and 31P-NMR analyses were conducted, resulting in the identification of an intermediate of MW 504. Finally, the surface was coated with a ZnS shell to obtain the emission wavelength from 530 nm to 570 nm and a maximum quantum efficiency of 42% when a ZnI$_2$ precursor was used.

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The Kirkendall effect: main growth mechanism for a new SnTe/PbTe/SnO2 nano-heterostructure

Attention to semiconductor nanostructures with a narrow bandgap energy and low production cost has increased in recent years, due to practical demands for use in various opto-electronics and communication devices. Colloidal nanostructures from the IV-VI semiconductors, such as lead and tin chalcogenides, seem to be the most suitable materials platform; however, their poor chemical and spectral stability has impeded practical applications. The present work explored the mechanism for formation of new nanostructures, SnTe/PbTe/SnO2, with a core/shell/shell heterostructure architecture. The preparation involved a single-step post treatment for the pre-prepared SnTe cores, which simultaneously generated two different consecutive shells. The process followed a remarkable Kirkendall effect, where Sn ions diffused to the exterior surface from a region below the surface and left a ring-like vacancy area. Then, Pb ions diffused inward and created a PbTe shell, filling the Sn-deficient region. Finally, the ejected Sn-ions at the exterior surface underwent oxidation and formed a disordered SnO2 layer. The reaction stages were followed by applying high angle annular dark field scanning transmission electron microscopy (HAADF-STEM) analysis with energy-dispersive X-ray (EDX) measurement. These intriguing processes were corroborated by a theoretical estimation of the relative diffusion length of the individual elements at the reaction temperature. The nanostructures produced were endowed with a relatively low toxicity, optical tunability, and chemical stability, which lasted more than a month at ambient conditions.


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Temperature-dependent luminescence properties of bright emitting InZnN colloidal quantum dots

III-V nitride quantum dots (QDs) offer a route to greener, heavy-metal free nanoparticles with intrinsic band gap energies tuneable from the infra-red, through the visible, into the ultraviolet by both alloying and quantum confinement effects. However, until now their utilisation has been significantly limited by highly volatile synthetic routes which typically lead to lower quality QDs and poor luminescence properties.[1-2] Realisation of a facile, solution-based synthesis technique for bright nitride QDs opens the route to studies which may prove as rich as the large volume of work performed on chalcogenide QDs over the past few decades. In this work we present optical studies of alloyed InZnN quantum dots of varying In:Zn content synthesised colloidally using sodium amide as the nitrogen precursor. This method merits tunable emission from the visible into the near infrared with quantum yields as high as 30%. This work includes presentation of the photoluminescence (PL) spectra and PL lifetimes of these QDs over a temperature range of 1.5-300 K. We identify a turning point in the temperature-varying Stokes shift, and observe the signatures of trap states in the temperature dependant PL lifetimes. We proceed to demonstrate a proof-of-principle organic light emitting diode (OLED) utilising these QDs as the active layer. These results lay the foundation for future studies of these nitride QDs which should be of particular interest for greener nanoparticle-based optoelectronics which exclude the use of heavy metals, and low-temperature phenomena in quantum confined systems.


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**Oxidation Impact on the Photoluminescence of Indium Phosphide Colloidal Quantum Dots**

Indium phosphide colloidal quantum dots is emerging as an efficient cadmium-free alternative for opto-electronic applications. Recently we have developed colloidal InP QDs syntheses based on easy-to-implement aminophosphine precursors.\(^1,2\) Here, we show by solid-state nuclear magnetic resonance spectroscopy that this new type of synthesis allows to make oxidation-free indium phosphide core or core/shell quantum dots. We reveal that the oxidation has an impact on the photoluminescence quantum yield of the obtained core/shell quantum dots, the oxidation rate seeming to be an advantage to obtain more luminous nanocrystals. This study demonstrates that the core/shell interface is a key parameter for making efficient InP-based quantum dots based luminescent devices.\(^1\) Tessier, M. D.; Dupont, D.; De Nolf, K.; De Roo, J.; Hens, Z. Chem. Mater. 2015, 27, 4893–4898.\(^2\) Tessier, M. D.; De Nolf, K.; Dupont, D.; Sinnaeve, D.; De Roo, J.; Hens, Z. J. Am. Chem. Soc. 2016, 138, 5923–5929.

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Green and narrow-band emission of Zn(Te, Se)/ZnS core/shell quantum dots

CdSe-based quantum dots (QDs) are currently applied in backlight downconversion in liquid crystal displays (LCD) because of their narrow-band emission and high quantum efficiencies. However, alternatives to CdSe-based QDs, which exhibit green and red emission, are strongly desired in order to avoid cadmium toxicity. Even though cadmium-free QD-phosphors such as InP and CuInS$_2$-based QDs have been vigorously investigated, these QDs have not achieved sufficient quality to replace CdSe with both high photoluminescence quantum yield and narrow-band emission. Recently, our group has theoretically and experimentally indicated that ZnTe QDs alloyed with ZnSe are good candidates as cadmium-free and green QD-phosphors, because the alloy system exhibits a considerable negative deviation of the energy band gap, i.e., large band gap bowing. In this study, we synthesized Zn(Te$_{1-x}$Se$_x$)/ZnS core/shell QDs with various composition x and sizes, and the formation of ZnS shell on Zn(Te,Se) core was safely confirmed. In the case of Zn(Te$_{0.77}$Se$_{0.23}$)/ZnS core/shell QDs with 4.3 nm of core diameter, we successfully demonstrated green photoluminescence emission at 535 nm and narrow emission-band with 30 nm of the full-width at half maximum. These results indicate that Zn(Te$_{1-x}$Se$_x$)/ZnS core/shell QDs have great potential as cadmium-free and green phosphors suitable to next-generation displays.

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ZnSe/ZnS Core/Shell Quantum Dots with Optimal Photoluminescence Properties for Blue LEDs

Traditional organic-based blue light-emitting diodes (LEDs) suffer from short lifespans in high-flux displays. Alternative robust fluorophores with high color purity are necessary for next generation high quality displays. Colloidal quantum dots (QDs) are unique candidates for LED applications due to their tunable emission, high photoluminescence quantum yield (QY), narrow photoluminescence emission linewidth, and high stability. However, QDs with a visible bandgap often contain toxic materials such as cadmium. With a bandgap of ~2.7 eV or ~460 nm, zinc selenide (ZnSe) is a promising material for LED applications in the ultraviolet to blue wavelength range. Here, we report the synthesis and characterization of zinc selenide/zinc sulfide (ZnSe/ZnS) core/shell quantum dots with tunable emission from 380 to 440 nm, solution QY up to 92%, and extremely narrow ensemble solution fluorescence linewidths reaching ~70 meV or ~10 nm. In addition, the solution Photon-Correlation Fourier Spectroscopy (s-PCFS) shows that the average spectral linewidth of the single nanocrystal matches the ensemble linewidth, indicating a highly uniform population of nanocrystals. These results suggest ZnSe/ZnS is a promising material for cadmium-free QDLED applications.

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Aqueous Based Reflux Synthesis of W-doped VO2 Delivering PVP-Coating with an Excellent Optical Property

Vanadium dioxide (VO2) responds thermochromically to environmental temperature changes by changing its crystal structure as a function of temperature. VO2 is transparent to infrared radiation (IR) when its temperature is below a so-called phase transition temperature (Tc), but IR light reflective above Tc, while retaining visible light transmittance. Such intrinsic property makes VO2 an attractive material for "smart" thermochromic coatings to efficiently utilize solar energy. However, to date, one of the major challenges that still limit the use of VO2(M/R) system for smart windows is to simultaneously and significantly improve both of solar modulation efficiency (ΔTsol) and luminous transmittance (Tlum) through an economical process. In this work we examined the synthesis of novel VO2(M) by using a low toxicity vanadium precursor, which was reduced with an electron donor, using a water-based approach. This material was shown stable against oxidation with simultaneous improvement of both Tlum (68.30 % at 22 °C) and ΔTsol (20.37 %) in a PVP coating.

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Study of transition metal (Fe, Co) ion doping in TiO2 Nano particles

In this research, we have studied the doping behaviors of two transition metal ion dopants on the crystal phase, particle sizes, XRD patterns, EDAX spectra, and photoreactivities of TiO2 nanoparticles. The crystalline size of TiO2 is close to 4 nm Calculated from (1 0 1) peak by using FWHM method in Scherrer’s equation. Test metal ion concentrations ranged from 1% to 4 at.%, we report the growth of [Fe, Co]xTiO2 nanocrystal prepared by Sol-Gel technique, followed by freeze-drying treatment at -30°C temperature for 12hrs. The obtained Gel was thermally treated at different temperature like 200°C, 400°C, 600°C, 800°C. The photoreactivities of transition metal ion-doped TiO2 nanoparticles under UV irradiation were quantified by the degradation of formaldehyde. Magnetic and Electrical Properties has been also carried out for application in field of Spintronics.


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Emission properties of single quantum dots in perovskite matrix.

Application-critical optical properties of single quantum dots, for example their emission wavelength and their fluorescence decay rate, can be influenced by incorporation in a suitable matrix. Hybrid lead halide perovskites are promising candidates for such matrices with respect to charge control and reduction of type-A blinking of chalcogenide quantum dots. Methylammonium lead bromide perovskite (MAPbBr$_3$) in particular not only exhibits a favorable and tunable band alignment, but furthermore a lattice parameter that is compatible with the formation of a crystalline matrix around the QD. We have established an experimental protocol for producing thin films of a hybrid QDs-MAPbBr$_3$ perovskite in which the QD concentration is adjustable. To this end, we first fabricated highly luminescent core-shell QDs with a CdSe/Cd$_x$Zn$_{1-x}$S gradient shell that are sufficiently resilient to the deleterious effects of mobile lead cations in the perovskite matrix. The second crucial step was the exchange of the organic ligands of the QDs with bromide ions under preservation of colloidal stability so that all precursors of the film can be mixed in solution without the presence of long carbon chains that would interfere with subsequent crystal formation. Finally, a method had to be identified that allowed to prepare compact and homogeneous perovskite films to ensure the successful embedding of the QDs. For this purpose, we found that thin-film creation by chlorobenzene injection showed the best results. The emission of single QDs could be detected in MAPbBr$_3$ perovskite films at low degree of doping. A red shift of the emission, monoexponential fluorescence decays and fluorescence antibunching could be observed, all of which indicates that the quantum efficiency of the QDs was largely preserved during the encapsulation process. These samples thus constitute a starting point for investigating and, possibly, controlling the blinking behavior of single nanocrystals in a semiconducting matrix with favorable band structure.


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Light-Induced Interface Energetics’ Modification at the Semiconducting Polymer and Metal-oxide Heterojunction in a Photodiode

With the increasing demand for alternative clean energy resources, and for flexible and cheaper electronics, organic-inorganic hybrid materials seem to show the potential to patch up the zone. In hybrid devices as these, the chemical interactions at the organic-inorganic heterojunction play critical role in determining the device performances. Depending on the conditions under which the heterojunctions are formed, the current injection/extraction or photogenerated exciton dissociation at the these interfaces could be severely affected. The unpredictable behaviour is attributed to unexpected surface states, adsorbants-induced defect states etc. In this work, we probe the interface energetic changes by light-soaking the photodiodes that are based of ZnO/P3HT and ZnO/PCDTBT as the active media in a bilayered configuration.

The steady-state current-voltage-electroluminescence, photocurrent/voltage characteristics, charge-extraction by linearly increasing voltage (CELIV), dark-injection current transients were monitored before and after the light-soaks in various spectral ranges. One of the stark differences noticed was the abrupt increase in the electroluminescence efficiency after soaking the device with UV portion of the light. The observed effects coherently indicate that there is a probable photodesorption of oxidizing groups such as O2, OH− etc. from the nanostructured ZnO surface, and therefore tuning the fermi-level of the metal oxide upon UV-light exposure.

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Selective Growth of Au onto ZnSe Nanorod Couples for photocatalysis

Noble metal-semiconductor hybrid nanocrystals are promising photocatalytic materials for solar energy harvesting. Compared with zero-dimensional (0D) quantum dots, one-dimensional (1D) semiconductor nanocrystals manifest enriched geometry and improved charge separation and transport capability. As an important semiconductor material, ZnSe, with a bulk band gap of 2.70 eV for wurtzite structure, is a good candidate as photocatalyst towards hydrogen generation in splitting water. Here we report the anisotropic selective deposition and growth of Au onto 1D ZnSe nanorod couples. The deposition location of Au tips can be controlled by the original concentration of the starting Au precursor and growth temperature. By sequential addition of Au precursor, the size of Au tips can be controlled. The Au-ZnSe nanorods couple hybrid nanostructures display morphology, location and size-dependent hydrogen production efficiencies under UV light irradiation.

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Colloidal Quantum Dot with Stabilized Inorganic Perovskite Matrix

For energy conversion applications, all-inorganic perovskite phases could be more stable than those containing organic cations. The cubic structure of cesium lead halide perovskites, having a suitable electrical conductivity, suffer from degradation under room temperature. Colloidal quantum dots (CQDs), emerging solution-processed materials for large-scale and low-cost photovoltaics, have just reached 12% certified photovoltaic performance. However, the closest-packed CQDs still have more than 30% voids, largely reduced the carrier transport mobility. Here we show a mixed absorber combining inorganic perovskites with lattice-matched colloidal quantum dots presents improved optical and thermal stability under room temperature. The perovskite matrix reduces the energy barrier for carrier hopping in quantum dot solids and increases the carrier diffusion length of quantum dot films. This contributes to a perovskite-shelled CQD solar cell with an efficiency over 12%, promising for light harvesting or for photodetectors.


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The Integration of Non-Heavy Metal Containing Quantum Dots into Polymer Films

Quantum dot nanoparticles have been intensely studied in the scientific community in the past decade due to a wide range of unique properties caused by the quantum confinement effect.

While the most common QDs in the literature incorporate toxic metals such as Cd or Pb, this project aims to synthesize QDs from earth-abundant friendly materials such as Zn, Si or Ti. To date we have synthesized novel families of ZnO, ZnS, TiO2, and Si Quantum dots at a laboratory scale. To this end the current aim of this project will be to find a sustainable green pathway for large scale quantum dot synthesis and a repeatable and feasible integration of quantum dots into polymer nanocomposites.

Synthesized quantum dots have been characterized via XRD and the changes to the physicochemical properties based on size, capping agent and synthetic methodology have been examined using optical spectroscopy. These dots have been melt mixed into ethylene-vinyl acetate and urethane polymer films and have been explored as possible encapsulants for photovoltaic (PV) technologies due to the down regulation of UV to Visible light potentially increasing efficiency. Furthermore the photo stability and large stokes shift of these nanoparticles allows for potential green house films (ZnO), Bioimaging (Si), and as antimicrobial polymers (TiO2)

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Combination of multiple functional molecules is of great importance to create materials with desired and new functions. Among functional molecules, colloidal semiconductor nanocrystals, quantum dots (QDs), with several nano diameter are attracted as a molecular-like smart nanomaterial. This is because the single QDs have the outstanding emission and photoelectronic properties and the emission wavelength can be tuned synthetically by changing the QD size. Since the synthetic methods to obtain QDs with uniform diameter were developed, the optical properties of single QD have been revealed in detail. Because the device performance can depend on the resulting non-covalently connected supramolecular structure of QDs in bulk state, understanding and controlling the self-assembly of QDs is important for construction of highly organized materials. Here we synthesized CdSe/ZnS core-shell QD with 3 nm in diameter and perylene bisimide (PBI) dye possessing thiol groups, and then attempted a coaggregation of QD and PBI. Photoluminescence spectra in diluted CHCl₃ solution exhibited a decrease in intensity over time upon mixing the QD and PBI, indicating the connection of PBI into QD surface. Interestingly, the absorption spectra showed a gradual decrease in intensity and the new band was emerged at longer wavelength regime, which is characteristic of a π-π staking between PBI chromophores. This result demonstrates the formation of PBI aggregates on QD surface despite non-aggregation condition of sole PBI molecule. Notably, upon changing the mixing condition of QD and PBI, different supramolecular structures were obtained. In this presentation, we will discuss the mechanism of QD-PBI coaggregation.

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Photoswitching Emission Behavior of a Single Quantum Dot Using Photochromic Reaction of Diarylethene

Diarylethene (DAE) is known as photoresponsive molecule indicating the photochromic reaction that is reversible transformation between open- and closed-ring isomers which have different absorption spectra. Fluorescence intensity of luminescent molecules can be controlled by the combination of DAE molecule because luminescent molecules work as a fluorescence donor in Förster energy resonance transfer (FRET) and the closed-ring DAE molecule, formed by ultraviolet (UV) light irradiation, work as an acceptor. This DAE-luminescent molecule system is applied to super-resolution microscopy and ultrahigh-density single optical memory. In previous works, fluorescent photoswitching molecules consisting of DAE and organic fluorescent molecules were synthesized, and the switching behavior was observed at single molecule level as well as at ensemble level in solution. Recently, semiconductor quantum dots (QDs) showing broad excitation, narrow emission, photostability and brightness are used as the fluorescent unit and the photoswitching depending on the photochromic reaction of DAE has been confirmed. However, observation at single molecule level was not reported so far. In this work, we prepared the sample in which QDs and DAE molecules are dispersed in polymethyl methacrylate (PMMA) thin film, and monitored the change in emission behavior of single QDs with the irradiation of UV and visible light. As a result, the emission intensity decreased and the lifetime was shortened upon the UV light irradiation. Irradiation with the visible light resulted in a recovery of the emission intensity and lifetime. The decrease in the emission intensity and lifetime was caused by the energy transfer from a QD to a closed-ring DAE.

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Band Occupation and Charge Transport in PbSe & CdSe Nanocrystal Superlattices

Colloidal semiconductor nanocrystals have gained interest since their optical and electronic properties can be tuned by varying their shape, size and composition. Recently, 2D square and honeycomb superlattice of lead- and cadmium-chalcogenide QDs have been prepared at the toluene air/interface after which the nanocrystals attach via their four vertical {100} facets\textsuperscript{1,2}. Afterward, cation exchange transforms PbSe into zinc blend CdSe. Theoretical studies show that these 2-D systems have distinct band structures compared to continuous nanosheets, with the appearance of Dirac cones in the case of the honeycomb\textsuperscript{3,4}. Strong electronic coupling via the atomic connections of the QDs in the superstructure may result in a higher mobility compared to the self-assembled QDs\textsuperscript{5}. In our research, we use electrolyte-gated transistors to study the optoelectronic properties of 2-D PbSe and CdSe superstructures\textsuperscript{6}. The gate electrode potential determines the Fermi level with respect to the conduction band (valence band). To monitor the stability of the superlattice under electron injection we measure the differential capacitance as a function of gate voltage. From the total injected charge and the number of nanocrystal sites in the gated part of the superlattice, we calculate the charge density of the superstructure. The conductivity of the network is measured as a function of the Fermi level position. The mobility of the system is calculated from conductivity and charge density. As an alternative method to quantify electron injection and band occupation into the superlattice, the optical absorption quenching employed. We studied the band occupation of PbSe and CdSe superstructures. In our recent work, we report the first study of electron transport in a 2-D PbSe system with a square geometry in which band occupation is assured by the high electron density of 8 electrons per nanocrystal site. The electron mobility between 5 and 18 cm\textsuperscript{2}/Vs is observed for these supersructures\textsuperscript{7}.


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On-Demand Control of Colloidal Quantum Dot Solids Thin-Film Formations and Their Electronic Transport Properties

Colloidal quantum dot solids render the flexibility and low cost of solution-processed materials; and the unique presence of the quantum confinement and the large surface-to-volume ratio due to the small size of the constituent individual quantum dot. These systems are attracting for broad range applications to exploits those merits, which are not only limited for the optical and optoelectronic devices (e.g. photovoltaic, light-emitters, photodetectors), but also in many other potential energy harvesting and energy storage devices. Every different devices require different types of material assemblies based on which properties of the QDs that are intended to be exploited. Assemblies for electronic transport (e.g. transistors, photodetectors, etc.) would be different from assemblies required for the application as battery electrodes, or water-splitting electrodes. Therefore, greater control on the formation of the QD thin-film and the optimization of the related charge carrier transport characteristics is vital to further advance the exploitation of this material system.

Here we demonstrate on-demand control of thin-film formation of highly crosslinked colloidal QD solids through the optimization of different deposition methods, which are: layer-by-layer spin coating, layer-by-layer (low-speed and high-speed) dip-coating, and liquid/air interface assemblies. Through optimization of deposition parameters, we are able to obtain a broad control of the film formation, from the large area superlattices for electronic device applications until to controllable holey structure that would be beneficial for battery and super-capacitor electrodes applications. The charge carrier transport on the films and the carrier accumulation capacities of these assemblies are thoroughly characterized by electric-double layer transistors and capacitors using ionic liquids. Depending on their arrangement of the assemblies, the quantum confinement nature of the PbS QDs strongly affects the capacitance and the charge carrier transport, which enable us to broaden the applications of colloidal QD assemblies for electronic and energy harvesting devices.


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Insights into the Role of Zinc–Phosphorus Complex as an Atomic Valve towards Monodisperse Colloidal Indium Phosphide Quantum Dots

Growth of monodisperse InP quantum dots (QDs) is pressingly required in display applications, as the size uniformity is directly converted to color purity in display products. Herein, we report the colloidal synthesis of InP QDs in the presence of Zn carboxylates, where size uniformity is markedly enhanced as compared to the case of InP QDs synthesized without Zn carboxylates. We monitor the appearance of metal-phosphorus complex intermediates in the growth of InP QDs using nuclear magnetic resonance spectroscopy and mass spectrometry analyses on aliquots taken during the synthesis. In the presence of zinc carboxylate, intermediate species consisting of Zn-P bonding appears. The Zn-P intermediate complex with P(SiMe3)3 shows lower reactivity than In-P complex, which is confirmed by our prediction based on density functional theory and electrostatic potential charge analysis. The formation of stable Zn-P intermediate complex results in lower reactivity, therefore monodisperse QDs. Insights from the experimental and theoretical studies would advance the mechanistic understanding and controlling of nucleation and growth of InP QDs, which are prerequisites to prepare monodisperse InP-based QDs in meeting the demand of display market.

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Tuning of sp-d Exchange Interactions in Wave Function Engineered Colloidal CdSe/(MnS)/Mn: CdS Hetero-Nanoplatelets

Recent improvements in synthesis techniques including the colloidal atomic layer deposition (c-ALD) have allowed for the synthesis of two-dimensional (2D) hetero-nanoplatelets composed of a variety of semiconducting materials, with a monolayer-by-monolayer precision in the layer thickness. The combination of different architectures and materials in a core-shell geometry now opens up the extraordinary possibility of precisely tuning the band alignment and thus the carrier wave function distribution in these colloidal quantum wells, creating tunable type I, quasi-type II and type II heterojunctions. Additionally, c-ALD allows the selective incorporation of transition metal dopants, such as manganese, into the host material adding magnetic functionality due to the sp-d exchange interactions between the magnetic moments of the dopant d-orbitals with the spins of the s-type electrons (s-d) and p-type holes (p-d). In this work, we use the absorption based technique of magnetic circular dichroism (MCD) spectroscopy to unambiguously prove the sp-d exchange interactions between the magnetic dopants and the host material carriers in the working model of shell-doped core-shell hetero-nanoplatelets. Varying the architecture of CdSe/Mn:CdS and CdSe/MnS/Mn:CdS quasi-type II hetero-nanoplatelets, on the one hand, allows to separately tune the p-d as well as the s-d exchange interactions\textsuperscript{1}. On the other hand, MCD not only enables access to the lowest exciton resonance, but also allows the observation of a variety of higher excitonic transitions. In combination with envelope wave function calculations, we are able to identify the contribution of different hole states to excited state transitions\textsuperscript{2}. Thus, this study not only demonstrates the possibility of separately tuning the s-d and p-d exchange interactions by a careful design of the nanoplatelet heterostructure, but also gives new insights into the excited state transitions in these novel 2D materials, which have not been investigated before. \textsuperscript{1}F. Muckel, S. Delikanli, P.L. Hernández-Martinez et al., Nano Letters, 2018, accepted (DOI:10.1021/acs.nanolett.8b00060).

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Whispering gallery mode lasing in supraparticles of luminescent nanocrystals

Since the first demonstration of amplified spontaneous emission and optical gain in systems composed of semiconductor nanocrystals (NCs) [1], these materials have attracted rising interest in the scientific community due to their unique opto-electronic properties. In particular, the tunable photoluminescent (PL) emission and, usually, high quantum yields are two of the main features promoting NCs as potential candidates for lasing applications. However, the large rate of non-radiative recombination through Auger processes is one of the main drawbacks of these systems, still preventing their extensive use in widespread applications. In this quest to realize efficient NCs based lasers, NCs have been implemented in a large variety of systems and cavities. Ordered assemblies of NCs, also known as supraparticles (SPs) [2], have recently emerged in the scientific field for their collective properties which differ from those of the composing NCs. SPs have been shown shape-dependent modulation of their PL emission originating from whispering gallery modes (WGMs) [3], thus opening the doors to their use as lasing cavities. Here we report, for the first time, on WGM lasing from SPs composed of luminescent NCs. We observe the appearance of lasing peaks on top of the WGMs (Q~1000), which then evolve into mode competition for higher pump pulse intensities. The WGM origin of the lasing is supported by coherence measurements, which show spatial coherence extending over the entire rim of the SPs. The SPs act in this case both as gain medium and lasing cavities. [1] V.I. Klimov et al., Science, 2000, 290, 5490, pp. 314-317, [2] de Nijs et al., Nat. Mat., 2015, 14, pp 56-60 [3] Vanmaekelbergh et al., ACS Nano, 2015, 9 (4), pp. 3942–3950.

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Charge Carrier Dynamics in Halide Perovskite Nanoplatelets

Halide perovskite nanoplatelets (NPls) show strong excitonic properties together with high photoluminescence quantum yields even at room temperature. A new solution-based synthesis route enables thickness control from bulk-like NPls down to a single monolayer [1]. This yields perovskite NPls with 3D or 2D character. With this reduction of dimensionality, the exciton binding energy increases mainly due to reduced Coulomb screening leading to higher exciton recombination rates. By performing transient absorption spectroscopy we have studied the relaxation scenarios of photogenerated charge carriers for the 2D and 3D case. We observe an intensity dependent cooling rate only in 3D, whereas in 2D a fast cooling takes place at all studied excitation densities. In addition, we have performed four wave mixing experiments to determine exciton dephasing times, i.e. the homogeneous linewidths of the 1s-exciton for different conditions. We compare these results to those obtained by transient absorption spectroscopy. We conclude that both reduced Coulomb screening and an increased surface to volume ratio drastically change the photogenerated dynamics of bound and unbound electron-hole pairs in thin halide perovskite NPls.


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**Noble metal seeded iron sulfide nanoplatelets**

Hybrid nanomaterials provide a liberty to not only achieve the unachievable complex heterostructures with controlled functionalities but also precisely define the materials with predictable linkages owing to the interfacial electron interactions. Rational and sequential integration of multiple domains at nanoscale can lead to the fabrication of materials with functionalities which are not attainable in separate materials. Here, we report the synthesis of noble metal-hexagonal iron sulfide nanoplatelets (NPLs) by means of simple heterogeneous seeded nucleation. Using thiol-capped noble metals such as Au and Ag, chemoselective nucleation was promoted in the presence of iron precursor while suppressing the homogeneous nucleation. These novel NPLs with a seed embedded inside exhibited strong plasmonic and magnetic response as well. For advance mechanistic understanding, the experimental conditions were tuned for the formation of well-defined hexagonal hybrid nanoplatelets which directed us towards the critical role of ligands attached on the surface of the seed. However, to generalize the mechanism, same approach was adapted for other noble metal seeds. Additionally, electrochemical testing was done to compare aforementioned heterodimer NPLs with iron sulfide and also to highlight the synergistic effect between two constituent materials. This work will provide a platform for the development of complex but sophisticated and high-quality heterostructures prone to manipulation for potential applications.

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Self-assembled semiconductor quantum dots (QDs) embedded in microcavities emitting at optical fiber communication wavelengths, particularly in the telecom C-band, which offers the lowest attenuation losses in silica fibers is highly interesting for quantum communication applications. Here, we report our recent efforts in molecular beam epitaxy growth of low density InAs/InP QDs, fabrication of InP-based microcavities emitting at telecom wavelengths, and their optical properties. Low QD density is obtained by careful control of the QD growth using a special capping technique and temperature processing after the dot formation. These QDs proved to have a nearly perfect single-photon emission and good coherence properties. The cavity processing is realized by electron beam lithography and different etching steps. The influence of geometrical parameters on the quality factor enhancement, light out-coupling and the mode profiles of InP-based photonic crystal (PhC) cavities are investigated. Suitable PhC are designed using finite difference time domain (FDTD) simulations. Low-temperature single-dot spectroscopy exhibits sharp excitonic emission lines from single QDs with very small fine-structure splittings. Signatures of the existence of exciton-biexciton states of a single QD are clearly identified. Sharp cavity modes with a weak coupling behaviour and relatively high quality factors were measured.

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Atomic scale dangling bond quantum dots on silicon surface

There is currently interest in extending silicon based microelectronics to quantum technologies, including silicon nanocrystals, gated quantum dots and dopants [1-3]. Recently, Wolkow and collaborators demonstrated atomic scale Quantum Dots created by removing a hydrogen atom from a hydrogen passivated Si(100)-(2x1) surface [4,5]. The removal of a Hydrogen atom creates a dangling bond (DB) of a Silicon atom with corresponding energy in the gap of bulk Si.

We present a new computational tool, Silicon-QNANO [6], to perform electronic structure calculations of million atom silicon nanostructures and apply it to DB qdots on a H-Si(100)-(2x1) surface. The surface of a silicon slab was reconstructed by using DFT as implemented in ABINIT program [7]. The top layer was reconstructed by the formation of Si dimers passivated with H atoms while the bottom layer remained unreconstructed and fully saturated with H atoms to simulate a seamless transition to bulk material. The slab Hamiltonian was approximated by a tight-binding (tb) Hamiltonian by expanding the electron wave functions as a LCAO. Using a genetic algorithm, we computed a set of tb parameters that reproduces the Si slab, including the reconstructed surface, band structure calculated with DFT.

The parameterized tb Hamiltonian was used to model large finite Si(100) slabs with reconstructed surface passivated with H atoms. We report calculations for systems with total number of Si atoms ranging from 32 (minimal slab) to hundred thousand. The accuracy of Si-QNANO was benchmarked w.r.t. analogous DFT calculations for systems with up to 600 atoms using the OCTOPUS code [8]. We next removed one hydrogen atom from the reconstructed surface and showed the emergence of an energy level in the gap with wave function strongly localized around the Si atom with a DB and showed the charging of a DB qdot with up to two electrons.


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Coherent single photon emission with suppressed inhomogeneous linewidth broadening from a site-controlled III-Nitride quantum dot

The III-Nitride semiconductor quantum dots (QDs) have received much attention for quantum information processes due to the ability to generate non-classical light with near unity polarizability at an elevated temperature.[1-5] However, the presence of large built-in electric field weakens not only the radiative recombination process, but also cause inhomogeneous linewidth broadening via fast and slow spectral diffusion. This linewidth broadening further could be magnified by defect density and interaction of permanent dipole moment with electrical field fluctuations nearby QD. Consequently, this randomizes the coherence property of III-nitride QD based single photon. Here, we demonstrate coherent single photon emission with suppressed inhomogeneous linewidth broadening from a site-controlled InGaN QD in a GaN nano-pyramid structure. The GaN nano-pyramid structures were grown on hole patterned GaN/sapphire substrate using metal-organic chemical vapor deposition. And then, we regrew InGaN/GaN layer on the GaN nano-pyramid structure. The single InGaN QD was successfully formed at the apex of nano-pyramid structure. Using high-resolution transmittance electron microscopy, the QD height was estimated to be < 2 nm, which could significantly reduce permanent dipole moment within QD. In addition, we observed superb crystal quality of InGaN/GaN interface layer, which indicates the reduction of defect density. Through the excitation power dependent photoluminescence, no measurable screening effect of single photon emission was observed beyond the saturation excitation power, which confirm the negligible quantum-confined Stark effect. Also, we investigate the variation in emission wavelength in time, where no measurable jittering with slow spectral diffusion can be seen. By suppressing these inhomogeneous linewidth broadening, we estimated the homogeneous linewidth to be 64 ± 8 μeV (corresponding time-scale: 20.5 ± 3.2 ps), which is the-state-of-the-art among InGaN single quantum emitters. This work presents improved coherent property of III-Nitride QD based on nano-pyramid structure and viable building blocks for solid-sate quantum information process.

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Control of ripening and size of PbS quantum dots with diglyme

PbS quantum dots effective emitters in the short-wave infrared, and are the base of the leading efforts in quantum-dot photovoltaic devices [1]. However, though size-uniformity is vital for many optoelectronic applications, the reproduceable synthesis of monodisperse PbS quantum dots at scale remains challenging. Recently, Hendricks et al. developed an elegant synthetic procedure involving substituted thioureas as the sulfur precursor [2]. The substituents tuned the reactivity of the precursor, resulting in size control via classic nucleation and growth kinetics. In a later publication, where substituted selenoureas were similarly employed, the claim was made that Ostwald ripening—which causes undesirable size-polymdispersity—was also suppressed [3]. This is in sharp contrast with widely-employed synthetic methods using TMS-S as the sulfur precursor [4], where the reaction must be rapidly quenched to suppress ripening and preserve monodispersity. Here, we identify the reason for these two different broadening behaviors, and show that it is lead complexation by the diglyme co-solvent that prevents rapid ripening, as opposed to the nature of the sulfur precursor. We also demonstrate that diglyme can be used to control nanocrystal size independently of temperature. Taken together, this method affords a highly monodisperse and reproducible synthesis of a range of PbS quantum dot sizes using a single, readily-available sulfur precursor.


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Synthesis and characterization of dual emitting white light NCs

The semiconductor nanocrystals (NCs) have been extensively studied for industrial applications. Light generation is one of the key areas of research, where ability to control light for useful applications in display devices, solid state lighting, and other light sources is expanding broadly. Colloidal semiconductor NCs are inexpensive compared to NCs grown with other techniques such as MBE, also colloidal synthesis allows for NC properties to be altered through ligand functional group modification. The ability to fabricate devices using colloidal NCs is also a mature field, but there still remains many challenges in characterization in order to study the structural aspects of small NCs. Physical characterizing of these small NCs is tough as the contrast in TEM is weak and line broadening in XRD studies is severe. It is not only hard to describe the surface of NCs without proper characterization but it also leads to irreproducibility with every slight variation surface chemistry and ligands. Sample preparation for every characterization techniques is the key to acquire better data. Here we show control over synthesis of very small NCs ranging from 1-2nm radius. We present characterization data for there ultra-small dual emitting NCs that emit white light. We address problems associated sample preparation to acquire better characterization data.

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Highly Stable Quantum Dot Films by Covalent Bonding of Designed Ligands and Polymers

Since quantum dots (QD) have been disclosed their unique semi-conductive properties, the studies for their applications have been accelerated. Photoluminescence (PL) of QDs was applied to display application by embedding QDs into polymer films. Although QD films have showed impressive color quality originated from narrow band emission in display applications, they still have issues of the efficiency reduction by energy transfers, low compatibility with polymer films and low stability against oxygen and moisture. The stability issue makes additional barrier layers necessary for protecting QDs and limits QD applications. In this study, we improved the stability of the QD film employing the newly designed ligand of QDs. The QD film was fabricated by covalent bonding among the reactive ligands and polymers via hydro-silylation. The reactive ligands of QDs were prepared by ligand exchange and an additional reaction at the end of the exchanged ligand. The reactive QD was not damaged during the ligand exchange and the additional reaction as well as the reactive ligand of the QD was characterized its structure and exact substituted ratio on the surface of the QD by FT-IR and $^1$H-NMR. The stability of QD films was evaluated by storage at the highly oxidative condition, 85°C/85% relative humidity (RH) and observed periodically the change of the optical property during storage in the harsh condition for 1 month. The novel QD and siloxane system showed 122% higher stability and better dispersity in the matrix than the QD embedded in a normal linear polymer such as poly(methylmethacrylate) caused by the alternate reaction sequences and regular distances among the QDs via the hydro-silylation.

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Colloidal Nanocrystal Heterostructures With Tunable Dual-Band Emission

Colloidal semiconductor nanocrystals displaying dual-band emission are of great interest due to complex multi-carrier dynamics in nanostructures having multiple emissive states, which have potential applications in white light generation, multimodal imaging, ratiometric sensing, and quantum light source. Here, we report a new dual-emitting nanostructure whose emission spectrum can be widely and continuously tailored by using a post-growth photoetching process. We synthesized a core/barrier/multi-branch nanocrystal heterostructure (NCH), that is a dual quantum system composed of a single quantum dot core and multiple quantum rod branches. A wide-bandgap barrier that electronically separates the two quantum systems allowed both core and branches to be brightly emissive (total QY up to ~40%) under ambient excitation fluence (1–10 nW/cm²). More importantly, the dual-band spectra were continuously tuned by controlling the branch emission by applying photoetching. The quantum size-selective photoetching enabled precise tuning of the branch emission in a broad visible range (500–585 nm) as well as significantly narrowing the bandwidth (FWHM ~20 nm). Time-resolved fluorescence decay and photoluminescence excitation spectroscopy studies respectively confirmed that the dual emissions were from the two distinct quantum systems (dot and rod) and significant non-radiative energy transfer was occurring from the wider-bandgap branches to the narrower-bandgap core. Our work suggests a novel and versatile strategy of designing dual-emitting nanocrystals with wide and precise spectral tuning by controlled photoetching.

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From Clusters to Nanocrystals: A Novel Growth Mode of Quantum Dots via Burst Dissolution of Magic-Sized Intermediates and Secondary Nucleation

Typically, the formation of colloidal semiconductor nanocrystals (NCs), or quantum dots (QDs), is described by applying classical nucleation theory (CNT) to their nucleation and growth. In this model, nucleation via aggregation of individual monomer units and subsequent continuous growth of nuclei by incorporation of monomer is assumed, resulting in a smooth free energy curve of nucleation. Various reports of so-called “magic-sized” clusters (MSCs), i.e. energetically highly stable clusters of a specific aggregation number, show the necessity to modify the CNT model, as the high stability of MSCs indicates their existence at local minima along the free energy curve. For these MSC intermediates, a sequential growth mechanism from one MSC size to the next has been reported, highlighting their extraordinary stability. In order to gain insight into the MSCs’ exact role during QD formation, which is currently not conclusively understood, we have employed our state-of-the-art continuous-flow device (CFD) to conduct in situ time-resolved investigations of the early stages of colloidal CdSe QD synthesis using on-line UV/Vis absorption spectroscopy and synchrotron-based small-angle X-ray scattering (SAXS). In our contribution, we show clear evidence of sequential growth of CdSe MSC intermediates during hot-injection-style synthesis at high temperature, which was previously reported only far below typical reaction temperatures due to the transience of the intermediates at reaction conditions. Moreover, we present a novel growth mode for CdSe MSCs via burst dissolution of smaller MSCs and subsequent secondary nucleation which was identified using our CFD. This growth mechanism, which exists beside traditional focussing and ripening growth, is highly relevant as it strongly affects final QD size distributions. While it intrinsically leads to significant broadening, it also possesses the potential to yield highly monodisperse QDs under the right reaction conditions.

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**Novel Techniques for Highly Stable and Luminescent Halide Perovskite Quantum Dots**

Colloidal lead halide perovskite quantum dots (QDs) currently have emerged as a new material with high photoluminescence and color purity which make them an ideal material, especially for display and light-emitting-diodes (LED) application. However, lead halide perovskite materials easily undergo degradation in the presence of heat, humidity and air. Therefore, it is required to provide protection on the lead halide perovskite QDs to obtain high stability for further use in the devices. Herein, we will discuss two novel techniques to obtain highly stable and luminescent lead halide perovskite QDs. The first technique is by using anti-solvent method at room temperature to synthesize 3D/0D (CsPbBr$_3$/Cs$_4$PbBr$_6$) composite. The obtained 3D/0D composite consist of 3D perovskite crystals embedded inside 0D matrix. Due to the higher stability of 0D matrix, the obtained composite has a higher stability compared to pristine CsPbBr$_3$ nanocrystals. The second technique is by embedding lead halide perovskite quantum dots (QDs) inside silica and alumina matrix. Silica and alumina layer provides double protection of perovskite nanocrystals from heat, humidity and air. Photodegradation, thermal behavior and temperature stability of the perovskite QDs will be discussed in more details. We believe that improving stability of perovskite QDs is the high priority to bring perovskite QDs into the real world applications.

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Cation exchange reactions: From Cu(2-x)S to CuInS2 nanorods

Properties of nanoparticles differ greatly, depending on their size, shape and crystal phase. Direct synthetic approaches are already available to obtain nanoparticles of various compositions, crystallographic phases and morphologies. However, postsynthetic modifications often offer a higher degree of possible adjustments for the desired nanomaterials. Furthermore, not all crystal phases and compositions are readily available to be synthesized in the desired morphologies. Thus, cation exchange (CE) reactions emerged as a way to obtain finely tunable compositions of complex nanomaterials. CE reactions are characterized by the substitution of cations in a host lattice by guest ions from the reaction solution, while the anionic sublattice is preserved and thus the initial morphology of the nanoparticles can be retained. The CE from Cu$_{2-x}$S to CuInS$_2$ in nanorods (NRs) offers the opportunity to investigate a reaction from a binary to a ternary system and study the underlying mechanism of such CE reactions, as this has not previously been reported for rod-like morphologies. We were successful in obtaining partially and fully exchanged NRs under shape retention at temperatures between 100 °C and 150 °C. The compositional changes were analyzed during the CE reaction with energy-dispersive X-ray spectroscopy (EDX), which was also used for kinetic analysis of the process. Moreover, X-ray diffraction (XRD) was employed to study the emergence of newly formed crystal phases and the undergoing change of the crystal lattice. Furthermore, the local environmental changes around Cu and In atoms could be probed with a synchrotron source by measuring EXAFS spectra. The crystallographic investigation suggested the formation of a reaction zone, which proceeds through the NR in a core/shell mechanism. Further mechanistic investigation suggested a diffusion mechanism via intrinsic vacancies.

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Synthesis and Characterization of PbS/ZnS Core/Shell Nanocrystals

The addition of a zinc sulfide (ZnS) shell to lead sulfide (PbS) nanocrystals is advantageous because it could lead to higher/more stable photoluminescence quantum yields, reduced non-radiative recombination from electron-phonon coupling, and long-term stability of PbS nanocrystals in aqueous buffer solutions for bio-imaging. However, while ZnS shells have been successfully added to several nanocrystals such as CdSe and CdS it has never been added directly (without a spacer layer) to PbS nanocrystals. This is because it is difficult to add shells to Pb chalcogenide nanocrystals due to their tendency to Ostwald ripen at moderate temperatures. We have overcome this roadblock and are the first to demonstrate the synthesis of PbS/ZnS core/shell nanocrystals using a “flash” type synthesis with Zn oleate and thioacetamide as the precursors. We have found that the reaction is self-limiting and deposits a single monolayer of ZnS per shell reaction without Ostwald ripening. High angle annular dark field scanning transmission electron microscopy (HAADF-STEM) verified the presence of the ZnS shell. Furthermore, the absorbance and photoluminescence peak energies were found to redshift upon adding the ZnS shell due to the relaxation of a ligand-induced tensile strain, as well as wave function leakage into the ZnS shell. This abstract fits in the “Material Growth and Fabrication Technique” track because it describes the growth of PbS/ZnS core/shell nanocrystals.

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Colloidal Cu2-\(x\)Se Nanocrystals Synthesized from Dodecaneselenol as Templates for Anisotropic Wurtzite CdSe and CuInSe2 Nanoparticles

Colloidal copper chalcogenide nanocrystals (NCs) are an emerging class of semiconductor materials with interesting optoelectronic properties, without containing heavy metals such as cadmium and lead. Binary copper sulfide NCs can be directly synthesized in a wide range of sizes, shapes, crystal structures and stoichiometries, which not only provides good control over their optoelectronic properties, but also makes them excellent template materials for post-synthetic cation exchange (CE) reactions in order to obtain other binary or multinary chalcogenide NCs. For the selenide analogue however, less precursors are available for direct synthesis, thereby limiting the size and shape control attainable for this class of materials. Here, we investigate the use of 1-dodecaneselenol as selenium precursor and ligand in the colloidal synthesis of Cu\(_{2-x}\)Se NCs. By tuning the reaction parameters, we are able to tailor the morphology of the obtained colloidal Cu\(_{2-x}\)Se NCs from quantum dots to nanoplatelets with varying aspect ratios, and ultrathin nanosheets. The Cu\(_{2-x}\)Se NCs are used as templates in post-synthetic CE reactions and successfully converted to CdSe and CuInSe\(_{2}\) quantum dots, nanoplatelets and ultrathin nanosheets. Interestingly, the crystal structure changes during the CE reactions from tetragonal umangite to hexagonal wurtzite, which preserves the hexagonal shape but changes the thickness and lateral dimensions of the nanoplatelets. The anionic sub-lattice reorganization that is underlying the crystallographic transformation and reconstruction of the product NCs is attributed to a combination of the unit cell dimensionality and an internal ripening process, through which atoms move from the non-polar (100) and (010) side facets to the polar (002) top facet. This work provides novel tools to design colloidal anisotropic Cu\(_{2-x}\)Se NCs, which can also be used as a new family of template materials for the synthesis of anisotropic NCs of wurtzite CdSe, CuInSe\(_{2}\) and other metal selenides.

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Aminoalkanoic Acids as Alternatives to Mercaptoalkanoic Acids for the Linker-Assisted Attachment of Quantum Dots to TiO2

Linear aminoalkanoic acids (AAAs) and mercaptoalkanoic acids (MAAs) were characterized as bifunctional ligands to tether CdSe QDs to nanocrystalline TiO2 thin films and to mediate excited-state electron transfer (ET) from the QDs to TiO2 nanoparticles. The adsorption of 12-aminododecanoic acid (ADA) and 12-mercaptopododecanoic acid (ADA) to TiO2 followed the Langmuir adsorption isotherm. Surface adduct formation constants (Kad) were \(\sim 10^4 \text{ M}^{-1}\); saturation amounts of the ligands per projected surface area of TiO2 \(\Gamma_0\) were \(\sim 10^{-7} \text{ mol cm}^{-2}\). Both Kad and \(\Gamma_0\) differed by 20% or less for the two linkers. CdSe QDs adhered to ADA- and MDA-functionalized TiO2 films; data were well modeled by the Langmuir adsorption isotherm and Langmuir kinetics. For ADA- and MDA-mediated assembly values of Kad were \((1.8 \pm 0.4) \times 10^6\) and \((2.4 \pm 0.4) \times 10^6 \text{ M}^{-1}\), values of \(\Gamma_0\) were \((1.6 \pm 0.3) \times 10^{-9}\) and \((1.2 \pm 0.1) \times 10^{-9} \text{ mol cm}^{-2}\), and rate constants were \((14 \pm 5)\) and \((60 \pm 20) \text{ M}^{-1} \text{ s}^{-1}\), respectively. Thus, the thermodynamics and kinetics of linker-assisted assembly were slightly more favorable for MDA than for ADA. Steady-state and time-resolved emission spectroscopy revealed that electrons were transferred from both band-edge and surface states of CdSe QDs to TiO2 with rate constants \(\langle \text{ket} \rangle\) of \(\sim 10^7 \text{ s}^{-1}\). ET was approximately twice as fast through thiol-bearing linker 4-mercaptobutyric acid (MBA) as through amine-bearing linker 4-aminobutyric acid (ABA). Photoexcited QDs transferred holes to adsorbed MBA. In contrast, ABA did not scavenge photogenerated holes from CdSe QDs, which maximized the separation of charges following ET. Additionally, ABA shifted electron-trapping surface states to higher energies, minimizing the loss of potential energy of electrons prior to ET. These tradeoffs involving the kinetics and thermodynamics of linker-assisted assembly; the driving force, rate constant, and efficiency of ET; and the extent of photoinduced charge separation can inform the selection of bifunctional ligands to tether QDs to surfaces.

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Direct probing and manipulation of the nuclear spins in individual II-VI CdTe/ZnTe quantum dots

Nuclear spin fluctuations in III-V quantum dots (QDs) have plagued the electron spin coherence in dense nuclear spin baths such as InGaAs/GaAs QDs. The II-VI QD system offers an attractive alternative, since the majority of nuclei have no spin (I = 0), while the electronic and optical properties are comparable with III-V QDs. Due to the small effective field from the nuclei felt by the electron (Overhauser field), experimental observations of nuclear spin effects are challenging and so far were limited to indirect detection via the electron spin polarization rate (Le. Gall et al., 2012). Understanding the properties and developing the techniques for nuclear spin manipulation is thus required in order to develop II-VI materials as a platform for QD spin qubits with dilute nuclear spin baths. Here we present the first direct observation of the Overhauser field in single CdTe/ZnTe self-assembled QDs, by detecting the change in splitting of the neutral exciton ($X^{0}$) Zeeman doublet. Overhauser shifts of up to ±2μeV are detected but only in a sample containing a quantum well (QW), which suggests that the QW states play a crucial role in the generation of DNP. The dilution leads to significantly faster nuclear spin dynamics: the nuclear spin build-up time is found to be 1.1ms and the lifetime of the resulting polarization is $T_1 \approx 1.6$ s. Finally, optically detected nuclear magnetic resonance (ODNMR) experiments were performed and resolution limited NMR peaks were observed for both Cd and Te isotopes. The peak was accompanied by a broad tail, which is a sign of strong inhomogeneous broadening likely due to charge fluctuations. Our experiments are the first demonstration of manipulating the nuclear spin bath in a single II-VI QD with both optical and radio-frequency fields and is a first step towards developing QD spin qubits in nuclear-spin-free materials.

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Nano-cathodoluminescence and Carrier Dynamics in All-inorganic Perovskite Nanocrystals

Nanocrystals of caesium lead halide perovskites (CsPbX₃, X = Cl, Br, and I) have emerged recently as a promising new type of optoelectronic material offering bright emission across the visible spectrum [1]. An understanding of carrier dynamics and recombination in these materials and their dependence on nanocrystal structure and composition will be essential to their exploitation and development.

In this study, we apply nano-cathodoluminescence in a scanning transmission electron microscope to characterise the nanoscale structure of individual CsPbCl₃ and CsPbI₃ nanocrystals, and relate it to their emission properties. We also compare these nanoscale optical properties to those of ensembles of the same nanocrystals and investigate the carrier dynamics by fluence-dependent transient absorption (TA) and photoluminescence (PL) spectroscopies, and quantum yield measurements.

It was found that all of the CsPbI₃ nanocrystals studied, which were of regular shape and had a narrow size distribution, exhibited efficient CL. In contrast, a larger size distribution was observed for the CsPbCl₃ nanocrystals with irregular shapes and higher aspect ratio nanocrystals; of these, only the larger, more regular nanocrystals exhibited efficient CL. Ensembles of these nanocrystals exhibited PL with a similar fluence-dependence; in both cases, the quantum yield initially grew as the fluence was increased before reducing again. This behaviour was attributed to the initial saturation of defect-related non-radiative recombination followed by the onset of Auger recombination, which occurs when the absorption of more than one photon per nanocrystal per excitation pulse becomes probable. Trapping and Auger recombination lifetimes were extracted from the TA transients and their scaling with the nanocrystal volume was used to explain why the CL efficiency is so sensitive to nanocrystal size.


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Vibrationally Modified Spectral Diffusion in CdSe/CdZnS Colloidal Quantum Dots

Colloidal quantum dots (CQDs), which are quantum dots synthesized in solution, have been of continued interest due to their wide range of applications in biological tagging, photovoltaics, and tunable light absorbers/emitters etc [1]. However, the inhomogeneous broadening of CQDs obscures certain dynamics such as spectral diffusion, and can usually only be studied by techniques such as single dot measurements and spectral hole burning. In this study, we circumvent this inhomogeneous broadening by performing two-dimensional coherent spectroscopy on CdSe/CdZnS core/shell CQDs at cryogenic temperatures. In the spectra obtained, features due to coupling with CdSe lattice LO-phonon modes are observed that provide information concerning their associated intraband coherences [2]. Surprisingly, the coherences that form the peaks observed do not dephase exponentially as expected from Markovian spectral diffusion. Instead, the peaks exhibit initial rises before decaying at longer times [3]. Further, the coherences involving vibrational states are found to exhibit longer rise times. Simulations of the acquired 2D spectra provide evidence that the LO-phonon mode coupling itself significantly modifies the dephasing lineshapes and their underlying spectral diffusion dynamics. Previous studies have focused on free surface charges and surface ligand rearrangement as possible causes for the band-edge Stark shift that lead to spectral diffusion. The above theories are not sufficient to explain our results however, which point to coupling with lattice vibrational modes as a major factor in the spectral diffusion dynamics. In conclusion, we acquired two-dimensional spectra that reveal differences between the dephasing dynamics of coherences depending on whether vibrational states are involved. In addition to a fundamental understanding of the material, these results will prove crucial for single-photon emitter and quantum information applications, in which dephasing due to non-Markovian coupling to a reservoir represents one of the largest challenges to practical implementation.


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Excitonic Energy Transfer in InP/ZnS core/shell Quantum Dot Langmuir-Blodgett films

Here, we fabricated heavy metal-free InP/ZnS core/shell quantum dot (QD) Langmuir-Blodgett (L.B) films and studied the excitonic energy transfer within the quantum dots. Two types of InP based core/shell quantum dot L.B. films, one with a thin ZnS shell and other with a thick ZnS shell, were prepared. The structural and optical properties of quantum dots reveal the successful multiple shell growth and atomic force microscopy study show the smoothness of the L.B. films. Time-resolved photoluminescence (PL) and fluorescence-lifetime imaging microscopy (FLIM) studies of thick-shell QD monolayer reveal decrease in the average life-time and narrowing life-time distribution compared to thin-shell QD L.B. film. The interparticle excitonic energy transfer was shown with spectrally resolved PL traces and higher energy transfer rate was observed for thin-shell QD L.B. film. Finally, we calculate the average exciton energy and indicated that after multiple shell growth, the energy transfer induced exciton energy shift decreased significantly from 130 meV to 20 meV.

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Single Nanocrystal Spectroscopy of Shortwave Infrared Emitters

Imaging in the shortwave infrared (SWIR; 1-2μm) has found applications in industries ranging from surveillance to remote sensing[^1,2]. Due to an influx of sensitive detector technology in the SWIR, this list of applications has been extended to include SWIR microscopy[^3]. Microscopy in any wavelength region depends on having bright, stable, and versatile emitters. Very few SWIR emitters have been developed and among the brightest and most stable are Indium Arsenide (InAs) and Lead Chalcogenide (PbX; X=S, Se, Te) nanocrystals. These materials have enabled high power conversion efficiency quantum dot solar cells, as well as in vivo video-rate imaging of physiological processes[^4,5].

In order to optimize an ensemble of SWIR emitting nanocrystals for narrow fluorescence linewidth, we must learn about the homogeneous and inhomogeneous broadening in these systems. In order to access the single nanocrystal fluorescence linewidths of InAs and PbX nanocrystals we have implemented the solution-Photon Correlation Fourier Spectroscopy (s-PCFS) method which allows us to extract an average single nanocrystal linewidth[^6]. We have learned that 1. There is significant inhomogeneous broadening in these systems suggesting room for synthetic optimization and 2. For InAs/CdSe core-shell nanoparticles, much like in CdSe/CdS nanoparticles and other quasi type-II systems, the thicker the shell, the more electron wavefunction leaks into the shell and the broader the fluorescence linewidth becomes[^7]. In order to access the true homogeneous linewidth of these materials we have affixed InAs/CdSe/ZnS core-shell-shell nanoparticles to a substrate and cooled them to 4K. At this temperature we can directly access exciton-phonon coupling and spectral diffusion at short time scales. Through these studies we have gathered a more complete picture of the factors that contribute to the broad fluorescence linewidth in this system.


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Ultrafast Exciton Dynamics in White Light Quantum Dots

The surface of CdSe quantum dots is often seen as a hindrance for applications because surface traps can decrease quantum yield and suppress optical gain. However, white light emission can be seen in ultrasmall quantum dots that emit from both core and surface states. These “white light dots” may aid in understanding how surface states influence exciton dynamics as well as how multi-excitons react far into the strong confinement regime. Ultrafast transient absorption spectroscopy experiments were conducted on CdSe quantum dots with two different sizes. Larger quantum dots with no surface emission were compared to ultrasmall quantum dots with considerable surface emission. Optical gain can be seen in the larger quantum dots but is completely suppressed in the ultrasmall sample. Two forms of excited state absorption compete against stimulated emission in both samples: formation of biexcitons from single excitons and excitation of excitons from lower excited states into the continuum. Both these features are more intense in the ultrasmall quantum dots resulting in minimal stimulated emission from both core and surface states. Further transient absorption studies on white light emitting quantum dots with various organic and inorganic surface treatments may help to understand the role of the quantum dot surface on excited state absorption.

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Photoemission studies of Band Bending in PbS Colloidal Quantum Dot Solids

A common design in colloidal quantum dot (CQD) solar cells and other optoelectronic devices is to use a film of CQDs. The modification of CQD surfaces, through ligand exchange, allows control over the doping level and carrier mobilities of the film [1]. Using doped CQD films allows for the engineering of band alignment between different CQD films, and CQD and oxide interfaces. This has led to increases in CQD solar cell efficiencies [2]. Still, there remains a question of whether band bending exists at the surface of a CQD film alone, interfaced with vacuum.

We have tested for band bending at the surface of CQD solids, in studies aimed at understanding the electronic structure of depleted heterojunction CQD solar cells. First we have used synchrotron-radiation-excited XPS to study the surface photovoltage (SPV) shift in the core levels and valence band of PbS CQD films photoexcited with white light. Unlike bulk semiconductors, there is no obvious mechanism for the creation of surface defect states to cause band bending at the surface. Nevertheless, we are able to observe a significant SPV shift, of the order of up to 70 meV in doped films.

Second we have used time-resolved laser-pump X-ray photoemission-probe spectroscopy at the TEMPO beamline at Synchrotron SOLEIL to study the carrier dynamics within the CQD films as the SPV shift relaxes. This technique has previously given insight into the quantum dot – ZnO heterojunction, commonly found in CQD solar cells [3]. Results from CQD films subjected to different ligand treatments, and different thicknesses are discussed.


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Two-Colour Fluorescence Intermittency Spectroscopy: Shedding Light on Blinking in Nanocrystals

Even under continuous excitation, single semiconductor NCs `blink'\textsuperscript{1}—exhibit periodic on/off photoluminescence switching on unusually long (milliseconds--seconds) timescales. This phenomenon hampers their performance as ensemble light-emitters, and particularly hinders their use in single-molecule fluorescence imaging. Here, we propose a new two-colour fluorescence intermittency measurement to gain insight into the mechanistic causes of blinking. We use a home-built epifluorescence microscope to monitor the emission of individual nanocrystals under above-bandgap photo-excitation.\textsuperscript{2} Then, we periodically expose the nanocrystals to sub-bandgap light to preferentially provide energy to long-lived non-emissive states, and shorten their duration. For example, long-lived trapped charges liberated by sub-bandgap excitation will no longer quench photon emission, manifesting as increased photoluminescence intensity, and altered single-nanocrystal emission statistics

Though blinking is visible directly only in single nanocrystals, the phenomenon is quantifiably tied to reversible ensemble photodimming (statistical aging)\textsuperscript{3} and, here, offers a more sensitive measure of modulation. To date, we have demonstrated optical photodimming and subsequent recovery of ensemble fluorescence, suggesting that long-lived trapped carriers thermally de-trap when not illuminated. Work is ongoing to characterize the statistical effect of the sub-bandgap modulation. Fundamental insight into photoluminescence blinking will motivate improvements in NC synthesis, and advance the use of NCs in a range of applications, such as luminescent down-shifting, LEDs, excitonic upconversion, biomedical imaging, and photovoltaics.


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The role of Quantum Confinement in Optical Gain Development in Lead Tri-Halide Perovskite Quantum Dots

Semiconducting lead halide perovskite materials are excellent candidates for realizing low threshold light amplification due to their large absorption/gain cross section, high luminescence quantum yield and ease of processing. Several solution processable lasers based on nanocrystal quantum dots (QDs) of CsPbBr$_3$ have been demonstrated, even operating under nanosecond optical pumping. Nearly all of these architectures use QDs with large average sizes. The carriers in these systems are subject to weak quantum confinement and hence there is no clear picture whether the use of QDs in the strong confinement regime would be beneficial, nor is there any insight in how optical gain would develop in this peculiar class of nanomaterials. Here, we show through a combination of quantitative transient absorption and femtosecond fluorescence spectroscopy, that optical gain in strongly confined perovskite QDs develops at remarkably low average exciton densities, yet is capped by a fast non-radiative biexciton recombination. We are able to explain our remarkable observations using a simple 3-level model that takes both exciton-exciton repulsion and strong electron-phonon coupling into account. The concepts shown here provide a rational approach to look for low threshold optical gain materials based on strongly confined QDs.

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Ultrafast Intraband Spectroscopy of Hot-Carrier Cooling in Lead-Halide Perovskite Nanomaterials

In conventional solar cells (SCs), above-bandgap “hot” carriers (HCs) rapidly lose their excess energy through vibrations of the semiconductor lattice. This HC cooling is a major contributor to the Shockley-Queisser limit for the efficiency of single-junction SCs. Semiconductors with retarded cooling rates are therefore highly desirable for the next generation of SCs. At the forefront of contemporary SC research are the lead-halide perovskites, which exhibit exceptional efficiencies and materials properties that can be tailored by altering the composition of the “APbX₃” structure (A = monocation; X = halide). Recent discoveries have revealed slow HC cooling in these materials at high carrier densities.<sup>1-3</sup> This phenomenon is expected to be more pronounced in nanostructures, owing to quantum confinement effects.<sup>4</sup>

Elucidating HC cooling dynamics in perovskite nanomaterials is therefore of critical importance to the development of high-performance, solution-processable SCs. Here we study HC cooling in a broad array of bulk and quantum dot lead halide perovskites (A = MA⁺, FA⁺, Cs⁺; X = I⁻, Br⁻) using ultrafast infrared intraband spectroscopy.<sup>5</sup> A visible pump pulse generates "cold" carriers, which are then elevated into higher-lying "hot" states by an infrared push pulse. By monitoring changes to the intraband absorption of an infrared probe in time, we selectively track the dynamics of HC cooling. For all samples, we find that the cooling is sensitive to both the HC density (higher push fluence gives slower cooling) and choice of cation (with the slowest cooling in Cs-based systems). These observations suggest the existence of a “hot-phonon bottleneck”,<sup>1-4</sup> and assert the role of the lattice vibrations towards HC cooling. The role of surface states is discussed when comparing bulk and quantum dot analogues.


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Using molecular dimers for excitonic upconversion

Colloidal quantum dots (QDs) are defined by their size-tunable optical energy gaps, and allow for very specific chemical functionalizations due to their easily modified surface ligands. Recently, it has been shown that QDs in both colloidal and solid-state architectures can be used as triplet sensitizer species for exciton-mediated photon upconversion\(^1,2\); a photophysical energy conversion process that yields one high energy emitted photon for two incident absorbed photons. This process takes advantage of the rapid spin-mixing of photoexcitations in QDs to populate the triplet (spin 1) excited states of organic acceptors via Dexter triplet energy transfer (TET). Triplet-triplet annihilation (TTA) on the organic acceptor yields a high-energy singlet (spin 0) excitation, allowing for photon emission.

In our current work, we explore upconversion via the use of novel, electronically decoupled organic dimers as triplet acceptors and annihilators. Our transient photoluminescence measurements show that surface-anchored dimeric triplet acceptors quench QD emission with an efficiency comparable to their monomeric counterparts. This offers a route towards a free-floating hybrid nanoparticle for excitonic upconversion, wherein the surface-anchored dimer will act as both the triplet extractor from the QD core, and the ultimate annihilator. Indeed, eliminating diffusion-controlled interactions from exciton transfer and upconversion could allow free-floating, biocompatible upconversion probes to be functionalized for background-free, in vivo fluorescence imaging.


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Mechanisms of photoluminescence intermittency in all-inorganic and organic-inorganic-hybrid perovskite nanocrystals

Lead halide perovskite nanocrystals (NCs) are highly luminescent materials and anticipated for use in photonic device applications such as light-emitting diodes (LEDs) and lasers [1]. Their reported luminescence quantum yields are as high as 90% and their emission wavelengths are tunable over the entire visible spectrum through halide exchange and NC size control [2]. Despite these promising characteristics, the exciton dynamics and intrinsic luminescent properties have not yet been clarified sufficiently, leaving implementation of the perovskite NCs difficult.

In this work, we study photoluminescence (PL) intermittency mechanisms in all-inorganic cesium lead bromide CsPbBr$_3$ and organic-inorganic hybrid formamidinium lead bromide FAPbBr$_3$ perovskite NCs. The single dot spectroscopy of CsPbBr$_3$ NCs clearly shows a stable light emission. On the other hand, for the FAPbBr$_3$ NCs we observed two types of NCs, i.e., those with a binary switching (blinking) and those with a gradual undulation (flickering) behavior of the PL intermittency. We clarified that blinking occurs in NCs with state-switching between neutral exciton and trion states. The NCs with PL flickering had no trion contribution, which indicates that flickering is caused by surface-localized electrons. We also clarified that the flickering behavior can be completely suppressed and the PL quantum yields are improved, by applying a postsynthetic surface treatment (with sodium thiocyanate) to the FAPbBr$_3$ NC ensemble. This result supports our interpretation. The intentional change of the surface conditions provides deep understanding of luminescent properties of perovskite NCs, leading to realization of bright LEDs and lasers.

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Optical couplings by dipole-dipole interaction in a single coupled-quantum-dot structure

As a building blocks of quantum bits, various coupled quantum dot (CQD) structures have been studied in terms of coupling and entangling of quantum states. When the interdot distance of a CQD is short enough ($d<10\text{nm}$) to result in a wave function overlap between the two quantum dots (QDs) via the tunneling effect, the optimal coupling is controlled by the interdot distance and any external DC electric field along the coupling direction. When the separation of two QDs is not small enough to give rise to a sufficient wave function overlap for tunneling to occur, the exciton dipole-dipole interaction can be used as an alternative method of coupling two separate QDs.

We find that the exciton dipole-dipole interaction in a single laterally coupled GaAs/AlGaAs quantum dot structure can be controlled by the linear polarization of a nonresonant optical excitation [5]. The exciton dipole-dipole interaction of a CQD becomes enhanced if the polarization of nonresonant excitation is parallel to the coupling direction. When the excitation intensity is increased with the linear polarized light parallel to the lateral coupling direction (H-polarized excitation), excitons ($X_1$ and $X_2$) and local biexcitons ($X_1X_1$ and $X_2X_2$) of the two separate quantum dots (QD$_1$ and QD$_2$) show a redshift along with coupled biexcitons ($X_1X_2$), while neither coupled biexcitons nor a redshift are observed when the polarization of the exciting beam is perpendicular to the coupling direction. The polarization dependence and the redshift are attributed to an optical nonlinearity in the exciton Forster resonant energy transfer interaction (FRET). The FRET interaction becomes dominant with increasing the H-polarized excitation, where redshifts, an exciton population transfer, and a coupled biexciton appear. We have also distinguished the coupled biexciton from local biexcitons by the large diamagnetic coefficient. One of the coupled biexciton transitions was found to be spectrally overlapped with $X_{2}$. 

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Measuring and Modeling the Temporal Diffusion of Exciton-Fluorescence Rates and Energies in Individual CdSe-Dot/CdS-Rod Nanocrystals

Low-temperature fluorescence spectra of individual chemically synthesized semiconductor nanocrystals typically show—besides the blinking—small fluctuations in the emission energy over time. We investigate this so-called spectral diffusion of individual CdSe-core/CdS-shell dot/rod nanocrystals at cryogenic temperatures. While recording spectral information of the emission, we synchronously measure the fluorescence decay dynamics. By that we can show that also the fluorescence decay rates are diffusing in time. We find that the single neutral exciton emission rate nearly linearly correlates to the emission energy. The experimental data is compared to self-consistent model calculations within the effective-mass approximation in which migrating surface charges account for the diffusion of rates and energies. The calculations reveal that two surface charges are sufficient to induce the experimentally observed effects, while the type-I character of the band alignment between core and shell is still preserved. (Lohmann et al., ACS Nano 11, 12185–12192 (2017)). We expand our theoretical model to include strain effects in heterostructured nanoparticles exploiting a valence force field approach. Within the effective-mass approximation, strain leads to changes in the conduction and valence band offsets, which directly influence the exciton energy and wave function. We find that strain cannot change the inherent type-I band structure in CdSe/CdS dot/rod heterostructures but leads to broader ranges in the diffusion of emission energies and rates and alters the slope of their correlation. Our approach principally allows for a simulation of all kinds of heterostructured nanoparticles and delivers insights into their excitonic properties, in particular the influenceability via external stimuli, like surface charges or electric fields. This is useful for the design of heterostructures to be utilized in both light-emitting and light-harvesting devices.

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Slow cooling and highly efficient extraction of hot-carriers in perovskite nanocrystals: towards hot-carrier perovskite solar cell

Thermodynamic calculations revealed that single junction solar cell conversion efficiencies can exceed the Shockley-Queisser limits and reach around 66% under 1-sun illumination if the excess energy of hot photogenerated carriers is utilized before they cool down to the lattice temperature (i.e., hot-carrier solar cells). Organic–inorganic lead halide perovskite semiconductors have recently emerged as the leading contender in low-cost high-performance solar cells. The key for the realization of hot-carrier solar cell include the slow hot-carrier cooling and effective extraction of hot-carrier energies which requires fast hot-carrier injection into charge collection layer before hot-carrier cooling down to the lattice temperature. Emulating semiconductor nanoscience, some interesting questions would be if the hot-carrier cooling rate in halide perovskites could be further modulated through confinement effects, and if these hot-carriers can be efficiently extracted. Here, the hot-carrier cooling dynamics and mechanisms in colloidal CH$_3$NH$_3$PbBr$_3$ nanocrystals of different sizes (with mean radius ~2.5–5.6 nm) and their bulk-film counterpart were compared using room-temperature transient absorption spectroscopy. Our results revealed that the weakly quantum confined CH$_3$NH$_3$PbBr$_3$ nanocrystals are very promising hot-carrier absorber materials (~2 orders slower hot-carrier cooling times and around 4 times larger hot-carrier temperatures than their bulk-film counterparts). This is attributed to their intrinsic phonon bottleneck and Auger-heating effects at low and high carrier densities, respectively. Importantly, we demonstrate efficient room temperature hot-electrons extraction (up to about 83%) by an energy-selective electron acceptor layer within ~1 ps from surface-treated perovskite nanocrystal very thin films (~30 nm). These new insights would allow the development of extremely thin absorber and concentrator-type hot-carrier perovskite solar cells.

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Slow Auger Relaxation in HgTe Colloidal Quantum Dots

The size-dependent biexciton Auger recombination lifetimes in HgTe colloidal quantum dots (CQDs) was measured by transient bleaching from 0.28 – 0.59 eV. In non-aggregated HgTe particles, the biexciton lifetime increases with particle size, from 10s of ps to >100 ps over the size range studied. This agrees with trends observed in other CQD systems, and extends them to these small-gap mid-infrared materials. The extracted Auger coefficient is three orders of magnitude smaller than for bulk HgCdTe materials with a similar energy gap and remarkably similar to that of CdSe CQDs, a visible-gap system. Such weak effects of electronic structure and the divergence between bulk and nanoscale Auger lifetimes for a given material remain unexplained by current theories. The slower Auger recombination rates in HgTe CQDs relative to bulk HgCdTe indicates that these CQDs possess an intrinsic advantage over bulk HgCdTe for applications in mid-infrared photodetection, where Auger recombination currently enforces cryogenic operation of the most sensitive detectors.

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Interplay between spontaneous emission, energy transfer and delayed emission in quantum dot supraparticles

Semiconductor quantum dots (QDs) have attracted increasing interest since their discovery due to their size-dependent opto-electronic properties. QDs are nowadays used in several opto-electronic applications, but a widespread use of QDs in commercial applications will require a thorough understanding of all processes that can occur once an exciton is generated in a nanocrystalline QD. Spontaneous decay of the exciton with emission of a photon is the main process and has been studied extensively, but recently it was found that the dynamics of spontaneous emission from individual nanocrystalline quantum dots (QDs) is strongly influenced by the temporary storage of the exciton in a non-emissive state, resulting in a considerable spreading of exciton decay times long after the typical spontaneous emission time [1]. Several opto-electronic applications require QDs assembled into a QD solid; then Förster exciton transfer between the QDs emerges in competition with spontaneous emission, and should evidently be affected by temporary exciton storage. Here, we present a study of the inter-relation between these three processes in CdSe(core)/CdS(shell) QD solids in the form of QD supraparticles [2,3]. By using monodisperse CdSe cores of constant size we were able to keep the intrinsic rate of spontaneous emission constant, while the rate of exciton transfer between dots was varied by the CdS shell thickness. Our results show the influence of temporary exciton storage on both spontaneous emission and exciton energy transfer.[1] F. T. Rabouw et al., Nano Lett., 2015, 15 (11), pp 7718–7725 [2] B. de Nijs et al., Nat. Mat., 2015, 14, pp 56-60 [3] F. Montanarella et al., ACS Nano, 2017, 11 (9), pp 9136–9142

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Revealing the Unique Electronic Band Structure of CuInS2 Quantum Dots

Ternary CuInS2 (CIS) quantum dots (QDs) have emerged as a promising new class of nanomaterial due to their unique characteristics, including the heavy metal free composition, a size tunable band gap across most of the visible spectra (500 – 800 nm), and a large Stokes shift, which can go up to 500 meV. Despite the relevance of this last property, the physical mechanism behind its origin is still under debate in the literature. In order to address this point, we present complete studies about both the non-linear optical properties and ultrafast carrier dynamics of CIS QDs with different sizes and shapes. By comparing the two photon absorption (2PA) and the linear absorption spectra of the samples, we show for the first time a sub-band gap 2PA transition originated from a unique inversion on the parity of the band edge states of the valence band, agreeing well with recent a theoretical prediction. However, the difference in energy of the photoluminescence (PL) and the lower 2PA transition can be as large as 200 meV, indicating that the parity inversion is not enough to explain the large Stokes shift observed, being necessary to consider the intermediate trap state in order to explain this feature in those nanomaterials. Furthermore, from ultrafast carrier dynamics studies, we demonstrate that biexciton Auger recombination is slower in CIS QDs than in Pb or Cd based nanocrystals, being comparable to the Auger lifetime for trion. This suggests that, when a biexciton is generated, one of the carriers is trapped on a localized state, from where the PL is originated. These results, together with the non-linear optical spectroscopy analysis, give us evidences to understand the unique electronic band structure of this kind of nanomaterial.


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Control of Quantum Well Width Distribution and Ultrafast Carrier Migration in Perovskite Quantum Wells

Metal halide perovskites have achieved photovoltaic efficiencies exceeding 22%, but their widespread use is hindered by their instability in the presence of water and oxygen. To bolster stability, researchers have developed low-dimensional perovskites wherein bulky organic ligands terminate the perovskite lattice, forming quantum wells (QWs) that are protected by the organic layers. In thin films, the width of these QWs exhibits a distribution that results in a spread of bandgaps in the material arising due to varying degrees of quantum confinement across the population. We show here that moving to the ligand allylammonium enables a narrower distribution of QW widths, creating a flattened energy landscape that leads to x1.4 and x1.9 longer diffusion lengths for electrons and holes, respectively. We attribute this to reduced ultrafast shallow hole trapping that originates from the most strongly confined QWs. We observe an increased PCE of 14.4% for allylammonium-based perovskite QW photovoltaics, compared to 11-12% PCEs obtained for analogous devices using phenethylammonium and butylammonium ligands. We also use ultrafast broadband pump-probe and two-dimensional electronic spectroscopy to uncover exciton transfer that occurs on timescales of 100 - 200 fs in these systems.

Proppe et al., Synthetic Control over Quantum Well Width Distribution and Carrier Migration in Low-Dimensional Perovskite Photovoltaics, J. Am. Chem. Soc. 140, 8, 2890-2896

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**Electron Delocalization and Gain in CdSe/CdS quantum dots-in-rods**

CdSe quantum dots are a promising platform for lasing applications, with good photo-stability, high quantum yield, and size-tunable emission. Gain applications have however been limited due to excited state absorption from single exciton states to biexciton states, which competes with stimulated emission. Quasi type-II band structure hetero-materials are one strategy for decoupling excited state absorption and stimulated emission. In this band structure either electrons or holes are delocalized over the interface, leading to small wavefunction overlap. This in turn leads to reduced Auger recombination rates and small biexciton binding energies.

CdSe quantum dots embedded in CdS rods are one such material. Size tunable emission is allowed by the CdSe core, while the CdS rod gives rise to electron delocalization in the conduction band. This system also displays polarized emission and large absorption cross sections for the rod that funnels excitons to the CdSe core. These properties make this system of particular interest to the quantum dot community.

Here ultrafast transient absorption measurements are performed to measure gain threshold, bandwidth, Auger lifetime, and biexciton binding energy in CdSe/CdS dots-in-rods. These are compared to ligand-passivated CdSe quantum dots and type-I CdSe/ZnS core/shell quantum dots. Auger rates, biexciton binding energy, and gain threshold are all lower in the CdSe/CdS dot-in-rod system, demonstrating the advantages of this band structure design as an active medium for gain.

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Hot Carrier Transfer in Semiconductor Nanocrystals: Effect of Quantum Confinements

Extraction of carriers from semiconductor nanocrystals (SNCs) is very important issue for the application to photovoltaic devices and solar energy conversion. Recent experimental and theoretical studies revealed the efficient electron transfer (ET) from semiconductor quantum dots (QDs) to acceptor molecules, in which the Marcus inverted region in the molecular ET theory does not exist [1, 2]. This anomalous behavior has been interpreted in terms of Auger-assisted ET. However, most of the studies on carrier transfer from SNCs are focused on ET dynamics from the lowest exciton state in SNCs. Hot carrier transfer from higher excited states in SNCs has not been systematically studied yet even though the crucial role of hot ET for solar energy conversion. In the present study, we have prepared several kinds of SNCs with controlled quantum confinement dimensionality such as QDs, nanorods (NRs), and nanoplatelets (NPLs), and examined hot carrier transfer dynamics from SNCs to acceptor molecules and noble metal nanoparticles by femtosecond state-selective excitation spectroscopy [3-5]. The effect of quantum confinements on hot carrier transfer dynamics will be discussed and compared with normal ET dynamics from the band-edge state of SNCs.


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Size dependent electron and hole cooling in CsPbBr3 perovskite nanocrystals

Over the last few years, lead-based inorganic perovskite nanocrystals (CsPbX₃, X = halide) have become important materials for several optoelectronic applications, such as sensitizers in solar cells and LEDs. Achieving high efficiencies is key for these materials to become realistic competitors for currently dominating materials. This is however not an easy feat, as processes such as electron and hole cooling happen on a rapid timescale, resulting in a major loss of energy. It is pivotal to understand these processes on an atomistic level to gain better insight on the driving forces behind this fast electron and hole cooling and how these cooling rates can be controlled. In this research, we focus on the dependency of electron and hole cooling rates on CsPbBr₃ nanocrystal size using density functional theory (DFT) calculations.

Using ab initio molecular dynamics (AIMD) simulations and nonadiabatic molecular dynamics (NAMD), we investigate electron and hole cooling in realistic CsPbBr₃ models ranging from roughly 2.0 nm to about 4.0 nm. These computational perovskite models were constructed by cutting a bulk structure along the (001) facets and charge-compensated by removing excess charge from the surface in the form of Cs⁺. From the AIMD trajectories, the molecular orbitals coefficients and nonadiabatic couplings are calculated using QMFlows while the cooling rates are calculated with PYXAID. In this study, we analyze the electron and holes individually and show that cooling rates depend on the size of the nanocrystal.

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Large Optical Stark Effect in Solution Processable CdSe Quantum Wells

Colloidal nanomaterials are considered as prime candidates for many diverse opto-electronic applications involving light-matter interaction such as light-emitting diodes and lasers or more exploratory polariton light sources. In particular, quasi-2D CdSe platelets are considered as ideal materials to realize this due to their supposedly large exciton oscillator strength and spectrally narrow excitonic features. However, to date no unambiguous estimation of the actual magnitude of the oscillator strength is given. Here, we show using polarization resolved ultrafast pump-probe spectroscopy, that excitons in CdSe platelets show strong light-matter coupling evidenced by a large optically induced stark effect (OSE). We can explain this OSE using a dressed atom picture which allows us to extract the dipole moment (oscillator strength) of the heavy-hole exciton, which matches other state-of-the-art excitonic materials. Finally, by comparison of the oscillator strength extracted from linear absorption and OSE spectroscopy, we are able to rationalize a model that is able to predict the maximum exciton oscillator strength of any given 2D material. We verify this approach using weakly confined CdSe quantum dots and variable temperature OSE measurements.

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Synthesis and optical spectroscopy of type-II semiconductor ZnSe/CdS dot-in-rod nanostructures

In type-II semiconductor heterojunctions the band offset leads to a spatial separation of electrons and holes. This charge separation makes type-II systems particularly useful, e.g., as light-harvesting materials in photovoltaic devices. Nanoparticles consisting of a spherical ZnSe core surrounded by a rod-shaped CdS shell, so-called dot-in-rods (DRs), represent a quantum system with an intrinsic type-II band alignment at the core/shell interface. Here, photogenerated electrons can delocalize within the shell, while holes are strongly localized in the core. The recombination energy of electrons and holes is mainly determined by the spatially indirect band gap at the interface. The type-II band structure should allow for an efficient manipulation of exciton properties by, e.g., external fields.

We wet-chemically synthesize highly luminescent type-II ZnSe/CdS DRs via a hot-injection method. Transmission-electron micrographs of the DRs reveal a radiused cylindrical geometry with average diameters and lengths of 7.3 nm and 29.9 nm, respectively. In solution, our ZnSe/CdS DRs exhibit an ensemble fluorescence energy of 2.16 eV with quantum yields above 35%. For comparison, the original ZnSe dots, acting as nucleation seeds for the CdS growth, fluoresce at 3.3 eV.

We use time-, energy-, and space-resolved confocal fluorescence spectroscopy at room and cryogenic temperatures to investigate individual ZnSe/CdS DRs. The DRs show a high photostability while emitting linearly polarized light. Compared to the more conventional type-I CdSe/CdS DR system [1], ZnSe/CdS DRs exhibit increased fluorescence lifetime. At 8 K, the lifetime is decreased compared to room temperature measurements. Furthermore, at low temperature, we observe abrupt spectral shiftings of the emission line over time, superimposed on smoother spectral diffusion processes. The spectral diffusion covers a larger range in energy than for type-I CdSe/CdS DRs, indicating the larger susceptibility of type-II structures to external stimuli, like surface charges.


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Sideband pump-probe technique resolves nonlinear modulation response of PbS/CdS quantum dots on a silicon nitride waveguide

Colloidal PbS/CdS quantum dots (QDs) have excitonic ground state energies that can be adjusted within the important C and O telecommunication wavelength windows. For applications, an ultrafast switching dynamics of sub-ps to few ps is desired with pulse powers of few fJ/pulse to target photonic switching energies of pJ/bit to sub-pJ/bit. We develop a highly sensitive side-band pump-probe technique to study the carrier dynamics in colloidal PbS/CdS quantum dots deposited on a silicon nitride waveguide after excitation by laser pulses with an average energy of few pJ/pulse. This technique combines an amplitude modulation of the pump pulse with phase-sensitive heterodyne detection. Differential changes in amplitude and phase of the order of 10⁻⁵, corresponding to arcseconds in the phase data, are well resolved by this sideband pump-probe technique. The PbS/CdS QDs are spin-coated onto a silicon nitride waveguide, which itself does not show any measurable nonlinear optical signal. Pump and probe pulse are guided co-linearly and co-polarized in the waveguide. We detect the signal using lock-in amplifier with two independent demodulators to track the heterodyne main band and the side-band imprinted by the pump amplitude modulation (for details see Ref. [1]). The simultaneous data fit in amplitude and phase of the decay traces yields biexponential behavior. We conclude that the nonlinear optical response of the system is governed by one process involving hot carriers, i.e. Auger recombination, and one process involving the biexciton. A modulation on a sub-nanosecond time scale is found caused by Auger processes and a fast biexcitonic decay component in PbS/CdS quantum dots. These fast electronic processes are promising for opto-electronic switching and modulation based on colloidal QDs in telecommunication.[1] M. Kolarczik et al., "Sideband pump-probe technique resolves nonlinear modulation response of PbS/CdS quantum dots on a silicon nitride waveguide," APL Photonics 3, 016101 (2018).

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Exploring the Exciton Dissociation in Quantum Confined CsPbBr₃ Nanosheet Films

All-inorganic perovskite CsPbBr₃ nanosheets (NSs) have demonstrated exceptional photophysical properties derived from analogous exciton electronic structure to quantum wells, such as increased exciton binding energy, enhanced absorption cross section and shortened PL lifetime. Despite the significant progress on the performance of optoelectronic devices, the understanding of the coupling between CsPbBr₃ NSs is still far less developed. Exciton generation and dissociation processes can be studied on the basis of photophysics analysis. We prepared 2D CsPbBr₃ NSs with an ultrathin thickness of 1.8 nm and large lateral dimensions of 700 nm, and large exciton binding energy of 231.6 meV was extracted by band edge absorption fitting. Enhancing inter-NSs coupling is one of the most efficient methods to promote exciton dissociation. Low temperature annealing was confirmed to cause the ligands rearrangement, resulting in decreased binding energy of 149.1 meV and high charge transfer rate of 0.15 ns⁻¹. Therefore, the tunneling is thought as a dominant way of exciton dissociation. Furthermore, a large carrier mobility and a high responsivity of 0.53 A/W were obtained for CsPbBr₃ NSs photodetector with strong inter-NSs coupling. In this work, we propose ligands rearrangement concept, which can be employed to explain two kinds of couplings, and thus the exciton dissociation mechanism is elucidated to help the researchers to design and prepare perovskite QDs optoelectronic devices with high performances. Some measured physical parameters including exciton binding energy, lifetime, charge transfer rate and mobility are essential to understand the perovskite QDs fundament deeply.

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**Influence of hot biexcitons on optical gain in CsPbI\(_3\) perovskite nanocrystals**

Nanocrystals (NCs) with high photoluminescence quantum yields have recently been synthesized for all-inorganic lead halide perovskites CsPbX\(_3\) (X=I, Br, Cl) [1]. Quantum confinement effects and the superior optical properties of their bulk counterparts make lead halide perovskite NCs unique laser materials with low-threshold optical gain. In high-density excitation regime, multiple excitons strongly affect the optical gain through exciton-exciton interactions. Although multiple excitons have been studied mainly in CsPbBr\(_3\) NCs [2, 3], the CsPbI\(_3\) NCs actually have a longer biexciton Auger lifetime than that of CsPbBr\(_3\) [4], and thus are being recognized as new candidates for laser materials. Therefore, it is important to reveal the exciton-exciton interactions in CsPbI\(_3\) NCs for material design of perovskite lasers.

In this work, we studied the electronic structure of hot biexcitons and their effects on the optical gain in CsPbI\(_3\) NCs by performing femtosecond transient absorption spectroscopy. Analyzing the induced absorption signal observed immediately after the pump excitation, we revealed that the biexciton binding energy has a peculiar dependence on the hot biexciton state, which is composed of an exciton at the band edge and a hot exciton generated by the pump pulse. We observed that the exciton-exciton interaction becomes stronger for hot excitons with larger excess energies. Furthermore, we found that under strong excitation conditions, optical gain is achieved and it can be controlled on the picosecond time scale by changing the excess energy of the hot excitons.

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Determining the Complex Refractive Index of Neat CdSe/CdS Quantum Dot Films

Understanding how structural and synthetic factors influence the complex refractive index of quantum dot (QD) solids is crucial to tailoring the light-matter interactions of QD-containing photonic and optoelectronic devices. However, neat QD films are challenging to accurately model as they are a mixture of inorganic core/shell materials and the surrounding organic ligands. Furthermore, both the QD film morphology and the complex refractive index vary due to particle size, ligand chain length, and the deposition process. We measure the complex refractive index of neat CdSe/CdS core/shell QD films by using variable-angle spectroscopic ellipsometry to derive the effective complex refractive index using Kramers-Kronig consistent dispersion models. We use this information in conjunction with intrinsic refractive index data of CdSe and CdSe/CdS QDs extracted from solution-state absorption data and a Bruggeman effective medium approximation (EMA) to describe neat QD films. We find that despite slight underestimations in the extinction coefficient of QD films, the Bruggeman EMA can successfully be used as a tool to approximate the complex refractive index of QD films. This information allows us to better understand packing variations between QD films and predict the absorption in QD thin films, including those made with core/shell heterostructures.

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Temperature Induced Gain Threshold Amplification and Ultrafast Trapping in Colloidal Quantum Dots

Continuous-wave (CW) lasing has recently been achieved in CdSe/CdS colloidal quantum dots (CQDs). The use of built-in biaxial strain helped reduce the gain threshold by ~1.4x in femtosecond regime and by ~3x in the quasi-CW regime compared to conventional core-shell CQDs. Here, we find that the difference in the impact on threshold for CW vs. femtosecond gain is caused by an increase in temperature that depopulates the excited state via a sub-picosecond recombination process, which we attribute to Auger-like ultrafast trapping. We combine theory and experiments to quantify the effect of this ultrafast trapping and reveal that it results in the superlinear improvement of the CW threshold vs. the respective changes in femtosecond threshold.


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Surface Plasmon Enhanced Quantum Dot Light-emitting Diodes using Sputtered Gold Nanoparticles

Noble metal nanocrystals have demonstrated strong ability in light engineering. Surface plasmon resonance of metal nanocrystals can effectively enhance the electroluminescence efficiency in traditional light-emitting diodes as well as newly developed organic light-emitting diodes and quantum dot light-emitting diodes devices. However, the metal nanocrystals synthesized through chemical methods usually involve large amount of surfactants, making the incorporation of the nanocrystals into device assembling quite difficult and non-universal. Here we reported the sputtered Au nanoparticles enhanced electroluminescence in inverted quantum dot light emitting diodes (ITO/Au NPs/ ZnMgO/ QDs/ TFB/ PEDOT:PSS/ Al). By combining the time-resolved photoluminescence, transient electroluminescence and ultraviolet photoelectron spectrometer measurements, the enhancement can be explained to the internal field enhanced exciton coupling to surface plasmons and the increased electron injection rate with Au nanoparticles incorporation. Phenomenological numerical calculations indicated that the electron mobility of the electron transport layer was increased from 1.39x10^{-5} to 1.91x10^{-5} cm^2/V·s for Au NPs modified devices. As a result, the maximum device luminescence is enhanced by 1.41 folds from 14,600 to 20,720 cd/cm^2 and maximum current efficiency is improved by 1.29 folds from 3.12 to 4.02 cd/A. These results demonstrate an effective method to apply plasmonic metal nanostructures for achieving super bright and efficient solid-state light emitting devices.


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Concurrences of photon pairs generated in the biexciton cascade of quantum dots in microcavities

The biexciton cascade in quantum dots can be used to generate entangled photon pairs. Here, we present analytical and numerical results and parameter dependences of different definitions of the concurrence used as measures of entanglement.

The concurrence as originally introduced by Wootters is in one-to-one correspondence to the entanglement of formation of a two-qubit state and can be directly calculated from the elements of a given density matrix. A natural choice is to use the time-dependent reduced two-photon density matrix, where the qubits are defined by the horizontal or vertical polarization of a photon. Like the two-photon density matrix, this concurrence is time-dependent. However, it is also common to extract concurrences from time-integrated two-time correlation functions that can be more easily obtained from experiments via quantum state tomography. One possible choice is the single-time-integrated concurrence obtained by integrating the two-time correlation function over the time of the click of the first detector and extrapolating towards zero delay time with respect to the click of the second detector. Alternatively, a double-time-integrated concurrence can be defined by additionally averaging over the delay time.

For quantum dots with large biexciton binding energies coupled to microcavities resonant to the two-photon transition to the biexciton state, analytic results for the three concurrences are obtained. We find that the time-dependent concurrence and the single-time-integrated concurrence quantitatively agree in a wide range of parameters. Therefore, the experimentally accessible single-time-integrated concurrence is found to be a good measure for the entanglement of formation of the two-qubit state described by the reduced two-photon density matrix. The double-time-integrated concurrence may exhibit different trends with respect to changes of the parameters. We find parameter ranges where upon variation of system parameters the time-dependent concurrence and the single-time-integrated concurrence increase while the double-time-integrated concurrence decreases, suggesting that both measure different physical properties.

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QD2018 – Poster Session 2 Abstracts

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The Role of Polyethylenimine in Enhancing the Efficiency of Quantum Dot Light-Emitting Devices

We investigate the use of polyethylenimine (PEI) as a wide band gap passivation layer between quantum dots (QDs) and the ZnO electron transport layer to improve the efficiency of quantum dot light-emitting devices (QDLEDs). The mechanism behind this efficiency enhancement has been disputed in literature due to the work function modification effect of PEI when deposited on ZnO films. We find that the improvement in QDLED efficiency primarily results from the passivation of exciton quenching sites at the ZnO/QD interface despite an increase in electron injection leading to further charge imbalance within the devices.

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Efficient Light-emitting Diodes Based on In-situ Fabricated FAPbBr3 Quantum Dots: The Important Role of Ligand-assisted Reprecipitation Process

Halide perovskites are desired light emitters with characteristics of brightly, color-tunable and narrow-band emission, which make them suitable materials for fabricating light-emitting diodes (LEDs) and laser technology. During the past three years, the external quantum efficiency (EQE) of perovskite based LEDs significantly increased from 0.01~0.1% up to 14.3%\[1-5\]. It has been realized that the dimensionality of perovskites plays an important role in determining the carrier transport and excitation recombination as well as the device performance. By introducing organic ligands, both of the molecular and size dimensionality of perovskites can be well-controlled through either in-situ fabrication on substrates via spin-coating a precursor solution or ex-situ fabrication via precipitation technique from colloidal solution. In-situ fabrication strategy provides an efficient and convenient way to control the dimensionality of perovskite toward efficient and brightly devices\[3-5\]. Typically, the in-situ fabrication on substrate for perovskite LEDs involves two steps: spin coating a precursor solution in polar solvents and then drop casting anti-solvent in a fixed time. Unlike the precipitation process in large volume flask, the in-situ fabricated perovskite nanocrystals experienced different nucleation and growth process. In this work, we investigate the crystallization of in-situ fabricated FAPbBr3 perovskite nanocrystals and illustrate the importance of ligand-assisted reprecipitation (LARP) synthesis process\[6\]. Highly luminescent and uniform FAPbBr3 perovskite nanocrystals based thin film with photoluminescence quantum yield up to 78% were obtained and the as-fabricated perovskite based LEDs show very high efficiency with peak EQE up to 16.26%.


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Hybrid Quantum Dot – Light-Emitting Electrochemical Cells

The next generation of lighting technologies gets more and more inspired by innovative design possibilities, overcoming the challenges of epitaxial LEDs in flexible and shapeable applications. Due to the easy and solution-based fabrication techniques, light-emitting electrochemical cells (LECs) represent a promising alternative for cost-effective large-area light emitters. The main challenge of LECs is the realization of long-term stable emitter molecules of different emission colors as well as the combination of differently colored molecules to generate white LECs. Colloidal quantum dots on the contrary particularly impress with their color tunability as well as their robustness due to their crystalline nature.

We present novel hybrid QD-LEC device architectures (QLECs), combining colloidal QDs with the ionic surrounding that is a fundamental component in LEC architectures. Combining a high-performance, ionic transition metal complex (iTMC)-based LEC with colloidal QDs, light emission from both QD and iTMC emitter is obtained. This can be either used to gain devices that change emission color with applied voltage or to generate superimposed emission of both light-emitting species at a given driving voltage [1]. By adding deep blue QDs in addition to a highly luminescent yellow iTMC emitter, white light-emitting hybrid devices with high luminance (> 850 cd m⁻²) and high color rendering index (80) are achieved. By replacing the organic iTMC emitter molecule completely with robust, crystalline QDs in an ionic surrounding, devices yielding up to > 350 cd m⁻² have been realized [2].


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Excitonic feature in perovskite quantum dots films

Cesium lead halide perovskite CsPbX$_3$ (X = Br, Cl, I) have been widely investigated for its high photoluminescence quantum yield and long carrier diffusion length, leading to applications in quantum dot (QD) LEDs, solar cells and photodetectors. However, the fundamental excitonic features and electronic structures of CsPbX$_3$ QDs are still not well understood. Here, we studied the excitonic signature of CsPbX$_3$ (X = Br, Cl) QD films by a unique approach: temperature dependent photoluminescence excitation (PLE) spectrum from 80K to 400K. In addition to the main bound excitonic emission, trion emission at lower energy and sub-band exciton state emission at higher energies are discovered under low temperature range. For CsPbCl$_3$ QD film, the sub-band emission peaks experienced blue shift and disappearance with increasing temperature, resulted from the effect of electron-phonon interaction. Different from the blue shift at low temperature, CsPbBr$_3$ excitonic peak shows red shift above 300K which results from the reduce of bandgap induced by the interatomic spacing increases. Moreover, a phase transition of CsPbCl$_3$ QDs was also observed at 360K. Our works contribute to the fundamental understanding of excitonic features and electronic structures of perovskite QD films, which is the platform of QD LEDs.

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All-solution-processed, full-color light-emitting device with sequential layer stacking of blue, green, and red quantum dots

During last decade performance of quantum dot-light-emitting diode (QLED) has dramatically progressed primarily based on a synergic effort on synthetic innovation of heterostructural quantum dots (QDs) and structural diversification/interfacial modification of multilayered device. Till now most of QLED work has been devoted the high-efficiency monochromatic devices with a single QD emitting layer (EML). However, for the ultimate application of QLED to next-generation display device, it should possess a full-color emissivity. For this, elastomeric stamp-based transfer printing and inkjet printing have been adopted for pixelating QD EMLs. Alternatively, full-colored white mode QLEDs could be realized by generating a spin-casted RGB QD-mixed EML or fabricating R,G,B unit-serially connected tandem QLED. As another conceivable EML design to attain a tricolored white emissivity one may consider an RGB QD-layered structure. However, spin-coating processing would be incompatible with such sequential stacking of different-colored QD EMLs, since the previously deposited EML should be severely damaged upon solution processing of the following one. Herein, we for the first time propose all-solution-processed full-color-capable white QLED, where stacked B/G/R QD EMLs deposited by a sequential spin coating are sandwiched by poly(9-vinylcarbazole) as a hole transport layer and ZnO nanoparticles (NPs) as an electron transport layer. To enable the formation of well-defined layered assemblies of B/G/R QD EMLs, an ultrathin ZnO NP buffer is inserted between different-colored QD layers, by which the physical deterioration of an underlying QD layer upon the subsequent deposition of another-colored upper QD layer is effectively prevented. The present full-color-capable QLED displays high electroluminescent figures-of-merit such as 16241 cd/m² in luminance, 15.9 cd/A in current efficiency, and 6.8% in EQE along with spectrally well-resolved RGB emission components. The promising results suggest that our novel EML design of ZnO NP buffer-mediated QD layer stacking may afford a viable means towards bright, efficient full-color-capable white device.

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Enhanced Device Performance by Improving Charge Balance with Y-Doped ZnO in Quantum Dot Light-Emitting Diodes

The performance of quantum dot (QD) light-emitting diodes (QLEDs) is critically dependent on the charge balance between electrons and holes injected into the QD emissive layer. Charging of QDs with excess carrier increases the possibility of non-radiative Auger recombination. In an inverted QLED structure with a ZnO electron transport layer (ETL), electron injection is more efficient compared to the hole injection because of nearly barrier-free electron injection from ZnO to the QD emissive layer, resulting in reduced device performance. To overcome this problem, inserting a thin insulating layer between the QDs and ETL has been introduced, but this approach requires elaborate efforts such as controlling precise thickness of inserted layer and adopting the layer without damaging bottom layer. Herein, we demonstrated efficient inverted QLEDs by improving the electron-hole balance using yttrium (Y) doped ZnO (YZO) ETL with a simple sol-gel process. Unlike other dopants such as Al, In, or Ga which increase conductivity of the doped ZnO film, the YZO film showed much reduced conductivity compared to the undoped ZnO film. Thus, YZO film significantly reduces leakage current and suppressed excessive electron injection into the QD emissive layer in QLEDs. As a result, the devices with the YZO as an ETL show significantly enhanced efficiency and lifetime. The maximum EQE and half lifetime of the QLED with YZO were 8.6% and 57.8 hours at 1000 cd/m² whereas those of the device with the undoped ZnO layer were 6.4% and 7.2 hours, respectively.

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Enhancement of stability of CsPbX3 perovskite quantum dots for white light-emitting diode applications

Recently, all inorganic CsPbX₃ (X = Cl, Br, and I) perovskite quantum dots (PQDs) have been spotlighted due to their excellent optical properties such as high photoluminescence (PL) quantum yield (QY), narrow PL band width, and full color emission from blue to red. However, the PQDs show low thermal and water stability. To improve the stability of the PQDs, polyhedral oligomeric silsesquioxane-coated CsPbX₃ PQDs were reported. Also, mesoporous silica particles integrated with CsPbBr₃ PQDs were reported. For further improvement of the stability of the CsPbX₃ PQDs, we synthesized PQD-inorganic SiO₂ composites. First, we synthesized highly efficient CsPbX₃ PQDs and we suggest facile route for the synthesis of CsPbX₂-SiO₂ (X = Br and I) composites. The synthesized CsPbX₃ PQDs showed high PL QYs of 44-94% and narrow emission band widths of 19-40 nm. By using the synthesized CsPbX₃ PQDs, perhydropolysilzane (PHPS)-derived CsPbX₃-SiO₂ composites were synthesized and they showed improved stability compared with bare CsPbX₃ PQDs. In particular, CsPbBr₃ PQD-embedded SiO₂ composites exhibited significantly improved thermal and water stabilities. Finally, CsPbX₃ PQD-SiO₂ composite-converted green and red light-emitting diodes (LEDs) were fabricated and the fabricated green and red LEDs showed high color purity. Moreover, blue LED-pumped CsPbX₃ PQD-SiO₂ composite-converted white LED was fabricated. The fabricated white LED exhibited natural white light and wide color gamut.

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Exceptional operating stability of color converting white light-emitting diode based on silica-mediated InP quantum dots

High-quality fluorescent quantum dots (QDs) with elaborately refined core/shell heterostructures have been highlighted as key materials for various optoelectronic devices. In particular, non-Cd InP QDs have been already commercially applied as color-converting emitters with a combination of blue light-emitting diode (LED) to LCD backlight unit. Due to still insufficient stability of InP QDs during operation, however, they should be packaged with high-cost oxide gas barrier films into a form of QD enhancement film (QDEF) for the securement of long-term reliability. The most viable strategy to protect QDs against degradable environments is to physically encase QDs with a chemically stable silica in the form of individual overcoating or collective embedding. Such a silica encapsulation has been conventionally explored via Stöber or reverse microemulsion process. However, both routes commonly encounter a critical issue of substantial loss in photoluminescence quantum yield (QY) compared to pristine QDs after silica growth. Herein, we implement the embedment of multishelled InP QDs, whose stability is quite inferior to CdSe counterparts, in silica matrix by means of a tetramethyl orthosilicate (TMOS) or 3-aminopropyl trimethoxysilane (ATPMS)-based, waterless, catalyst-free synthesis in a thermohygrostat. It turns out that under the present silica reaction the original QY of QDs is nearly fully retained. To the best of our knowledge, this is the first result demonstrating the formation of InP QD-silica nanocomposites with the original QY nearly intact. On-chip-packaged white QD-LED integrated with green and red InP QDs-silica composites is fabricated and placed in continuous operation at 60 mA for a prolonged period of time in order to verify the practical efficacy of silica embedding on QD stability, resulting in exceptional device stability over time. Thus, we believe that the present silica embedding strategy affords a practical means of not only maximally retaining QY of QDs but effectively passivating QDs.

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Stability Improvement of Quantum Dot Light Emitting Diodes with Atomic Layer Deposited Metal Oxide Films

Colloidal quantum dots (QDs) are highlighted as a promising light emitting material for optoelectronic devices due to advantages such as high color purity, high quantum yield and low cost solution process. In addition, the emission wavelength of QDs can be easily shifted by controlling their size, shape, and composition. Many researchers applied QDs in the display and lighting technologies. Nowadays, the device performance of blue, green, and red quantum dot light emitting diodes (QLEDs) are comparable to those for the state of the art organic light emitting diodes (OLEDs). However, the stability of QLEDs lags behind OLEDs, and it should be overcome for commercialization. In this work, we introduced 1nm thick Al₂O₃ layer as electron blocking layer between emitting layer (EML) and electron transporting layer (ETL) to suppress the leakage current and reduce exciton quenching induced by charge imbalance. Al₂O₃ interlayer was deposited by spatial atomic layer deposition (ALD) process that allows for precise thickness control. We investigated the effect of Al₂O₃ interlayer with inverted structure device consisting of ITO/ZnO/Al₂O₃/QDs/PEIE/poly-TPD/MoO₃/Al. It is known that electron transporting materials have higher carrier mobility than hole transporting materials and it causes charge imbalance in QLEDs. Charge imbalance causes Auger recombination and a detrimental effect on the devices. The Al₂O₃ interlayer serves to lower the mobility of electrons and Auger recombination causing the reduction of device performance was reduced, thereby improving the device efficiency about 1.1 times and stability by controlling the charge balance in the light emitting layer. The device with maximum current efficiency 49.4 cd/A and external quantum efficiency (EQE) 11.8% was fabricated by inserting Al₂O₃ interlayer. Al₂O₃ interlayer reduces the leakage current and results in better charge balance in the EML. Also, it improves operational stability of device and reduces EQE roll-off about 30% by suppressing exciton quenching by Auger recombination.

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Grafting of titanium and zirconium species onto multi-shelled InP QDs towards their enhanced stability

To date, InP quantum dots (QDs) appear to be sole substitutes for Cd-QDs with respect to nontoxic composition as well as narrow photoluminescent (PL) capability for the application to next-generation display devices. Owning to the ceaseless synthetic efforts particularly on heterostructural multi-shelling engineering, PL quantum yields (QYs) of InP QDs became comparable to those of synthetically matured Cd-QD counterparts. Although various QDs with satisfactorily high QYs for their industrial applications are presently available, their fluorescent stability against degradable environments is still a key issue. The most common approach toward QD stability is to physically encapsulate QDs typically with chemically stable oxides such as sol-gel silica, ZnGa$_2$O$_4$, and In$_2$O$_3$ and proved effective in preventing or retarding the oxidation of QDs upon a long-term exposure to photon and heat. In this work, we for the first time develop a simple but novel strategy to encapsulate the surface of InP QDs through in-situ grafting with titanium isopropoxide (Ti(i-PrO)$_4$) or zirconium propoxide (Zr(PrO)$_4$). First, highly fluorescent green or red InP QDs with an absolute QY over 80% are synthesized by adopting a multi-shell architecture of ZnSeS/ZnS. And then, the resulting InP/ZnSeS/ZnS QDs are successively reacted with an appropriate amount of Ti(i-PrO)$_4$ or Zr(PrO)$_4$ for QD surface grafting. Such grafting is presumed to stem from hydrophobic interaction. Then, comparative QDs without versus with grafting are identically subjected to UV irradiation and thermal aging for prolonged periods of time, evidently resulting in superior PL retention and colloidal stability from grafted QDs. These results clearly show that the present grafting strategy would be a promising means to effectively passivate QD surface by suppressing desorption of surface ligands as well as photooxidation.

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Demonstration of bicolored white electroluminescent devices based on Cu-X-S (X=Ga, In) quantum dots

Over the past decade the performance of quantum dot (QD)-based electroluminescent (EL) device or light-emitting diode (QLED) has been remarkably improved mostly through innovative attempts such as compositional/structural designing of QD, development of charge transport layer (CTL), and diversification of device architecture. Such substantial progress is mainly based on well-developed Cd-containing II-VI QDs targeting on a high-color reproducibility display device. Meanwhile, the realm of white emissive QLED as a general lighting planar device remains still unexplored. Herein, we report the first demonstration of efficient white QLED based on non-Cd I-III-VI family QDs of copper gallium sulfide (Cu-Ga-S, CGS) and copper indium sulfide (Cu-In-S, CIS). Band gap and photoluminescence (PL) of CGS QDs are successfully tuned by Zn alloying. After ZnS shelling, the resulting Zn-alloyed CGS (ZCGS) QDs exhibit excellent PL quantum yields (QYs) over 75% and tunable emission wavelengths of 470-490 nm, depending on the degree of Zn alloying. Meanwhile, PL wavelengths of CIS/ZnS QDs with high QYs over 80% are tailored in the range of 560-600 nm by varying Cu deficiency, which plays critical roles in controlling their absorption and PL properties. Then, these two types of ZCGS and CIS QDs are solution-blended with different weight ratios to generate randomly-mixed QD emitting layers (EMLs) for the following QLED fabrication. White QLED with a multilayered sequence of poly(9-vinlycarbazole) hole transport layer, QD-blended EML, and ZnO nanoparticle electron transport layer is fabricated via all-solution processing. White EL that originates only from EML of ZCGS and CIS QDs without any contribution of CTL emissions is successfully obtainable. The best device displays not only color rendering index of 80 but peak quantities of 1500 cd/m$^2$ in luminance, 3% in external quantum efficiency, and 3.1 lm/W in power efficiency, which correspond to the record values in general lighting QLEDs.

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Enhanced Performance and Stability of InP Quantum Dot Light Emitting Diodes through Al-doped ZnO Electron Transport Layer

Quantum dot light emitting diodes (QLEDs) have attracted a lot of attention in display and lighting industry due to their superb characteristics such as narrow emission peak, size dependent bandgap tunability and low cost solution process. Although cadmium (Cd)-based QLEDs show excellent device performance, the toxicity of Cd is a serious concern for practical applications. Thus, recent research is focused on developing eco-friendly quantum dot (QD) materials. Among them, indium phosphide (InP) QDs are most promising for developing high-performance displays with a wide color gamut. However, the performance of InP-based QLEDs is still far behind compared to the Cd-based QLEDs. It is necessary to reduce nonradiative recombination processes in InP QLEDs. In this work, we show that efficiency and stability of InP QLEDs are greatly improved by adopting Al-doped ZnO (AZO) as an electron transport layer (ETL). By optimizing the Al doping concentration, we can achieve a peak external quantum efficiency (EQE) of about 4% and 10 times longer lifetime compared to the device with an undoped ZnO ETL. Systematic analysis on the optical, electronic and electrical properties of AZO films is carried out to understand the underlying mechanism for the improved device performance. Compared to the undoped ZnO film, the AZO film has enhanced electron mobility and reduced energy barrier for the electron injection into the InP QD emissive layer. As a result, the device with AZO shows better charge balance, leading to improved device performance. We believe that the analysis based on the Al-doped ZnO will give further insight into improving and understanding InP QLEDs.

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A hybrid inorganic-organic multilayered EL devices for white emission

For last 10 years, the performance of colloidal quantum dot light-emitting diodes (QLEDs) has been dramatically improved. However, most of QLED works has been investigated in the form of monochromatic device, while white QLED still remains a long way to go. After solution-processed deposition of hole transport layer and emitting layer based on colloidal quantum dots, electron transport layer and emitting layer based on organic host-dopant system were deposited using thermal evaporation. The generation of white light was accomplished by the sequential structure of QLEDs and OLEDs. The unique hybrid electroluminescent devices generated a high-quality white light with a high color rendering index (CRI). The detailed I-V-L characteristics were studied to get efficient and high CRI white emission.

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Effect of Electron-Hole Balance on Quantum Dot Light-Emitting Diodes by Tuning Electron Transport Properties

Colloidal quantum dots (QDs) are one of the most highly interesting materials for light-emission owing to the quantum confinement effect. Among a variety of light-emitting applications, QD-based light-emitting diodes (QLEDs) using the electroluminescence of QDs are of interests for the next-generation display technology. However, there are several remaining issues of improving the QLEDs for utilizing them practically. One main problem in QLEDs is the electron-hole charge balance which is related to the device efficiency, efficiency roll-off, and operational lifetime. Thus, optimizing the charge balance in a QLED is an important research area. Herein, we will present our recent results on controlling the electron-hole charge balance of QLEDs using ZnO as an electron transport layer (ETL). We have investigated the methods to tune the electron transport ability of the ETL by i) adopting an interfacial layer between ZnO and QD, ii) doping impurities in ZnO, and iii) forming a multilayered ETL structure. We believe that our analysis and understandings in this presentation will be meaningful to advance this research field.

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Enhanced Light Out-Coupling Efficiency in CdSe/CdS Dot-in-Rod Light-Emitting Diodes

The nanocrystal-based light-emitting diodes have narrow full width at half maximum, capacity of controlling the emission wavelength, and stability compared to the organic fluorescent materials. In particular, light-emitting diodes based on quantum dots, which are 0-dimensional nanocrystals, reported external quantum efficiency close to 20%, which is the theoretical limit of out-coupling efficiency. However, the light-emitting diodes based on nano-rods, which are one-dimensional nanocrystals, have not been studied much as compared with quantum dots.

One of the characteristics of the nano-rods is dipole effect. Since the light extraction efficiency of the light-emitting devices is increased by arranging the dipole of the nano-rod in parallel to the light-emitting surface, the theoretically high efficiency can be achieved as compared with the device using isotropic nanocrystals based light emitting diodes.

We researched polarized emission and dipole effects of dot-in-rod nanocrystal based light-emitting diodes controlling the morphology of nanocrystals. In order to achieve high external quantum efficiency of the device, dot-in-rod nanocrystals with high photoluminescence quantum yield were used. We synthesized dot-in-rod nanocrystals with precisely controlled aspect ratio and characterized how the dipole of them affects the performance of devices. The structural characteristics of the dot in rods were analyzed by TEM and XRD, and optical properties were measured by absolute quantum efficiency, UV-visible absorption spectra and ellipsometry. We fabricated a red light-emitting diodes with high external quantum efficiency and brightness through optimized device fabrication, and confirmed that the light out-coupling efficiency has a dependence on the morphology of the dot-in-rod nanocrystals.

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Polyethylenimine-ethoxylated Dual Interfacial Layers for High-efficient Quantum Dot Light-Emitting Diodes

We controlled the electron injection rate in inverted quantum dot light-emitting diode (QLED) by inserting polyethylenimine ethoxylated (PEIE) layer between a ZnO electron transport layer (ETL) and quantum dot (QD) emitting layer and successfully demonstrated high efficiency and brightness of QLEDs. The inverted QLED has the layer structure of ITO (cathode)/ZnO NPs/PEIE/QDs/PEIE/P-TPD/MoO3/Al (anode). The PEIE layer was introduced between ZnO and QD to balance charge within QD emissive layer in the device, which serves as an effective electron blocking layer without changing turn-on voltage and emissive spectra. As a result, the optimized QLED with 5nm PEIE shows a ~36% improved current efficiency of 70cd/A and external quantum efficiency (EQE) of 17.3% compared to the QLED without PEIE. In particular, the maximum brightness of the optimized QLED dramatically improved by a factor of 2.3 relative to the QLED without PEIE. The main reasons for these QLED performance improvement are due to the suppressing the leakage current across the device and well confined exciton by inserting PEIE layers.

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Bright Colloidal Quantum Dot Light-Emitting Diodes Enabled by Efficient Chlorination

The external quantum efficiencies (EQEs) of state-of-the-art colloidal quantum dot light-emitting diodes (QLEDs) are now approaching the limit set by the out-coupling efficiency. However, the brightness of these devices is curtailed by the use of poorly conducting emitting layers, a consequence of present-day reliance on long-chain organic capping ligands. Here, we report how conductive and passivating halides can be implemented in Zn chalcogenide-shelled colloidal quantum dots (CQDs) to enable high-brightness green QLEDs. We use a surface management reagent, thionyl chloride (SOCl2), to chlorinate the carboxylic group of oleic acid and graft the CQD surface with passivating chloride anions. This results in devices with an improved mobility which retain high EQE in the high injection current region and also feature a reduced turn-on voltage of 2.5V. The treated QLEDs operate with a brightness of 460,000 cd m\(^{-2}\), significantly exceeding that of all previously-reported solution-processed LEDs.

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Ligand exchange of quantum dot induced by thiophenol derivatives for all-solution processed inverted quantum-dot light emitting diode

Quantum dot light emitting diodes (QLEDs) are referred as promising new display devices because of high color purity, wide color gamut and solution processability. Inverted structure QLEDs can be suitable for display applications because they can be fabricated on the cost-effective n-type integrated circuit. However, reported inverted structure QLEDs have shown lower external quantum efficiency (EQE) than conventional structure. In this work, native oleic acid (OA) ligands on CdSe@ZnS/ZnS QDs are partially exchanged to 4-(dimethylamino)thiophenol (4-DMATP) ligands in order to enhance EQE of inverted QLED. QDs with 4-DMATP ligands show similar quantum yields compared to pristine QDs with OA ligands while color purity and emission wavelength are maintained. Negative dipole of 4-DMATP ligands results in up-shifted valence band maximum of QDs. QDs with 4-DMATP ligands have 0.36 eV higher valence band maximum than QDs with OA ligands. QLEDs were fabricated with the layer structure of ITO/ZnO/QDs/PEIE/Poly-TPD/MoOx/Al. PEIE results in further up-shifted valence band maximum of QDs with 4-DMATP ligands. QLED structure shows reduced energy barrier between QDs and Poly-TPD to close to 0.04eV for hole transport. QDs with 4-(dimethylamino)thiophenol show improved EQE of 16.3 % and maximum luminance of 43500 cd/m² while QDs with oleic acid ligands show 5.29 % and 33300 cd/m². These results suggest that proper ligand exchange is an effective way to improve efficiency in solution-processed inverted QLEDs.

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Improved Performance of Cadmium-free Quantum Dot Light Emitting Diodes by Hybrid Quantum Dot with Semiconducting Polymer

Quantum dot light emitting diode (QLED) has emerged as one of candidates for next generation display with various advantages such as narrow emission bandwidth, great photo-stability, and tunable emission wavelength owing to size dependency. Despite rapid improvement and promising outlook, most of researches of QDs and QLEDs characteristics have been focused on cadmium-based QDs which have toxicity and are strictly restricted for use. For that reason, researchers started to focus on cadmium-free QDs such as InP, ZnSe and CuInS2, but limited studies were reported [1]. In this work, we demonstrated green QLEDs with the hybrid quantum dot which composed pf InP/ZnSeS/ZnS QD and semiconducting polymer. The semiconducting polymer is consisted of the carbazole group which has superior hole conducting properties and relatively low highest occupied molecular orbital (HOMO) and the disulfide moieties which have high affinity to Zn centers. The hybridization proceeds as the ligand exchange procedure. The oleic acid ligands which are initially coordinated to the QDs were replaced by the semiconducting polymer. Compared to the origin QDs, increased inter-dot distance in the hybrid QDs effectively suppresses the energy transfer (ET) among QDs. Furthermore, hybrid QDs have better nanoscale morphology [2]. As a result, we succeeded in fabricating cadmium-free InP QLEDs with the improved EQE of 2.5. We believe that our approach will provide insights into developing highly efficient cadmium-free QLEDs.


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Silicon nanocrystals in polymers and in Transparent Wood for light converting applications

Silicon nanocrystals largely preserve indirect bandgap structure of the bulk material. Corresponding light emission properties, such as long emission lifetime and a large Stoke shift, combined with high quantum efficiency, make them interesting potential candidates for a number of light-converting applications.

Here we present fabrication and characterization of bulk nanocrystal/polymer hybrids. Si QD/PMMA composites prepared using different size silicon nanoparticles show luminescence in deep red, orange, yellow and blue spectral ranges, consistent with quantum confinement [1]. Quantum efficiency was in the range of 30-40% and does not change upon transition from solution to solid phase.

Successful dispersion of Si QDs in PMMA led us towards application of this nanocomposite to transparent wood matrix [2]. After removing lignin-containing components wood material becomes colorless. Subsequent impregnation of cell wall cavities with a refractive index matching polymer, such as PMMA, makes this bio-composite transparent, while maintaining its mechanical properties. When quantum dot-rich PMMA is used a luminescent transparent wood material is formed. It reveals a high degree of scattering, which is fiber orientation dependent.

Finally, to explore quantum efficiency and integration limits we tested off-stoichiometry thiol-ene polymers as a host matrix. It is known for aggressive radicals, fast polymerization rate (seconds), and facile integration with glass due to low polymerization shrinkage [3]. Stable quantum efficiency of 60-70% for Si QDs was obtained and smooth QD/polymer laminates on glass fabricated.


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Stability Enhanced Benzyl-Alcohol Treated CH$_3$NH$_3$PbBr$_3$ Nanocrystals for Optical Gain Applications

Solution processed Organic-Inorganic hybrid Perovskites (OIHP) nanocrystals are a new class of novel light-emitting materials featuring high photoluminescence quantum yields (PLQY), low amplification thresholds and cheap synthetic routes. While highlighting the promise of cost-effective, narrow-width emissions and spectral tunability, the low reaction yield and structural instability of these nanocrystals unfortunately exacerbates their sensitivity to ambient moisture (that negatively impacts the PLQY and limits the integration into “daily-use devices”). In this work, we attempt to address the stability issues of OIHP CH$_3$NH$_3$PbBr$_3$ nanocrystals by considering Benzyl-Alcohol (BnOH) treatment in the Ligand-Assisted Re-Precipitation (LARP) protocols. Our main hypothesis was to strengthen the ligands bound onto the nanocrystal’s surface so as to form a strong passivation layer that protects the nanocrystal. This feat was achieved by allowing the nanocrystals to directly precipitate under the presence of BnOH in the “Poor-solvent”. As a result, Monodisperse and ambient-stable CH$_3$NH$_3$PbBr$_3$ nanocrystals of ~11nm were formed. Through detailed spectroscopic studies, we observed near-unity PLQY, high optical modal gain ($520$cm$^{-1}$), consistent absorption-cross sections with reported values and consistent ASE thresholds from drop-casted films of BnOH-treated nanocrystals without significant degradation of material quality up to 4 months in ambient conditions (>60 %RH).

Due to the consistency of optoelectronic properties and various improvements to ASE thresholds as compared to untreated (control) nanocrystals, we conclude that BnOH-treatment do not alter the intrinsic properties but simply play the pivotal role in surface passivation by steering certain preferred ligand binding which likely point to an enhanced surface passivation scheme. With the enhanced ambient stability, it renders the treated nanocrystals ideal for low-threshold lasing, upconversion lasing and other light-emitting optoelectronic applications.

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Fabrication of quantum dot/photoresist nanocomposites using the wrinkled silica-quantum dot hybrid particles

Quantum dots (QDs) have been widely used as phosphors in the liquid crystal display (LCD) using light emitting diodes (LED) as a backlight. In many ways to incorporate QDs within display panel, ‘in panel’ type which embeds QDs into the glass panel has recently drawn many researcher’s interest. It has advantages over other types because QDs are located away from heat source (blue LED) to prevent reduction in efficiencies. Therefore, it is expected to increase efficiencies of the device by 50% compared to the conventional LCDs. However, there still remains some problems. Although photoresist must be used for patterning, bare QDs cannot be distributed in the photoresist. To overcome this limitation, we use wrinkled silica nanoparticles (WSNs) as template particles for not only dispersing QDs but also enhancing recycle of light. WSNs can be synthesized using the water-oil surfactant system.1 This unique surface structure scatters light more than the spherical silica nanoparticles. To use WSNs as host particles to disperse QDs into film, their surface was modified with glycidoxypropyl trimethoxysilane. Then, QD-WSN hybrid particles (WSQs) were prepared by embedding QDs into WSN using swelling method.2 Thus, we could fabricate photoresist nanocomposites containing WSQs. Using various techniques such as electron microscopy, UV-vis spectrophotometer and fluorometer equipped with integrating sphere, we characterized particles and nanocomposites. We believe this study suggest new ways to disperse QDs in photoresist film and enhance the efficiencies of nanocomposites.


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New Technologies for Colouring in Displays

Colloidal quantum dots (QDs)-based light-emitting diodes (QLEDs) has been actively researched due to the potential impacts to the display and lighting industry base on the unique properties of QDs itself such as size-dependent bandgap tuneability, narrow emission spectrum, and low-cost solution-based processing. However, the most of promising results in the past used cadmium (Cd) contained II-VI semiconductor nanocrystals. Therefore, a considerable future task is to substitute Cd-containing QDs with less toxic materials. InP-based QDs is a promising material among the III-V semiconductor nanocrystals and it has been already applied in conventional and inverted QLED structure.

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Cd-free quantum dot sheet for flexible illumination with the high color rendering index

The next generation light source is not just simple illumination of the light but it should be in harmony with its surrounding environment and have a decorative effect when it is off, and it should provide the sunlight-like or moonlight-like lighting when turned on. To achieve this, the illumination style is expected to be design-in or built-in light. Also it would be a flexible illumination with light quality being glare-free, tunable color temperature, thin and lightweight. In this study, cadmium-free quantum dots were used as the light source of flexible illumination. Quantum dots hybrid illumination is optimal for indoor lighting as it allows high color rendering index. we optimized the synthesis technique of non-cadmium nanoparticles. By using the ligand exchange technique of the nanoparticles, the dispersion property with the monomer was improved and the compounding ratio was optimized to develop a nano-sheet having excellent quantum efficiency. The quantum dot hybrid lighting was fabricated by applying a quantum dot sheet onto a blue LED. The luminance uniformity and the color rendering index of the flexible sheet were 98% and Ra 92 or more, respectively.

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High Efficiency Solid-State-Ligand-Exchange Free Quantum Dot Ink-based Solar Cells

Colloidal quantum dot based solar cells (CQDSCs) promise low-cost, bandgap tunability, solution-processed, low temperature, and air-stable photovoltaics for next generation power source. Surface chemistry and device engineering optimization lead to power conversion efficiencies (PCE) up to 11.3%. However, CQD device fabrications were almost exclusively by a combined process of in-situ solid-state ligand exchange (SSE) with multiple layer-by-layer (LbL) deposition, especially thiol-passivated CQD layer for hole transport layer (HTL), which has been a major obstacle to high-throughput processing. In this work, we developed hole transport layer that can be directly coated on top of CQD ink active layer for highly efficient CQDSCs without using either SSE or LbL deposition method. The SSE-free device exhibits a PCE of ~11%, which comparable to the current state-of-the-art device performance. Owing to SSE-free deposition methods, the device fabrication by high-throughput processing was feasible with preliminary result of >10% efficiency. This result suggests its potential use in various CQD based optoelectronic devices with industrially-friendly processes.

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The role of hydro/oxo-phobic top hole-selective layer for efficient and stable colloidal quantum dot solar cells

Here, we explore the underlying mechanisms by which doped organic thin films as a top hole-selective layer improve the performance and stability of colloidal quantum dots (CQDs) solar cells. The molecular dynamics-based theoretical studies prove that the hydro/oxo-phobic properties of the organic hole-selective layer serve to efficiently cover the CQDs solid. Moreover, the robust and outstanding electrical properties of the hole-selective layer, simultaneously ensures a high power conversion efficiency (PCE) and increases the stability performance of CQDs solar cells. As a result, a best PCE of 11.7 % in a lead sulfide (PbS)-based CQDs solar cell is achieved and over 90% of initial performance is ensured after 1 year storage under ambient conditions.

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Degeneracy of Discrete Electronic States of Colloidal Quantum Dots for Thermoelectric Device

Investigation of carrier transport and electronic properties of highly-crosslinked ligand-exchanged colloidal quantum dot (QD) assemblies is vital for the utilization of these class of materials in diverse emerging practical applications. So far, their tunability of energy bandgap and the discrete energy levels formation have attracted great interest in their applications in many devices where optical properties are primarily important (e.g. photovoltaics, displays [QLEDs] and even laser). On the other hand, QD assemblies are prospective for thermoelectric applications, since low-dimensionality leads to enhanced Seebeck effect and suppressed thermal conductivity. For those applications, some certain doping levels are required; thus knowledge on how many electrons can sit for each QD discrete energy levels is vital.

To understand electronic state degeneracy in QD solids is challenging since it will be strongly influenced by the energetic landscape of the quantum confinement, in addition to the compound type. So far, limitations exist in elucidating degeneracy information only from the optical characterization of the QD solids.

Here we demonstrate the probing of the electronic state degeneracy of QD solids via measurement of electronic transport. High carrier density accumulation in ionic-liquid-gated PbS QD transistors enable sufficient of the Fermi level beyond quantum dots’ HOMO-LUMO, thus accessing the discrete energy levels to fill each of them. From the correlation of the carrier density and QD density in the assembly, the number of accumulated electrons per individual QD can be counted to obtain the numbers of electronic state degeneracy for each discrete energy levels. Through this probing, we can know how much dopant is necessary to dope the QDs up to some certain levels required for many applications and to formulate various doping strategies. This includes the realization of thermoelectric devices with enhanced Seebeck coefficient due to the filling of the discrete energy levels.


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Quantum Dot-Based Systems for Photocatalytic Hydrogen Generation

Colloidal semiconductor quantum dots (QDs) have shown great promise as photocatalysts for the production of chemical fuels by sunlight. The production of H₂, a clean-burning fuel, is investigated with CdSe QD photosensitizers and molecular catalysts. The catalyst concentration dependence of H₂ production was studied with large diameter (4.4 nm) CdSe QDs and a nickel-dihydrolipoic acid (Ni-DHLA) catalyst. While exhibiting excellent robustness and longevity, the H₂ production efficiency was relatively poor at low catalyst loadings, but the efficiency was substantially increased with increasing Ni-DHLA:QD molar ratios. Ultrafast transient absorption was used to probe the electron transfer (ET) dynamics in order to better understand the trends in H₂ production efficiency. Though the ET rate showed no substantial change with catalyst concentration, the efficiency of ET was found to increase at increased catalyst concentrations, consistent with H₂ production activity. Together, these results suggest that higher catalyst concentrations increase the probability of ET from QDs to Ni-DHLA, overcoming the relatively small driving force for ET and decreased surface electron density for large diameter QDs. A photocatalytic system using 2.8 nm CdSe QDs and a biologically inspired Co-microperoxidase-11 (Co-MP11) catalyst demonstrated excellent activity for H₂ production, yielding well over 100,000 turnovers (with respect to the catalyst) and exhibiting activity for over 100 hours. Interestingly, with DHLA-capped QDs, the Co-MP11 catalyst exhibited poor activity, but with the use of glutathione (GSH) or mercaptopropionic acid (MPA) ligands, this enhanced activity was observed. Additionally, high activity was observed for GSH- and MPA- capped QDs in the presence of CoCl₂, suggesting the formation of an active catalyst in situ. Altogether, these studies highlight the importance of catalyst-ligand compatibility, allowing for robust activity and long-lived performance in QD-based photocatalytic systems.

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Hybrid passivated colloidal quantum dot inks for tailoring the band edge positions

Colloidal quantum dots (CQDs) are of interest for optoelectronic applications due to their bandgap ($E_g$) tunable through the quantum size effect and their solution-processing, ambient stability, and uniform film formation without lattice-matching requirements. Rapid progress on surface passivation and device structure has shown great improvements on optoelectronic devices based on CQDs such as photovoltaics, light emitting diodes, and photodetectors. Energy level of semiconductors determine the functionality of optoelectronic devices. By engineering the energy level of the semiconductors, the efficiency of the devices varies greatly. Recent development of CQD inks by solution-phase ligand exchange enables the high performance and single-step process of CQD solids. However, most of CQD inks are halide-passivated, and CQD inks with organic ligands showed poor performance and air stability. Here, we develop the hybrid passivation strategy for CQD inks that involves introducing the organic ligands as a post-treatment which can tune the band edge position of CQD inks through ligand dipoles.

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Chloride Passivated ZnO Electron Transport Layer for Efficient Colloidal Quantum Dot Photovoltaics

Colloidal quantum dots (CQDs) are promising materials for solution-processed optoelectronics. State-of-art CQD device architectures rely on a planar heterojunction between the CQD absorber layer and an n-type metal oxide semiconductor, typically ZnO. This heterojunction establishes a depletion region for efficient charge transport and separation. Unfortunately, several features of the ZnO material today limit overall device performance. These include abundant surface defects; and an unfavorable electronic band alignment. These lead to interfacial recombination that harms both current and voltage, and simultaneously achieving good passivation of the ZnO and a favorable band alignment with the photoactive CQD layer for efficient charge extraction has – until now – been an unresolved problem.

Here we report a new and facile surface passivation method for ZnO nanoparticles that employs solution-phase chloride (Cl) treatment. The surface defect sites of the ZnO nanoparticles are efficiently passivated using this Cl-treatment strategy, thereby reducing the surface trap density. In addition, the Cl passivated ZnO (Cl@ZnO) exhibits excellent electrical properties such as a deep conduction band and an upwards shift of the Fermi level compared to conventional ZnO, both of which are favorable to electron extraction. With these benefits, the CQD photovoltaics using Cl@ZnO demonstrates a superior power conversion efficiency (PCE) of 11.6% (11.4 ± 0.2%) compared to 10.6% (10.4 ± 0.2%) for a device with conventional ZnO.


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**Industrially Feasible, Air-stable Solar Cell by Using Low-cost, Monodisperse, Green PbS Quantum Dots**

Recently, Quantum dot sensitized solar cells (QDSSCs) have received much attention among the alternatives of dye-sensitized solar cells (DSSCs). Between the QDSSCs, near-infrared (NIR) QDs attracted significant interest in the past decade due to their exceptional advantages. As a NIR QDs, PbX (X = S, Se, Te) QDs is mostly used materials for QDSSCs due to their properties such as multiple exciton generation, wide bandgap, strong absorbance and luminescence in the NIR region. [1] Due to this strong confinement regime, PbS QDs exhibit high quantum yield (QY) up to 90%, which is hardly possible with traditional semiconductors. [2] Therefore, small effective masses of charge carriers and the large Bohr radius of PbS QD provides charge delocalization in QD films to increase charge carrier mobility and the conductivity.[3] Up to yet most of the studies on PbS QDSSCs have been relying on synthetic routes involving highly toxic (TMS)2S and TOP precursors. Generally, thioacetamide (TAA) is used for aqueous PbS QD synthesis, due to the low solubility of TAA in most used organic solutions in PbS synthesis, such as 1-octadecene (ODE) and oleylamine (OLA). [4, 5] In this work, we describe a facile and optimized approach based on PbO as lead and TAA and sulfur precursors to synthesize NIR PbS QDs. This combination of precursors is the first for the solar cells in the literature, which makes synthesis unique and instead of using toxic and airsensitive precursors we used relatively green synthesis approach. Moreover, 3.5 g PbS QDs obtained from one pot reaction, which is relatively huge amount of QDs with small synthesis volume of the PbS QDSSC literature. Our purpose is produce colloidal PbS QDs and QDSSC by using non-air sensitive and relatively low toxic chemicals and without using a glove box in these processes. Obtained PbS QDs used to fabricate industrially feasible, stable, efficient QDSSC. Unlike the works in the literature, both QD synthesis and device fabrication are done without glovebox use, which is a proper fabrication for big scale manufacturing.

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Eco-friendly colloidal quantum dot solar cells fabrication

Recently, Pb- and Cd-based colloidal quantum dot solar cells are attracting attention with high efficiency (over 11%) and stability. These quantum dots, however, should be replaced as the non-toxic materials due to its high reactivity and high surface area to volume ratio. The size effect of Pb and Cd nanostructure should be much fatal to human body. The ternary I-VI type quantum dots are worthy substitute which has eco-friendly materials and low-cost fabrication. The AgBiS₂ nanostructure solar cell was already reported showing 5% power conversion efficiency.<sup>[1]</sup> Herein, we investigated the characteristics of various eco-friendly quantum dots and fabricated the full solar cell devices. Ternary and other Pb- and Cd-free colloidal quantum dot is used for high efficiency green solar cells.<sup>[2]</sup><sup>[3]</sup>


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Solution-Phase Ligand Exchange of Colloidal Quantum Dots for Enhanced Photovoltaics

Colloidal quantum dots (CQDs) are a promising semiconductor for a variety of electronic and optoelectronic applications such as field-effect transistors, light-emitting diodes, photodetectors, and photovoltaics because of their size-tunable bandgap and solution processibility. In particular, lead sulfide (PbS) CQDs have promise in their potential to absorb and harvest an infrared (IR) light energy from the full solar spectrum, and therefore are attractive as a light-harvesting material for solar cells. The development of surface chemistry-based modifications in CQDs has enabled tremendous progress in CQD solar cells with high photovoltaic performance. Recently, lead halide-passivated CQD ink, which is prepared using the solution-phase ligand exchange, is so far known as the most favorable way for single-step, large-scale fabrication and high performance of CQD solar cells. Here, we show the developed approaches for CQD solar cell fabrications based on solution-phase ligand exchanged CQD inks, enabling the improvement of photovoltaic performance. As a first, we demonstrate a new solution process for the fabrication of nanostructured CQD solids, which can be used as a back interface in bottom-illuminated CQD solar cells. The nanostructured CQD solid has a densely and uniformly packed morphology of round-shaped motifs composed of only CQDs, and shows improved optical absorption at the interface of between the CQD layer and gold electrode. Next, we show a reactivity-controlled solution-phase ligand exchange of CQDs, resulting in improved polydispersity in the CQD solids and open-circuit voltage in the CQD solar cells.

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Optically engineered multi-junction Infrared quantum dot solar cells augmenting power points of perovskite- and crystalline silicon solar cells

Colloidal quantum dots (CQDs), which can harvest the infrared (IR) part of the solar spectrum, are ideal tandem partners to augment the performance of crystalline silicon (cSi) and perovskite solar cells. However, state-of-the-art CQD solar cells incorporating a single layer of light harvester have hindered efficient solar-to-energy conversion due to the limited light absorption. Here, by developing a new strategy to disperse PbX (X = I, Br)-passivated PbS CQD in non-polar solvents, we successfully demonstrated efficient multi-junction (MJ) IR devices with optically engineered combinations of CQDs. The optimized optical engineering and cascade charge transfer between different bandgap \( E_g \) CQDs enabled the efficient IR multi-junction solar cells exhibiting among the highest ‘additive’ power conversion efficiency of 3.2% using a perovskite filter (for \( E_g \approx 0.97 \text{eV}/1.13 \text{eV} \) MJ CQDs) and 1% using a cSi filter (for \( E_g \approx 0.90 \text{eV}/0.97 \text{eV} \) MJ CQDs), respectively. The method provides a new pathway to overcome the Shockley-Queisser efficiency limit of mature Si-based photovoltaics (PVs) as well as emerging low-cost perovskite PVs, thereby to further reducing the cost per watt of solar electricity.

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Colloidal quantum dot-based photovoltaics with highly transparent short-wave infrared conductive oxides

Development of ultra-high efficiency solar cells using low-cost solar cell technology such as low temperature and solution-based methods is the ultimate goal of solar energy uses. The utilization of photon energy in a wide range of solar spectrum is also a critical issue. PbS colloidal quantum dots (CQDs) are strong candidates for the active materials for near infrared and short-wave infrared photovoltaics (0.8 μm - 2 μm). However, in the wavelength region, the free carrier absorption of transparent conductive oxide (TCO) layers such as F-doped SnO₂ (FTO) causes non-negligible energy loss of the incident solar energy [1]. Therefore, highly transparent near infrared conductive oxides are required to increase the efficiency of the solar cells. We explored low free carrier density but high mobility TOCs in order to keep as high a conductivity as that of the FTO substrate. We then focused on high-mobility Ta-doped SnO₂ (TTO) thin films grown on glass substrates by pulsed laser deposition using polycrystalline anatase TiO₂ seed layers. The TTO films showed a transmittance of approximately 80% in the near infrared and short-wave infrared regions, and a sheet resistance of 12 Ω/sq.

PbS CQD/ZnO NW solar cells (TCO/ZnO NW-PbS QD hybrid (1.2 μm) layer/PbS QD overlayer (0.3 μm) /Au (0.1 μm)) were fabricated by using PbS CQDs that give the first exciton absorption peak at 1.6 mm [1]. The EQE of the exciton peak obtained on the solar cells with FTO substrates was approximately 20%, whereas the solar cells fabricated with the TTO substrates reached approximately 40% EQE. This 50% EQE enhancement is associated mainly with the high transmittance of the TTO layers used in our solar cells. The TTO films were confirmed to be promising TCO materials for PbS CQD-based infrared photovoltaics.


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Quantum dot derived catalysts

Atomic vacancies contribute to electronic defects in quantum dots, a notorious challenge in solution-processed optoelectronics\textsuperscript{1-4}. In contrast, defect sites are often proposed as key active sites in the design of catalysts as they may lower the energy barrier for the electrosynthesis of desired chemical products\textsuperscript{5-11}. For metal-based catalysts such as those employed in CO\textsubscript{2} electroreduction, a promising strategy to improve activity would be to achieve a high density of homogeneously dispersed atomic defects; yet it is seldom achieved in pure metals\textsuperscript{10}. We hypothesized that vacancy-rich catalysts could be obtained by synthesizing quantum dots (QDs) and electrochemically reducing them during the CO\textsubscript{2} reduction reaction (CO\textsubscript{2}RR). Here we report quantum dot derived catalysts (QDDCs) with up to 20 vol\% of vacancies that - compared with previously reported CO\textsubscript{2}RR catalysts - achieve the highest current density in electrosynthesis of formate, carbon monoxide (CO) and ethylene at the low potentials of \textendash 0.2 V, \textendash 0.3 V and \textendash 0.9 V versus reversible hydrogen electrode (RHE), respectively\textsuperscript{12-16}[PDL1]. The materials are stable following 80 hours of CO\textsubscript{2}RR. This performance in CO\textsubscript{2}RR electrosynthesis in aqueous solution surpasses by 5x previously-reported metal and metal oxide catalysts\textsuperscript{12-16}, showing promise for practical industrial applications. X-ray absorption spectroscopy (XAS) and computational studies, taken together, reveal that the vacancies produce a local atomic and electronic structure that enhance CO\textsubscript{2}RR.

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Luminescent quantum dots in the cellulose nanocrystal matrix for solar energy harvesting

Luminescent solar concentrator (LSC) is promising for harvesting solar energy thanks to its capability of collecting both direct and diffused sun light without any optical component. Colloidal quantum dots are of interests for use in LSC for their broadband absorption above semiconductor bandgap and tunable Stokes-shift for minimizing the light reabsorption in LSC. Recently, cellulose nanocrystals (CNCs), wood-derived eco-friendly materials, have been demonstrated as matrix and waveguide materials in LSC. Here we propose to incorporate quantum dots into CNC films for efficiency solar energy harvesting. Previously reported luminescent quantum dots in LSC contain non-polar organic ligands that can be easily incorporated into organic matrix, such as PMMA, however, these organic ligands put challenges in processing quantum dot/CNC films since no solvents are available to dissolve both quantum dot and CNC simultaneously. In this paper, we have developed a method to produce quantum dot/CNC suspension by applying solution-phase exchange to replace non-polar organic ligands to the metal halide complex as well as performing acetylation on CNC surfaces. It is worth noticing that the fabricated quantum dot/CNC film keeps similar photoluminescence quantum yield and lifetime compared to as-synthesized quantum dot in non-polar organic solvents. The developed method here has the potential to be widely used to mix quantum dot with other materials having polar functional groups.


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**Direct nanostructuring of PbS quantum dot films for improved solar cells**

Lead sulphide quantum dots (PbS QDs) are promising materials for optoelectronic devices due to their tuneable bandgap, earth abundant constituents and solution processability. In particular, they are attractive candidates for solar energy conversion, as their bandgap can be tuned over the optimal range for the incident solar spectrum. Solar cells made from PbS QDs have improved steadily since their inception, and have achieved a record certified efficiencies over 12%. In quantum dot solar cells (QDSCs), the thickness of the QD absorbing layer is limited by the charge extraction length, which is typically much shorter than the light absorption length. Hence it would be desirable to increase the path length in the absorbing layer while retaining or improving charge extraction from the film. Reports of ordered bulk heterojunctions incorporating nanostructured n-type layers, such as ZnO nanowires, have shown great potential for improving charge extraction and allowing thicker absorbing layers. However, they suffer from lack of control over the growth process and increased series resistance due to the extended interface. In this work, we develop a direct nanoimprinting technique to pattern QD active layers in QDSCs. Directly patterning the QD film with wedge structures can increase the path length of light within the film and improve charge extraction. We obtain well defined wedge patterns as seen under SEM, and characterise the films after imprint. We then investigate the performance of imprinted QDSCs.

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**Water-Resistant Property of AgBiS2 Film for Next Generation Solar Cells**

Although organic, quantum dots and perovskite solar cells, have been regarded as next generation solar cells because of high device efficiency and low cost for device fabrication, they are challenging for device stability under ambient condition. Additional encapsulation process should be applied to improve the device stability. Therefore, it is required to develop intrinsically water-resistant semiconductors. We confirmed AgBiS\(_2\) has water resistant nature through measuring X-ray diffraction spectroscopy (XRD) and X-ray photon spectroscopy (XPS). These results supported water resistance of AgBiS\(_2\) by showing no changes in crystallinity and chemical bonding energy. To evaluate its water resistance in real device, we fabricated solar cell by using AgBiS\(_2\) as the light harvesting layer. In device fabrication, we compared water treated AgBiS\(_2\) film with non-treated one. The device including water treated AgBiS\(_2\) film showed comparable power conversion efficiency of 4.02 % to that of the control device, 3.97%. Finally, through x-ray spectroscopy and device fabrication, we found that AgBiS\(_2\) film have a water-resistance as a candidate for stable next generation solar cell.

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Ink manipulation for efficient large-area colloidal quantum dots solar cells

Colloidal Quantum Dots (CQD) are one of the most promising materials used to fabricate solar cells, thanks to their relatively easy band-gap manipulation and consequently well-controlled absorbance spectrum [1]. Nonetheless, one of the issues CQD-based devices face is lack of large-scale fabrication technologies that would help in commercializing this exciting technology. This research focuses on creating CQD inks and development of fabrication methods that offer excellent large-scale fabrication capabilities without compromising the performance of the solar cells. The proposed solution provides a framework to fabricate large-scale solar cells (>49cm²), and involves a blade-coating printing technique that allows fabrication of thick active layer devices in a fast and inexpensive manner [2]. Specifically, the influence of various ink compositions and their influence on fabrication and the final device performance is analyzed. In this work, n-Butylamine is used as the main solvent, which is mixed with various higher boiling-point co-solvents in order to improve the drying process. Among a wide variety of co-solvents, N-Methyl-2-pyrrolidone demonstrated the best results in terms of creating thick and smooth active layer films. Detailed chemical and physical analysis of the fabricated films confirms that careful tuning of the ink composition together with appropriate blade-coating techniques allows fabrication of densely packed films of quantum dots that serve as an active layer of the CQD solar cells. Finally, this research includes fabrication of the full solar cell stack and analysis of the interactions between the various layers. This work provides an invaluable set of information on how to fabricate large area CQD solar cells that could be utilized by both scientists and engineers in the field of quantum dots-based devices.

2. Hongta Yang and Peng Jiang, Large-Scale Colloidal Self-Assembly by Doctor Blade Coating, Langmuir 26, 16, 13173-13182

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Pseudohalide-Exchanged Quantum Dot Solids Achieve Record Quantum Efficiency in Infrared Photovoltaics

Colloidal quantum dots (CQDs) are promising materials for use in solar cells due to their tunable bandgap, via the quantum size effect, that enables broadband harvesting of the solar spectrum. Although only thin films of CQD absorber layers are required for capturing most of the solar spectrum, extraction of photo-excited carriers generated throughout the entire length of CQD films is limited by carrier trapping. Since as-prepared PbX (X = S, Se) CQDs are nonstoichiometric and lead-rich, they are particularly susceptible to having mid-gap trap states. In the latest record-performing devices, improvements have arisen mainly from judicious management of the CQD surface stoichiometry. Halide anions (Cl\(^-\), Br\(^-\), I\(^-\)) and molecular halides (Cl\(_2\), I\(_2\)) have been used to reduce trap state density, leading directly to improved PCE. We took the view that the success of halides in chalcogenide quantum dot passivation merited renewed explorations of the potential for pseudohalides (SCN\(^-\), CN\(^-\)) to achieve greater performance by reducing trap states. Preliminary evidence also indicated that mixed-anionic-passivant strategies can outperform strategies that rely only on a single class of ligands. In our study, we combined the thiocyanate anion with halides during solution exchange to achieve a hybrid surface passivation. Pseudohalides, as hybrid passivating agents, offer strong chemical interactions with the CQD surfaces, as evidenced by observations of the thiocyanate stretching transition in the infrared spectra for exchanged CQDs. The hybrid surface passivation effects on CQD films were investigated using field-effect transistor studies. We extracted the density of trap states from transfer characteristics and found that the trap density of the hybrid film was \(~4\) times lower than that of CTL films. The reduced densities enhanced transport lengths and so enabled record-thickness high-performing CQD films. These devices exhibited the highest external quantum efficiency (EQE) of 80% at the excitonic peak. The corresponding PCE reached 11.2%.

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Cost-effective and Large-area Luminescent Solar Concentrator based on Colloidal Carbon Dots

Luminescent solar concentrators (LSCs) can serve as large-area sunlight collectors for photovoltaic (PV) cells, reducing the cost of PV generated power. Typical LSCs consist of optical waveguides doped with highly emissive fluorophores e.g. quantum dots (QDs) or dyes/polymers which are required to exhibit high optical efficiency and long-term stability. Compared to conventional fluorophores, carbon dots (C-dots) have superior advantages of non-toxicity, environmental friendliness, low-cost and simple preparation using abundant carbon based feedstock. Here, we demonstrate cost-effective large-area LSCs (up to 100 cm²) using colloidal Cdots.

Two types of LSCs were fabricated by either incorporating oil-soluble oleylamine-treated Cdots into photo-polymerized poly(lauryl methacrylate) (PLMA) or spin-coating the water-soluble C-dots/polyvinylpyrrolidone (PVP) mixture on the glass substrate. LSCs based on C-dots/PLMA exhibit a quantum efficiency of 5% (geometric (G) factor of 45) and a 1.1% optical conversion efficiency (100 cm² , G factor of 12.5) of thin-film LSCs based on C-dots/PVP was achieved under one sun illumination. This performance is comparable to those of LSCs based on inorganic QDs with similar G factor. The LSCs based on C-dots are highly air-stable without any noticeable variation in photoluminescence under UV illumination (1.3 W/cm² ) for over twelve hours.

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MidGap-State-Mediated Photoelectrochemical Hydrogen Generation in CdSe Quantum Dots/β-Pb0.33V2O5 Nanowires Heterostructures

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Size Dependence of Photocatalytic Hydrogen Generation for CdTe Quantum Dots

Colloidal semiconductor quantum dots (QDs) have shown great promise as photocatalysts for a variety of chemical transformations including oxidations, reductions, and most recently carbon-carbon bond forming reactions. QDs inherently possess several desirable properties for photocatalysis including size tunable oxidation and reduction potentials, which are largely independent of surface chemistry. We recently found that CdSe QDs and simple aqueous Ni2+ salts in the presence of a sacrificial electron donor form a highly efficient and robust system for photochemical reduction of protons to H2. CdTe QDs have attracted growing interest for solar photochemistry applications due to the possibility for near-infrared absorption with larger diameter particles, thus absorbing covering more of the solar spectrum. The photocatalytic H2 generation from different sizes of dihydrolipoic acid (DHLA) capped CdTe quantum dots in aqueous solution was evaluated and compared to CdSe QDs. Even for the optimum size, CdTe QDs were 10 times less active for H2 production than CdSe QDs, which we attributed to rapid oxidation of the CdTe QD surface. We also found an unusually size dependence with CdTe QDs; specifically, a decrease in the H2 production efficiency was observed below a critical QD size. The origins of this size dependence will be presented.

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Improving the performance of organic solar cells through the addition of silver nanoparticles

We report the fabrication and performance of organic solar cells incorporating silver nanoparticles of different sizes and the comparison of their performance with the polymer solar cells of standard multilayered structure. The bulk-heterojunction photovoltaic devices adopted in this study consist of a basic structure including poly(3-hexylthiophene) (P3HT) as a donor, [6,6]-phenyl-C61-butyric acid methyl ester (PCBM) as an acceptor, and a highly conductive polymer as the anode, i.e., poly(3,4-ethylenedioxythiophene): poly(styrenesulfonate (PEDOT:PSS). All devices have been coated with aluminium layers as the cathode. To demonstrate the effectiveness of the silver nanoparticles (Ag-NPs) in the performance of the polymer solar cells, the following aspects have been studied: (1) Investigation of the performance of the organic solar cells with the active layer doped with Ag-NPs (ITO/PEDOT:PSS/P3HT:PCBM:Ag-NP/Al cells) to reveal the changes in the power conversion efficiency (PCE) and the fill factor (FF) of the devices from the current density to voltage (J-V) characteristics. It was found that an increase of 5.4% and 21% in the PCE and FF, respectively, has been achieved for the solar cells doped with 5 nm Ag-NPs. (2) Probing the size effects experimentally, in which Ag-NPs of different sizes, ranging from 5 nm to 30 nm, had been adopted in the active layer of the solar cells. It was found that the cells doped with 5 nm Ag-NPs gave the highest maximum PCE in this study. (3) Analysis on the mechanisms corresponding to the changes in the performance of the organic solar cells with the inclusion of the Ag-NPs.

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Tapered fiber interferometer coated with gold nanoparticles for biosensing

With salient advantages of compact size, light weight, immunity to electromagnetic interference and remote sensing, optical fiber sensors have been recognized as powerful devices to achieve biosensing and environmental monitoring. In this study, tapered fiber Mach-Zehnder interferometers coated with silica core-gold shell nanoparticles have been proposed and demonstrated to achieve biosensing. Two fiber tapers, fabricated with fusion splicing technique, have been used to form a tapered fiber Mach-Zehnder interferometer. The effects of varying different specifications of the tapers, such as waist diameter, separation distance between the tapers, and propagation direction, have been investigated. By using the layer-by-layer dipping method, multiple thin films have been deposited on the fiber structure for the detection of streptavidin which is a common target material used to test the effectiveness of a biosensor. The sensing mechanism here relies on the electrostatic attraction between cationic and anionic materials, in which the cationic material used in this study is poly (allylamin hidrocloride) (PAH), while the anionic material adopted here is SiO2 core/Au shell nanoparticles. The fabrication of the multilayered structures is followed by the functionalization with biotin which is a vitamin that possesses a strong binding ability with streptavidin and functions as an adhesive material to streptavidin. The sensing response of the interferometer on detecting aqueous solutions of streptavidin has been observed by measuring the shift in the wavelength of the transmission spectrum of the tapered fiber interferometer. The effects of depositing (PAH/SiO2:Au NPs) in multilayered structures, as well as the influences of different device specifications are revealed.

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Seeded Emulsion Polymerization as a Powerful Tool for the Biofunctionalization of Quantum Dots

With the polymer encapsulation of quantum dots via seeded emulsion polymerization, we present a powerful tool for the preparation of fluorescent nanoparticles with an extraordinary stability in aqueous solution. The method of the seeded emulsion polymerization allows a straightforward and simple in situ functionalization of the polymer shell under preservation of the optical properties of the quantum dots. These requirements are inevitable for the application of semiconductor nanoparticles as markers for biomedical applications. Polymer encapsulated quantum dots have shown only a marginal loss of quantum yields when they were exposed to copper(II) ions. Under normal conditions the quantum dots were totally quenched in presence of copper(II) ions.

The encapsulation techniques are based on the micellar encapsulation of quantum dots with PI-b-PEG on one hand and seeded mini emulsion polymerization on the other. In the first step, a partial ligand exchange with diethylenetriamine functionalized polyisoprene (PI-DETA) is performed, followed by addition of PI-b-PEG as surfactant and controlled transfer into an aqueous system. The inner hydrophobic part of the thereby formed micelles is crosslinked using radical initiators. Typical molar masses of the PI-b-PEG vary between 5 and 15 kDa. Various functional groups can be introduced as terminal endgroups of the PEG block allowing a large variety of bioconjugation techniques. Moreover, we were able to introduce branching of the PEG chain at the interface to the PI block.

Furthermore, a broad range of in situ functionalized polymer-coated quantum dots were obtained by addition of functional monomers or surfactants like fluorescent dye molecules, antibodies or specific DNA aptamers. This modification makes quantum dots highly attractive for biomedical applications.

The emulsion polymerization can also be used to prepare multifunctional hybrid systems, combining different species of nanoparticles within one construct without any adverse effects of the properties of the combined starting materials.

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**Development and Investigation of Ultrastable PbS/CdS/ZnS Quantum Dots for Biological Applications**

Near infrared (NIR) PbS-based quantum dots (QDs) have attracted much attention due to their unique optical properties and promising applications in many fields. In the past several years, our group has developed high quality PbS QDs, PbS/CdS core/shell QDs.1 Recently, we synthesized water dispersible PbS/CdS/ZnS core/shell/shell QDs for the first time by growing a biocompatible ZnS shell on the PbS/CdS core/shell QDs surface and simultaneously functionalizing the surface with mercaptopropionic acid ligands.2,3 These QDs demonstrated strong emission and excellent stability. In collaboration, they were applied for deep tissue imaging and exhibited interesting results: the penetration depth as deep as ~1.5 cm was achieved.

1) Ren F.; Zhao H.; Vetrone F.; Ma D. Nanoscale 2013, 5, 7800.

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Deep sub-wavelength excitation of single quantum dot using two-photon plasmonic nanofocusing

The large-scale mismatch between a single quantum dot (<10 nm) and the diffraction limit of light (~several hundred nanometers) prevents focusing an excitation laser at the scale of a single quantum dot. As a result, a huge number of carriers are excited, not only in the quantum dot, but also in the surrounding environment (e.g., a barrier), which leads to large background emissions and numerous dephasing processes. We present a demonstration of site-selective two photon plasmonic nanofocusing on a single quantum dot. Highly enhanced local electric field at the position of each single quantum dot, generated by the self-aligned plasmonic nanofocusing, leads to a large increase in the optical nonlinear effect (i.e., two-photon excitation) by a factor of 5,000 [1]. In addition, two photon plasmonic nanofocusing overcomes the limitation of diffraction limit of light by providing a deep sub-wavelength excitation of a single quantum dot. Selective excitation of quantum dot enables us to suppress background noises and dephasing processes, caused by surrounding carriers, which have hindered numerous practical applications. Here, as a result of suppressed background emissions, we demonstrated that single InGaN quantum dot emission can be investigated at near room temperature, which was not possible using conventional excitation methods. Our approach opens promising perspectives for quantum optics experiments with highly reduced background emissions and dephasing process.


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