SPECIAL ISSUE: Optical Gain Materials towards Enhanced Light-Matter Interactions

Progress in semiconductor quantum dots-based continuous-wave laser

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ABSTRACT Continuous-wave (CW) operated laser is a key component for photonic chips. It has been highly desired to develop low-threshold, high-efficiency, long-term operational, low-cost and easily integratable CW laser. As excellent optical gain materials, quantum dots (QDs) have been intensively investigated for on-chip CW lasers owing to their high photoluminescence quantum yields, tunable emission wavelengths and easy integration. Base on the difference of preparation processes, QDs can be classified into epitaxial QDs and solution-processed colloidal QDs (CQDs). In this mini-review, we summarize the research progresses of epitaxial III–V semiconductor QD and solution-processed CQD-based CW lasers. The challenges associated with the realization of CQD CW lasers are discussed in detail. In particular, the emerging perovskite-based CW lasers are highlighted. Finally, a short perspective of QD-based CW lasers is presented.

Keywords: quantum dots, continuous-wave laser, perovskite, photonics

INTRODUCTION

In response to the explosive growth of global information, the high-speed, high-density and low-cost information processing technique is intensively needed [1]. Photonic chips hold great promise to be the next generation data processing technology due to their fast responsibility. As described by Paul [2], a photonic chip at least includes the components with functionalities of light generation, transportation, and detections. Over the past decades, great success has been made in the on-chip light sources, high-bandwidth detectors, fast modulators and waveguides [3–8]. In comparison, the development of on-chip light source lags behind the other counterparts, which has

become a great challenge for the field of integrated photonics.

Continuous-wave (CW) lasers have been considered a remarkable breakthrough to achieve on-chip integrated light source. The epitaxial growth of III–V semiconductor quantum wells (QWs) on silicon (Si) substrates has enabled CW laser for chip-to-chip optical communications. However, the performance is limited due to the lattice mismatch and the thermal expansion coefficient difference between III–V materials and Si substrates [3]. The III–V quantum dots (QDs) have ability to overcome these hindrances [9–14]. The performance of III–V QD CW laser has been greatly improved in the past few years, which approaches the requirements of on-chip light source in near infrared (NIR) region. Along with that, the demand of CW laser also promotes the research of colloidal QDs (CQDs) as economical and color-tunable alternatives for CW laser applications [15]. The gain thresholds of CQD-based CW lasers have been greatly reduced by means of materials engineering. However, the thresholds are still far from the practical application. Recently, the halide perovskites are emerging as alternative gain media to reduce the threshold to achieve CW lasers [16–23]. The primary results have encouraged the entire community to pursue perovskite-based room-temperature CW lasing.

In this mini-review, we summarize the recent developments of CW lasers based on epitaxial QDs and solution-processed CQDs. In the first part, we describe the progress of III–V semiconductor QD-based CW lasers on different substrates, including germanium (Ge), Ge-on-Si and Si substrates with a following discussion about micro-disk (MD) CW lasers. In the second part, we review

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the advancement in the field of solution-processed QCD-based CW lasers and discuss the current challenges for room-temperature-operated QCD CW lasers. In the third part, the recent perovskite-based CW lasers are presented and discussed. Finally, we give a short conclusion with our perspectives for the future investigation. Instead of a comprehensive description, we try to give the milestones and future trends of QD-based CW lasers.

III-V SEMICONDUCTOR QD CW LASERS

The room-temperature-operated semiconductor CW lasing was firstly achieved using AlAs-GaAs heterostructure QW by Alferov *et al.* [24] in 1971. Since then, CW lasers based on QWs have been extensively explored. However, their thresholds are temperature sensitive. To solve this problem, Arakawa *et al.* [25] theoretically proposed the use of QDs as gain media to decrease the temperature sensitive threshold in 1982. Twelve years later, Kirstaedter *et al.* [26] fabricated the first QD-based CW laser using the Stranski-Krastanow growth method. They observed room-temperature lasing from (InGa)As QDs with a threshold current density of 950 A cm^{-2} . From then on, the development of QD-based CW lasers has drawn a great number of attentions from both of scientific and industrial fields.

The zero-dimensional (0D) QDs can more easily

achieve population inversion compared with 2D or 3D materials. Semiconductor QDs represent particle-in-a-box-like quantum-confined structures, exhibiting atom- or molecule-like properties [27]. At quantum length scale, the electronic spectra of QDs show discrete density of states due to the quantum confinement effect, tending to delta-function-like energy levels. Similar to atoms, the ground state of QDs is doubly degenerated, which has a maximum occupancy of two electrons. Therefore, the 0D QDs have the lowest achievable gain threshold. Because of the high energy level spacing between the discrete energy levels, the carrier population is restrained to broaden into higher states [27]. Besides, the thermal spreading of carriers in QDs is decreased due to the delta-function like state density. These characters make the threshold of QD lasers be far less temperature sensitive than that of conventional QW lasers [25].

With the efforts from all over the world, the performance of III-V QD-based CW laser has been greatly improved. Table 1 summarizes the milestones in the developments. In the following, we will describe the details in the material innovations on the growth.

III-V semiconductor QD CW lasers on Ge or Ge-on-Si substrates

In 2000, Kazi *et al.* [28] fabricated the self-formed InGaAs

Table 1 Representative milestones of epitaxial III-V semiconductor QD-based CW lasers

Authors	Description	Wavelength (nm)	Thresholds (A cm^{-2})	Operation condition	$T_{\text{max}}^{\text{b}}$ ($^{\circ}\text{C}$)	Lifetime (h)	Publication year
Arakawa <i>et al.</i> [25]	Theoretically proposed						1982
Kazi [28]	$\text{In}_{0.2}\text{Ga}_{0.8}\text{As}$ 'QD like'	854	1320	RT ^a		78	2000
On Ge or Ge-on-Si substrates							
Liu <i>et al.</i> [29]	InAs/GaAs QDs On Ge	1305	55.2	RT	60		2011
Lee <i>et al.</i> [30]	InAs/GaAs QDs On Ge/Si	1280	163	RT	30		2012
Liu <i>et al.</i> [31]	InAs on Ge/Si	~1250	~426	RT	119		2014
Liu <i>et al.</i> [32]	InAs/GaAs on Ge/Si	1267	2000	RT	30	4600 (extrapolated)	2015
On Si substrates							
Chen <i>et al.</i> [3]	InAs/GaAs on Si	1315	62.5	RT	75	10,0158 (extrapolated)	2016
Liu <i>et al.</i> [9]	InAs/GaAs on GaP/Si	1280	860	RT	90		2017
Norman <i>et al.</i> [12]	InAs on V-groove Si	1250	333	RT	80		2017
Chen <i>et al.</i> [13]	InAs/GaAs on Si	1292	425	RT	36		2017
Jung <i>et al.</i> [10]	InAs on GaP/Si	1270	132	RT	80		2017
Wang <i>et al.</i> [33]	InAs/GaAs on Si	~1300	550	RT			2018

a, b) RT and T_{max} represent room-temperature and the maximum lasing temperature, respectively.

QD-like structures on Si substrate by the metalorganic chemical vapor deposition (MOCVD) method. The atomic force microscopy (AFM) characterizations show that the as-formed InGaAs QD-like nanocrystals have a diameter of about 20 nm with an average height of ~6 nm. The laser structure was composed of a QD-like active region between 70-nm-thick $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ layers. An electrically pumped CW laser at wavelength of ~860 nm with a threshold of 1.32 kA cm^{-2} was achieved. CW laser at optical communication wavelength has been of great interest. The breakthrough of III-V QDs at about 1300 nm was accomplished by Liu *et al.* [29] in 2011. They proposed the growth of single-domain GaAs buffer layers on the Ge substrate using Ga pre-layer technique. The AFM and transmission electron microscopy (TEM) images show that the obtained GaAs layer has a quite smooth surface morphology (Fig. 1a). With that, InAs/GaAs QDs can be fabricated directly on the pre-fabricated high-quality GaAs buffer layer. By using this method, the antiphase boundary (APB) issues were resolved, and the room-temperature-operated CW lasing at wavelength of 1305 nm with a low threshold of 55.2 A cm^{-2} was observed (Fig. 1b and c).

Soon after the demonstration of InAs/GaAs QD CW laser on Ge substrate, the room-temperature InAs/GaAs QD CW laser on Ge-on-Si substrates was reported by the same group. The threshold was about 163 A cm^{-2} [30]. Using a similar method, Liu *et al.* [31] fabricated p-doped InAs QDs epitaxially grown on Ge-on-Si substrate in 2014. High temperature CW lasing emission with a threshold of ~255 mA was obtained at a temperature up to 119°C. Furthermore, they studied the InAs/GaAs self-assembled QD lasers epitaxially grown on the Ge-on-Si substrates. The obtained CW laser was able to operate

over 2700 h at 30°C [32].

III-V semiconductor QD CW lasers on Si substrates

Although the QD CW lasers on Ge or Ge-on-Si substrates show excellent performances, the QD CW lasers directly grown on Si substrates without the intermediate Ge layer are more attractive because the Ge layer restricts the range of Si circuits and has large optical absorption coefficient at telecommunication wavelengths [3,33]. However, it is difficult to grow CW lasers directly on Si due to the lattice mismatch and the thermal expansion coefficient difference between III-V materials and Si substrates [3]. The threading dislocation (TD) in the gain layers forms a number of non-radiative recombination centers and cuts down the laser performance. QDs are less sensitive to defects. A breakthrough of QD CW lasers directly grown on Si was firstly accomplished by Chen *et al.* [3] in 2016. In their work, they developed several important strategies to realize highly efficient monolithic QD lasers. Initially, they used the phosphorus-doped Si (100) wafers with 4° offcut to the [011] plane to prevent the formation of antiphase domains when growing III-V materials on Si substrates. Then, they applied a thin AlAs layer to provide a good interface for III-V QDs growth. Furthermore, the $\text{In}_{0.18}\text{Ga}_{0.82}\text{As}/\text{GaAs}$ strained-layer superlattices were grown as dislocation filter layers on the GaAs buffer layer. Taking advantages of these efforts, they successfully fabricated room-temperature-operated QD CW lasers with a low threshold of 62.5 A cm^{-2} , high output power of > 105 mW, and long lifetime operation of 3100 h (Fig. 2a–c).

The standard Si-based complementary metal-oxide-semiconductor (CMOS) foundry process requires nominal (001) Si substrates. To adapt CMOS processing,

Figure 1 (a) AFM images (top) of the surface morphology for 1.2 nm GaAs on Ge substrates with As prelayer (left) and Ga prelayer (right) growth techniques. TEM images (bottom) of the interface between the GaAs buffer layer and the Ge substrate with As prelayer (left) and Ga prelayer (right) growth techniques. (b) Room-temperature emission spectra of the Ge-based InAs/GaAs QD laser for different drive currents below and above threshold. (c) Light output power *versus* current and voltage *versus* current for InAs/GaAs QD laser diodes grown on Ge and GaAs substrates. Reproduced with permission from [29]. Copyright 2011, Springer Nature.

Figure 2 (a) Schematic of the layer structure of an InAs/GaAs QD laser on a Si substrate. (b) Emission spectra for a $50\ \mu\text{m} \times 3200\ \mu\text{m}$ InAs/GaAs QD laser at various injection current densities under CW operation at 18°C . (c) Aging data for the InAs/GaAs QD laser on Si at a constant heatsink temperature of 26°C and CW drive current of 210 mA. Reproduced with permission from [3]. Copyright 2016, Springer Nature.

it is of great industrial importance to grow the III–V semiconductor CW lasers on on-axis (001) Si substrate [9]. In 2017, Liu *et al.* [9] reported the electrically pumped III–V InAs/GaAs QD CW lasers epitaxially grown on on-axis (001) Si without offcut or Ge layers. The CW laser was developed on an on-axis p-doped Si substrate with an n-doped Si homo-epitaxial buffer and n-doped GaP nucleation layer. A threshold around $860\ \text{A cm}^{-2}$ and an output power of 110 mW were achieved. Subsequently, Jung *et al.* [10] improved the GaAs templates with a TD density of $7.3 \times 10^6\ \text{cm}^{-2}$ and achieved CW lasing emission with lowest threshold current density of $132\ \text{A cm}^{-2}$. In 2018, the group further improved the performance and reliability of QD lasers by introducing a low TD density GaAs buffer layer. The CW laser with a threshold of 6.2 mA and an extrapolated mean-time-to-failure of more than one million hours was demonstrated [11].

Another alternative method to realize the CW lasers directly grown on Si is using patterned on-axis (001) Si substrate. The patterned on-axis (001) Si could circumvent the issue of antiphase domains and reduce the TD density in the GaAs buffer layer. In 2017, Norman *et al.* [12] demonstrated the electrically injected CW QD lasers grown on on-axis (001) Si patterned with (111) V-grooves. They realized the CW lasing with a threshold of 36 mA and operation temperature up to 80°C . The electrically pumped InAs/GaAs QD CW laser monolithically grown on on-axis Si (001) substrates without any buffer layers was also reported [13]. In this work, a 400 nm APB-free epitaxial GaAs film with a thickness of 400 nm was first deposited on a standard industry-compatible on-axis Si (001) substrate by MOCVD, and then the laser structure was grown on this APB-free GaAs/Si (001) substrate by molecular beam epitaxy [13]. The CW lasing emission with a threshold of $425\ \text{A cm}^{-2}$ was achieved.

III–V semiconductor QD MD CW laser on Si substrates

MD can provide whispering-gallery (WG) lasing modes with high quality. In 2016, Wan *et al.* [34] first demonstrated the optical pumped $1.3\ \mu\text{m}$ room-temperature QD MD CW lasers epitaxially grown on pre-patterned on-axis (001) Si substrate without any buffer layer. The MD CW laser consisted a five-layer InAs/InGaAs dot-in-a-well structure in a disk region. The tilted and top-down scanning electron microscopy (SEM) images of the disk reveal the smooth sidewall surface of the MD (Fig. 3a). The high quality MD guarantees the achievement of single mode laser with low threshold of $200\ \mu\text{W}$, as shown in Fig. 3b and c. In 2017, Kryzhanovskaya *et al.* [35] demonstrated an electrically pumped single-mode MD laser that was monolithically grown on Si (001) substrate with 4° offcut to the (011) plane. The laser structure, CW lasing emission spectra and threshold behavior are shown in Fig. 3d–f. By applying direct current injection, the minimum threshold current density of room-temperature-operated lasing was about $600\ \text{A cm}^{-2}$. Using V-grooved GaAs-on-Si template and V-groove (001) Si substrates, Wan *et al.* [36,37] further demonstrated the electrically pumped multi-mode QD MD lasing around $1.3\ \mu\text{m}$ with low power consumption and high temperature stability. These CW lasers can operate at high temperature up to 100°C .

COLLOIDAL NANOMATERIAL-BASED CW LASERS

Although III–V semiconductor QD CW lasers have been intensively studied and gained great progress, the fabrication techniques, e.g., molecular beam epitaxy, CVD and liquid phase epitaxy, are expensive and complex. Moreover, it is difficult to realize the fabrication on flexible substrates. This drawback hinders the integration with flexible and low-cost photonic device.

Solution-processed high-gain media have been pursued

Figure 3 (a) SEM images of a 4- μm fabricated MD with 90° tilted view and 70° tilted view. (b) Emission spectra and (c) integrated PL intensity of the dominant mode as a function of pump power. Reproduced with permission from [34]. Copyright 2016, The Optical Society (OSA). (d) Schematic of the layer sequence. The left inset shows a SEM image of the structure cross section; the right inset shows a bright-field TEM image of the QD active layers. (e) Room-temperature CW emission spectra, and (f) the threshold behavior. Reproduced with permission from [35]. Copyright 2017, The Optical Society (OSA).

for decades because of their easy synthesis and integration. CQDs are very competitive gain materials for solution-processed laser applications [15,38]. In contrast to the epitaxial III–V semiconductor QDs, CQDs have potential advantages of easy fabrication and integration. First, CQDs can be synthesized easily and inexpensively. Generally, CQDs are obtained through wet-chemical methods, e.g., hot-injection synthesis, one-pot non-injection organometallic synthesis, aqueous synthesis, and flow microreactor synthesis [39,40]. These methods do not rely on high-temperature, expensive vacuum-based processing and the expensive rigid substrates. Second, the high-quality CQDs have a high degree of monodispersity in solutions. They are compatible with almost all kinds of laser cavities. Third, the integration process of CQDs on Si or other platforms is swift and cost-effective. Using spin coating, Langmuir-Blodgett deposition or other non-vacuum techniques, we can transfer CQDs from a dispersion to a thin film easily [15]. Lastly, the absorption and the emission properties of CQDs can be tuned in the range from visible to infrared according to the composition and size control.

In comparison with the excellent performances of electrically pumped III–V QD-based CW lasers, solution-processed CQD CW lasers are still in their preliminary

stage currently. There is no report on electrically pumped CQD CW lasers. The optically pumped CQD CW laser is a significant step toward reducing the threshold for integrated photonics [39].

Challenges for CQD CW lasers

The main obstacles for CQD achieving CW lasing emission are the large Auger recombination, the heat effect, and the poor uniformity of QD films. The Auger recombination is the most important factor affecting QD lasing threshold. Because of the non-unity degeneracy of band-edge states, the optical gain normally requires excitation of more than one exciton in each QD. However, the multiple excitons undergo rapid, nonradiative Auger recombination, prohibiting the sustained population inversion [41]. Another important factor is the heat effect. The optical performance of the QDs is temperature-dependent [42]. The high temperature will reduce the radiative efficiency, and increase the nonradiative Auger recombination [43,44], following increasing the threshold of the amplified spontaneous emission (ASE) and lasing. The heat mainly comes out of two sources. One is the rapid light injection of the pump source, and the other is the quantum defect and nonradiative recombination of the QDs. The heat effect increases the nonradiative re-

combination, which in turn generates more heat. What's more, the QD films normally suffer from low thermal conductivity. It accelerates the accumulation of heat in the QD films. The third factor is the poor uniformity of QD films on the laser cavities [45,46]. The rough surface of QD films causes much light scattering. It will consume the excited photons and produce more heat, resulting in the increase of the lasing threshold.

To reduce the lasing threshold and achieve CW lasing in CQDs, we need to decrease the defects of QDs, so as to suppress Auger recombination and improve the quantum yield (QY) of QDs. The elongated nanocrystals and heteronanocrystals can suppress the Auger recombination rates [47]. Several studies have achieved the quasi-CW and CW lasing in CdSe-based core/shell CQDs and 2D nanoplatelets [42,45,47].

CdSe and HgTe CQD-based quasi-CW or CW lasers

Table 2 shows the major findings and key parameters of CdSe and HgTe CQD-based lasers. The investigation of CQDs for lasing application originates from Klimov's work in 2000 [48]. They proposed that the optical gain in CQDs did not rely on single-exciton emission, but on biexciton or mutiexciton emission. To illustrate this point, they modeled the band-edge emission in QDs using a two-level system with twofold spin-degenerate states and showed that the population inversion was due to two electron-hole pair states. The fast Auger recombination

was the major obstacle preventing optical gain in CQDs. Klimov *et al.* [48] partially resolved this problem by employing close-packed QD films as the gain media and using femtosecond pulsed laser as the excitation source. The two experimental designs guaranteed the first observation of ASE in CQDs. After this work, the use of CQDs as gain medium for lasing has been widely studied. For example, the CQD lasers based on WG, distributed feedback (DFB) and random cavity were demonstrated [49–52].

Based on the optical gain models, the population inversion in CQDs can only be achieved if the average number of excitons is greater than one [48]. In 2004, Ivanov *et al.* [53] further developed a model to achieve optical gain with single exciton, and they successfully demonstrated the single exciton ASE from specially designed type-II CdS/ZnSe CQD later [54]. The type-II CdS/ZnSe CQDs can spatially separate electrons and holes between the core and the shell, inducing a positive biexciton binding energy and shifting the absorption spectrum with respect to the luminescence line of CQDs. Five years later, Dang *et al.* [55] demonstrated that the single exciton gain could also be realized in pyramidal shaped type-I CdSe/ZnCdS core/shell CQDs.

It took more than fifteen years to achieve the optical pumped CQD-based CW laser. In 2013, using the type-I CQDs with CdSe core and thin ternary $\text{Zn}_{0.5}\text{Cd}_{0.5}\text{S}$ shell as gain media, Dang *et al.* [56] first demonstrated the single

Table 2 Representative milestones of colloidal CdSe and HgTe-based quasi-CW or CW lasers

Authors	Materials	Type	Color	Thresholds	Operation condition	Publication year
Pulse lasers						
Klimov <i>et al.</i> [48]	CdSe QDs	ASE	Red	~8 mW	80 K	2000
Eisler <i>et al.</i> [50]	CdSe, CdSe(ZnS)QDs	Laser	Red	~0.9 μJ (80 K)	RT and 80 K	2002
Klimov <i>et al.</i> [54]	CdS/ZnSe QDs	ASE	Red	2 mJ cm^{-2}	RT	2007
Dang <i>et al.</i> [55]	CdSe/ZnCdS QDs	Laser	Red	~60 $\mu\text{J cm}^{-2}$	RT	2012
Quasi-CW lasers						
Dang <i>et al.</i> [56]	CdSe/ $\text{Zn}_{0.5}\text{Cd}_{0.5}\text{S}$	Laser	Red	250 $\mu\text{J cm}^{-2}$		2013
Wang <i>et al.</i> [57]	CdZnS/ZnS	Laser	Blue	30.2 μJ per pulse	RT	2015
Adachi <i>et al.</i> [42]	CdSe/CdS/ZnS QDs	Laser	Red	52 kW cm^{-2}	RT	2015
CW lasers						
Fan <i>et al.</i> [47]	CdSe/CdS CQDs	Laser	Red	6.4–8.4 kW cm^{-2}	253 K	2017
Yang <i>et al.</i> [45]	CdSe/CdS nanoplatelets	Laser	Red	220 W cm^{-2}	RT	2017
Wu <i>et al.</i> [60]	Charged CdSe/CdSe _x S _{1-x} /CdS QDs	ASE	Red	500 W cm^{-2} (calculated value)	RT	2017
Geiregat <i>et al.</i> [61]	HgTe QDs	ASE	Infrared	40 mW cm^{-2}	RT	2017

mode lasers operated in quasi-CW regime. The laser was pumped by a solid-state laser with a pulse duration of 270 ps. It was 2.25 times longer than the multiexciton Auger recombination lifetime of the CdSe/Zn_{0.5}Cd_{0.5}S QDs, thus entering a quasi-steady state regime. The threshold of single mode lasing emission was about 250 $\mu\text{J cm}^{-2}$. This achievement was mostly attributed to the high-quality densely packed CQD films. Later on, Sun's group from Nanyang Technological University [57] demonstrated quasi-CW blue liquid lasers from the solution of CdZnS/ZnS alloyed core/shell QDs. The pulse width of the pump laser used in this work was 5 ns, which was one order of magnitude longer than the biexcitonic Auger recombination time constant (~ 350 ps) of the CdZnS/ZnS QDs.

In 2015, Sargent's group [42] realized that the main cause of the nanosecond-only operation of CQD lasers was thermal runaway. This can be attributed to the combination of rapid heat injection from the pump laser, poor heat removal and the temperature-dependent threshold. Using the ultra-compact CdSe/CdS/ZnS core/shell/shell CQD film as gain media and the thermally conductive MgF₂ material as substrate, they achieved microsecond-sustained CQD laser with a threshold of 52 kW cm^{-2} . To further decrease the band-edge degeneracy and the single-dot photoluminescence (PL) linewidth of QDs, they created asymmetric CdSe/CdS core/shell CQDs [47]. According to their method, CdS shell was grown on the (0001) facet of CdSe core, producing an asymmetric compressive shell in an oblate shape that created built-in biaxial strain, while still maintaining good surface passivation. The excitonic fine structure in biaxially strained dots could be spread uniformly and the thermal depopulation of the valence-band-edge was prevented. By incorporating the asymmetric biaxially strained CQDs film into a photonic crystal DFB cavity, they realized the CW lasing emission with a threshold of about 6.4–8.4 kW cm^{-2} with linewidth of about 3 meV. High-quality MD provides strong carrier confinement and high-quality-factor WG modes. Chien *et al.* [58] introduced a SiO₂ sandwiched colloidal CdSe/ZnS QD MD laser structure. The sandwich structure not only increased the confinement factor but also reduced the photo-oxidation of QDs under intense optical extraction. Based on this laser structure, they realized CW lasing with a threshold of 500 kW cm^{-2} and narrow linewidth of 0.18 nm.

The 2D colloidal QWs (CQWs) hold several properties to achieve low threshold lasing. Firstly, the elongated nanocrystals can suppress Auger recombination due to

the energy levels quantized in only one direction. It imposes a strict momentum conservation rule for Auger recombination. Secondly, the CQWs have larger spontaneous and stimulated emission cross sections. Thirdly, the loss due to sample heterogeneity can be largely avoided. In 2014, Grim *et al.* [41] fabricated 2D CdSe CQWs and showed that these CQWs possessed large exciton and biexciton binding energies, giving rise to room-temperature-operated CW laser. The threshold of the lasing was about 440 W cm^{-2} . Using colloidal CdSe/CdS core/shell nanoplatelets as gain media, Yang *et al.* [45] demonstrated a room-temperature-operated CW nanolaser with a threshold input power of less than 1 μW .

As of now, the CQD CW lasers are still at the laboratory stage. One of the most important challenges is the difficulty to realize the electrical pumped lasing. A key obstacle to realize that is the large Auger decay. The thick-shell with a continuously graded shell of QDs can suppress the Auger decay and preserve the strong confinement effect. Lim *et al.* [59] fabricated these thick-shell, type-I CdSe/Cd_xZn_{1-x}Se QDs (Fig. 4a). In these nanocrystals, the confinement potentials are 'smoothened' for both electrons and holes, thus suppressing the hole- and electron-related Auger pathways. By applying a current-focusing device (Fig. 4b), which allows one to obtain a high current density up to $\sim 18 \text{ A cm}^{-2}$ without damaging the QDs and the injection layers, they obtained direct-current electrical pumped optical gain with a threshold of 3–4 A cm^{-2} (Fig. 4c).

To achieve population inversion, multi-excitons are normally required because of the non-unity degeneracy of the lowest-energy conduction band and valence band levels of QDs. It indicates that an intense pump pulse or high-power CW excitation is necessary to achieve lasing threshold. In 2017, Wu *et al.* [60] proposed a charged CQD concept to overcome this fundamental problem. By employing negatively charged QDs, the optical gain threshold can be reduced to sub-single-exciton or even zero-threshold level. Fig. 5a depicts the concept of zero-threshold optical gain, assuming the twofold degeneracy of the band-edge levels [60]. In neutral QDs, the single exciton state is optically transparent because the stimulated emission is exactly compensated by electron absorption. When the dot is occupied by an electron permanently, the ground-state absorption is reduced. A single-exciton excitation can produce optical gain. When a QD is doped with two or more electrons, the ground-state absorption can be completely bleached. 'Zero-threshold' gain can be achieved. Based on this idea, they realized the optical gain at extremely low excitation levels

Figure 4 (a) Continuously graded core/shell CdSe/Cd,Zn_{1-x}Se/ZnSe_{0.5}S_{0.5} QDs (cg-QDs). (b) The light-emitting diode design used in this work features a tapered hole-injecting layer with a small-area contact (70–100-μm wide) to the cg-QD layer which allows boosting of the current density up to 18 A cm⁻². A photograph of a real-life 8-pixel device under applied bias is shown as an inset. TCTA: tris(4-carbazoyl-9-ylphenyl)amine; ITO: indium tin oxide. (c) A color plot of electroluminescence (EL) spectra as a function of current density. At low currents, the device shows usual single-band EL due to the band-edge 1S transition (orange states in the left inset). When the current density exceeds ~1 A cm⁻², the second higher-energy band (~2.15 eV) due to 1P transition (green states in the right inset) emerges in the EL spectra (green shading). Reproduced with permission from [59]. Copyright 2017, Springer Nature.

(Fig. 5b and c).

Apart from the visible region, the CQD CW laser at the NIR telecom window is also very attractive because it can be applied in Si photonics directly. Geiregat *et al.* [61] first reported the infrared CW ASE with ultralow threshold of 40 mW cm⁻² from colloidal HgTe QDs. The average excitation density $\langle N \rangle$ of the threshold value was as low as 0.005 per QD. The low gain threshold was attributed to the stimulated emission between the lowest conduction-band and an empty state in the bandgap of the HgTe QDs. Their results propose an approach for low threshold QD CW lasers based on intentional trap states.

Perovskite-based quasi-CW or CW lasers

During the last few years, colloidal metal halide perovskite semiconductors have become a hot research field in light sources owing to their superior properties such as easy fabrication process, tunable PL wavelength and narrow band emission. Metal halide perovskites have general chemical formula of ABX₃, A₂BX₅ or A₄BX₆, where A is Cs, CH₃NH₃ (MA), NH₂-CH=NH₂ (FA), etc., B is Pb, Sn, Ge, etc., and X is Cl, Br, I [17,18,62,63]. 2D Ruddlesden-Popper perovskite QWs with a chemical form of L₂A_{n-1}B_nX_{3n+1} can also be formed. Here L is large long organic chains that act as spacer between BX₆⁴⁻ octahedral layers and *n* is the number of these layers. Perovskite nanomaterials are a class of excellent robust gain materials with high absorption coefficient and low density of defects [18,64–67]. They renew hope for a solution-processed CW lasing and set up a worldwide race to this goal.

Table 3 shows the major findings and key parameters of

perovskite-based lasers, especially the quasi-CW or CW lasers. In 2014, Sum's group from Nanyang Technological University [68] demonstrated optical gain from MAPbX₃ perovskite thin films for the first time. The wide wavelength tunable ASE with low threshold was observed at low temperature. Later on, using distributed Bragg reflector as Fabry-Pérot cavities, Deschler *et al.* [69] demonstrated the optically pumped room-temperature-operated laser from solution-processed mixed halide perovskite semiconductors. From then on, the tunable ASE/lasing emission covering the entire visible range and NIR in perovskite nanowires (NWs), QDs or QWs are rapidly developed [16–18,39,65–67,70–97].

In 2015, Cadelano *et al.* [43] systematically discussed the conditions needed for realizing the true CW lasing form perovskite polycrystalline films. In this work, they first measured the ASE threshold of MAPbI₃ and MAPbBr₃ films as a function of temperature. They found that the threshold density was quadratically decreased with decrease of temperature, which was due to the temperature dependence of the radiative recombination rate and loss rate. They explored the ASE emission by varying the duration of the pump pulse from 4 to 300 ns, and indicated that the runaway heating limited the long-duration of ASE in perovskite films. The feasible improvements in heat management would allow the ASE to be sustained continuously at room temperature. Later, Jia *et al.* [98] proposed that the lasing duration of perovskite polycrystalline films were not just limited by thermal runaway. The lasing ceased mainly due to the photo-induced structural change in the perovskite. In the report, they fabricated high thermal conductivity Au/Si gratings.

Figure 5 (a) Concept of zero-threshold optical gain. In a neutral QD with twofold spin-degenerate band-edge states. The gain threshold corresponds to $\langle N \rangle = 1.15$. In QDs with one extra electron, the gain threshold is reduced to $\langle N \rangle = 0.69$. In QDs doped with two extra electrons, the ground-state absorption is completely bleached, implying that optical gain is threshold-less. Filled and open blue circles are electrons and holes, respectively. Black and red arrows correspond to absorption and stimulated emission, respectively. Red wavy arrows are incoming and outgoing photons. (b) Pump-fluence-dependent PL spectra of the QD film before and after photochemical charging ($\langle n_e \rangle = 1.1$) with $\langle N \rangle = 0.67, 1.02, 1.36, 1.69$ and 2.04 . (c) Modelling of time-integrated photon densities (Φ) in the cavity as a function of $\langle N \rangle$ for neutral, singly charged and doubly charged QDs. Reproduced with permission from [60]. Copyright 2017, Springer Nature.

Optically pumping the device with an InGaN diode laser at 160 K, they achieved the quasi-CW lasing emission with a threshold of 5 kW cm^{-2} and duration of $\sim 25 \text{ ns}$.

The first demonstration of CW lasing from perovskite was made in NWs, where lasing was attributed to polariton lasers. Polariton laser is one type of laser source whose coherent light emission is from the Bose-Einstein

condensation of exciton-polaritons in semiconductors [99,100]. A polariton laser can emit coherent light at ultra-low pump power below the threshold for population inversion. In 2017, Evans *et al.* [101] reported the low temperature (77 K) operated CW polariton laser from CsPbBr_3 perovskite NWs at excitation density of 6 kW cm^{-2} . According to analysis of the cavity modes and

Table 3 Representative milestones of perovskite-based quasi-CW or CW lasers

Authors	Materials	Type	Color	Thresholds	Operation condition	Publication year
Pulse lasers						
Xing <i>et al.</i> [68]	CH ₃ NH ₃ PbX ₃ ^a film	ASE	Visible	~10 μJ cm ⁻²	RT	2014
Deschler <i>et al.</i> [69]	CH ₃ NH ₃ PbI _{3-x} Cl _x film	Laser	Red	~0.2 μJ per pulse	RT	2014
Xing <i>et al.</i> [67]	CH ₃ NH ₃ PbX ₃ NWs	Laser	Visible, near-infrared	11 μJ cm ⁻²	RT	2015
Yakunin <i>et al.</i> [65]	CsPbX ₃ QDs	ASE/Laser	Visible	~5 μJ cm ⁻²	RT	2015
Quasi-CW lasers						
Cadelano <i>et al.</i> [43]	MAPbX ₃ film	ASE	Green, red		220 K	2015
Jia <i>et al.</i> [98]	MAPbI ₃ film	Laser	Red	5 kW cm ⁻²	160 K	2016
CW lasers						
Evans <i>et al.</i> [101]	CsPbBr ₃ NWs	Laser	Green	6 kW cm ⁻²	77 K	2017
Jia <i>et al.</i> [103]	MAPbI ₃ film	Laser	Red	~17 kW cm ⁻²	102 K	2017
Li <i>et al.</i> [104]	MAPbI ₃ film	Laser	Red	13 W cm ⁻²	RT	2018
Brenner <i>et al.</i> [105]	Cs _{0.1} (MA _{0.17} FA _{0.83}) _{0.9} Pb _{0.84} (I _{0.84} Br _{0.16}) _{2.68} film	ASE	Red	387 W cm ⁻²	80 K	2019
Wang <i>et al.</i> [106]	MAPbX ₃ QDs	Laser	Green, blue, red	Green: 15 W cm ⁻² Red: 24 W cm ⁻² Blue: 58 W cm ⁻²	RT	2019

a) X represents Cl, Br, I.

their temperature dependence, they revealed that the CW lasing originated from polariton modes near the bottleneck region on the lower polariton branch. This work indicates that perovskite NWs are promising materials for polariton lasers. Later on, Jiang *et al.* [102] further reduced the linewidth of CsPbX₃ NW CW nanolasers to 0.09 nm in low temperature condition.

The first report on the real photon CW lasers was from Giebink's group [103]. They fabricated a DFB laser structure by depositing a ~120-nm-thick MAPbI₃ polycrystalline film on the alumina grating that lied on a high-thermal-conductivity sapphire substrate. The lasing structure, lasing emission and lasing threshold behavior are shown in Fig. 6a–c. At temperatures below its tetragonal-to-orthorhombic phase transition, obviously polarized lasing emission with a threshold of ~17 kW cm⁻² was observed. The continuous gain originates from tetragonal-phase inclusions that are photogenerated by the pump within the bulk orthorhombic host matrix. Li *et al.* [104] directly patterned DFB gratings on MAPbI₃ polycrystalline films using thermal nanoimprint lithography and raised the operation temperature of the CW laser to room-temperature. Fig. 6d shows the schematic illustration of the nanoimprint process of the imprinted MAPbI₃ DFB resonator. The direct patterning of perovskite not only provided a high-quality-factor cavity with large

mode-gain overlap but also improved emission characteristics of the perovskite films. The ultra-low lasing threshold of 13 W cm⁻² was achieved, just as shown in Fig. 6e and f. Brenner *et al.* [105] employed a phase-stable perovskite film by spin-casting a precursor solution containing triple cation system with a non-stoichiometric ratio of Cs_{0.1}(MA_{0.17}FA_{0.83})_{0.9}Pb_{0.84}(I_{0.84}Br_{0.16})_{2.68} and demonstrated CW ASE at temperatures from 80 to 120 K. They also studied the temperature-dependent ASE properties and showed that the material degradation prohibited the ASE at high temperatures.

All the perovskite CW lasers mentioned above are based on perovskite NWs or polycrystalline films. These materials still need to be rationally designed toward easy integration and large-scale fabrication. Perovskite QDs (PQDs) have better optical performance compared with the NWs and polycrystalline films [20,64]. Moreover, the PQDs can be *in situ* fabricated in polymeric matrix, which provides highly luminescent composite films with advantageous features of easy fabrication and integration. Very recently, by carefully choosing polyacrylonitrile (PAN) as polymeric matrix, our group obtained *in situ* fabricated PQD-embedded PAN films and realized ultralow-threshold and color-tunable PQD-based CW lasers operated at room-temperature for the first time [106]. The PQD-embedded PAN films had outstanding

Figure 6 (a) Cross-sectional SEM of the second-order DFB laser architecture consisting of a MAPbI₃ film deposited on an 80-nm-thick etched alumina grating on a sapphire substrate. (b) The emission spectra and (c) input–output characteristics of the laser under different continuous excitation powers at 102 K. The emission spectrum above threshold is strongly s-polarized. Reproduced with permission from [103]. Copyright 2017, Springer Nature. (d) A schematic illustration of the nanoimprint process and perspective view SEM image of the imprinted MAPbI₃ DFB resonator. (e) CW lasing characteristics at room-temperature and (f) lasing threshold behavior. Reproduced with permission from [104]. Copyright 2018, American Chemical Society.

optical and thermal properties. They had high transmittance ($\sim 90\%$), near-unity PLQYs (up to 97%), very smooth surface (root-mean-square roughness: 0.784 nm), and high thermal conductivity ($\sim 1.2 \text{ W m}^{-1} \text{ K}^{-1}$) (Fig. 7a–c). Moreover, the PQDs show unique features of charge spatial separation effects due to the interactions between the *in situ* fabricated PQDs and the PAN matrix. Combining those films with DFB cavities, we obtained green, blue and red room-temperature-operated CW lasers with thresholds as low as 15, 58, and 24 W cm^{-2} , respectively (Fig. 7d–f). This progress opens up a door to achieve perovskite CW lasers for photonic application.

Although perovskite-based lasers have made great progress and several groups have achieved CW laser, it remains an open question whether perovskites are excellent gain materials to achieve room-temperature-operated CW lasing. A recent comment by Brenner *et al.* [107] expressed their reservations about the validity of room-temperature CW laser.

CONCLUSION AND OUTLOOK

In this work, we review the recent developments of QD-based CW lasers for photonic applications. The QDs can be classified into two categories: the epitaxial QDs and the solution-processed CQDs. We start with the summary of

epitaxial III–V semiconductor QD-based CW lasers on Ge, Ge-on-Si and Si substrates, and then discuss the difficulties and challenges associated with the realization of room-temperature-operated CQD CW lasers. In the last part, we also review the progress toward perovskite nanocrystal-based CW lasers.

To meet the commercial on-chip application, CW lasers with high performance, easy integration, and low cost are expected. Epitaxial III–V QD-based CW lasers, to some extent, are a promising candidate as on-chip light source. They are compatible with the semiconductor fabrication infrastructure and can realize electrical pump easily. Owing to the huge effort, the electrically pumped epitaxial QD CW lasers on Si substrates have achieved great advance. The CW lasing emission with low threshold of 62.5 A cm^{-2} , high output power of $>105 \text{ mW}$, long lifetime operation of 3100 h and high operation temperature to 100°C were achieved. However, a few roadblocks remain unresolved. One big challenge is the reduction of the defect density caused by the dissimilarity between III–V materials and Si substrates. Deep understanding about the growing process is still necessary to achieve precise control over the interface.

CQDs have good solution-processability and easy integration properties. The CQD CW lasers can provide a

Figure 7 (a) Transmittance and PL spectra of the *in situ* fabricated MAPbBr₃ PQD-embedded PAN film. The inset shows an optical image of the film under daylight and ultraviolet light. (b) 5 μm × 5 μm AFM image of the 1.9-μm-thick film spin-coated on quartz glass. The root-mean-square roughness of the film is 0.784 nm. (c) Thermal conductivities of the film at different temperatures. (d) Schematic setup of the room-temperature CW lasing measurement. The inset shows an SEM image of the grating. (e) Emission spectra of the film on the DFB cavity. (f) Lasing emission spectra of the blue and red PQD-embedded PAN films on DFB cavities. Reproduced with permission from [106]. Copyright 2019, American Chemical Society.

promising “hybrid approach” that combines the CQD lasers directly with Si substrates. Besides, CQDs have advantages of tunable emission wavelengths and low fabrication cost, making them appealing for on-chip applications. However, compared with the advance of epitaxial QD CW lasers, the solution-processed CQD CW lasers are still in an initial stage, although mill-stone-breakthroughs were made recently.

The biggest barrier for CQD CW lasers is the high threshold. Up to date, the electrically pumped CQD CW lasing has not been reported. It is because the lasing threshold is too high and CQDs cannot withstand high current injection. Sustaining large optical gain under strong pumping power is another challenge. High output power and long lifetime are the requirements for commercial laser products. That means the CQDs must possess the capability of high-power and long-duration pumping. However, the optical gain of CQDs is extremely sensitive to the pump power and heat due to the temperature-dependent properties of Auger recombination [44]. The next challenge is the fabrication quality of CQD films. Currently, the CQDs in solution play a dominant role in photonic applications. Transferring CQDs from a

dispersion to a high-quality film is not quite easy.

To overcome these barriers, the CQDs need more in-depth research from fundamental chemical physics and preparation technology. Achieving optical gain down to sub-exciton, suppressing Auger recombination at high pump power, improving stability at high temperatures and high current injection require huge efforts.

Perovskites would probably make a breakthrough in the field of electrically pumped CQD lasers. Perovskite materials have excellent gain properties, such as high absorption coefficient and low density of defects [18]. A high absorption coefficient guarantees a large optical gain. It is reported that the net optical gain of MAPbI₃ perovskite thin film is about 3200 cm⁻¹, which is higher than that of traditional CQDs [108]. The optical loss, including phonon scattering and edge scattering, will consume the photons and increase the laser threshold. The low density of defects in perovskite is helpful to reduce the optical loss. Miao *et al.* [109] have proved that the perovskite-based light-emitting diodes are able to keep high external quantum efficiency at a high current density of 300 mA cm⁻². This fact demonstrates that the perovskite materials have the ability to withstand high

current densities. In addition, *in situ* fabricated PQDs provide an effective route to achieve high-quality QD film without using solution process. Very recently, the PQDs-embedded glasses were fabricated by applying laser irradiation [110].

Although CQD-based CW lasers are in an initial stage, they show a remarkable potential to integrate into on-chip photonics. With the understanding of the physics of gain building and annihilating, there are plenty of rooms to realize the required CW lasers. Maybe the answer is perovskites.

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半导体量子点连续泵浦激光研究进展

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摘要 连续激光器是光子芯片的关键部件。低阈值、高效率、长寿命、低成本、易集成的连续激光器一直是科学界和工业界追求的目标之一。量子点具有荧光量子产率高、发光波长可调、易于集成等优点,是一种优异的激光增益材料。根据制备工艺的不同,量子点可分为外延生长的量子点和溶液制备的胶体量子点。本文综述了使用外延生长方法制备的III-V半导体量子点和胶体量子点连续激光器的研究进展,详细讨论了胶体量子点实现连续激光所面临的困难和挑战,总结了钙钛矿连续波激光器研究的进展。最后,讨论了量子点连续激光器实现光子芯片集成应用所面临的挑战。