Quantum dots for optoelectronics

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1 Introduction: Development of Quantum Dot Materials

Four decades ago, Ekimov and Efros embarked on their research into semiconductor-doped glasses and the formulation of theories to understand their characteristics.[1,2] Meanwhile, Louis Brus was delving into semiconductor particles within liquid colloids.[3] These two distinct research endeavors, as well as the efforts of synthetic chemists represented by Bawendi et al.,[4,5] eventually led to the development of fascinating quantum dot (QD) materials. QDs are tiny semiconductor structures that exhibit unique optical and electronic properties due to their nanoscale size which leads to the quantum confinement effects.[6] The invention of QDs, which truly harnesses quantum phenomena through size effects rather than purely material effects, has ushered in a new dimension for manipulating quantum phenomena. This innovation has sparked a revolution in physics, chemistry, and materials science, leading to a huge impact on the academic fields including optoelectronics and quantum technology. Today, the remarkably bright and pure luminescent properties of QDs make them a star in both academic and industrial contexts. Their significance is particularly evident in optoelectronic devices, encompassing applications such as light-emitting diodes (LEDs), lasers, solar cells, biosensors, and quantum light sources.

2 Two Classes of Quantum Dots: Colloidal and Epitaxial QDs

Confined by full-space potential, QDs exhibit well-defined quantized energy levels different from bulk materials.[6] It shows outstanding quantum efficiency, stability, and scalability. Setting them apart from non-quantum-confined nanocrystals, the emission wavelength can be tuned by controlling QD size, shape, and material composition. This tunability holds the potential for diverse QD-based optoelectronic devices used for various application scenarios. There are two well-established classes of optically active QDs, colloidal QDs and epitaxial QDs, according to their different preparation methods.

Colloidal QDs refer to tiny semiconductor particles dispersed in a solvent using a stabilizing ligand.[7] The most famous synthesis strategy for colloidal QDs to date is the hot injection method.[8] On the other hand, epitaxial QDs are nanoscale heterostructures embedded in a solid-state environment made of III–V semiconductors. They are typically grown through molecular beam epitaxy or metal-organic chemical vapor deposition on a semiconductor substrate under strict vacuum conditions. The most studied epitaxial QDs are self-assembled QDs, grown by the Stranski–Krastanow (S-K) mode.[9] Notably, colloidal QDs are room-temperature operable and much easier to prepare compared to most epitaxial QDs, which only work at low temperatures (except for GaN-based QDs). This gives colloidal QDs meaningful advantages in everyday light-emitting applications. However, the fluorescence linewidth of a single exciton in self-assembled QD can be as narrow as a few μeV at liquid helium temperature, rendering single epitaxial QD an ideal quantum emitter. Epitaxial QDs provide an ideal solid-state interface to bridge photons and electrons at the quantum level and a great platform to explore deep light–matter interactions.

Both kinds of QDs have been the focus of extensive research spanning over three decades, yet there has been limited collaboration and interconnection between the two research communities, which might be attributed to their different origins in physics and chemistry. For colloidal QDs, the main interest has been focused on improving luminescence yields and stability through chemical methods, while research on epitaxial QDs has predominantly focused on their subtle physical properties. Therefore, colloidal QDs have found extensive use in LEDs, optically pumped lasers, and solar cells, whereas epitaxial QDs have made strides in the development of sophisticated single-photon sources, entangled photon pair sources, and electrically pumped on-chip lasers.

3 Colloidal QDs

3.1 Colloidal QD Display and Lighting

Colloidal QDs shine brightly in the field of display and lighting devices, given their flexible bandgap control by the adjustment of QD size and composition, highly efficient emission due to the direct transitions, and excellent color purity derived from their discrete energy levels.[10] Importantly, colloidal QDs also hold cost-effective scalability and compatibility with typical semiconductor fabrications.

The first QD-LED was achieved in 1994,[11] when CdSe QDs were used. Although the first-generation device had a low external quantum efficiency (EQE) ranging from 0.001% to 0.01%, it underscored the importance of aligning energy levels between the emitters and electrodes for effective charge recombination. In 2002,[12] Coe-Sullivan and his colleagues successfully realized a QD-LED by sandwiching a QD layer between two distinct organic layers, TPD and Alq_3. The device exhibited a 25-fold enhancement in luminescence efficiency over the previous results. In 2004,[13] researchers successfully applied CdS/ZnS QDs to 460–480 nm LED devices with photoluminescent quantum yields (PLQYs) of 20% to 30%, though only with a low EQE of 0.1%. Further advancements were made with the introduction of an alloy structure, specifically CdSe/ZnS, which enabled a pure blue electroluminescence (EL) at 470 nm in QD-LEDs, with an improved EQE of 2.1%.[14] Since then, specialized QD structures like core/shell designs have been crafted to enhance PLQYs for the emission of three primary display colors – red, green, and blue.[15–18] The significantly

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developed PLQYs, coupled with efforts on device structure optimization, have led to state-of-the-art QD-LEDs with EQEs exceeding 30% to date.[16]

3.2 Cadmium-Free Colloidal QD Emitters

Due to environmental toxicity concerns associated with Cd, extensive research has been directed towards Cd-free QD emitters. The first InP-based QD-LEDs were introduced by Lim et al., exhibiting an EL peaking at 532 nm with a low EQE of 0.008%.[19] Cd-free blue QD-LEDs have also drawn attention. Defect-controlled ZnTeSe/ZnSe/ZnS QDs, especially after chloride passivation, and bulk-like ZnSe/ZnS QDs have achieved nearly perfect PLQYs, leading to QD-LED devices with EQEs close to their theoretical maximums.[20,21] Abundant research has been conducted to explore environmentally friendly materials in line with practical applications. However, to bring the technology of colloidal QDs into future commercial products, there is a pressing need for enhanced operational stability and reliable manufacturing processes.

3.3 Colloidal QDs for Laser Technology

With the ability to generate coherent and focused high-intensity light beams, lasers are crucial for a wide range of scientific, industrial, and medical applications. High-performance on-chip lasers have been a longstanding objective and the emergence of QDs has brought new possibilities for laser technology. In the 1980s, the state density of confined carriers in low-dimensional structures, in particular QDs, was investigated for use as optical gain of semiconductor lasers. Interestingly, what we know as QDs now were originally referred to as 3D quantum wells or quantum boxes.[22,23] In 1982, Arakawa proposed the concept of 2D and 3D quantum wells and predicted their improved temperature insensitivity for lasing.[22] Better optical gain coupled with a lower threshold of current density for quantum box laser was then analyzed.[23] Besides the delta-like state density of electron states, the tunable wavelength of QD also allows for the lasing to cover a broad spectrum.

Solution-processable gain materials are appealing for laser chips due to their versatility and scalability. In comparison to other materials like organic semiconductors and perovskites, colloidal QDs with 0D properties are more desirable for laser. Despite the early success of experiments in luminescence with colloidal QDs,[24] there still existed several problems such as rapid nonradiative Auger recombination to realize lasing with colloidal QDs. These issues were eventually overcome. Amplified spontaneous emission (ASE), a precursor to lasing, was achieved with CdSe QDs in 2000.[25,26] Later progress in controlling Auger decay more effectively through methods like compositional grading of the nanocrystal interior has led to significant improvement.[27,28] These developments include the demonstration of optically pumped CW colloidal QD lasers (with a threshold of 6.4 kW/cm^2),[29] dual-function devices that can operate as high-current-density (~200 mA/cm^2) LEDs, and optically excited lasers (with a threshold of 18.8 $\mu J/cm^2$).[30] While lasers based on colloidal QDs are typically driven by optical pumping schemes, a noteworthy breakthrough was made recently by electrically pumped ASE (with a threshold of 13 A/cm^2) using colloidal QDs.[31] In contrast, epitaxially grown QDs are inherently suitable for electrical pumping, despite their relatively higher cost.

3.4 Colloidal QD Solar Cells

Colloidal QDs have also paved the way for another direction—solar cells. To date, an expanding community of researchers across engineering, chemistry, physics, and materials science is dedicated to the development of colloidal QD solar cells with the primary objective of achieving high efficiency at an economical cost. The earliest colloidal QD solar cell can be traced back to 1998,[32] when InP QDs were used as sensitizers within a dye-sensitized solar cell (DSSC) configuration. However, this device showed limited power conversion efficiency (PCE). To improve the device performance, various kinds of colloidal QD solar cells have been proposed, including Schottky QD solar cells,[33] depleted heterojunction QD solar cells,[34] quantum funnels QD solar cells,[35] and bulk–nano heterojunction QD solar cells,[36] while the PCE of these devices only slightly increased. Recently, the community has developed size-tuned CQDs on both sides of a p-n junction, necessitating the development of both n-type and p-type films within the CQD stoichiometry.[37] This design, known as the quantum junction, has eliminated the band offset challenge seen in previous structures and finally optimized the PCE of colloidal QD solar cells to 18.1% in 2021.[38]

4 Epitaxial QDs

4.1 Quantum Light Sources Based on Epitaxial QDs

During the concurrent development of colloidal QDs, epitaxial QDs also came into the spotlight. With the progress of vacuum thin film epitaxy technology, the first epitaxial QDs of Ge/Si and InGaAs/GaAs based on S-K mode were realized in 1990.[39,40] In the past two decades, the integration of epitaxial QDs into optical micro-nano structures has led to the rich development in sophisticated single photon sources, entangled photon sources, and low-threshold on-chip lasers. When single epitaxial QD is weakly coupled with a carefully designed optical microcavity, a triggered single-photon source with near-unity photon purity and indistinguishability and highly entangled photon pairs can be achieved. For the long-dreamed optical quantum network, epitaxial QD-based active devices can generate flying quantum bits, photons, for the transfer of quantum information, which can also serve as solid quantum nodes where writing and reading of information in QDs can be performed through strong coupling with optical microcavities.

Epitaxial QDs have been acknowledged as the preeminent solid-state quantum emitters compared with the common parametric down-conversion in a bulk crystal, ideal for integrable quantum light sources. Since the first observation of photoluminescence in a single QD in 1994,[41,42] an on-demand single photon generated by a single QD under pulsed resonant excitation with single photon indistinguishability of 97% and purity of 98.8% was reported in 2013.[43] So far, embedding QDs into an elaborated micropillar cavity based on a distributed Bragg reflector together with resonant excitation brings to truly near-perfect single photon sources.[44–46] Employing an open cavity equipped with a Gaussian top mirror, a QD single photon source successfully operated with single photon purity of 97.9%, photon indistinguishability of 97.5%, and fiber end-to-end collection efficiency up to 57% in 2021.[47]

The recombination of the biexciton state in a single self-assembled QD is a cascade decay process, which will create a pair of photons entangled in polarization degree of freedom. Purcell effects for entangled photon pairs from highly symmetric QDs can be realized through the coupling with broadband optical structures, such as nanowires,[48] micro-lenses,[49] and microcavities with low quality factor.[50–52] Looking back to the initial experimental demonstration in 2006,[53] it is great progress that by 2019 QD entangled photon pair source with entanglement fidelity up to 90% was reported.[50] For another aspect, colloidal QDs can achieve photon emission with anti-bunching characteristics at room temperature.[54] However, challenges such as bleaching, blinking, and limited stability restrict their further application in quantum light sources.

4.2 Epitaxial QDs for Laser Chip

On the other hand, high-density epitaxial QD ensembles are ideal active mediums for integrated lasers, coupled with high reliability, high modulation speed, and extremely low lasing threshold even under high temperatures. To address the challenge of low luminescence efficiency of silicon-based light sources and the combination of III–V materials with silicon substrates, the integrated epitaxial QD/Si laser offers an effective solution that agrees well with large-scale manufacturing and ensures compatibility with current CMOS technology. Lasing emissions from epitaxial QDs were realized in 1994 for the first time.[55] However, detrimental factors such as the large spectral inhomogeneous broadening of QD ensemble initially limited the performance of quantum dot lasers, far from the theoretical limits. At room temperature, the threshold current density of the self-assembled QD laser was 950 A/cm^2 in the beginning and improved rapidly to 26 A/cm^2 in 1999 and to 10.4 A/cm^2 in 2009.[56,57] Accompanied by the significant development of QD epitaxial growth techniques, laser device design, and material patterning process technology, GaAs-based QD lasers have eventually emerged as better alternatives to traditional semiconductor double heterostructure laser and quantum well laser for their stunningly low threshold current density and wide-range operation temperature performance.[58] Recently, low threshold lasing of QD-pumped novel optical cavities has sparked great interest, such as engaging the higher-order topological corner state or bound states in the continuum.[59,60] Over the same period, representative progress has also been made in the field of QD detectors.[61,62] Due to the intrinsic sensitivity to infrared irradiation at normal incidence, theoretical lower dark current, prolonged carrier lifetime, and higher operating temperatures, novel QD detectors promise to outperform infrared photodetectors based on quantum wells, while ongoing efforts are focused on noise reduction and responsivity improvement.[63]

Along with the development of microelectronics, efficient light sources in silicon photonics have always been attractive. To date, the monolithic integration through direct epitaxy III–V materials on Si or Ge substrate has been in the spotlight compared to hybrid or heterogeneous integration methods.[64] Since the initial formation of self-assembled InGaAs/Si QD in the late 1990s,[65,66] such monolithic QD lasers have made considerable progress.[67–69] However, there is still room for further improvement. Their limitations mainly stem from defects including anti-phase boundaries, threading dislocations, and microcracks.[70] Simultaneously, III–V QDs grown epitaxially on Ge also show potential for high-performance laser.[71] In addition, epitaxial QDs also offer versatile functionality, serving as amplifiers, superluminescent diodes, infrared photodetectors, and solar cells.

The availability of high-quality colloidal QDs and epitaxial QDs provides great flexibility, allowing researchers to choose the system most suitable for specific requirements. It is also worth considering the fabrication processes of the two QDs and devices based on them. The formation of colloidal QDs involves mixing corresponding solutions in a flask at moderate temperatures, which is certainly a more cost-effective process than epitaxial growth, which necessitates ultrahigh vacuum equipment like molecular beam epitaxy. In epitaxial growth, the evaporation cells need to be filled with relatively expensive high-purity materials. However, epitaxial QDs are grown with well-controlled precision, allowing for tunable emission wavelengths and high optical quality. Furthermore, epitaxial fabrication naturally enables the integration of additional functional devices such as optical waveguides and resonators, also assisting in electrically pumped devices when doping layers are added. For colloidal QDs, such components need to be integrated separately, which can be achieved relatively easily through the spin coating method but typically with arbitrary QD orientation.

5 Conclusion

Colloidal QDs and epitaxial QDs will undoubtedly be the key materials to light up future optoelectronic devices. Rather than being direct competitors, these two classes of QDs are more complementary when targeting different devices. Epitaxial QDs have already found commercial applications in light sources, which have presented superior optoelectronic properties for solid-state applications over the past 30 years. In the future, epitaxial QDs are expected to play a central role in quantum optical networks as quantum light sources and information nodes. Efforts will focus on improving structural symmetry, optical quality, and position determination for single epitaxial QDs, as well as developing epitaxial QD lasers with reduced thresholds and enhanced performance. This technology holds promise for both scientific and commercial applications. However, it remains unclear whether and when colloidal QDs will reach a similar level in terms of quantum technologies, which would require substantial efforts.

On the other hand, colloidal QDs are alluring for future mass-producible illumination products that could operate across a wide spectral range in the visible or near-infrared. LEDs using colloidal QDs have shown outstanding performance, and recent developments have brought colloidal QDs closer to practical active QLED displays. The wavelength tunability stemming from controllable size variations of colloidal QDs opens the door to high-brightness, multicolor displays like laser TVs. Furthermore, colloidal QDs hold promise for other optoelectronic devices such as high-performance photodetectors and solar cells, particularly taking advantage of their environmental compatibility and cost efficiency. The enormous commercial potential of QDs is becoming increasingly evident. Over the next decade, we look forward to witnessing the emergence of more colloidal QD applications for everyday use in the market.

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