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Advances and prospects of lasers developed from colloidal semiconductor nanostructures

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Abstract: Since the first observation of stimulated emission from colloidal quantum dots (CQDs) in year 2000, tremendous progress has been made in developing solution-processed lasers from colloidal semiconductor nanostructures in terms of both understanding the fundamental physics and improving the device performance. In this review paper, we will start with a brief introduction about the fabrication of CQDs and the corresponding electronic structures. The emphasis will be put on the discussion about the optical gain and lasing from colloidal nanostructures including the gain mechanism, the main hurdles against optical gain and lasing as well as strategies to optimize the lasing performance. Afterwards, the recent advances in CQD lasers, exemplified by the achievement of continuous wave lasing, will be presented. Finally, the challenges and a perspective of the future development of lasers based on the colloidal semiconductor nanostructures will be presented.

Keywords: Lasers, colloidal semiconductor nanostructures, Auger recombination

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1. Introduction

Lasers, which are the devices that produce coherent light by the process of stimulated emission, have found an ever-increasing number of applications in various areas, such as scientific research, industry, medicine and military defense and have become an essential component in modern technology. The first semiconductor laser diode was realized from a bulk GaAs homojunction formed by Zn diffusion in 1962 [1, 2]. Such achievement is very exciting but it is not practically useful because of the high threshold current densities ($\sim 10^4 \text{ A cm}^{-2}$ at liquid nitrogen temperature) of these initial lasers [1, 2]. Tremendous follow-up efforts have been devoted to improving the laser performance from then on. The most striking advance in opening the commercial market of laser diodes is the invention of the epitaxial quantum well lasers [3]. Due to the presence of the quantum confinement effect (QCE) in 1D, the quantum wells exhibit step-like density of electronic states that is nonzero at the band-edge, which leads to the reduction of laser threshold by approximately three orders of magnitude compared to those of the bulk analogues [3, 4].

The success in the quantum mechanics-governed quantum well lasers provides strong motivation for the development of lasers based on the even lower dimensional semiconductors. Quantum dots (QDs) represent the ultimate nanostructure that exhibit QCE in all of the three dimensions [5, 6]. Theoretically, the discrete atomic-like density of electronic states in QDs will promise the further reduction of laser threshold [7]. Moreover, the energy spacing between the electronic levels in strongly confined QDs can be much larger than the thermal energy, which results in a temperature-insensitive optical gain, a much desired characteristic for laser applications [8, 9]. Indeed, improved lasing performance was achieved from the QDs grown by epitaxial techniques [8, 10, 11]. However, these epitaxial QDs possess relatively large lateral

sizes (~ 10 nm) and have difficulty in accessing the smaller dimensions, leading to the loss of various advantages of QCE [8-11].

Colloidal quantum dots (CQDs), or nanocrystals, are tiny semiconductor particles developed in the early 1980s (Fig. 1) [12, 13]. The size of CQDs can be well controlled in sub-nanometer precision thanks to the versatile wet chemistry technique, such that the lasing wavelength can be tuned in a wide range of spectrum leveraging on QCE [14, 15]. In contrast to the epitaxial semiconductors, CQDs essentially have no request on the substrate and thus are compatible with almost all kinds of the laser cavities [16, 17]. More importantly, the CQD suspensions enable the fabrication of devices by the solution-based methods, such as spin-coating and inkjet printing, which renders the development of cost-effective lasers [18-20].

However, it turns out that the development of optical gain and lasing action in CQDs is much more complex than expected because the unexpected issues stand out as the obstacle, exemplified by the nonradiative Auger recombination (AR) [21, 22]. It was not until year 2000 that the first observation of stimulated emission from CdSe CQDs has been achieved [23]. Since then, there has been an explosion of research activities on developing lasers from colloidal nanostructures motivated by the potentially superior lasing performance and the low-cost solution-processability [24, 25].

In this paper, a review focused on the development of lasers based on the colloidal semiconductor nanostructures will be presented. We will start with a brief introduction on the synthesis of CQDs by the wet-chemistry method, followed by a short description of the corresponding electronic structure and optical transitions. The emphasis will be put on the discussion of the optical gain in CQDs including the gain mechanism, the major complication of AR and the evolvement in addressing the AR issue. Thereafter, the progress in developing lasers

from colloidal nanostructures with various geometries and the most recent advance in CQD lasers, such as the single-exciton gain, multi-photon pumped lasers, continuous wave operation and the emergence of perovskite nanocrystals for lasing, will be presented. Finally, the challenges and a perspective of the future development of lasers based on the colloidal semiconductor nanostructures will be put forward.

2. Synthesis of CQDs by wet-chemistry method

Pyrolysis of metal-organic precursors in hot solvent as termed “hot-injection method” is the most commonly used approach to synthesizing the high-quality CQDs with a high degree of monodispersity (particle size distribution < 5%) and high photoluminescence quantum yield (PLQY) (>50%) [26, 27]. Fig. 2 shows the typical synthesis setup and reaction process for growing CdSe-based CQDs, where a three-necked flask equipped with a thermocouple and temperature controller represent the key components [28]. The growth kinetics of CQDs has been well investigated by La Mer and Dinegar [29]. In general, the growth of CQDs consists of three steps, that is, the particle nucleation process, particle growth process and Ostwald ripening [29]. The nucleation process is usually very fast and is activated by the supersaturation of the monomers derived from the thermal decomposition of the precursors. In the meantime of nucleation, the monomers are depleted and the nucleation ceases when the monomer concentration stays below the critical magnitude. Afterwards, the particle growth process starts to dominate. Based on the diffusion theory, the monomers first approach the particles and then trigger the chemical reaction at the surface, which indicates that the particle growth rate is diffusion-limited [30]. According to the Fick’s diffusion law, the variation of CQD radius (R) with time is derived by:

$$\frac{\partial R^2}{\partial t} = \frac{2VD}{N_A}(C_t - C_s)$$

where D is the diffusion coefficient, V is the volume of the solution, C_t is the monomer concentration of the bulk solution at time t and C_s is the monomer concentration at the surface of the particle. The equation indicates that the particle size distribution will get narrower as the particles grow, which is known as the “self-focusing” stage [31]. As the monomer concentration decrease and become mostly depleted, the Ostwald ripening process will take over, where tiny nanocrystals will shrink and dissolve into the solution and the larger nanocrystals will continue to grow. As a result, in the Ostwald ripening regime, the size distribution will become worse, which is known as the “defocus” stage. Therefore, one should suppress the Ostwald ripening process in order to obtain the highly monodisperse CQDs [32]. Further progress in the hot-injection method is associated with the search for new high-temperature solvents, precursors and ligands, which makes it possible to achieve CQDs with high PLQY. Moreover, colloidal nanostructures with other geometries, such as the nanorod, tetrapod, and nanoplatelet, can be fabricated by deliberately controlling the reaction temperature, time, and surfactant ligands [33, 34].

Besides the hot-injection method, the one-pot non-injection organometallic synthesis methods have also been applied to fabricate various kinds of CQDs [35]. The main advantage of the non-injection synthesis lies in the feasibility of fabricating CQDs in large quantities. As mentioned above, the hot-injection method necessitates the swift injection of a precursor solution into a hot mixture of organic solvents, which makes it challenging to control the reaction temperature upon the injection of the precursor solution, resulting in poor batch-to-batch reproducibility [35]. As for the non-injection approach, all the precursors are put into the reaction

vessel and the chemical reaction is triggered by controlling the temperature or introducing nucleation initiators without any further solvent injection. Hence, it can be easily scaled-up to yield CQDs on the gram scale. However, until now, the quality of the CQDs based on non-injection approach is still inferior to those made by hot-injection method, which may be due to the more difficult control of the nucleation process in the former [36].

In recently years, the flow microreactor synthesis of CQDs has been developed, where the CQD precursors are conveyed by the microfluidic channels and reactions take place in the continuous microreactors equipped with heaters and fluid control elements [35]. In this method, the nucleation and growth of CQDs can be made to occur at different areas, enabling the optimization of CQD fabrication to obtain CQDs with extremely narrow size distribution. Moreover, this emerging CQD fabrication method also provides a favorable platform to study the mechanism of nucleation and the subsequent growth of CQDs, offering clues for the fabrication of other CQDs which are not feasible yet. So far, the synthesis of CQDs in microfluidics is used solely for research purposes due to the limitation of mass-production.

The introduction of an inorganic shell onto the CQDs has fueled significant advance on the development of these colloidal nanosystem [37]. On one hand, the shell could offer an efficient passivation of the surface trap states, which leads to the greatly enhanced PLQY. On the other hand, by selecting the shell with proper alignments of the conduction band and valence band with respect to those of the core, the optical properties the CQDs including the emission spectra and dynamics can be strongly modulated (More details will be given in **Section 5.1** Engineering the spatial distribution of electron and hole wave function). In general, the core/shell CQD systems are fabricated in a two-step procedure, that is, the initial synthesis of core nanocrystals and the subsequent shell growth [37]. For the shell growth, typically, a small number of

monolayers of the shell semiconductors are deposited on the cores and the amount of the shell precursors shall be predetermined on request. It should be noted that in order to avoid nucleation of the shell material and suppress the ripening of the core nanocrystals, the temperature adopted for the shell growth is normally lower than that for the synthesis of the cores. We refer the interested readers to prior publications [29, 35-37] for more comprehensive description about the fabrication of CQDs and the engineering of core/shell CQDs.

3. Electronic structure and optical transitions in CQDs

As the physical dimension of CQDs is comparable or less than the free travelling length of the electron-hole pair or the exciton, the elementary excitation in CQDs, the exciton will adjust itself to adapt to the presence of the rigid boundary in the nanoparticle, which results in the quantization of its energy and momentum [13]. This phenomenon is known as the “quantum confinement effect”, the most popular term in the nano-world [13, 38, 39]. The optical and electronic properties of the CQDs are strongly associated with the QCE, which makes the CQDs unique and interesting from the perspective of both fundamental physics and practical applications. In general, the band gap of a certain semiconductor material is a constant. In contrast, by virtue of the QCE, the effective band gap of the CQDs can be tuned by more than 1 eV by facilely changing the dot size, enabling the broadband emission color tunability [38]. Fig.1b shows the colorful CdSe CQDs in solution with different sizes, vividly manifesting the wide range band gap tunability as a result of QCE. Another favorable result induced by the QCE is the collapse of the continuous density of states in bulk semiconductor towards the discrete levels in CQDs as shown in Fig. 3a and b [13]. On one hand, such atomic-like electronic levels will lead to the increased density of the band-edge states in CQDs and hence reducing the gain threshold. On the other hand, such isolated levels in CQDs may endow them with the

temperature-insensitive optical gain due to the suppression of the thermal depopulation of the band-edge states [7].

The size-dependent electronic properties in QDs can be quantitatively described by the well-known “particle-in-a-sphere” model [14, 40]. Based on this model, the photo-excited charge carrier can be regarded as a particle located in the infinite spherical potential well:

$$x < r; V_x = 0$$

$$x > r; V_x = \infty$$

where r is the potential well radius or the radius of the QD. The wave function of the charge carrier can be obtained by solving the Schrödinger equation taking into account the potential well in the formula, which has been addressed by Flügge in 1971 [41] and the wave function of the charge carrier is given by:

$$\psi_{nlm}(x, \theta, \phi) = Cx^{-1}J_l(k_{nl}x)Y_l^m(\theta, \phi)$$

where C is constant, $Y_l^m(\theta, \phi)$ stands for the spherical harmonic, $J_l(k_{nl}x)$ stands for the spherical Bessel function of l -th order and $k_{nl} = \alpha_{nl}/x$, where α_{nl} is the n -th zero of $J_l(k_{nl}x)$. According to the above analysis, the energy of the charge carrier is given by [42]:

$$E_{nl} = \frac{h^2 \alpha_{nl}^2}{8\pi m_0 r^2}$$

where m_0 is the mass of the charge carrier. We can see that the energy of the charge carrier confined in QDs is quantized, which is different from the kinetic energy of a carrier in the free space and elucidates the energy level splitting in QDs. It is also noted that the energy of the charge carrier in QD is reversely proportional to the square of the dot radius, which explains

the size dependent band gap energy in CQDs, that is, the band gap increases as the size of the CQD decreases.

The above “particle-in-a-sphere” model satisfactorily sheds light on the increased band gap with the reduction of size and the presence of the structured absorption spectra due to the quantization of the energy. However, it turns out that the real electronic structure of semiconductor CQDs is much more complicated and one has to consider the multi-subband character of valence band in order to explain the experimentally observed energy levels or optical transitions in CQDs [40]. Here we will take the most well studied system, the CdSe CQDs, as the example. In the case of CdSe semiconductor, the conduction band is relatively simple since it is constructed by the Cd 5s atomic orbitals which contain only double degeneracy. By comparison, the valence one arises from the Se 4p orbitals which contains 6-fold degeneracy [43]. The 6-fold degenerate valence band breaks up into a 4-fold band with angular momentum of $J=3/2$ and a 2-fold band with $J=1/2$ due to the well-known spin-orbit interaction [40]. Furthermore, the 4-fold $J=3/2$ band breaks up into 2-fold heavy-hole and 2-fold light-hole band, known as the A–B splitting. In quantum confined CdSe nanocrystals, the QCE mixes the above discussed subbands in valence band and the hole states are actually characterized by the total angular momentum, T , which takes into account both the unit cell contribution and the envelop function contribution. In terminology, the valence bands in CQDs are described as $nL_T(h)$ [44]. Based on the calculation by Ekimov *et al.* [6], the energy levels arrange in a rising order is $1S_{3/2}(h)$, $1P_{3/2}(h)$, $2S_{3/2}(h)$, $1P_{1/2}(h)$ and $1S_{1/2}(h)$ (Fig. 3c). The optical transitions from these valence subbands to the conduction bands have been experimentally observed and agree with the theoretical calculation very well [45]. Fig. 3d shows the absorption spectrum of high-quality CdSe CQDs, where the first four optical transitions are well-resolved.

Until now, we have only considered the band-mixing among the valence subbands and it matches the featured absorption spectra of CQDs measured at room temperature very well. However, when dealing with the exciton emission associated with the optical transition $1S(e)-1S_{3/2}(h)$, the electron-hole exchange interaction has to be taken into consideration because of the enhanced overlap between the electron and hole wave function in CQD, which leads to a large splitting between the optically active (bright) and optically passive (dark) excitons [46, 47]. Especially, the experimentally observed temperature-dependent exciton dynamics and the single CQD emission at low temperature can only be interpreted by taking into account the dark-bright splitting in the band-edge $1S(e)-1S_{3/2}(h)$ transition [47, 48].

4. Optical gain in CQDs

4.1 Optical gain mechanism

Optical gain describes the optical amplification in the semiconductor material, which is the prerequisite to build a laser [23, 49]. In general, optical gain requires the population inversion condition such that the light generation by stimulated emission dominates over the light absorption in the material. In bulk semiconductors, the optical gain is generally achieved when the injected carrier density is high enough to make the separation between the quasi-Fermi levels for the electrons and holes to be larger than the band gap, which is known as the Bernard-Duraffourg inversion condition [50]. In the case of CQDs, the situation is different because of the quantized atomic-like energy levels. Due to the typically multifold degeneracy of electron and hole states involved in the band-edge optical transition in CQDs, the requirement for optical gain is not that straightforward, where the radiative recombination from multiexciton state is usually involved [23, 47]. In fact, the degeneracy of the lowest transition level in CQDs depends on a variety of parameters, such as the crystal structure, the shape of the nanocrystal, core/shell

structure and so on, which has been discussed in the previous section. Taking the canonical CdSe CQDs as the example, in order to facilitate the analysis of optical gain and give a generalized gain model, the lowest transition in CdSe CQDs is typically treated as 2-fold degeneracy given that all the above-mentioned parameters take effect in lifting the degeneracy of the optical transition [46]. It is noted that lifting of degeneracy is critically important to reduce the gain threshold, which has previously been adopted as the effective strategy in epitaxial quantum well lasers. Fig. 4a shows the simplified optical transition state in CdSe CQDs, featuring a 2-fold two-level system. In the ground state, two electrons are located in the bottom level. When one of the electrons is excited to form the single exciton state in the CQD, the optical transparency is attained due to the compensation of the stimulated emission by absorption in the CQD (Fig. 4b) [51]. In order to develop optical gain, the other electron in the CQD has to be excited such that the stimulated emission outplays the optical absorption (Fig. 4c). This means that biexciton state or even high-order multiexciton states are responsible for the optical gain in CQDs. Indeed, it is found that the optical gain can only be developed with $\langle N \rangle > 1$ (Fig. 5a), where $\langle N \rangle$ is the average number of excited excitons per CQD, based on the transient absorption measurement [23]. Moreover, the gain spectrum is red-shifted with respect to that of the single exciton state and coincides with biexciton spectral position (Fig. 5b), further confirming that the optical gain in CQDs originates from the multiexciton recombination.

The characteristics of multiexciton emission in CQDs have been investigated by both steady-state and time-resolved spectroscopy under high excitation intensities such that multiexciton states are generated [52, 53]. It is found that the time-integrated PL from CQDs with strong excitation ($\langle N \rangle = 10$) are almost identical to that with weak excitation ($\langle N \rangle < 1$), suggesting that the efficiency of multiexciton emission is very poor. This is because that the

multiexciton decay in CQDs is dominated by the fast nonradiative AR rather than radiative recombination (More detailed discussion on AR will be presented later) [21]. In order to resolve the multiexciton emission from the PL spectra, the femtosecond PL upconversion technique has been employed thanks to the sub-picosecond time resolution. The transient PL spectrum recorded at time delay of 1 ps ((Fig. 5c) clearly manifests the appearance of a shoulder on the low-energy side of the singe-exciton PL peak and a new emission band on the high-energy side, which correspond to the biexciton emission and the higher-order multiexciton emission, respectively, according to the pump intensity dependent PL analysis (Fig. 5d). The large red-shift of the biexciton emission spectrum with respect to that of the single exciton indicates the strong attractive exciton-exciton interaction in CQDs [52, 53].

4.2 Understanding challenges underlying the optical gain of CQDs

Although CQDs have been developed in the early 1980s, they failed to show any sign of stimulated emission or lasing action after a decade of research. In fact, it has experienced a long process to figure out the major challenges underlying the development of optical gain in CQDs. In the following, we will discuss them one by one.

4.21 Surface trapping

Initially, the challenge in achieving optical gain and lasing from CQDs was thought to be the efficient carrier trapping by the abundant surface defects because of the huge surface-to-volume ratio in nanoparticles [27]. This was reasonable since the original CQDs had vulnerable surface which is capped with organic ligands, resulting in poor PLQY ($< 10\%$) and presence of strong defect-state emission band [54]. Later on, the idea of a core/shell structure was adopted [27, 55]. By surface passivation with a larger band gap semiconductor shell, the PLQY of the CQDs is

significantly enhanced to be higher than 50% and the deep level emission associated with the defect states is effectively eliminated. However, the CQDs still failed to lase, which suggests that there might be other hidden culprits that blocked the optical gain or lasing action.

4.22 Inefficient carrier cooling-phonon bottleneck

In general, the pump photon energy is larger than that the band gap of the gain media, so that the nonequilibrium electrons and holes are generated upon photoexcitation [56]. These hot carriers have to relax to the lowest excited state (known as the carrier cooling) in order to produce optical gain [57]. In bulk semiconductors, the high-energy carriers can undergo efficient carrier cooling by carrier-phonon and carrier-carrier interactions, which gives rise to sub-picosecond relaxation duration [56]. In contrast to the bulk semiconductors with nearly continuous energy states, the CQDs feature the isolated atomic-like energy levels. Due to the restrictions of energy and momentum conservation, the probability of the carrier-phonon interaction in CQD is speculated to be greatly reduced with respect to that in bulk counterpart, which is the so-called “phonon bottleneck” [57, 58]. The phonon bottleneck effect in CQDs will lead to the inefficient carrier flow to the emissive state, thereby plaguing the buildup of optical gain.

The carrier cooling dynamics in CQDs was intensively investigated in late 2000s by the ultrafast transient absorption measurement [59, 60]. In contrast to the theoretical prediction, the experimental results reveal a rapid carrier cooling process in CQDs which is comparable to those in bulk semiconductors [60], indicating that the hot carrier relaxation is not the limiting factor for the development of optical gain in CQDs. Moreover, the electron cooling rate is found to increase with the decrease of the dot size, indicating that cooling mechanisms other than phonon emission are dominated in CQDs [61]. Finally, the comprehensive spectroscopic studies disclose

that the intrinsic electron-hole interaction is responsible for the fast carrier cooling in CQDs, which involves the energy transfer from electron to hole and the subsequent rapid hole relaxation thanks to the dense available states [62, 63].

4.23 Photoinduced absorption

Photoinduced absorption (PA) denotes the sequential absorption process from the excited state after the ground state absorption, which is a common issue for the gain media, particularly, those with broadband gain spectra [64]. The PA and its impact on the optical gain in CQDs have been investigated based on the white-light pump-probe technique [22, 65]. It is found that PA signal increases with the decrease of the dot size and totally stifles optical gain in small-sized CQDs. Moreover, the amplitude and the structure of PA change with surface condition and the solvent of the CQDs, indicating that the PA in CQDs is an extrinsic property associated with the environment, which also explains the enhanced PA competition in small-sized CQDs because of the large surface-to-volume ratio [22]. The above conclusion is instructive since it means that one can suppress or eliminate the PA in CQDs by properly engineering the surface and the environment of the CQDs so as to achieve the optical gain. For example, it is demonstrated that the CdSe CQDs with better surface passivation exhibited suppressed PA for a given excitation fluence and that the CQD dispersed in trioctylphosphine or in the form of solid-state films did not show any PA signal [23].

4.24 Auger recombination-the limiting factor

AR is a nonradiative many-body carrier decay process, in which the energy released by the annihilation of a pair of electron and hole is utilized to stimulate the other electron or hole to the higher energy state. With the in-depth understanding of optical gain in CQDs based on the

comprehensive spectroscopic studies, it is revealed that the AR is the main hurdle against the development of stimulated emission and lasing in CQDs. AR is actually a very common carrier decay pathway in materials and does not bring big trouble for the bulk semiconductors [66, 67]. However, it becomes the efficient gain depletion channel in CQDs as the AR rate is significantly enhanced in CQDs compared to the bulk analogue [21, 68, 69], which can be attributed to twofold reasons. On one hand, the carrier-carrier interaction is strengthened in CQDs due to the spatial and dielectric confinement; On the other hand, the conservation for translational momentum which mitigate the AR in bulk semiconductors is relaxed in CQDs (Fig. 6a and b), thus facilitating the AR in CQDs. In fact, AR is a very important topic in CQD research, which not only closely relates to the optical gain and lasing, but also has significant impact on the light-emitting diode (LED) performance and blinking behavior [70, 71]. In the following part, we will discuss the representative characteristics of AR in CQDs, which benefits for better understanding the optical gain in CQDs.

4.241 Quantization of Auger recombination rate

In bulk semiconductors, the AR is not efficient due to the conservation rule for translational momentum as depicted in Fig. 6a and the rate of AR is simply proportional to the cubic of the carrier density stemming from the character of a three-particle process, which is given by [21, 72]:

$$dn_{eh}/dt = -Cn_{eh}^3$$

where C is the Auger constant. In stark contrast, the AR becomes extremely fast in CQDs as a result of the enhanced Coulomb interaction and the relaxation of the momentum conservation. For example, the AR lifetime in CdSe CQDs is as short as several to tens of picoseconds, which is much smaller than the corresponding spontaneous emission lifetime (~tens of nanosecond)

[21]. Moreover, as the recombination of excitons in CQDs happens in steps from the N to N-1, and eventually to the single exciton state, the AR rate in CQDs has to be described by a set of discrete values, corresponding to the decay of different exciton numbers in CQDs, instead of following the rule described by the above equation [21, 73]. In other word, the AR rate should be quantized in CQDs rather than being continuous as in bulk semiconductor. Klimov *et al.* for the first time investigated the AR rate in CQDs as a function of the photoexcited exciton number [21, 73]. The exciton dynamics under varied excitation intensities, corresponding to different number of excitons in CQDs, were interrogated by recording the bleaching decay of 1S optical transition in transient absorption experiments. Based on the subtraction strategy, the discrete time constants associated with the 4-, 3- and 2-exciton states can be obtained (Fig. 6c) [21]. As can be seen, the AR rate monotonously accelerates with the increase of exciton number. Moreover, the AR lifetime is found to be inversely proportional to the square of the average number of excitons (the ratio of the AR lifetimes corresponding to 4-, 3- and 2-exciton states is 10 ps : 21 ps : 45 ps = 0.22 : 0.47 : 1), consistent with the three-particle process, indicating that the AR in the quantum confined CQDs is dominated by the path of X^- or X^+ (the released energy from the annihilation of an exciton is transferred to an electron or a hole, respectively) as described in Fig. 6b rather than in the means of bimolecular decay [74].

4.242 Size dependent Auger recombination rate

As is known that the optical and electronic properties of CQDs are strongly dependent on the radius of the nanocrystals, it is natural to ask how the AR rate changes with dot size. Based on the systematic AR measurement as a function of the size of CdSe CQDs, the AR rate is unraveled to greatly increase with the reduction of the dot size (Fig. 6d) and the AR lifetime shows a linear dependence on the volume of the CQDs (Fig. 7a) [21, 73]. Furthermore, the size-

dependent AR rate in other kinds of CQDs in addition to II-VI group semiconductors have been comparatively investigated, which includes the InAs, PbSe, PbS, Si, and Ge CQDs (Fig. 7b). The interesting finding is that no matter the CQDs are made of direct-gap semiconductors (CdSe, InAs, PbSe, PbS) or indirect-gap (Si, Ge) semiconductors, they follow the same size-dependent trends in AR rate. The underlying mechanism is attributed to the admixture between electronic states with different translational momenta by confinement effect, resulting in the independent carrier-carrier interactions in CQDs on the band structure of the bulk semiconductor [75]. Such general linear dependence of AR lifetimes on CQD volume is denoted as the “universal volume scaling” [76].

4.243 Superposition principle in Auger recombination rate

Previously, most of the AR investigation was carried out on the neutral and the negatively charged CQDs. With the advance of control on the charge states in CQDs (e.g. chemical treatment), it becomes feasible to investigate the AR in either negatively charged or positively charged CQDs [77-79]. By separately measuring the Auger lifetimes of both neutral and charged multicarrier states, it is found that the Auger decay of the multicarrier states in CQDs can be described as a superposition of individual three-particle Auger processes. For example, the neutral biexciton Auger decay can be presented as a superposition of independent negative and positive trion recombination pathway (negative and positive trion denotes an exciton with an extra electron and an extra hole, respectively) [80]. This phenomenon is known as the superposition principle in AR in CQDs. It is also demonstrated that the AR lifetime of trions in CQDs scales with the cubic of the dot radius, indicating the “universal volume scaling” is also applicable to trions, as illustrated in Fig. 7a. Moreover, the superposition principle is found to be quite general, which may be applicable to any kinds of the quantum-confined CQDs. Notably,

the superposition principle in AR in QDs can be exploited as an enabling tool for evaluating AR time constant of random compositions.

5. Optimizing optical gain and lasing in QDs by Auger recombination suppression

As AR is a nonradiative many-body recombination process, it represents a carrier loss mechanism under high excitation intensities. Due to the much enhanced AR rate in QDs, the AR has been recognized as the dominating channel that depletes the population inversion, thus hindering the optical gain and lasing in QDs [19, 23]. As a result, it becomes the subject of much research to find out approaches to suppressing or even killing AR in QDs, which is attractive to develop cost-effective solution-processed QD optical amplifier and lasers [81-83]. In the following part, the major progress on this topic will be presented.

5.1 Engineering the spatial distribution of electron and hole wave function

The “universal volume scaling” rule tells us that the increase of dot size can serve as a method to reduce the AR rate in QDs. However, it is noticed that even for the large QDs with size beyond twice of the excitonic Bohr radius, the AR is still too fast with typical sub-nanosecond time constant [21, 73]. Moreover, the change of the dot size will be accompanied with the shift of the emission and gain spectra, which makes it infeasible to simultaneously control the lasing wavelength and suppress AR in QDs. Therefore, other effective AR suppression methods without satisfying the emission color tunability are highly desirable.

Recalling the physical reasons for the enhanced AR in QDs, it would be apparent that one can suppress the AR by weakening the carrier-carrier interaction in QDs which can be realized by spatially decoupling the electron and hole wave functions. Heterostructures have been overwhelmingly employed to control the spatial distribution of electrons and holes as

exemplified by the quantum wells and superlattices [84, 85]. As for the canonical CdSe QDs, different types of heterostructures can be obtained by selective coating of shell semiconductors, such as the type-I CdSe/ZnS, type-II CdSe/ZnTe or ZnTe/CdSe (Fig. 8) and quasi-type-II CdSe/CdS QDs [86]. Although the suppressed AR has been observed in the type-II CdSe/ZnTe and ZnTe/CdSe QDs by virtue of the almost entirely spatial separation of the electron and hole wave functions, they were demonstrated to be unsuitable for optical gain or lasing applications because the significant decouple of the electrons and holes not only mitigates the AR but also reduces the optical oscillator strength, making it difficult to achieve stimulated emission [87-89]. In this situation, the CdSe/CdS QDs with quasi-type-II band alignment becomes the most intensively studied system, which offers the opportunity for obtaining both suppressed AR and strong oscillator strength, thus benefiting for low-threshold optical gain. As shown in Fig. 9a and b, due to the much smaller conduction-band offset than that of the valence band, the holes are confined in the core, while the electrons are delocalized over the whole QDs. As a result, the overlap between the electron and hole wave functions can be effectively reduced. Such phenomenon is more pronounced for the QDs with thick shells which are termed as the “giant” nanocrystals or g-NC (Fig. 9c) [90-92]. Fig. 9f presents the pump-power dependent PL intensities of the g-NC with different integration time durations. It is noticed that the multiexciton recombination in the g-NC contributes to the total emission signals at much longer time delay (>50 ns) as compared to the CdSe/ZnS QDs with similar core size (Fig. 9g), suggesting the greatly increased multiexciton lifetime and hence suppressed AR in the quasi-type-II g-NC [93]. According to the time-resolved PL measurement, the AR lifetime of the g-NC is determined to be even longer than 10 ns, in sharp contrast to that (~ 0.2 ns) of the CdSe/ZnS QDs (Fig. 9d and e), which directly indicates that the AR has been effectively suppressed in the

thick-shell CdSe/CdS CQDs [93]. Importantly, these g-NC exhibits dramatically improved optical gain performance. In particular, the stimulated emission threshold is demonstrated to be as low as $\sim 26 \mu\text{J cm}^{-2}$ under femtosecond laser excitation (pulse-width: 100 fs), whereas the contrast experiments on the traditional type-I CdSe/ZnS CQDs reveals the high stimulated emission threshold of $\sim 300 \mu\text{Jcm}^{-2}$ under the same conditions (Fig. 10a), representing a more than one order of magnitude reduction in the pump threshold. Furthermore, with the increase of pump power, stimulated emission involving the higher-order multiexcitons besides the biexciton was observed in the g-NC, giving rise to a much broader optical gain bandwidth ($\sim 500 \text{ meV}$) (Fig. 10b and c), which again stems from the suppressed AR in the g-NC. Interestingly, the Coulombic exciton-exciton interactions in quasi-type-II CdSe/CdS CQDs can be tuned to be either attractive or repulsive by deliberately controlling core-shell dimensions, and hence, the optical gain spectrum can be on-demand adjusted to be red-shift (type-I-like) or blue-shift (type-II-like) relative to spontaneous emission maximum (Fig. 10d) [94].

5.2 Engineering the shape of the confinement potential

The initial theoretical work about the impact of shape of the confinement potential on AR was explored by Cragg *et al.* and Climente *et al.* [81, 82]. Based on their calculation, the alloyed interface with smoothed confinement potential in the heterostructure can dramatically diminish the AR rate by more than three orders of magnitude with respect to that with abrupt interface (Fig. 11a). The underlying physical mechanism is that by adjusting the confinement potential from a sharp terminating boundary to a graded parabolic profile, it is able to significantly decrease the overlap between the initial and the final state involving in the AR process, thereby reducing the AR probability [81]. Inspired by Cragg's work, García-Santamaría *et al.* speculated that the unintentional interfacial alloying may be the additional reason for the surprisingly

suppressed AR in g-NC because only considering the effect of spatial distribution of electron and hole wave functions cannot fully explain the reduced AR rate [93, 95]. From the fluorescence-line-narrowing measurement, they clearly observed the signal from the alloyed $\text{CdS}_x\text{Se}_{1-x}$ layer, which suggests the formation of alloyed core-shell interface and supports their hypothesis (Fig. 11d). With the advance of chemical synthesis of CQDs, it becomes more and more feasible to control the profile of the core-shell interface on-demand, which provides an ideal platform to study the impact of interfacial alloying on AR. Based on the comparative investigation of carrier dynamics of the CdSe/CdS CQDs with sharp and alloyed core-shell interface, it was directly demonstrated that the interfacial alloying plays an important role in mitigating AR in CQDs (Fig. 11b and c) [96, 97].

Wang *et al.* for the first time explored the effect of core-shell interface on optical gain and lasing performance in CQDs via side-by-side measurements [19]. Specifically, three kinds of CdZnS-based CQDs, namely, CdZnS core, CdZnS/ZnS CQDs with sharp core-shell interface (SS) and CdZnS/ZnS CQDs with alloyed core-shell interface (AS), were investigated in the same breath. It is manifested that the CQDs with alloyed interface exhibit significantly reduced stimulated emission threshold with respect to those with sharp interface (Fig. 12a) and the improved gain performance is unambiguously attributed to the suppressed AR in these interface-gradient CQDs by comprehensive steady-state and time-resolved spectroscopy (Fig. 12 b, c and d) [19]. Soon afterwards, Park *et al.* performed the comparative experiments on CdSe/CdS core-shell CQDs with sharp and smooth interfaces, which similarly disclosed the improved gain and lasing performance in the CQDs with compositional smooth interface [98]. The above two works and follow-up works [99-101] highlight the importance of surface/interface engineering toward optimization of CQDs for solution-processed lasers.

6. Beyond spherical CQD based lasers: non-spherical colloidal nanostructures for lasing

Aside from the nearly spherical CQDs, colloidal nanostructures with different morphologies, such as nanorod, nanoplatelet and nanotetrapod, can be obtained by the facile wet-chemistry method. By virtue of the unique electronic structures, the absorption and emission characteristics of these non-spherical nanostructures can be radically different from the spherical CQDs, which offers new freedom to achieve improved gain and lasing performance. The major findings in this aspect will be discussed in the following part.

6.1 Colloidal nanorods

Different from CQDs where the carriers are confined in 3D, the excited carriers can move freely along the length of the nanorods while being confined within the diameter direction [33, 102]. The distinct confinement dimensions between colloidal nanorods and CQDs will lead to different carrier dynamics. Htoon *et al.* systematically studied the effect of confinement dimension on the multiexcitonic AR by tuning the diameters and lengths of the nanorods [74]. It is revealed that the transition from 3D to 2D confinement has significant impact on the multiexcitonic AR. For the colloidal nanorods with sufficiently large aspect ratio ($>\sim 8$), the AR follows the quadratic, two-particle decay, in contrast to the cubic, three-particle process as observed in the quantum-confined CQDs, suggesting that the AR takes place in the means of bimolecular, exciton-exciton interactions in these nanorods [74]. A direct benefit from the bimolecular AR is the suppressed AR of higher multiparticle states, leading to the prolonged optical gain duration and occurrence of stimulated emission from higher-order multiexcitons (Fig. 13a) [74, 103]. The first observation of lasing action from CdSe/ZnS core-shell nanorods was reported by Kaze et al [104]. Interestingly, they observed a linearly polarized laser emission directly associated with the

symmetry of the nanorods, while for CQDs the lasing emission was unpolarized, indicating the anisotropic optical gain from the colloidal nanorods (Fig. 13b) [104, 105]. This work demonstrates that the control of the shape or the confinement dimension of the nanostructures could provide additional advantages for lasers.

6.2 Colloidal quantum wells

With the further progress of chemical synthesis of semiconductor nanostructures, 2D colloidal quantum wells, analogous to those fabricated by the costly molecular beam epitaxy or metallo-organic chemical vapor deposition techniques, can be attained by the solution-phase method (Fig. 14a) [34, 106, 107]. Different from the epitaxial quantum wells, the colloidal quantum wells typically possess small lateral sizes with length and width of tens of nanometers as shown in Fig. 14a. Hence, this class of colloidal quantum well is also termed as nanoplatelets. Although colloidal quantum wells with larger lateral dimensions in the micrometer range have been synthesized, the PLQY of them is rather poor at this stage [107]. Strikingly, the thickness of the colloidal quantum wells can be precisely controlled at the atomic level, which results in the absence of inhomogeneous spectral broadening in the quantum well ensemble and hence extremely narrow PL linewidth [34, 108]. Such optically uniform quantum wells could inhibit the unfavorable inter-particle nonradiative energy transfer, which is advantageous for optical gain and lasing applications [109, 110]. Similar to the colloidal nanorods with large aspect ratio, the mutiexcitonic AR in colloidal quantum well is unraveled to follow the second-order kinetics, indicating that AR is dictated by bimolecular fashion as a result of the negligible quantum confinement effect in the width and length direction [111, 112]. More importantly, the AR in colloidal quantum wells is demonstrated to be greatly suppressed compared to that of the CQDs with similar emission wavelength, which can be attributed to the large volume and the strict

restriction on momentum conservation in colloidal quantum wells [111]. As such, the colloidal quantum wells are expected to function as favorable lasing materials. Indeed, optical gain and lasing action have been observed from the CdSe-based colloidal quantum wells (Fig. 14c and d) [109, 113]. The stimulated emission threshold was measured to be as low as $6 \mu\text{J}/\text{cm}^2$ under femtosecond excitation (pulse-width: 100 fs) and the model gain coefficient as high as 600 cm^{-1} . The systematic studies reveal that, with the decrease of the lateral dimensions, the PL quantum efficiency increases and stimulated emission threshold decreases due to the reduced number of the defected sub-population and possibly less scattering loss in the small-sized quantum wells (Fig. 14b) [107]. Leveraging on the superior gain performance, Grim *et al.* succeeded in observing room-temperature continuous-wave lasing from the colloidal quantum well [114], which represent an important milestone in developing lasers from colloidal semiconductor nanostructures (see more details in **Section 8.3** Continuous-wave lasing).

6.3 Colloidal nanostructures with mixed dimensionality

The colloidal nanocrystals with mixed dimensionality could provide new possibilities in photonic applications, such as providing simultaneous dual-color emission [115]. CdSe/CdS dot-in-rod and CdSe/CdS dot-in-tetrapod heterostructures are the most representative nanocrystals of mixed dimensionality [116-118]. In these heterostructures, the absorption and the emission can be designed to occur in different part, namely, the former mainly takes place in the CdS rod or tetrapod, while the later takes place in the CdSe core. On one hand, the CdS rods/tetrapods are able to serve as the light antenna, which significantly enhances the effective absorption cross-sections of the CdSe/CdS heterostructures. On the other hand, this unique property endows these heterostructures with the additional functionality termed “Stokes-shift engineering”, such that the detrimental reabsorption effect can be significantly suppressed, contributing to low-threshold

stimulated emission and lasing [118-120]. Moreover, the above-mentioned strategies for AR engineering can also be applicable in these heterostructures, which can further reduce the gain threshold (Fig. 15a and b) and enable the achievement of stimulated emission involving higher-order multiexciton states (Fig. 15c) [118, 121, 122].

7. CQD lasers with various resonator configurations

One of the most important advantages of CQDs lies in the solution processibility, which promises the development of cost-effective lasers. Furthermore, CQDs essentially have no request on the substrate, whereas the strict lattice matching condition is usually required for the epitaxial semiconductor lasers. As a result, the CQDs are compatible with almost all kinds of the cavity architectures. Since the first observation of stimulated emission from CQDs, tremendous efforts have been devoted to attaining the true lasing behavior using various resonator configurations. Until now, nearly all kinds of optical feedback mechanisms have been realized from CQD lasers. Malko *et al.* pioneered to coat the CQDs solid film onto the inside wall of microcapillary tubes and claimed the observation of lasing in whispering-gallery modes at 80 K [123]. However, due to the scattering from the inhomogeneous films, the Q-factor is very low and the lasing peaks are not clear, which makes the assignment of lasing mechanism inexplicit. After that, a number of follow-up studies have been done to improve the device performance. Wang *et al.* greatly improved the lasing performance by infiltrating the CQD solution into the hollow fiber to realize the liquid CQD lasers at room temperature (Fig. 16a) [124]. The detailed lasing characterization and analysis unambiguously attribute the longitudinal laser modes to whispering-gallery-mode lasing [124]. Bawendi *et al.* have demonstrated a facile approach to covering the CQDs onto the surface of silica or polystyrene microspheres, where the individual microsphere actually serves as a laser resonator, offering the whispering-gallery-mode optical

feedback (Fig. 16b) [25]. As a consequence, hundreds of CQD-based microlasers can be produced by a single spin-coating process, enabling the low-cost and easy mass-productive laser fabrication. Interestingly, the CQD liquid droplet can act as an oscillating resonator itself such that the high-Q whispering-gallery-mode lasing can be achieved from the levitated micro-droplets (Fig. 16c) [125]. These microsphere whispering-gallery-mode lasers can be also pumped evanescently by the tapered optical fibres (Fig. 16d), which allows efficient in-coupling and out-coupling of the pumping and emitting light, so as to constitute the compact and integrated laser systems [126, 127].

More handy random lasers can be realized from the close-packed CQD films by deliberately introducing suitable scattering centers (Fig. 16e) [16, 128, 129]. Although the CQD random lasers are very efficient light sources and low-cost, the lasing spectra and the output beams are of poor quality exhibiting multi-mode lasing peaks and non-directional lasing emission [16, 128]. In order to achieve the single longitudinal mode operation, the distributed feedback (DFB) lasers are fabricated from the CQD/grating hybrids (Fig. 16f) [20, 130-133]. The DFB lasers can also be made mechanically flexible by using the ultra-thin glass membrane substrate, enabling new functionalities of CQD-based lasers [131]. Furthermore, the vertical cavity surface emitting lasers featuring desirable coherent output beams made from CQDs were demonstrated by confining the CQDs inside two parallel reflective distributed Bragg mirrors consisted of alternating layers of different refractive indices (Fig. 16h) [134, 135].

By virtue of the small size of the CQDs, they can be facilely integrated with the extremely miniaturized laser cavities, exemplified by the photonic-crystal nanobeam cavity (Fig. 16g) [136], such that the energy-efficient CQD-based nanolaser can be made benefiting from the small mode-volume cavities adopted that reduce the dosage of the gain media and enhance the

spontaneous emission coupling into the cavity mode by Purcell effect [137]. To fully manifest the advantage of colloidal nature (e.g. particle suspension) in the CQDs, the ink-jet printing technique has been employed to fabricate CQD microlasers [18, 19]. As a result of the “coffee ring” effect, the CQDs will self-assemble into circular quasi-toroid shape which serves well as the high-Q laser cavities and hence either the whispering-gallery-mode or the Fabry–Pérot lasing can be obtained [19, 138]. Furthermore, with the advent of electrohydrodynamic printing, the CQDs can be precisely placed at on-demand positions with nanometer precision [139]. Taking advantage of the technological advance, Kress *et al.* have demonstrated the CQD–based spaser (surface plasmon amplification by stimulated emission of radiation) that allow controlled generation, extraction, and manipulation of plasmons by locating the CQDs inside the high-quality plasmonic cavities [140].

The achievements in CQD lasers in the past decades highlight the attractive solution processability of CQDs in addition to the wide range emission-color tunability, which promise the complement to the technologically mature epitaxial semiconductor lasers.

8. Recent advance in CQD lasing

8.1 Novel concept of single-exciton gain

As discussed above, the AR is recognized as the major hurdle against stimulated emission and lasing from CQDs. Thus, tremendous efforts have been devoted to reducing AR rate for optimization of optical gain. However, if the optical gain could develop from the single exciton state rather than the multiexciton states, the detrimental AR would not be of any trouble at all. This is the motivation for the advent of novel concept of single-exciton gain in CQDs [141]. In principle, the single exciton state can only give rise to the situation of optical transparency where

the stimulated emission is completely consumed by the ground-state absorption (Fig. 17a). Yet, when the carrier-induced Stark effect is taken into consideration, the situation becomes different and the optical gain from single exciton state might be possible. Specifically, the photoexcited single exciton state will generate a local electric field which lifts the transition energy of the ground-state absorption (Fig. 17b), which is known as the Stark shift. For conventional type-I CQDs, the ground-state absorption energy will shift downward due to the attractive exciton-exciton interaction upon single exciton formation. In contrast, in type-II CQDs, the electrons and holes are distributed separately in the core and the shell, such spatial imbalance of electron and hole wave functions will lead to the repulsive exciton-exciton interaction; therefore, the ground-state absorption energy will shift upward for the singly excited CQDs (Fig. 17b). In the ideal case, the theoretical limit for single exciton gain in terms of average exciton per CQD is $\langle N \rangle = 2/3$ [141]. The repulsive exciton-exciton interaction in CQDs was first proposed and observed by Ivanov *et al.* in 2004 based on the type-II ZnSe/CdSe heterostructure [142]. Unfortunately, due to the intrinsically smaller repulsive energy (Δ_{XX}) of exciton-exciton interaction than the transition line-widths in ZnSe/CdSe CQDs, they failed to achieve the single-exciton optical gain but only observed blue-shifted stimulated emission peak with respect to the spontaneous emission maximum for the first time [142]. Three years later, Klimov *et al.* engineered the novel type-II CdS/ZnSe core-shell CQDs (Fig. 17e and f), from which strong repulsive exciton-exciton interaction with $\Delta_{XX} > 100$ meV was realized according to both theoretical and experiment analysis [141]. Such large repulsive energy effectively hinders the detrimental absorption from singly excited CQDs and enables the first observation of single-exciton gain [141]. In contrast to the conventional type-I CQDs, the stimulated emission peak from CdS/ZnSe CQDs coincides with that of spontaneous emission (Fig. 17c), indicating that

optical amplification originates from single-exciton state. Moreover, the second stimulated emission band corresponding to the biexciton recombination appears as pumping power increases, further confirming the achievement of single-exciton gain from these CdS/ZnSe CQDs (Fig. 17d). Following the same idea, the single-exciton gain was realized in the quasi-type-II CdSe/CdS nanorods with engineered strong repulsive inter-exciton interaction in 2013 (Fig. 17g) [126]. Notably, the type-I CdSe/Zn_{0.5}Cd_{0.5}S core-shell CQDs were claimed to exhibit single-exciton gain by Dang *et al.* based on the fact that the calculated average number of excitons per CQD at threshold was significantly lower than unit [135]. The authors proposed that the valence band degeneracies may be lifted in the CdSe/Zn_{0.5}Cd_{0.5}S CQDs, which rationalizes the realization of single-exciton gain in their type-I heterostructures. Importantly, this work achieved the low-threshold blue, green and red stimulated emission and lasing from the CQDs with the same composition (Fig. 17h), which represents impressive progress toward solution-processed full-visible-color CQD lasers [135].

8.2 Multi-photon pumped CQD lasing

Multi-photon pumped laser means that the lasing action is induced by the simultaneous absorption of several photons with sub-band gap energy (Fig. 20b) [143]. Compared to the one-photon excitation, the multi-photon counterpart adopts much longer pumping wavelength in infrared range and exhibits a super-linear dependence of absorption on incident power, which results in a larger penetration depth into samples and a higher spatial resolution [144]. The pioneering work on the theoretical prediction of two-photon absorption was done by Maria Goeppert-Mayer [145]. The generation rate of electron-hole pairs in an atom or a molecule by absorbing two photons with identical energy and momentum at the same time can be derived according to the second-order perturbation theory, and the solution is given by [146]:

$$W^{(2)} = \frac{2\pi}{\hbar} \sum_{u_1, u_0} |M_{u_1, u_0}| \delta(E_{u_1} - E_{u_0} - 2\hbar\omega),$$

$$M_{u_1, u_0} = \sum_{u_2} \frac{V_{u_1, u_2} V_{u_2, u_0}}{E_{u_2} - E_{u_0} - \hbar\omega - i\hbar\gamma_{u_2}}$$

where u_0 , u_1 and u_2 denote the entire set of initial, final and intermediate states with the quantum numbers n , l and m , γ_{u_2} is the reciprocal lifetime and V is the interaction operator represented by [42]:

$$V_{f,i} = \left\langle \psi_f \left| \frac{e}{mc} \vec{A} \cdot \vec{P} \right| \psi_i \right\rangle$$

From above analysis, it can be found that the two-photon absorption coefficient which can be facilely determined by the well-established Z-scan technique (Fig. 18a) is much smaller than the above band gap single-photon absorption. Therefore, extremely high pumping densities on the order of GWcm^{-2} are anticipated for activating two-photon pumped lasers. Nevertheless, thanks to the relatively large two-photon absorption cross-sections and favorable photostability of CQDs compared to those of organic dyes, two-photon pumped stimulated emission and lasing have been demonstrated from a variety of CQDs but with impractically high pump thresholds [147-149]. Leveraging on the antenna effect which has been known to boost the effective absorption cross-sections in CdSe/CdS dot-in-rod heterostructures, Xing *et al.* realized the low threshold ($\sim 1.5 \text{ mJ/cm}^2$) stimulated emission from the CdSe/CdS quantum rods, which is nearly one-order of magnitude smaller than those of the typical CQDs [150]. The carrier dynamic studies reveal an extremely fast decay process ($\sim 7 \text{ ps}$) when pump intensity exceeds certain value, which is much faster than the spontaneous emission and the Auger recombination (Fig. 18d), indicating the achievement of stimulated emission [150]. Even lower pump threshold ($\sim 1.2 \text{ mJ/cm}^2$) for the two-photon induced optical gain and lasing have been reported in CdSe-based colloidal quantum wells,

which is ascribed to the high two-photon absorption cross-sections as a result of the large volume [151]. Furthermore, Wang *et al.* exploited the CdSe/CdS/ZnS core-multi-shell CQDs with quasi-type-II band alignment to achieve the three-photon pumped stimulated emission and coherent random lasing, which represents a significant step toward high-order nonlinear optically pumped CQD lasers [128]. Through comprehensive spectroscopy analysis, the success is ascribed to the enhanced three-photon absorption cross-sections, suppressed nonradiative AR, strengthened photostability and reduced reabsorption effect with respect to that of one- and two-photon analogues (Fig. 18e). The feasibility of adopting CQDs as optical gain media based on multi-photon pumping is not only attractive for fundamental physics, but also could offer new possibilities in photonics and biophotonics where long excitation wavelengths and high-order nonlinear excitation are strongly desired, such as laser-assisted disease diagnostics and the development of infrared optically pumped compact lasers.

8.3 Continuous-wave lasing

The realization of continuous-wave (CW) lasing from solution-processable gain materials is highly desirable for practical applications in various fields including data communication, spectroscopy, and sensing [152]. Moreover, optically pumped CW lasing is generally accepted to be an important stepping stone towards the powerful electrically pumped laser diode [153-155]. Ever since the first demonstration of stimulated emission from CQDs, the pursuit of CW CQD lasing has become the subject of intense research, which, however, is demonstrated to be challenging due to the relatively high pump threshold and the short optical gain lifetime limited by the fast AR in CQDs [23]. The first report of CW lasing from colloidal nanocrystals was made by Grim *et al.* [114]. The authors take advantage of the large absorption coefficient and relatively low AR rate of CdSe colloidal quantum wells to achieve the stimulated emission with

extremely low threshold of $\sim 6 \mu\text{Jcm}^{-2}$ under femtosecond laser excitation (pulse-width: 70 fs). Leveraging on the superior gain properties, they move forward to demonstrate the surface-emitting lasing in CW operation from CdSe colloidal quantum wells, representing an important breakthrough in developing lasers from colloidal nanostructures [114]. Later on, Yang *et al.* drop-casted the CdSe/CdS core-shell colloidal quantum wells onto the silicon nitride photonic-crystal cavity and claimed the achievement of stable CW nanobeam laser with an extremely low threshold power of less than $1 \mu\text{W}$ [137]. These reports about the CW lasers from colloidal CdSe nanoplatelets are very inspiring. However, one should be very cautious to recognize the real lasing action, especially in the form of nanolasers (e.g. in Yang's work) where it is difficult to distinguish the lasing radiation from the spontaneous emission [156]. Moreover, the results in Grim's work may require further verification. In particular, according to the reported threshold ($6 \mu\text{Jcm}^{-2}$) under femtosecond pulse pumping, the required pump threshold in CW operation can be estimated to be $\sim 5 \times 10^4 \text{ W/cm}^2$, which, however, is several orders of magnitude higher than that observed in the experiment (6.5 W/cm^2) [76, 114].

Very recently, the CW lasing from CdSe CQDs have been realized by exploiting the strategy of decreasing the level degeneracy using strain (Fig. 19a,b). The strain engineering has been previously demonstrated to be an enabling method to reduce the pump threshold in epitaxial quantum well lasers [157-159]. In the work, Fan *et al.* developed the so-called facet-selective epitaxy to grow asymmetric compressive shells on CdSe cores so that the splitting of the band-edge states can be larger than the thermal energy (Fig. 19b) due to the introduction of the biaxial strain [159]. Accordingly, the effective level degeneracy of the CQDs is reduced, resulting in greatly decreased optical gain threshold as compared to those without biaxial strain (Fig. 19d and e). Eventually, by coating these biaxially strained CQDs onto the 2D photonic crystal distributed

feedback optical cavity, optically pumped CW laser was achieved with low thresholds of 6.4–8.4 KW/cm² (Fig. 19c) and favorable spatial coherence (Fig. 19f). This work represents an excellent example to optimize the optical gain from CQDs by borrowing experiences from the mature epitaxial semiconductor lasers and may inspire more skillful schemes for tuning the electronic structure of CQDs in view of developing more practical CQD-based CW lasers.

8.4 Emerging perovskite nanocrystals for lasing

The all-inorganic cesium lead halide (CsPbX₃, X =Cl, Br, I) nanocrystals are emerging as the attractive light-emitting materials ever since the report of the seminal work by Protesescu *et al.* in 2015 [160]. In contrast to the CdSe-based CQDs which necessitate the additional shell coating to ensure the high PLQY (>50%), these bare CsPbBr₃ perovskite nanocrystals show even higher PLQY (>90%) without any deliberate surface passivation, which may be attributed to the defect-tolerant nature, large exciton binding energy, equal extent of confinement for charge carriers and self-formed surface protective layer [160-162]. Besides, the fabrication of CsPbX₃ nanocrystals can be carried out at room temperature and free from inert environment, whereas the high temperature and protective gases are generally indispensable for fabrication of CdSe-based CQDs [161, 163-165]. Most importantly, the CdSe CQDs are known to suffer from photodegradation and relatively poor PLQY in the short visible wavelength range (410–530 nm), while the emerging CsPbX₃ perovskite nanocrystals exhibit superior optical properties in the blue-green spectral regime, rendering them as the appealing complement to the traditional metal-chalcogenide CQDs [160, 166].

The superior optical gain properties and lasing action from the CsPbX₃ perovskite nanocrystals were first reported by Wang *et al.* and Yakunin *et al.* independently in 2015 [167, 168]. By that time, the overwhelmingly investigated CQDs for developing lasers have been made

from metal-chalcogenide semiconductors with crystal structure of either zinc blend or wurtzite [24]. Although materials with perovskite structure have long been disclosed to show interesting physical properties, such as superconductivity and ferroelectricity, the perovskite nanocrystals have not yet manifested impact in photonics and optoelectronics [169-171]. The above two works have brought a new material system into the laser field, and, as expected, they attracted extensive attention immediately. Fig. 20a shows the development of stimulated emission from the thin film of CsPbBr₃ nanocrystals with the increase of pump density. The threshold is extracted to be as low as $\sim 20 \mu\text{J cm}^{-2}$ under femtosecond laser excitation (pulse-width: 100 fs) (Fig. 15b), which is several times smaller than those of CdSe CQDs [135]. An even lower threshold of $\sim 6 \mu\text{J cm}^{-2}$ was detected by Yakunin *et al.* [168], where the difference may come from the quality of the samples, the film thickness as well as the experimental errors. Based on the comprehensive spectroscopic analysis, the low-threshold stimulated emission from CsPbBr₃ nanocrystals is attributed to the large absorption cross-section ($\sim 2.5 \times 10^{-14} \text{ cm}^2$ for 9 nm-sized CsPbBr₃ nanocrystals), the suppressed reabsorption due to the large biexciton binding energy ($\sim 50 \text{ meV}$), high PL QY ($\sim 90\%$) and the relatively mild Auger loss ($\sim 100 \text{ ps}$ for 9 nm-sized nanocrystals) [167, 172]. The model gain of $\sim 450 \text{ cm}^{-1}$ was determined from CsPbBr₃ nanocrystals according to the variable stripe length technique (Fig. 20e). Notably, the stimulated emission and lasing can be easily tuned from blue, green to red colors benefiting from the facile composition and size dependent band gap energy (Fig. 20d) [167, 168].

By coating a thin film of CsPbBr₃ nanocrystals onto the inner wall of a capillary tube, the high-quality whispering-gallery-mode microlasers with Q-factor of ~ 2000 were demonstrated (Fig. 20f) [167]. Through a similar approach, the CsPbBr₃ nanocrystals can be covered onto the silica microspheres and whispering-gallery-mode laser was realized by virtue of the total internal

reflection around the circumference of the microsphere (Fig. 20g). In general, the whispering-gallery-mode lasers are favorable in obtaining high Q-factor and small mode volume [173]. However, the output beam suffers from the poor spatial coherence, which greatly limits the practical application. Bearing this in mind, Wang *et al.* inserted the thin film of CsPbX₃ nanocrystals between two distributed Bragg reflectors where the gain profile of the perovskite nanocrystals are designed to match well with the high reflectivity band of the reflectors, and, eventually, realized the single longitudinal mode vertical cavity surface emitting lasers (VCSEL) [174]. The representative lasing spectra from green-, blue- and red-emitting CsPbX₃ nanocrystals enabled VCSELs are shown in Fig. 20h and i. Note that VCSEL is a much desired laser type which have found important applications throughout our daily life including optical communication, high density optical storage, laser display, signal processing and parallel optical computing [175]. The achievement of CsPbX₃ nanocrystals activated VCSELs highlights the feasibility of independent manipulation of the resonant cavity and solution-processable nanocrystals, which represents great progress toward cost-effective solution-processed lasers.

Despite such remarkable progress, several concerns have to be addressed before the perovskite nanocrystals can be used successfully in practical laser devices. It is noted that the AR rate of the CsPbX₃ nanocrystals is much faster than the relatively mature CdSe-based heterostructures, which is unfavorable for the operation in long pulse duration and continuous wave mode [176]. To address this issue, one may adopt the core-shell heterostructure engineering as used in conventional QDs to suppress the Auger loss, which, however, is not yet applicable for perovskite nanocrystals due to the spontaneous anion-exchange reactions [162]. Moreover, although the CsPbX₃ nanocrystals have demonstrated much better stability than the organic-inorganic hybrid perovskites, they still suffer from the relatively poor stability against

the moisture and heat, which is even worse for the red-emitting CsPbX₃ nanocrystals, known as the “red wall” [162].

9. Issues and Challenges

9.1 Toxicity

Until now, the most intensively investigated CQDs for lasers that cover the technologically important visible and near-infrared spectral windows are CdSe- and PbS-based CQDs, respectively [177-179]. However, the presence of toxic heavy metals in these CQDs is a big concern for the environment and health safety, which may stifle their use in consumer applications. The same problem exists in the emerging CsPbX₃ nanocrystals, which have been recognized as the advantageous lasing materials, especially in the short visible range. The European Union’s Restriction of Hazardous Substances Directive explicitly limits the amount of cadmium-, lead- and mercury-containing compounds that can be adopted in electronic devices [180, 181]. Particularly, cadmium is restricted to 100 ppm, which is ten times less than that of the other heavy metals [180, 182]. As a consequence, research into lasers made from the heavy metal-free CQDs has become imperative. Carbon dots are a kind of well-known environmental friendly materials [183-185]. Zhang *et al.* and Qu *et al.* have claimed the observation of lasing action from the carbon dots [186, 187]. However, in addition to the impractically high pump thresholds, the lasing action is found to be extremely sensitive to extrinsic conditions. For instance, Qu *et al.* reported that the sp² C and N atom doping is critical to achieve stimulated emission from carbon dots [187]. Moreover, only the carbon dots dispersed in mixed ethanol aqueous solution can develop stimulated emission, while changing the solution to pure water or pure ethanol will completely extinguish it [187]. As a matter of fact, the carbon dots are a much

more complex system than expected and the emission mechanism of carbon dots has remained an open question [188], which may make them ineligible for lasing media at this moment. InP-based CQDs have notably emerged as an attractive candidate for developing the heavy metal-free CQD lasers due to its broad spectral tunability and maturing synthesis [189-191]. Although there have been a number of reports on InP CQD-based LEDs with rapidly improved performance in the past few years, the studies on the InP CQD lasers are far laggard [191-193]. Up to date, lasing action from InP/ZnS CQDs has only been reported by Gao *et al.* in 2011 [191]. However, despite that the pump threshold reported in that work was more than one order of magnitude higher than those of CdSe-based CQD lasers, there is no follow-up work reported for more than 7 years, which makes the prospect of InP CQD lasers elusive. The InGaP CQDs have also been reported to support lasing action, but the gain threshold is relatively high [134]. More issues remain in other heavy metal-free CQDs for laser applications. For example, the ternary I–III–VI₂ chalcogenide CQDs, such as CuInS₂ and, CuInSe₂, suffer from the intra-band defects due to the wide range of stoichiometric variations and complex crystal structure [181, 194, 195]. As for the earth-abundant silicon-based CQDs, the different synthetic methods, precursors, and post-treatment will result in greatly distinct optical properties, and the emission mechanism from silicon nanocrystals still remains unclear to date [196-198].

In short, although several reports have demonstrated the success in achieving heavy metal-free CQD lasers, none of them could show satisfactory performance that matches those of the CdSe-based CQD lasers. Therefore, there is much work to be done in pursuing the high-performance metal-free CQD lasers, which may require the combined efforts from the theory, wet-chemistry fabrication and characterization.

9.2 Stability

Despite that the CQDs are much more stable than the typical laser dyes, they are still vulnerable to oxygen/water and will degrade upon exposure to air due to the existence of labile organic ligands [199]. As such, the poor long-term stability of CQDs has become a critical issue towards practical laser applications. To solve this problem, researchers have attempted to embed the CQDs into a protective matrix so as to attain robustness against the environment [200-203]. Initially, the sol-gel derived titania have been exploited as the host matrix and the hybrid CQD/titania composites show long-term stable stimulated emission and lasing at room temperature [25, 52]. However, the CQD/titania system exhibit fast structural and photophysical degradation, such as cracking, when exposed to water and short-chain alcohols, making them unsuitable for applications in moisture circumstances [204]. Later, Chan *et al.* disclosed that the sol-gel derived silica are better host matrix for CQDs, which results in robust optical gain and lasing even under different chemical environments including polar solvents, making it suitable for integration with microfluidic systems [204]. Nevertheless, the complex ligand exchange is requisite to incorporate the large volume CQDs into the silica matrix and the PLQY of the CQDs is largely compromised.

Poly(methyl methacrylate) (PMMA), a kind of organic polymer, has been recognized as the favorable matrix for CQDs due to the low cost, optical transparency and chemical/physical inertness [132]. The hybrid CQD/PMMA nanocomposites have been widely employed in different kinds of optoelectronic devices including the active waveguides, solar concentrators and LEDs [205-208]. However, the loading concentrations of CQDs are typically low ($< \sim 10\%$) so as to form the homogeneous nanocomposite films with less optical scattering loss. Interestingly, Sun *et al.* realized a novel type of CQD-based whispering-gallery-mode microlasers based on the highly concentrated CQD/PMMA (~ 45 wt%) nanocomposites [209].

The microbubbles with varied sizes can be formed during the drying process of the nanocomposite droplets due to the synergetic effects of CQD-assisted bubbling and stabilizing by PMMA, which naturally provide the low-loss whispering-gallery-mode optical feedback, leading to the development of low-threshold and high-Q microlasers. Compared to the matrix-free CQDs, these CQD/PMMA composites show much improved long-term robustness against environment and the microlasers can operate favorably under water and other polar solvents [209]. Despite that the microbubble lasers are attractive for photonic and biophotonic applications, the thin composite films with high CQD concentration stand for a more versatile optical gain medium, which allows the fabrication of different types of lasers [204]. However, until now, it is still challenge to produce the hybrid CQD/polymer thin films with high optical gain, which may be attributed to the easy aggregation of the CQDs and the poor compatibility between the CQDs and the conventional polymers [210-212].

In addition to provide the protection by the matrix, it would be another effective approach to replacing the labile organic ligands by the more robust inorganic ligands [24, 213]. These inorganic ligands can also improve the conductivity of the CQDs, enabling the additional functionality in light detection [213, 214]. However, the research in this aspect is still in its infancy. Currently, the inorganic ligands cannot reach satisfactory surface passivation for CQDs, manifested by the poor PLQY, and the dispersibility in solution is poor [214].

The stability issue represents one of the most important concerns for practical applications. At this moment, the robustness of CQDs obtained by the wet chemistry method is much inferior to that of the technologically mature epitaxial semiconductors. As a result, more efforts should be devoted to improving the long-term stability of CQDs in addition to merely demonstrating novel and fancy CQD lasers.

9.3 Electrically pumped lasing

Until now, the lasing demonstrations from CQDs are exclusively activated by optical pumping. However, in most of the lasing applications, the electrical pumping is more practically desirable [215]. The challenges to achieve electrically pumped CQD laser are multifold and complex, but have been much better understood now thanks to the comprehensive optical studies. As discussed above, the optical gain from CQDs originates from the multiexciton states due to the double degeneracy of the band-edge level. Therefore, the Auger recombination which is recognized as the main gain depletion pathway under optical pumping will even more seriously plague the electrically driven CQD lasing since the carriers are injected into CQD one-by-one [23, 216-218]. Furthermore, the organic ligands are inevitably adopted to passivate the CQD surface and attain the ink stability in solution. However, these surface organic ligands generally exhibit poor electric conductivity and act as an insulating layer around the CQDs which hinders the carrier injection and transport [217, 218]. In contrast to the epitaxial semiconductor lasers which adopt a p-n junction to infuse electric current, an electron transport and a hole transport layer are required to inject the excess carriers. However, up to date, it is still difficult to realize efficient current injection into CQDs due to the limited material selection [217, 219]. Last but not the least, even if electric current can be well injected into CQD film, the high flowing current (tens of A cm^{-2}) required to achieve the population inversion in CQDs will easily damage the laser device by Joule heating induced temperature increase during electrical injection [220]. Very recently, Lim *et al.* reported the achievement of optical gain from CQDs under electrical pumping [221]. The authors exploited the $\text{CdSe/Cd}_x\text{Zn}_{1-x}\text{Se/ZnSe}_{0.5}\text{S}_{0.5}$ CQDs with alloyed core-shell interface as the gain medium (Fig. 21a) such that extremely long biexciton Auger lifetime of 2.4 ns was obtained, and then they employed the a p-i-n architecture with optimized

organic/inorganic charge transport layers and inserted a deliberately shaped insulating spacer between the thin CQD film and charge-injection layers (Fig. 21c), which not only enables the high current injection (18 A cm^{-2} , sufficient for the development of optical gain) (Fig. 21b), but also allows effective heat dissipation from the active area. This work represents the first achievement of optical gain from CQDs by electrical pumping, which represents another milestone in developing practical CQD lasers. However, it has not yet been accomplished to demonstrate a true electrically injected CQD laser because it may not be straightforward to introduce a suitable cavity to provide the optical feedback [221, 222]. Moreover, despite that the electrically driven optical gain has been demonstrated from CQDs in the red spectral range, it is more challenging to shift the gain wavelength to green and blue regime due to the more serious Auger issue and inferior current injection, which has been similarly troubling the CQD-based LEDs [21, 217].

10. Summery and Outlook

In the past two decades, remarkable progress has been made in realizing lasers from colloidal semiconductor nanostructures in terms of both understanding the fundamental physics and improving the device performance. In this Review, we start with a brief introduction about the wet-chemistry method for synthesizing the highly luminescent CQDs, followed by the description of the electronic structure and optical transitions in CQDs. More efforts are focused on the discussion about the optical gain and development of lasers from colloidal nanostructures including the gain mechanism, the main hurdles against the development of optical gain and strategies to enhance the gain performance. Fig. 22 shows the timeline of the major findings in development of CQD lasers since the first demonstration of stimulated emission from CQDs in year 2000. We highlight the understanding of Auger recombination in the quantum confined

colloidal nanostructures, its impact on the optical gain and lasing performance as well as the progress in reducing the lasing threshold by AR suppression and shape engineering of the nanostructures. Especially, the recent advance of CQD lasers is very inspiring, such as the achievement of continuous wave lasing and the first demonstration of optical gain from CQDs by direct electrical pumping. Nevertheless, there is still a wealth of open questions to be answered and challenges to be addressed in order to bring the CQD lasers out from the laboratory to consumers.

First of all, the optical gain threshold of CQDs is still too high for practical applications. Table 1 summarizes the representative optical gain thresholds from different kinds of colloidal nanostructures. Despite that the continuous wave lasing has been achieved from the deliberately designed biaxially strained CQDs, the threshold of several KW/cm^2 may not be easily affordable by the cheap coherent light sources [159]. Moreover, until now, the continuous wave stimulated emission and lasing have only been demonstrated in red colors, extending the lasing wavelength down to blue and green region may not be straightforward because the Auger loss will become more serious for smaller-sized CQDs [21]. In order to further decrease the gain threshold, a much more comprehensive knowledge about the parameters that impact the performance of CQD-based gain media has to be gained, which requires a much deeper understanding of the gain mechanism and the gain dynamics from both theoretical and experimental investigations. For example, it is still not clear how the phonons will influence the optical gain in colloidal nanostructures despite that the exciton-phonon coupling plays an important role in many optical processes [223]. Moreover, the surface-localized states within the band gap which are usually treated as the detrimental issue may be beneficial for building a laser because it could help to

construct a 4-level system under certain conditions, which contribute to an extremely low lasing threshold [224].

Second, so far, most of the progress in developing CQD lasers (see Fig. 22) is made in CdSe-based or PbS-based CQDs which contain the toxic heavy metals. With the rapid increasing demand for sustainable and economical technology, it has become an inevitable trend to study the non-toxic CQDs and developing the heavy metal-free CQD based lasers. In this regard, more efforts should be devoted to investigate the existing non-toxic or less toxic CQD, such as the InP CQDs, carbon dots, ZnO CQDs and so on. On the other hand, there is a plenty space to explore the novel kinds of CQDs which consist of earth abundant and eco-friendly elements. Last three years have witnessed the booming development of lead halide perovskite nanocrystals. However, these emerging CsPbX_3 nanocrystals are similarly challenged by the issue of possessing toxic lead elements despite that they exhibit superior optical properties [162].

Third, although both optically and electrically pumped lasers have been extensively used in fields ranging from fiber optic communication to medical therapeutic, it would be more convenient that these lasers developed from the colloidal semiconductor nanostructures can be compatible with the ubiquitous electric grids and even the portable batteries. The ideal case is to develop the CQD lasers by direct electrical pumping, which, however, is demonstrated to be extremely challenging and have not yet been achieved [221]. A compromise proposal is to realize the CQD laser by the so-called “indirect electrical pumping” [225]. In this situation, the CQD lasers are integrated with the semiconductor LEDs which are commercially available with pretty low price. If the pump threshold of the CQD lasers can be reduced to be sufficiently low and is affordable by the incoherent radiation from the common LEDs, then the indirect

electrically pumped lasing in the hybrid QD laser can be realized [226]. Given the mature technology in InGaN LEDs and the advantages of QDs [227], these hybrid lasers can be made compact and cost-effective, which may be practically beneficial in various photonic and optoelectronic fields.

Last but not the least, almost all the research efforts to date are focused on the demonstration of lasing action from QDs by exploiting different kind of architectures, but too little attention has been paid to the exploration of real applications of QD lasers. Although some proof-of-concept applications have been demonstrated, such as the refractive index sensing and gas sensing [131, 132, 209, 228], the unique advantages of QD lasers are far from being fully exploited. Taking advantages of the solution processibility and the compatibility with nearly all types of substrates, these QD lasers can be envisaged to serve as the building blocks for the development of CMOS-compatible on-chip integrated photonic circuits which have been recognized as the key element for modern technology [17, 229, 230]. Moreover, the lasers made from the colloidal nanostructures could contribute more to the existing and emerging fields where the epitaxial semiconductor lasers are unable to penetrate, such as the ultrathin displays made from QD laser arrays on flexible surfaces and nanolaser sensor that can be implanted into bodies [231-233].

In summary, we believe that with further advances through a combination of innovative steps in materials science, spectroscopy and device technology, the solution-processed lasers made from the colloidal semiconductor nanostructures will play an important role in a variety of photonic and optoelectronic applications and may serve as an attractive complement to the epitaxial semiconductor lasers in the near future.

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Fig. 1. (a) Representative illustration of core-shell colloidal quantum dots. Adopted from Nanotech-Dubna. (b) Solution of CdSe CQDs with different sizes, manifesting the band gap tunability by size control. Adopted from Evident Technologies, Inc.

Fig. 2. Hot-injection method for the synthesis of colloidal CdSe-based quantum dot. Typically, the three-necked flask is put inside a heating mantle and furnished by a thermocouple and temperature controller components. Reprinted with permission from Ref. [28].

Fig. 3. (a) Variation of band gap energy of quantum dot by the quantum confinement effect as well as the emergence of discrete band levels as the dot size decreases. (b) The relationship between density of electronic state and the dimension of the bulk and nanostructures. Adopted with permission from Ref. [39]. (c) Optical transitions in quantum confined CdSe colloidal quantum dots. Adopted with permission from Ref. [45]. (d) Representative linear optical absorption spectrum of 4.1 nm-sized CdSe CQDs manifesting the well-resolved optical transitions. Reprinted with permission from Ref. [45].

Fig. 4. (a) Schematic illustration of optical absorption from the ground-state quantum dot (b) Schematic illustration of optical transparency from the singly-excited quantum dot. (c) Schematic illustration of optical gain from doubly-excited quantum dot.

Fig. 5. (a) Transient absorption spectra from colloidal quantum dot, showing the optical gain with average number of excited excitons larger than unit. Reprinted with permission from Ref. [23]. (b) Transient absorption signal at different energies derived from the transient absorption spectra, showing the optical gain located at the lower energy side of the absorption edge. Reprinted with permission from Ref. [23]. (c) Normalized time-integrated PL spectrum and time-resolved upconversion PL spectra measured at time delay of 1 ps and 200 ps. Reprinted with permission from Ref. [45]. (d) Single exciton and multiexciton upconversion PL spectra at fixed time delay of 1 ps and different excitation densities. Reprinted with permission from Ref. [53].

Fig. 6. (a) Schematic illustration of limitation of Auger recombination in bulk semiconductors by the energy and momentum conservation. Reprinted with permission from Ref.[86]. (b) Schematic illustration of Auger recombination in quantum confined quantum dot involving a negative trion (X^-) (left), a positive trion (X^+) (middle) and a biexciton (right) which can be considered as combination of a negative trion and a positive trion Auger recombination. Reprinted with permission from Ref. [76]. (c) Variation of Auger lifetime as a function of average number of excitons in 2.3 nm-sized CdSe colloidal quantum dots. (d) Size dependent Auger lifetime in CdSe quantum dots. Reprinted with permission from Ref. [21].

Fig. 7. (a) Size dependent Auger lifetimes of biexcitons (black open symbols), negative trions (orange solid symbols), and positive trions in CdSe quantum dots. Reprinted with permission from Ref. [76]. (b) Size dependence of biexciton Auger lifetimes for different kinds of colloidal quantum dots showing the universal volume scaling. Reprinted with permission from Ref.[76].

Fig. 8. Schematic illustration of spatial distribution of electron and hole wave function in type-I CdSe/ZnS, quasi-type-II CdSe/CdS, and type-II CdSe/ZnTe or ZnTe/CdSe core/shell quantum dots. Reprinted with permission from Ref. [86].

Fig. 9. (a) Asymmetric conduction and valence band offset in CdSe/CdS quantum dots, manifesting the localized hole in the core and the delocalized electron across the whole dot volume. (b) Schematic illustration of Auger recombination in CdSe/CdS quantum dots. Reprinted with permission from Ref. [83] (c) Calculated electron (black curves) and hole (red curves) wave function distribution in CdSe/CdS quantum dots with core radius ($R_0 = 1.5$ nm) and thin CdS shell ($H = 0.4$ nm, corresponding to 1 monolayer, dashed lines) and thick CdS shell ($H = 4.0$ nm, corresponding to 10 monolayers, solid lines). (d) Time-resolved PL intensity of reference

CdSe/ZnS core/shell nanocrystals (NCs). (e) Time-resolved PL intensity of the CdSe/CdS “giant” nanocrystals (g-NCs), revealing the rapid multi-exciton recombination due to Auger decay and the slow single exciton recombination. (f) PL intensity for CdSe/ZnS NCs with different average number of excited excitons, $\langle N_0 \rangle$, extracted at different time delays after excitation. (g) The same for CdSe/CdS g-NCs. (c-g) Reprinted with permission from Ref.[93].

Fig. 10. (a) Stimulated emission spectra from the CdSe/CdS g-NCs manifesting the broadband optical gain. (b) Emission intensity as a function of pump fluence for CdSe/CdS g-NCs and the reference CdSe/ZnS NCs. (c) Normalized transient absorption spectrum of CdSe/CdS g-NCs showing broadband optical gain. (a-c) Reprinted with permission from Ref.[93]. (d) Exciton interaction control for tuning the stimulated emission position. Reprinted with permission from Ref. [94].

Fig. 11. (a) Plot of the Auger recombination rate as a function of the confinement potential width. Reprinted with permission from Ref. [81]. (b) Time-resolved PL intensity of CdSe/CdS quantum dot with sharp core-shell interface. Reprinted with permission from Ref. [96]. (c) Time-resolved PL intensity of CdSe/CdS quantum dot with alloyed core-shell interface. Reprinted with permission from Ref. [96]. (d) Breakdown of universal volume scaling in Auger lifetime in CdSe/CdS g-NCs with alloyed core-shell interface. Reprinted with permission from Ref.[95].

Fig. 12. (a) Pump threshold of CdZnS/ZnS quantum dots with different core-shell interfaces. (b) Time-resolved PL intensity of CdZnS/ZnS quantum dots with alloyed interface, showing the increased Auger lifetime (inset). (c) Stimulated emission spectra of CdZnS/ZnS quantum dots with alloyed interface. (d) Lasing spectra from CdZnS/ZnS quantum dots. (a-d) Reprinted with permission from Ref. [19].

Fig. 13. (a) Comparison of Auger lifetime of nanorods with nearly spherical dots. Reprinted with permission from Ref. [74]. (b) Polarized lasing emission from CdSe/ZnS nanorods. Reprinted with permission from Ref. [104].

Fig. 14. (a) Representative TEM image of colloidal quantum wells. (b) Lateral size dependent quantum yield and stimulated emission spectra. (a-b) Reprinted with permission from Ref. [107]. (c) Colloidal quantum well based laser. (d) Lasing emission from colloidal quantum wells. (c-d) Reprinted with permission from Ref.[113].

Fig. 15. (a) Stimulated emission from CdSe/CdS dot-in-tetrapod nanostructure. (b) Stimulated emission from CdSe/CdS dot-in-rod nanostructure. (c) Comparison of pump thresholds of CdSe/CdS dot-in-tetrapod and dot-in-rod nanostructure. (a-c) Adopted with permission from Ref. [118]

Fig. 16. Lasers developed from colloidal semiconductor nanostructures with different kinds of configurations. (a) Liquid whispering-gallery-mode laser. (b) Silica microsphere solid-state laser. (c) Liquid droplet laser. (d) Microsphere laser with evanescent pumping. (e) Random laser (f) Distributed feedback laser. (g) Photonic crystal nanobeam laser. (h) Vertical cavity surface emitting laser. Reprinted with permission from Ref. [124], [25], [125], [126], [128], [131], [137], [135].

Fig. 17. (a) Simplified two-level system to show the balance between the absorption and stimulated emission. (b) Concept of single-exciton gain enabled by Stark effect. (c) Stimulated emission spectra from single-exciton gain and biexciton gain. (d) Observation of both single-exciton gain and biexciton gain from type-II quantum dots. (e) Electron and hole wave function distribution in CdS core. (f) Electron and hole wave function distribution in type-II CdS/ZnSe quantum dot. (a-f) Reprinted with permission from Ref. [141]. (g) Single exciton lasing from CdSe/ CdS quantum dots. Reprinted with permission from Ref. [126]. (h) Red, blue and green stimulated emission from CdSe/CdZnS quantum dots. Reprinted with permission from Ref.[135].

Fig. 18. (a) Setup of Z-scan technique for nonlinear absorption measurement. (b) Schematic illustration of linear and multiphoton pumped stimulated emission. (c) Stimulated emission by simultaneous three photon absorption. Reprinted with permission from Ref. [128]. (d) Transient PL of CdSe/CdS nanorods. Reprinted with permission from Ref. [150]. (e) Comparison of one-, two-, and three-photon pumped stimulated emission spectra from CdSe/CdS/ZnS core-multishell quantum dots. Reprinted with permission from Ref. [128].

Fig. 19. (a) Absorption and emission of hydrostatically strained quantum dots. (b) Absorption and emission of biaxially strained quantum dots. (c) Continuous wave lasing spectra from biaxially strained quantum dots. (d) Transient absorption spectra from hydrostatically strained quantum dots. (e) Transient absorption spectra from biaxially strained quantum dots. (f) Continuous wave lasing output beam from biaxially strained quantum dots. (a-f) Reprinted with permission from Ref. [159].

Fig. 20. (a) Stimulated emission spectra from CsPbBr₃ perovskite nanocrystals. (b) PL intensity and linewidth as a function of pump intensity. (c) Stimulated emission intensity as a function of pump pulse number. (a-c) Reprinted with permission from Ref. [167]. (d) Composition tuned stimulated emission. (e) Model gain coefficient of CsPbBr₃ perovskite nanocrystals. (d-e) Reprinted with permission from Ref. [168]. (f) Whispering-gallery-mode lasing from CsPbBr₃ perovskite nanocrystals. Reprinted with permission from Ref. [167]. (g) Microsphere lasing from CsPbBr₃ perovskite nanocrystals. Reprinted with permission from Ref. [168]. (h) Vertical cavity surface emitting lasing from CsPbBr₃ perovskite nanocrystals. Reprinted with permission from Ref. [174].

Fig. 21. (a) Schematic illustration of CdSe/Cd_xZn_{1-x}Se/ZnSe_{0.5}S_{0.5} quantum dots. (b) A simplified band diagram of the device structure. (c) The p-i-n architecture adopted for the electrically driven optical gain from colloidal quantum dots. (a-c) Reprinted with permission from Ref. [221].

Fig. 22. Timeline showing the major progress in the research field of lasers developed from colloidal semiconductor nanostructures. (a) First achievement of lasing from colloidal quantum dots in 2003. (b) Concept of single-exciton gain in 2007. (c) Theoretical prediction of Auger suppression by potential profile in 2009. (d) Breakdown of volume scaling in Auger lifetime in 2011. (e) Three-photon induced stimulated emission in 2014. (f) Stimulated emission from colloidal quantum well in 2014. (g) Optical gain from colloidal quantum dots by electrical pumping in 2017. (h) First achievement of stimulated emission from colloidal quantum dots in 2000. (i) Stimulated emission from nanorods in 2003. (j) Stimulated emission by two-photon absorption in 2008. (k) Stimulated emission from the heterostructured nanocrystals in 2009. (l) Continuous wave lasing from colloidal nanostructures in 2014. (m) Stimulated emission and lasing from perovskite nanocrystals in 2015. Reprinted with permission from Ref. [45], [141], [81], [95], [128], [109], [221], [23], [74], [147], [93], [224], [167].

Table 1. Representative pump threshold of stimulated emission (SE) from colloidal nanostructures

Materials	Shape of the nanostructure	SE Threshold	SE Wavelength	Pump Source	Temperature	Publication Date	Ref.
CdSe/ZnS	Nearly spherical dot	~8 mW	~540-610 nm		80 K	2000	[23]
CdSe/ZnS	Nearly spherical dot	~300 $\mu\text{J m}^{-2}$	~620 nm	400 nm, 100 fs	295K	2009	[93]
CdSe/CdS	“Giant” nanocrystal	~26 $\mu\text{J cm}^{-2}$	~620 nm	400 nm, 100 fs	295K	2009	[93]
CdSe/CdZnS	pyramid-like	~90 $\mu\text{J cm}^{-2}$	~625 nm	400 nm, 100 fs	295K	2012	[135]
CdSe	Nanorod	~0.08 mJ cm^{-2}	~686 nm		295K	2003	[74]
CdSe/CdS	Dot-in-rod	0.15–1.5 mJ cm^{-2}	~590-620 nm	400 nm, 100 fs	295K	2012	[119]
CdS/CdSe/CdS	nanoplatelets	~6 $\mu\text{J cm}^{-2}$	~650 nm	400 nm, 100 fs	295K	2013	[109]
CdZnS/ZnS	Nearly spherical dot	~50 $\mu\text{J cm}^{-2}$	~430-470 nm	400 nm, 100 fs	295K	2014	[19]
PbS	Near spherical dot	~1 mJ cm^{-2}	~1300 nm	800 nm, ~1.7 ps	295K	2005	[179]
CsPbBr₃	nanocube	~6 $\mu\text{J cm}^{-2}$	~520 nm	400 nm, 100 fs	295K	2015	[167, 168]
CdSe/CdS	Oblate	14 $\mu\text{J cm}^{-2}$	~635 nm	355 nm, 250 fs	295 K	2017	[159]
CdSe	nanoplatelet	6.5 W cm^{-2}	~525 nm	444 nm, CW laser	295K	2014	[224]