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All-inorganic metal halide perovskite nanostructures from photophysics to light-emitting applications

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Abstract: All-inorganic metal halide perovskite nanostructures (IHPNs) have sparked intense research interest by virtue of the superior optical properties and the cost-effective synthesis. Since the first report in 2015, striking development of IHPNs has been witnessed in terms of fabrication strategy, optical characterization, and optoelectronic applications. In this review article, we begin with a brief survey of the mainstream fabrication routes. The emphasis will be put on the understanding of the photophysics of IHPNs including the electronic structures, steady-state and transient absorption and emission, nonlinear optical properties and representative exotic photophysical phenomena. Afterwards, we summarize the light-emitting applications of IHPNs in lasers and light-emitting diodes. Finally, the challenges and a perspective of the future development of IHPNs will be presented.

1. Introduction

The past few years have witnessed the impressive performance of organic-inorganic halide perovskites (OIHPs) in photovoltaics since their first being used as the sensitizer for dye-sensitized solar cell in 2009.^[1, 2] Besides the low-cost solution-processibility, the primary advantages of these OIHPs for photovoltaics lie in the high absorption coefficients ($>1 \times 10^4 \text{ cm}^{-1}$), long balanced diffusion lengths ($>100 \text{ nm}$), small effective masses for electrons and holes ($0.069\text{--}0.25m_0$), and relatively low carrier trap density.^[3, 4] Until 2014, the OIHPs were discovered to be also promising cost-effective light-emitting materials with wide emission color tunability,^[5-11] wherein Tan *et al.* reported the bright light-emitting diodes (LEDs) made from the OIHPs and Xing *et al.* demonstrated low threshold optical gain from the OIHPs.^[5, 6] However, the later findings pointed out that these bulk OIHPs may not be ideal candidates for LED and laser applications. Firstly, it is unraveled that the light emission from the OIHPs at room temperature primarily originates from the free carrier recombination due to the relatively low exciton binding energy. The bimolecular radiative recombination in the OIHPs is very slow such that the nonradiative decay arising from the defect-state trapping dominates the carrier recombination, which results in the typically low photoluminescence quantum yield (PLQY) ($< 30\%$), especially under low carrier injection densities ($< \sim 10^{15} \text{ cm}^{-3}$) comparable to that under LED working conditions.^[5, 6, 12-15] Hence, the spontaneous free carrier generation following light absorption in the OIHPs, which explains the high photoconversion efficiency, becomes the obstacle of improving their light-emission performance.^[5] Secondly, these OIHPs are extremely sensitive to the moisture, oxygen and heat, which was found to be closely related to the existence of organic component, thereby incurring constraint of a critical environment for fabrication, storage and device operation, commonly notorious in organic photonic materials.

Fortunately, the above limitations of OIHPs can be simultaneously overcome by exploiting the all-inorganic metal halide perovskite nanostructures (IHPNs), a closely related family of materials. The all-inorganic constituents with intrinsically higher melting and decomposition points allow for the improved photo- and thermo-stability as well as higher resistance to the atmosphere, and the enhanced exciton binding energies in the perovskite nanostructures ensure the stable excitonic emission at room temperature. In early 2015, Protesescu *et al.* reported the pioneered work on the all-inorganic cesium lead halide (CsPbX_3 , X = Cl, Br, I) light emitting nanocrystals.^[16] Immediately after their report, there has been an explosion of research activities on IHPNs motivated by their superior optical properties and the low cost of synthetic methods.^[16-18] Compared to the canonical CdSe quantum dots, which have developed for more than two decades,^[19] the emerging all-inorganic CsPbBr_3 perovskite nanocrystals exhibit even better emission characteristics including the higher PLQY (PLQY of 100% has been recently achieved^[20]) and narrower emission line-width (<20 nm).^[16, 18, 21, 22] Moreover, while the high temperature (280 °C) and protective atmosphere are required for synthesis of CdSe based quantum dots, the fabrication of CsPbX_3 nanocrystals can be performed at room temperature and free from inert environment.^[18] Leveraging on the halide composition control, the emission wavelength of CsPbX_3 nanocrystals can be finely tuned across the whole visible spectral range, enabling wide color gamut.^[16] Especially, the facile fabrication of high quality blue-green (410–530 nm) emitting CsPbX_3 nanocrystals makes them more competitive against the traditional metal-chalcogenide nanocrystals which typically suffer from photodegradation and relatively poor PLQY in the visible short wavelength range.^[16] Naturally, the superior optical properties of CsPbX_3 nanocrystals render them highly appealing for a plethora of optoelectronic applications including the high efficiency LEDs,^[18, 23-26] high color purity displays,^[18, 27] low-threshold optical amplifier and

lasers.^[28-32] In addition to the device engineering, the photophysics of CsPbX₃ nanocrystals have been investigated by both steady-state and transient optical spectroscopy, which helps in understanding their fundamental properties and performance optimization. It can be envisaged that these CsPbX₃ nanocrystals are emerging as a new research paradigm and would play a significant role in the modernization of nanoscience and nanotechnology in the near future.

With the rapid development of these novel materials, we timely present an up-to-date review on the development of the IHPNs. In the first section, the mainstream fabrication routes of the perovskite nanocrystals are briefly introduced. Thereafter, we will emphatically discuss the photophysics of the IHPNs focusing on the electronic structures, steady-state and transient absorption and emission, nonlinear optical properties and several representative exotic photophysical phenomena, where we will put forward the plausible mechanisms for the superior optical performance of the perovskite nanocrystals. The third section covers the light-emitting applications of the IHPNs in lasers and LEDs. Finally, the challenges and a perspective of the future development of the IHPNs will be presented.

2. Synthesis of CsPbX₃ nanostructures

The investigation of bulk metal halide perovskites can be traced back to 1950's when Möller reported photoconductivity in the CsPbX₃ crystals.^[33] However, it failed to attract broad attention due to the poor optical properties and unhandy fabrication methods.^[33] Until the year of 2015, Protesescu *et al.* reported the facile synthesis of the high-quality colloidal nanocrystals of cesium lead halide perovskites (CsPbX₃, X = Cl, Br, and I) by the hot injection method (**Figure 1a**),^[16] the CsPbX₃ research then exploded in a variety of directions. **Figure 2a** shows schematic illustration of the hot injection approach for CsPbX₃ nanocrystals. As a typical recipe, the powder of lead halide salt (PbX₂) is dissolved in the hot solvent (octadecene (ODE)) with the surfactants

of oleylamine (OAm) and oleic acid (OA). And then, the nanocrystals are formed subsequently by injecting the cesium precursor into the hot solvent according to the chemical reaction equation:

The reaction temperature in combination with reaction time and precursor concentration can be manipulated to obtain nanocrystals with varied sizes. The insets of Figure 2a present the representative photograph of the as-grown CsPbBr₃ nanocrystals and the corresponding transmission electron microscope (TEM) image, which indicates the brightness under room light and the monodispersity of nanocrystals with cubic shape.

Soon after the report of hot-injection synthesis of CsPbX₃ nanocrystals, many other kinds of fabrication methods have been proposed with the aim to improve the fabrication procedures and overcome the limitations of hot injection approach.^[18, 34-41] Li *et al.* demonstrated the room temperature synthesis of CsPbX₃ nanocrystals based on the supersaturation-induced recrystallization which has been observed in natural minerals, alloys, and ion solutions.^[18] Besides the advantage of the low temperature operation, this recrystallization method is free from inert environment and injection condition, implying the feasibility of commercialization. In brief, all of the precursors including CsX and PbX₂ are initially dissolved in the aprotic polar solvent of dimethyl sulfoxide (DMSO), together with the organic ligands of OAm and OA. Then, the above mixture solution is transferred into toluene which acts as the poor solvent for the ions of Cs⁺, Pb²⁺, and X⁻. The recrystallization takes place upon the liquid transfer and CsPbX₃ nanocrystals grow rapidly due to the presence of a highly supersaturated state for the ions. The organic ligands are indispensable here as they play an important role in controlling the crystal size and dispersing the nanocrystals in the nonpolar solvent. Figure 2b displays the schematic illumination of the recrystallization fabrication flow. Noticeably, despite the low reaction temperature and absence of

inert atmosphere, the CsPbX_3 nanocrystals obtained by the recrystallization method show comparable optical properties to those of the hot injection analogues. Later, Dirin *et al.* developed the templated growth of ligands-free CsPbX_3 nanocrystals by simply infiltrating the perovskite precursor solutions into the pores of mesoporous silica and allowing the solution dry naturally (Figure 2c).^[35] Surprisingly, the synthesized CsPbX_3 nanocrystal powders exhibit excellent emission performance with PLQY surpassing 50%. The size of the nanocrystals can be facilely controlled by choosing the silica templates with proper pore sizes, while the composition is determined by the preset precursors. Furthermore, Zhu *et al.* reported the solvent-free synthesis of CsPbX_3 nanocrystals by mechanically milling or grinding raw CsX and PbX_2 salts with stoichiometric combinations at room temperature.^[37] Obviously, this mechanosynthesis method represents the greenest strategy toward CsPbX_3 nanocrystals as it avoids the routine use of toxic organic and inorganic solvent besides the absence of high reaction temperature and inert gas protection. The last but not the least, in order to facilitate the in-situ photoluminescence and absorption measurements so as to readily map the reaction parameters, such as the molar ratios of cesium, lead, and halide precursors, reaction times and temperatures, Lignos *et al.* employed the droplet-based microfluidic platform to fabricate the CsPbX_3 nanocrystals (Figure 2d), where they demonstred that the microfluidic systems could serve as the unique tool to optimize the synthesis parameters of the CsPbX_3 nanocrystals.^[36]

Aside from the quasi-zero dimensional CsPbX_3 nanocubes, it is highly intriguing to fabricate the CsPbX_3 nanostructures with different morphologies including the one-dimensional nanowires and two-dimensional nanoplatelets. On one hand, the CsPbX_3 nanostructures with varied dimensions can provide a platform to comprehensively study the photophysics of the emerging perovskite materials, such as the quantum confinement effect and carrier dynamics. On the other

hand, in terms of optoelectronic applications, CsPbX₃ nanocrystals with certain morphology may favor the specific requirement of the device.^[42, 43] In the past couple of years, a number of reports have been devoted to the morphology engineering of CsPbX₃ nanostructures.^[22, 44-50] Bekenstein *et al.* firstly demonstrated the fabrication of CsPbBr₃ nanoplatelets by temperature control. They found that the hot injection method used for synthesizing the CsPbX₃ nanocrystals (nanocubes) can also be employed to synthesize the two-dimensional nanoplatelets by simply lowering down the reaction temperature (90-130 °C).^[44] As shown in **Figure 3a**, the green-emitting CsPbBr₃ nanocubes with edge length of 8-10 nm are produced under reaction temperature of 150 °C; In contrast, the cyan-emitting nanoplatelets with lateral sizes of ~20 nm and thickness of 3 nm) are produced at temperature of 130 °C (Figure 3b) and the blue-emitting nanoplatelets with even thinner thickness are formed at 90°C (Figure 3c).^[44] Notably, these CsPbBr₃ nanoplatelets show bright emission with high PLQY of up to 84% and exhibit unique self-assembly behavior. The perovskite nanoplatelets with other halide composition (X=Cl, Br and I) can be obtained by the facile anion exchange process, such that the emission wavelength of the CsPbX₃ nanoplatelets can be tuned to cover the whole visible spectral range. Zhang *et al.* are the first to report the synthesis of CsPbX₃ nanowires by reacting cesium oleate with PbX₂ in the hot solvent of ODE with OAm and OA at temperatures of 150–250 °C.^[48] They carefully investigated the formation dynamics of nanowires by analyzing the intermediate products at different reaction times. Take the reaction at temperature of 150 °C as an example, the perovskite nanocubes are the main reaction products at the early stage (< 10 min). With increasing the reaction time, the thin perovskite nanowires appeared and the amount of the nanowires increased, accompanied by the diminishing of the perovskite nanocubes. For even longer reaction times (>90 min), the nanowires vanish and the products are mostly irregular large crystals.^[48] They also noticed the formation of the square

nanosheets during the reaction (10-40 min) and the different intermediates are able to co-exist in the reaction.^[48] Figure 3d shows the sketch of the different concentration of the perovskite nanostructures along with the reaction time. Their observations unambiguously manifest the importance of reaction time control for synthesizing CsPbX₃ nanostructures with preferred morphologies.^[48] In addition to the reaction temperature and time, the surfactant ligands have been known as an effective tool to manipulate the size and shape of the nanocrystals.^[51, 52] Sun *et al.* reported the facile shape control of the CsPbX₃ nanocrystals by employing proper surfactant combinations of acid and amine, from which they demonstrated the fabrication of quasi-zero-dimensional nanospheres and nanocubes, one-dimensional nanorods, and two-dimensional nanoplatelets with few-unit-cell thickness.^[51]

3. Photophysics of CsPbX₃ nanocrystals

3.1 Crystal and electronic structures

It is well-known that the optical properties of semiconductors are closely related to the crystal and electronic structures.^[16, 53] The first investigation on the crystal structure of CsPbX₃ was performed in 1958.^[33] It is disclosed that CsPbX₃ typically shows three types of crystal polymorph, that is, the orthorhombic, tetragonal, and cubic phases.^[33, 54] The phase transitions can occur when the environment temperature changes.^[54-56] In the cubic structured CsPbX₃ crystals which, in general, are only stable at high temperatures, the Pb²⁺ and X⁻ ions form a three dimensional net of corner sharing [PbX₆] octahedral and the Cs⁺ ions occupy the octahedral voids created by the halogens, see inset in Figure 1b. As the temperature decreases, the [PbX₆] octahedral will tilt regarding the orthogonal geometry and the crystal structure will transform from the cubic to the tetragonal and to the orthorhombic gradually, adopting the distorted perovskite structures (see **Table 1** for the typical phase transition points).^[33, 54, 57, 58] Noticeably, Protesescu *et al.* found that all of the hot-

injection prepared CsPbX₃ nanocrystals except the CsPbI₃ ones exhibit stable cubic phase at room temperature, which is ascribed to the high reaction temperature and the contribution from the surface energy of the nanocrystals.^[16] For the case of CsPbI₃ nanocrystal, although it initially shows the cubic phase with bright emission, it converts to the emission-inactive orthorhombic phase upon extended storage (weeks or months).^[16, 59]

The electronic structures of CsPbX₃ perovskites with different halide compositions are found to show qualitatively common features.^[60-62] As shown in Figure 5a, the valence band is constituted by the admixture of *np* orbitals from the halogens (Cl: *n* =3, Br: *n* =4, I: *n* =5) and 6s orbitals from the lead, while the conduction band consists of admixed Pb(6p) and the halide *np* orbitals with leading contributions from the Pb(6p) orbitals, see the density of states of the cubic CsPbBr₃ in Figure 4b as a representative.^[18, 34] **Figure 4a** displays the calculated electronic structures of cubic CsPbCl₃, CsPbBr₃ and CsPbI₃, respectively, based on the density functional theory taking into account the relativistic corrections and spin-orbit interactions.^[59] We can see that the band structures are almost independent on the halide compositions except the difference in the bandgap values and all the CsPbX₃ perovskites show a direct bandgap, indicating the potential application in optoelectronics.^[18, 34, 59, 63] Moreover, small and comparable effective masses of the electrons and holes can be predicted from the electronic band dispersion around the band edges, which contributes to the high carrier mobilities.^[16, 61] As the crystal phase is transformed to tetragonal and orthorhombic polymorphs, the symmetry is lowered with respect to the cubic one, along with the increase of the bandgap and the effective carrier masses.^[16, 53, 57, 59, 64] The discussion about the theoretical calculations predicting the formation of various defects and their respective thermodynamic stability in CsPbX₃ will be provided in the following part (Section 3.2.1).

3.2. Steady-state optical properties of CsPbX₃ nanocrystals

3.2.1 High photoluminescence quantum yield

It is surprising that the as-grown CsPbX₃ nanocrystals exhibit bright emission with PLQY of up to 90%, which makes them highly promising in light-emitting applications. For traditional metal-chalcogenide nanocrystals, exemplified by the CdSe, the as-grown nanocrystals without deliberate surface treatment typically show low PLQY of <20% and manifest broad mid-gap trap state emission.^[19] Such advantageous optical properties of CsPbX₃ nanocrystals with respect to the traditional ones attract the researchers' attention promptly. Up to now, several plausible and instructive mechanisms have been put forwarded to rationalize the high PLQY values of the “naked” CsPbX₃ nanocrystals including large exciton binding energy, equal extent of confinement for charge carriers, surface self-passivation, self-formed type-I core-shell structure and defect-tolerant nature. We will judge them one by one in the following.

(1) Large exciton binding energy and equal confinement of charge carriers

Exciton is the electron-hole pair formed by electrostatic Coulomb attraction, which was first proposed by Yakov Frenkel in 1931.^[65] The bond energy value of the Coulomb force is called exciton binding energy (E_b). Excitons with large E_b can withstand the thermal disturbance and survive at room temperatures (thermal energy of ~26 meV at 300 K), which is definitely beneficial for light emission applications. The calculated E_b of bulk cubic phase CsPbCl₃, CsPbBr₃ and CsPbI₃ are 75, 40 and 20 meV, respectively, according to the formula:^[16]

$$E_b = \frac{2\hbar}{m^* a_0^2},$$

where m^* is the reduced carrier mass and a_0 is the Bohr diameter of the excitons. The Bohr diameter of excitons can be derived based on the effective mass theory, which are 5 nm, 7 nm, and 12 nm for CsPbCl₃, CsPbBr₃ and CsPbI₃, respectively. It can be seen that the calculated E_b of

CsPbCl_3 and CsPbBr_3 is larger than the thermal energy at room temperature, which indicates that the CsPbCl_3 and CsPbBr_3 are optically favorable semiconductors. In fact, it is not straightforward to precisely measure the E_b in experiment. One may estimate the E_b from the temperature dependent absorption and photoluminescence measurements. For example, Zhang *et al.* have roughly evaluated the E_b of CsPbBr_3 and CsPbCl_3 to be 38 and 72 meV,^[66] respectively, which are close to the theoretical values. As the size of the crystals decreases to be comparable or smaller than the Bohr diameter of the excitons, the well-known quantum confinement effect and dielectric effect will come into play. These effects endow the nanocrystals with even larger E_b than the corresponding bulk counterparts. As a result, the excitons of CsPbBr_3 nanocrystals can be quite stable at room temperature, which will contribute to the high PLQY of CsPbBr_3 nanocrystals. In stark contrast to the traditional CdSe-based nanocrystals which require a stronger confinement of carriers to ensure high PLQY, the CsPbBr_3 nanocrystals are able to exhibit high PLQY in weak confinement regime where the size of the nanocrystals is comparable or even larger than the Bohr excitonic diameter.^[16, 17] This distinction in optical behaviors between the traditional CdSe-based nanocrystals and the CsPbBr_3 nanocrystals may arise from the intrinsic electronic structures, whereas the similar effective mass of electron and hole of CsPbBr_3 nanocrystals ($m_e=0.15$ electron mass; $m_h=0.14$ electron mass) will give rise to an nearly identical extent of confinement for both charge carriers, which, in turn, will increase the radiative transition probability by virtue of the enhanced overlap between electron and hole wave function.^[16, 23, 24] However, due to the size inhomogeneity and the existence of labile organic surface ligands, it is not reliable to estimate the E_b of colloidal CsPbBr_3 nanocrystal ensembles based on the temperature dependent absorption or emission measurements. More precise and reliable approach is required to directly derive the

exciton binding energies of the halide perovskite nanocrystals in terms of size, composition and shape.

(2) Surface passivation and type-I core-shell structure

Due to the large surface-to-volume ratio for nanocrystals, the surface will affect or even dominate the optical properties of nanocrystals.^[19] It is found that most of the defects are formed at the surface of the nanocrystals, therefore, surface passivation by organic ligands and surface coating by larger bandgap semiconductor with type-I alignment can kill out the defects and ensure essentially trap-free nanocrystals.^[67] Although there is no deliberate surface passivation or shell coating for the CsPbX₃ nanocrystals, these effects may play a part by themselves. Li *et al.* for the first time investigated the surface chemical states of CsPbX₃ nanocrystals by X-ray photoelectron spectroscopy (XPS) technique.^[18] They found that all of the CsPbX₃ nanocrystals possess halogen-rich surface with no exception. The surplus halogen atoms will connect with cations and suppress the formation of halogen vacancy, which effectively passivate the surface. On the other hand, due to the abundant halogen element, PbX_n compound with larger bandgap may be self-formed on the surface of CsPbX₃ nanocrystals, which effectively serves as the shell, reminding the beneficial type-I core-shell structure, see schematic illustration in Figure 5b.

(3) Defect-tolerant nature

Defect tolerance means that the intrinsic defects do not act as electronic trap states,^[35] which is enabled by the unique electronic band structures.^[68] In lead halide perovskites, the valence band maximum (VBM) is antibonding in nature arising from the hybridization between the Pb(6s) and X(np) orbitals (**Figure 5a**), which is in stark contrast to those of II–VI metal-chalcogenide semiconductors whose VBM is constituted by bonding orbitals. Moreover, the strong spin–orbit coupling effect in lead halide perovskite brings down the conduction band minimum (CBM) to be

lower than the energy of the Pb(6p) atomic orbitals, rendering the defect-related dangling bonds ineffective to generate a deep trap center for carriers. Kang *et al.* have calculated the formation energies and charge-transition levels of intrinsic point defects in CsPbBr₃ by first-principles calculations.^[68] It is found that most of the intrinsic defects only induce shallow transition levels, indicating CsPbBr₃ virtually exhibit intact electronic quality despite the existence of defects. In addition, the effect of the surface organic ligands on CsPbBr₃ nanocrystals has been evaluated by Brinck *et al.* by density functional theory,^[69] which disclose that the surface ligands do not induce additional trap states and the presence of dangling bonds due to evaporation of surface ligands do not quench the PL of CsPbBr₃ nanocrystals as depicted in Figure 6c.

Besides the above possible mechanisms, the high emission color-purity can also contribute to the high PLQY. In traditional metal-chalcogenide colloidal nanocrystals, the emission line-width lies in the range of 25-35 nm due to the size and composition inhomogeneity. While for CsPbBr₃ nanocrystals, the FWHM is much narrower typically ranging from 15 nm to 25 nm, which can be attributed to the superior size uniformity and weak quantum confinement effect. As a comparison, Figure 7b shows the typical PL spectra of Rhodamine 6G, CdSe/CdS/ZnS core-shell nanocrystals and CsPbBr₃ nanocrystals. The narrow PL line-width can effectively suppress the detrimental reabsorption process and the inter-particle Forster resonance energy transfer (FRET) in the nanocrystal ensemble, hence favoring the high PLQY as manifested in Figures 7c and d.^[17]

3.2.2 Composition and size tuned optical properties

As a close relative to the organic-inorganic hybrid perovskites, the all-inorganic CsPbX₃ nanocrystals share the common property of composition controllable bandgap energy and emission spectrum.^[16] By changing the halogen elements (Cl, Br and I), the PL wavelength can be finely tuned across the whole visible range from ~400 to ~700 nm (**Figures 6a and e**), maintaining the

high PLQY above 50%. As discussed above, the CsPbX_3 nanocrystals feature a narrow PL line-width, varying from ~ 12 nm for CsPbCl_3 nanocrystals to ~ 42 nm for CsPbI_3 nanocrystals. The narrow PL line-width suggests that the CsPbX_3 nanocrystals possess a wide gamut of pure colors, which is favorable for high quality lighting and displays. Figure 6g plots the color gamut of CsPbX_3 nanocrystals in CIE chromaticity coordinates introduced by the Commission Internationale de l'Eclairage. The selected triangle constituted by red-, green-, and blue-emitting CsPbX_3 nanocrystals well covers the conventional color standards of LCD TV (the Liquid Crystal Display) and NTSC TV (the National Television System Committee). Interestingly, the compositional modulation in the CsPbX_3 nanocrystals can be fulfilled either by simply mixing the CsPbCl_3 , CsPbBr_3 , and CsPbI_3 nanocrystals in solution or by adding compounds serving as halide sources due to the fast ion exchange processes.^[21, 70] Hence, desired emission spectra can be achieved by simply applying appropriate ratios of the parent CsPbX_3 solutions and suitable amount of halide sources.^[21, 70] In order to elucidate the composition dependent band edge energies of CsPbX_3 nanocrystals, the nature of VBM and CBM have been investigated as a function of halide element.^[71, 72] Cyclic voltammetry measurements unravel that the VBM dramatically changes to higher energies by 0.80 eV with the change of X from Cl to Br to I, whereas the shift in CBM is only as small as 0.19 eV, indicating that the VBM is more sensitive to the halogen elements, which further confirms the dominate role of halogen for constituting the VBM.

As the hallmark of colloidal quantum dots, the bandgap energy is strongly size-dependent due to the quantum confinement effect. Figure 6f shows the absorption and PL spectra of CsPbBr_3 nanocrystals with different sizes varying from 3.8 to 11.8 nm. It is found that the absorption edge and the PL wavelength blue-shift as the size decreases, which can be well-explained based on effective mass approximation (EMA).^[16] In particular, the confinement energy (ΔE) is given by:

$\Delta E = \frac{\hbar^2 \pi^2}{2m^* r^2}$, where the r is the nanocrystal radius. The size dependent bandgap of CsPbBr₃ nanocrystals can be well reproduced by the theoretical calculation.^[16] Another interesting phenomenon can be observed from Figure 6f is the size dependent Stokes shift in CsPbBr₃ nanocrystals.^[16, 73] The presence of size dependent Stokes shift in luminescent nanocrystals is thought-provoking, which may deliver rich information about the photophysics and electronic structures.^[73] Previously, similar size dependent Stokes shift have been observed in CdSe quantum dots, which is attributed to the existence of band edge excitonic fine structure based on the detailed theoretical modeling.^[74] However, it is unknown whether comparable band edge excitonic fine structure is responsible for the size dependent Stokes shift of the CsPbBr₃ nanocrystals because many other possible reasons can give rise to the similar phenomenon, such as the deteriorated size uniformity for small sized nanocrystals.^[73] Therefore, further in-depth investigation is required to shed light on the size dependent Stokes shift of the CsPbBr₃ nanocrystals, which is of importance from view of both fundamental physics and practical applications.

3.2.3 Single particle optical properties

In general, the colloidal nanocrystals suffer from the size inhomogeneity, which results in broadening of the emission and absorption spectrum. Although the CsPbX₃ nanocrystals exhibit relatively better color purity compared to the conventional CdSe-based quantum dots, the size inhomogeneity is inevitable.^[17] For instance, a pronounced band edge excitonic absorption peak is observed in bulk CsPbBr₃, while it diminishes in nanocrystal absorption spectra with sizes of ~10 nm, which may suggest the size non-uniformity of the nanocrystal ensemble.^[16, 66, 75] As a result, single particle characterization becomes necessary in order to dig out the cloaked information in the nanocrystal ensemble, such as excitonic fine structure, spectral diffusion and charged emissive states. **Figure 7a** presents the fluorescence image of single CsPbBr₃ nanocrystal manifested by the

individual bright spots. The corresponding PL peaks is found to vary from ~ 505 nm to ~ 515 nm, which is a clear demonstration of the size-dependent energy gaps of the CsPbBr₃ nanocrystals.^[76] Fluctuations or blinking in PL emission from individual CsPbBr₃ nanocrystal are unambiguously observed under continuous illumination, while a large proportion of the CsPbBr₃ nanocrystals ($\sim 90\%$) exhibit greatly suppressed blinking behavior with on-time (emissive state) larger than 85%.^[17] The low temperature single particle measurements further unveil the narrow PL spectrum, suppressed blinking and minor spectral diffusion from CsPbX₃ nanocrystals.^[77] By using the high-resolution spectroscopy, asymmetric PL spectra with narrow line-widths of < 2 meV are observed. Notably, some of the nanocrystals show multiple emission peaks even under low excitation intensities ($\langle N \rangle \ll 1$) (Figure 7c). The polarization dependent PL measurement (Figure 7d) discloses that the three resolved peaks exhibit different degrees and axes of polarization. Moreover, the line-widths of the peaks and the relative polarization orientations change from one particle to another, indicating that the fine structure of CsPbBr₃ nanocrystal is related to anisotropies or defect complexes. With the increase of excitation intensity, the charged exciton states are determined, which, in turn, indicates that the charged or multiexcitons are still emissive. Importantly, several groups reported the achievement of single photon emission from the single CsPbX₃ nanocrystal, which is evidenced by strong photon antibunching behavior as shown in Figure 7b, indicating that the CsPbX₃ nanocrystals are promising quantum emitters.^[76-78]

Moreover, Lin *et al.* employed the sophisticated valence-loss electron spectroscopy to directly observe the bandgap variations of CsPbBr₃ nanocrystals from single particle level.^[79] It turns out that bandgap energy of the individual CsPbBr₃ nanocrystal is dictated by the smallest of the three edges of the cubic nanocrystal, which strictly follows the quantum confinement effect. They also demonstrated that neighboring CsPbBr₃ nanocrystals will effectively interact with each

other, giving rise to the modification of band structure for the individual nanocrystal.

3.3. Carrier dynamics of CsPbX₃ nanocrystals

3.3.1 PL dynamics of single exciton state

The time resolved PL measurements of colloidal nanocrystals can provide a wealth of information about the photophysics. **Figure 8a** shows the PL dynamics of CsPbX₃ nanocrystals with different halide compositions in solution at room temperature. It is noticed that the PL decay rate decreases monotonically as the halogen changes from Cl to Br to I with 1/e lifetime changing from several nanoseconds to several tens of nanosecond. The regular change of the PL decay dynamics as a function of halogen substitution in CsPbX₃ nanocrystals can be qualitatively explained based on the dipole model.^[80-82] For a dipole oscillator or emitter, the radiative transition rate R_r is proportional to the product of the bandgap energy E_g and the square of the momentum matrix element p (other factors like the dielectric screening and fine structure splitting can be ignored for the CsPbX₃ nanocrystals).^[16, 58, 83] The momentum matrix element p responsible for the transition from conduction band to valence band in semiconductor is given by: $p = m_0 E_p / 2$, where E_p represents the Kane energy. In general, the Kane energy is nearly a constant of about 20 eV for a wide range of semiconductors of II–VI, III–V and group-IV families. Therefore, as the bandgap energy increases with the change of anion from I to Br to Cl, the radiative recombination becomes faster. However, the increasing rate of the PL decay is much faster than that of the bandgap change with halide substitution, indicating that the Kane energy may be sensitive to the composition of lead halide perovskites. This is reasonable because the unique electronic band structure of lead halide perovskites, especially, the antibonding nature of the VBM, whereas the constant Kane energy is only applicable for the conventional semiconductors with bonding nature of VBM.^[81, 84] Comprehensive study on the anion-dependent Kane energy in lead halide perovskites is highly

desired. On the other hand, as shown in Figure 8a, the PL transients of the CsPbX₃ nanocrystals are not single exponential decay, indicating the multi-channel recombination. Given that the excitation intensity is sufficient low thus most of the CsPbX₃ nanocrystals contain less than one electron-hole pair, the PL exclusively comes from the single exciton recombination. Such multi-channel PL decay from single exciton regime was also observed in CdSe nanocrystals and was ascribed to the recombination from the core and surface area of the nanocrystals.^[85] Similarly, the multi-channel PL transient of CsPbX₃ nanocrystals suggests the radiative recombination from multiple carrier states, which also implies the presence of carrier trapping processes in these CsPbX₃ nanocrystals. In turn, the multi-exponential PL decay explains the subunit PLQY of these CsPbX₃ nanocrystals. Bearing this in mind, Koscher *et al.* successfully achieved the “perfect” CsPbBr₃ nanocrystals with PLQY of unity by postsynthetic thiocyanate surface treatment and tentatively attribute to the repair of under-coordinated lead atoms on the surface as schematically depicted in Figure 8b.^[20] Concurrently, the PL decay kinetics transform from the multi-exponential curve with a distribution of trap sites to a single exponential decay (Figure 8b), which is the representative of single energy level emitters. Unfortunately, this surface-repair strategy only works for CsPbBr₃ nanocrystals and have little effect for other halide nanocrystals, such as CsPbBr_xCl_{3-x} and CsPbBr_xI_{3-x} compositions, which indicates the complexity of the emission process in CsPbX₃ nanocrystals.^[20] It requires the synergic effort of theory, synthesis and optical characterization to reach a clear picture about the emission process of CsPbX₃ nanocrystals, which is definitely essential to make full exploitation of their potential.

3.3.2 PL dynamics of multiexciton states

With the increase of the excitation intensity, more than one excitons may occupy the same individual nanocrystal. The multexcitonic recombination is closely correlated with various

photophysical phenomena and practical application of colloidal nanocrystals and turns out to be radically different from that of the single exciton.^[28, 83] Hence, spectrally and temporally resolving the multiexciton state emission is crucial in terms of understanding and utilizing of the CsPbX₃ nanocrystals. In general, the photocarrier recombination kinetics over a range of excitation intensities can be described by the rate equation:

$$\frac{dn(t)}{dt} = -k_1n - k_2n^2 - k_3n^3,$$

where n is the photocarrier density, t is time and k is the recombination constant.^[86] For CsPbBr₃ nanocrystals whose excited state populations are primarily excitons and defect trapping is insignificant as manifested by the high PLQY, each term in right-hand side of rate equation can be ascribed to physical carrier annihilation processes of the single exciton radiative recombination, the exciton–exciton annihilation and the three-body Auger recombination, respectively.^[87, 88] Figure 8c displays the pump intensity dependent PL dynamics of CsPbBr₃ nanocrystals. It is noticed that a much faster decay than that of single exciton recombination appears under high excitation intensities, corresponding to the nonradiative biexciton Auger recombination. Similar phenomena are also observed for CsPbX₃ nanocrystals of other halides.^[83, 89, 90] By subtraction of the single exciton recombination, the biexciton Auger lifetime is determined to be less than 100 ps for CsPbBr₃ nanocrystals with size of ~10 nm. Based on the pump intensity dependent carrier dynamics, the degeneracy of the band-edge states are demonstrated to be 2-fold, indicating that both the electron and the hole band-edge states accommodate at most two carriers.^[83, 87] As a result, the carrier dynamics under high excitation levels are exclusively governed by biexcitons, while higher-order multiexcitons can only show emission from the higher-energy levels. It is worth noting that precise determination of the biexciton Auger recombination rate in CsPbX₃ nanocrystals is not straightforward because it is extremely fast and relies on a number of

parameters, such as the excitation intensity, size, shape, composition and charging effect.^[91] By comparing the PL decay transients recorded for stirred and unstirred CsPbX₃ nanocrystals solution, Makarov *et al.* have demonstrated the presence of charged exciton or trion emission in the perovskite nanocrystals and determined the trion lifetime of ~235 ps for CsPb(Br/I)₃ dominated by the three-body Auger recombination.^[83] Wang *et al.* used the quasi-continuous wave pumping and ultrafast spectroscopic techniques to spectrally resolve the biexcitonic emission and extract the biexciton binding energy to be ~50 meV.^[28] Later on, multiexciton interaction in CsPbX₃ nanocrystals was investigated as a function of nanocrystal size and composition by Castañeda *et al.* as displayed in Figures 8d-h.^[90] The biexciton binding energy of CsPbX₃ nanocrystals is found to be significantly enhanced compared to the conventional CdSe nanocrystals, and the largest value of the biexciton binding energy reaches about 100 meV (Figure 8i). Such strong biexciton Coulombic interaction is also consisted with the much faster Auger recombination rate than that of CdSe nanocrystals with the same volume. The strong multiexciton Coulombic interaction indicates the CsPbX₃ nanocrystals are promising candidates for the Coulomb assisted applications, such as carrier multiplication and lasing.^[91]

3.3.3 Transient absorption of CsPbX₃ nanocrystals

The ultrafast transient absorption (TA) spectroscopy could provide complementary information to the time resolved PL experiments. Until now, the TA measurements of CsPbX₃ nanocrystals have been widely investigated by several groups.^[83, 87-89, 92-95] Wu *et al.* for the first time investigated the carrier dynamics of CsPbBr₃ nanocrystals using the femtosecond white-light pump-probe system.^[88] **Figure 9a** shows the typical TA spectra of CsPbBr₃ nanocrystals with excitation wavelength of 400 nm, where three well-resolved features are observed, that is, one band of photoinduced bleach (XB) centered at 480 nm and two bands of photoinduced absorption (PA).

The energy of XB agrees well with the lowest energy excitonic band, suggesting the origin of state-filling-induced bleach. The PA₁ with energy slightly below the lowest excitonic band is attributed to the so-called “biexciton-induced level shifting”^[87, 92, 96] based on the following two characteristic behaviors. First, this peak only appear at short time range of <1 ps; Second, this peak can only be observed by excitation well above the energy of the lowest optical transition as unveiled by Chung *et al.* (Figures 9c and d) and Mondal *et al.*^[92, 93] The PA₂ covers the whole visible region and the amplitude is much weaker than that of XB. Such broadband PA₂ has been widely observed in other kinds of colloidal nanocrystals, while the exact origin of this broadband PA is still unknown.^[87, 97] Importantly, in combination with the time resolved PL measurements, they demonstrated that the trapping of the excited electrons and holes is nearly negligible in the CsPbBr₃ nanocrystals, in which ~94% of lowest excitonic states recombine with a single exponential time constant of 4.5±0.2 ns, in agreement with the high PLQY of the samples.^[88]

Chung *et al.* have comprehensively studied the hot carrier relaxation dynamics in the CsPbX₃ nanocrystals.^[93] **Figure 10** shows the intraband hot carrier relaxation process as a function of anion composition. All the samples irrespective of halide element exhibit ultrafast cooling dynamics in the time scale of sub-picosecond (<0.6 ps). Moreover, they observed a regular change of the cooling rate when substituting Br by I: CsPbBr₃ (310 fs) > CsPbBr_{1.5}I_{1.5} (380 fs) > CsPbI₃ (580 fs), which is ascribed to the larger overlap integral between the hole states and phonon modes in CsPbBr₃ nanocrystals than that of CsPbI₃ nanocrystals. The sub-picosecond hot carrier cooling dynamics indicate the breakdown of phonon bottleneck in the CsPbX₃ nanocrystals, which favors the buildup of the lowest transition states and hence the light-emitting applications.^[93] However, the underlying mechanisms of the absence of phonon bottleneck in the CsPbX₃ nanocrystals has

not yet been positively determined, whereas Chung *et al.* tentatively attributed it to the efficient multiphonon emission process.^[93]

The pump intensity dependent TA measurements have also been performed for CsPbBr₃ nanocrystals. **Figure 11** shows the TA spectra of CsPbBr₃ nanocrystals at different excitation densities and different probe delays under excitation wavelength of 400 nm. With the increase of excitation densities, the XB shows a carrier density dependent blue-shift and broadening, which is attributed to the charge carrier accumulation at the band edges for the optical transition and can be elucidated on the basis of a dynamic Burstein–Moss shift, which has also been similarly observed in the organic-inorganic hybrid perovskites.^[86–88] The PA₁ signal to the red of XB feature greatly increases with excitation density, which can be attributed to the enhanced impact of multiexciton states.^[87, 88] By extracting the transient XB kinetics at different excitation intensities for CsPbBr₃ nanocrystals, a fast decay channel corresponding to the biexciton Auger recombination is detected, the Auger lifetime is estimated to be 44±8 ps, which is comparable to the value derived from the time resolved PL measurements.^[28, 83, 88, 95] Moreover, Yarita *et al.* demonstrated the existence of charged excitons or trions with increasing excitation densities in the CsPbBr₃ nanocrystals, whose lifetime is determined to be ~200 ps, which, again, is consistent with the time resolved PL results.^[95]

Time-resolved terahertz (THz) spectroscopy used to probe transient photoconductivity within the CsPbBr₃ nanocrystals was done by Yettapu *et al.*, from which they unraveled the remarkably high carrier mobility (~4500 cm² V⁻¹ s⁻¹) and large diffusion length (>9.2 μm) in the CsPbBr₃ nanocrystals owing to the negligible influence of surface defects in trapping charge carriers. The above values are even comparable to or better than the best quality bulk single

crystalline semiconductors, indicating that the colloidal CsPbBr₃ nanocrystals are promising candidates for optoelectronic and photovoltaic device applications.

3.4 Nonlinear optical properties of CsPbX₃ nanocrystals

With the development of high power lasers, the nonlinear optics has gained increasing importance in both fundamental physics and practical application. Wang *et al.* are the first to investigate the multi-photon absorption and emission properties of CsPbX₃ nanocrystals.^[29] As shown in the inset of the **Figure 12a**, the CsPbBr₃ nanocrystals exhibit bright emission upon irradiation at optical wavelength of 800 nm by a femtosecond laser source, which indicates that the CsPbBr₃ nanocrystals are highly multi-photon active since there is no linear absorption at the near infrared region. The spectrally integrated PL intensity shows quadratic and cubic dependence on the excitation density for excitation wavelength of 800 nm (Figures 12a and b) and 1250 nm, respectively, which indicates that the CsPbBr₃ nanocrystals undergo the corresponding two-photon and three-photon absorption and emission processes. To gain more insight into the nonlinear optical process, the PL spectra and transient PL dynamics of CsPbBr₃ nanocrystals in solution induced by optical wavelengths of 400 nm, 800 nm and 1250 nm, which correspond to one-, two- and three-photon excitation, were measured. The nearly the same PL spectra and PL decay curves (Figure 12d) suggest that the photo-carriers induced by both linear absorption and nonlinear multi-photon absorption finally relax to the identical lowest excited state in order to generate the spontaneous emission, which is schematically depicted in the inset of Figure 12d. The Z-scan technique was employed to quantitatively determine the two-photon absorption cross section of the CsPbBr₃ nanocrystals with carbon disulfide as a reference. Figure 12c shows that the nonlinear absorption totally originates from the CsPbBr₃ nanocrystals rather than the solvent as manifested by the deep groove and flat response for the CsPbBr₃ sample and solvent, respectively. The two-

photon absorption cross-section of the CsPbBr₃ nanocrystals is determined to be 1.2×10^5 GM, which is orders of magnitude larger than those of traditional quantum dots and dyes, suggesting their potential as efficient nonlinear absorbers and nonlinear photonic media. Leveraging on the efficient nonlinear absorption and emission in the CsPbX₃ nanocrystals, the low-threshold two- and three-photon pumped stimulated emission or amplified spontaneous emission are achieved from the close packed thin films of the perovskite nanocrystals with wavelength tunable across the whole visible spectral range, which will be elaborated later. Soon after, several groups reported the strong nonlinear absorption and emission from CsPbX₃ nanocrystals with different compositions, shapes and sizes.^[32, 98-103] In particular, Chen *et al.* systematically investigated the two-photon absorption in CsPbBr₃ nanocrystals with varied sizes from 4.6 to 11.4 nm by monitoring the two-photon induced ground state bleaching signals,^[101] from which a power law dependence with exponent 3.3 ± 0.2 of the two-photon absorption cross-section on nanocrystal size was determined. Moreover, the two-photon absorption cross-section of the perovskite nanocrystals was found to scale as the linear photon absorption amplitude. The revealed power-law dependence and the relationship between the linear and two-photon absorption will benefit for developing high-performance nonlinear optical devices based on CsPbX₃ nanocrystals.

3.5 Exotic photophysical properties

Compared to the conventional covalent metal chalcogenide nanocrystals whose crystal structure is either zinc blend or wurtzite, CsPbX₃ nanocrystals feature the perovskite structure, the unique electronic structures, and the ionic crystal nature. Hence, these CsPbX₃ nanocrystals may exhibit intrinsically different photophysical properties, which we term “exotic photophysical properties”. In the following, we will present several exotic phenomena observed in CsPbX₃ nanocrystals,

which are physically interesting and essential for practical applications. In fact, these “exotic” phenomena are not exotic at all after the underlying mechanisms are elucidated.

3.5.1 Temperature-insensitive PL peak position above room temperature

For the device applications including LEDs and lasers, the temperature in the operating active region will inevitably increase due to the generated heat. For the traditional CdSe-based nanocrystals, the bandgap typically decreases with increasing temperature, thereby deteriorating the device chromaticity.^[104] Interestingly, CsPbBr₃ nanocrystals are found to show nearly temperature-insensitive PL peak position in the temperature range of ~20-100 °C.^[17, 39, 100, 105, 106]

In general, the bandgap change of semiconductors as a function of temperature stems from the combined effects of crystal lattice expansion and electron-phonon coupling. In the case of the traditional CdSe-based nanocrystals, both the lattice dilation and the electron-phonon interactions give rise to the decreased bandgap as the temperature increases. In contrast, as a result of the anti-bonding nature of the VBM in CsPbBr₃, the thermal expansion of the crystal lattice will result in the increase of the bandgap due to the thermal induced decrease of the VBM. In the temperature range of ~20-100°C, the lattice expansion effect and the electron-phonon interaction in CsPbBr₃ nanocrystals cancel each other out, resulting in the nearly color-drift free emission. The unique temperature-insensitive chromaticity of CsPbBr₃ nanocrystals may provide new opportunities in light-emission applications, such as the LED, amplifiers and lasers.

3.5.2 Pressure induced isostructural phase transformation

Pressure, as a unique thermodynamic variable, may dramatically affect the electronic structure and optical properties of active materials. Investigation of the pressure dependent photophysical behavior of CsPbX₃ nanocrystals would help to understand the relationship between the atomic/mesostructures and the associated properties.^[107] Very recently, Nagaoka *et al.* reported

the pressure induced conversion of CsPbBr₃ nanocrystals from the nanocube superlattices into uniform 2D nanoplatelets.^[108] Notably, the pressure-formed CsPbBr₃ nanoplatelets show enhanced PLQY and prolonged lifetime with respect to the as-grown CsPbBr₃ nanocrystals. Besides the surface modification by the applied pressure, the improved optical property of the CsPbBr₃ nanoplatelets was also attributed to the perfect single-crystallinity phase with minimal defects. Their results indicate that pressure processing can be exploited as the practical approach for the rapid conversion of lead halide perovskites into preferred structures with enhanced properties. Deeper understanding of the pressure effects on CsPbBr₃ nanocrystals from the view of atomic energy level was performed by Xiao and co-workers.^[107] To avoid the non-hydrostatic pressure resulted from the anisotropic superlattices, they adopted the isolated CsPbBr₃ nanocrystals as the target sample. As shown in **Figure 13a**, the PL peak wavelength shows redshift and the PL intensity decreases severely upon compression to 1.21 GPa, after which the emission undergoes an abrupt quenching. Based on their systematic in situ high-pressure-absorption, -PL, -lifetime, and -angle dispersive X-ray diffraction measurements as well as the first-principles density functional theory calculation, they unraveled the occurrence of an isostructural phase transformation of the CsPbBr₃ nanocrystals at ~1.2 GPa (Figure 13b: stage I to stage II) due to the repulsive force impact arising from the overlap between the valence electron charge clouds of neighboring layers. Moreover, with further increasing the pressure (> 7 GPa), the strong distortion of lead–bromide octahedron as a result of the Jahn–Teller effect eventually gives rise to an electronic transition from direct to indirect bandgap (stage III). Their findings disclose the fundamental relationship between structural variations and optical properties of CsPbBr₃ nanocrystals under high pressure.

3.5.3 Light induced structure transformation

By virtue of the ionic crystal nature and the dynamic ligand binding of the CsPbX_3 nanocrystals, the inter-particle ion diffusion should be applicable under external driven force. Wang *et al.* reported the light induced transformation from the quantum confined IHPNs including the few-monolayer nanoplatelets and the nanocubics to the bulk phase due to the fusion among the nanoparticles (Figures 13c and d).^[109] Taking advantage of the remarkable emission color change during the transformation, they realized the fancy multi-color luminescence photopatterns and established the security label photo-encoding from the CsPbBr_3 nanoplatelets and polymethyl methacrylate (PMMA) composite films (Figures 13e-g). Especially, by virtue of the lamellar self-assembly in the CsPbBr_3 nanoplatelets, the triggering light intensity for the photopatterning reaches as low as $\sim 20 \text{ mW/cm}^2$, which is affordable by cheap and commercially available lamps. The distinctive light induced transformation in the CsPbX_3 nanostructures is attributed to the highly dynamic ligand binding and facile ion inter-diffusion by virtue of the ionic crystal nature.^[21, 109-111] The findings not only present an interesting photoinduced structure conversion in the emerging all-inorganic perovskite nanostructures, but also demonstrate the application potential of the perovskite nanostructures in security protection, such as anti-counterfeiting and information encryption. As a matter of fact, light induced structure modification occurs not just in inorganic halide perovskites, it was recently found that light has a strong influence on tuning morphology and structuring hybrid halide perovskite films, which opens up a new way of optimizing materials for device applications.^[112]

3.5.4 Spontaneous linearly polarized emission

Linearly polarized light emitters are highly desirable in a variety of applications including the lighting and displays.^[113, 114] Interestingly, Wang *et al.* reported the highly polarized emission from the CsPbX_3 ($\text{X} = \text{Br}, \text{I}$ and mixed halide systems Br/I) nanocrystal ensembles in solution and solid

film without any deliberate alignment.^[115] Especially, the polarization of CsPbI₃ nanocrystals is revealed to be as high as 0.36 in hexane solvent and 0.40 in solid film. They explained the observed linearly polarized emission from the CsPbX₃ nanocrystals in terms of the crystal structure of the perovskite, the contribution of the halide ion immigration, and the spatial arrangement of nanocrystals by self-assembly. However, there are still many unknowns in the polarized emission phenomenon from the CsPbX₃ nanocrystals. For example, it is not clear why only the iodine-rich nanocrystals show strong linearly polarized emission and why the CsPbBr₃ nanocrystals lose the polarized emission in solid film while the polarization of CsPbI₃ nanocrystals even increases. Without doubt, further comprehensive studies are required in order to reach a clear picture behind the phenomenon, among which the single particle polarization measurements may provide valuable information.^[113]

4. Light-emitting applications

4.1 Lasers

Lead halide perovskite materials are actually not new to researchers, the study of cesium lead halide perovskite materials dates back to the 20th century. As a matter of fact, the optical gain and stimulated emission from CsPbBr₃ films has been firstly observed by Kondo *et al.* in 2005.^[116] However, the films required high temperature annealing process and exhibited poor microcrystalline quality, rendering the high pump threshold, hence the lead halide perovskites have not attracted much attention at that time. Quantum dots or nanocrystals are new class of advantageous optical gain material over bulk and quantum well counterparts by virtue of the size-tunable emission wavelength, well-separated density of states, large optical oscillator strength and solution processibility.^[117-121] Nearly simultaneously, two groups (Wang *et al.* and Yakunin *et al.*) first independently reported low threshold stimulated emission and lasing from the CsPbX₃

nanocrystals, representing a breakthrough in the research field of inorganic perovskite semiconductor lasers.^[28, 30] In addition, Wang *et al.* have studied the mechanism of stimulated emission and demonstrated the observation of the stability. **Figure 14a** shows the pump intensity dependent PL spectra from the close packed thin film of CsPbBr₃ nanocrystals. The appearance of narrow band emission at the red side of the spontaneous emission peak indicates the occurrence of stimulated emission with low pump threshold of $\sim 20 \mu\text{J cm}^{-2}$ (Figure 14c). Even lower pump threshold of $\sim 6 \mu\text{J cm}^{-2}$ was reported by Yakunin *et al.*, where the difference between the two reports may arise from the quality of the nanocrystals and the films as well as the experimental errors. The low stimulated emission threshold for CsPbBr₃ nanocrystals can be attributed to the large absorption cross-section ($\sim 2.5 \times 10^{-14} \text{ cm}^2$ for 9 nm sized CsPbBr₃ nanocrystals^[28]), high PLQY, the suppressed reabsorption due to the large biexciton binding energy and the relatively small Auger loss.^[28] The stimulated emission from the CsPbBr₃ nanocrystals is found to be very stable under uninterrupted laser irradiation as can be seen from Figure 14d. The model gain of the CsPbBr₃ nanocrystal thin film was estimated to be up to 500 cm^{-1} by the variable stripe length (VSL) method (Figure 14e). It should be noted that obtained model gain values from VSL may show lab-to-lab variance due to the impact of film surface roughness and detailed pumping conditions.^[28] As a result of the bandgap tunability of the CsPbX₃ nanocrystals, the optical gain can be finely tuned spanning the whole visible spectrum by either composition and/or size manipulation (Figure 14b), suggesting the CsPbX₃ nanocrystals as highly promising lasing materials.

Figure 15a shows the whispering gallery mode (WGM) lasing emission made from the CsPbBr₃ nanocrystals infiltrated into a hollow capillary tube. Similarly, the CsPbBr₃ nanocrystals can also be carefully coated onto the microspheres to realize high-quality WGM microlasers (Figure 15b). Although WGM lasers represent one of the easiest configurations, they suffer from

the lack of directionality, one of the most important advantages of a laser.^[122] To this regard, Wang *et al.* sandwiched the solution-processable CsPbX₃ nanocrystals between two high reflective dielectric reflectors and achieved both single longitudinal mode and multiple longitudinal mode vertical cavity surface emitting lasers (VCSELs), manifested by the spectral narrowing, nonlinear increase of the PL intensity, drastic reduction of PL lifetime and remarkable decrease of output beam divergence.^[31] Figures 15c and d show the typical single longitudinal mode lasing spectra from VCSELs made from green-, blue- and red-emitting CsPbX₃ nanocrystals, which unambiguously confirm the feasibility of the CsPbX₃ nanocrystals as practical laser media. VCSELs hold great promise in many important applications including optical communication, high density optical storage, laser display, parallel optical computing and signal processing. The successful demonstration of CsPbX₃ nanocrystals based VCSELs highlighted the versatility of independent adjustment of the cavity and solution-processable nanocrystals, thus representing a significant step towards CsPbX₃ nanocrystal-based practical laser devices.

Besides the one-photon pumped lasers, the nonlinear multi-photon pumped stimulated emission and lasing have been demonstrated from the CsPbX₃ nanocrystals thanks to the favorable nonlinear absorption and emission processes.^[29, 32, 99] Given that the multi-photon induced emission and lasing exhibit many intrinsic advantages over the one-photon counterparts,^[123, 124] such as the large penetration depth, high spatial resolution and little damage to the targeted samples, the realization of low threshold two- and three-photon pumped stimulated emission and lasing from the CsPbX₃ nanocrystals may offer new opportunities for the nonlinear photonics and revive the nonlinear optical devices.

Later on, lasers without additional resonant cavities are made from the single crystalline CsPbX₃ nanostructures, such as the nanowires and nanoplatelets.^[50, 66, 125-129] These micro- or

nanolasers are extremely compact because the CsPbX₃ nanowires and nanoplatelets serve as both the gain media and the resonators, but with the sacrifice of solution processibility as for the colloidal nanocrystals. Figure 16e displays the pump intensity dependent PL spectra from the single CsPbBr₃ nanowire with well-defined rectangular end facets under pump wavelength of 400 nm. With the pump intensity exceeding the threshold, the emission spectra are dominated by closely spaced lasing peaks. Based on the nanowire length dependent free spectral range (*FSR*) investigation, the lasing peaks correspond well to Fabry–Pérot modes, indicating the development of Fabry–Pérot lasing from the CsPbBr₃ nanowires. Concurrently, a periodic emission pattern is observed from the PL image of the nanowire (**Figures 16a-d**), originating from the interference of the coherent emission from nanowire end surfaces. Figure 16f presents the lasing spectra from CsPbX₃ nanowire with varied halide compositions. We can see that the lasing emission can be broadly tuned from ~420 nm to ~720 nm by simply controlling the anionic stoichiometry. Moreover, the CsPbX₃ nanowires exhibit quite stable lasing emission for more than 8 h (150 fs, 250 kHz), indicating that these inorganic perovskite nanowires are very robust, as opposed to the organic-inorganic hybrid perovskite nanowires.^[50, 127, 130] Zhang *et al.* demonstrated high-quality WGM lasing from the CsPbX₃ nanoplatelets with thickness ranging from several tens of nanometers to several hundreds of nanometers synthesized by the vapor transport chemical vapor deposition method.^[66, 131] The lasing mechanism from the CsPbX₃ nanoplatelets is attributed to the WGM lasing based on the size dependent *FSR* measurement and the theoretical simulation. Notably, the lasing threshold is found to be as low as ~2 $\mu\text{J cm}^{-2}$ and the line-width of the lasing peak is as narrow as 0.14 nm, indicating the high performance of the CsPbX₃ microlasers. In addition, two- and three-photon pumped lasing has been realized from the CsPbX₃ nanostructures, such as the nanorods with triangular cross sections and nanocubes,^[126, 132, 133] which suggests that

these CsPbX₃ nanostructures are highly appealing in frequency up-converted lasing devices and on-chip integrated photonics. **Table 2** summarizes the major findings of stimulated emission and lasing from CsPbX₃ nanostructures.

Surprisingly, Park *et al.* reported the observation of strong coupling phenomenon in the CsPbX₃ nanowires grown by the chemical vapor transport method.^[125] They detected the unusual spacing of the Fabry–Pérot lasing modes and these lasing modes were demonstrated to be the same fundamental transverse mode, thereby implying the occurrence of strong light–matter interactions in the CsPbX₃ nanowires. A large group refractive index of 14–17 was extracted from the dispersion curve according to the exciton–polariton mode. The observation of exciton-polariton in the CsPbX₃ nanowires is quite inspiring. On one hand, it points out that the CsPbX₃ nanowires are promising candidates for the “threshold-less” polariton lasers. On the other hand, the large group refractive index induced by the strong light-matter interactions allows for the realization of sub-wavelength waveguide, which is attractive for the integrated photonic circuits and miniaturized photonic devices.^[91]

4.2 Light-emitting diodes

By virtue of the facile emission color tunability, high PLQY, narrow emission line-width, and solution processability, the CsPbX₃ nanocrystals have attracted intense interest in applications of solid-state lighting and display upon the emergence.^[18, 23, 26, 134, 135] In generally, the colloidal nanocrystal-based LEDs can be divided into two categories based on the different excitation mechanisms, namely the phosphor-conversion LEDs and the electrically driven LEDs. For the phosphor-conversion LEDs, an additional near-UV or blue LEDs, such as the commercially available GaN and InGaN semiconductor LEDs, are exploited as the pumping source for the colloidal nanocrystals via indirect injection of electron–hole pairs by means of the photon

reabsorption and/or Förster resonance energy transfer.^[136-138] Thanks to the high color purity of the CsPbX₃ nanocrystals, the emission spectra of the corresponding color-conversion LEDs can be adjusted to mimic that of the natural sunlight by integrating different colored perovskite nanocrystals, which is comfortable for the eyes.^[18, 25, 27, 139, 140] The first electrically driven CsPbX₃ nanocrystal-based LED is reported by Song and co-workers.^[26] By using the typical stacking layered LED structure composed of ITO/PEDOT:PSS/PVK/QDs/TPBi/LiF/Al (**Figures 17a and b**), they achieved bright electroluminescence (EL) from the CsPbX₃ nanocrystal-LEDs. Figure 17c shows the representative current density–luminance–voltage responses of the green-emitting CsPbBr₃ LED. The turn-on voltage is determined to be ~4.2 V, which is larger than the bandgap of the corresponding CsPbBr₃ nanocrystals. A high luminescence of 946 cd m⁻² can be achieved at voltage of 8.8 V. It is found that that the EL of the device shows a narrow line-width, comparable to that of the nanocrystals in solution (Figure 17d), while the peak position slightly redshifts, which may be attributed to the inter-particle interactions between the nanocrystals and the impact of the applied electric field.^[26] The inset of Figure 17d displays the photograph of the LED device with bright and uniform green emission. Taking advantage of the facile emission color tunability, the multi-colored LEDs based on CsPbX₃ nanocrystals with varied halide compositions were realized (**Figures 17e, f and g**). However, the initial non-optimized perovskite nanocrystal-LEDs show relatively poor stability and low external quantum efficiency (EQE) of 0.07%, 0.12%, and 0.09% for the blue, green and red LEDs, respectively.

Inspired by the pioneering work, tremendous efforts have been devoted to improving the device performance of the CsPbX₃ nanocrystal-LEDs, mainly starting from the manipulation of active materials and device structures.^[23, 134, 141-145] Regarding the active CsPbX₃ nanocrystals, Pan *et al.* employed the ligand-exchange strategy to replace the long chain ligands on the as-grown

CsPbX₃ nanocrystals by a relatively short ligand (e.g., di-dodecyl dimethyl ammonium bromide), which resulted in the enhanced carrier transport and increased emission efficiency of the perovskite nanocrystal films.^[143] As a result, a greatly boosted EQE of up to 3% was achieved from the green-emitting halide-ion-pair-capped CsPbBr₃ nanocrystal-LEDs.^[143] They further applied the ligand-exchange method on the mixed anion CsPbBr_xCl_{3-x} nanocrystals and achieved the high EQE of 1.9% for blue-emitting LEDs. At the end of 2016, Li *et al.* developed the surface purification approach to simultaneously optimize the surface passivation and carrier injection efficiency of the CsPbX₃ nanocrystals, which is much more convenient as compared to the ligand-exchange strategy.^[144] The idea is quite straightforward but effective: on one hand, abundant ligands help to passivate the surface, hence ensuring high PLQY and favorable colloidal stability. On the other hand, these surface ligands typically possessing poor conductivity will act as insulator which blocks the carrier injection and transportation in the active layer. As shown in **Figure 18a**, the hexane/ethyl acetate mixed solvent was adopted to control the surface ligand density. By optimizing the balance between the surface passivation and carrier injection, the current efficiency was improved from 0.43 to 13.3 cd/A, and finally, they obtained a record high EQE value of 6.27% from the purification treated CsPbBr₃ nanocrystal-LEDs (Figure 18c).^[144]

In general, additional hole- and electron-transporting layers are required for colloidal nanocrystal based LEDs due to the relatively poor injection efficiency. For example, in the first demonstration of CsPbX₃ nanocrystal-LEDs, Song *et al.* employed the TPBi film as electron-transporting layer, while the PVK film was used as the hole-transporting and electron-blocking layer.^[26] However, it is found that the turn-on voltages for the devices were exclusively larger than the bandgap energy of the corresponding CsPbX₃ nanocrystals as the emitter, which indicates the inefficient carrier injection from the transporting layers. To address this issue, Zhang *et al.*

employed the ultraviolet photoelectron spectroscopy (UPS) to confirm that the poor hole injection was the main limiting channel.^[128] After checking out this problem, they incorporated a thin film of perfluorinated ionomer (PFI) in the LED structure between the hole-transporting layer and CsPbX₃ nanocrystal film, which would have two-fold functions: facilitating the hole injection and maintaining the charge balance of the active CsPbX₃ nanocrystals. As a result, an efficient and barrier-free charge injection was achieved as evidenced by the bright green emission from the CsPbBr₃ nanocrystal-LED under a voltage as low as 2.5 V. Accordingly, the current efficiency and the EQE was greatly improved compared to those of the PFI-free analogues. However, despite that the all-inorganic CsPbX₃ nanocrystals are quite robust, all of the above CsPbX₃ nanocrystal based LEDs suffer from poor stability (the EL intensity reduces to <40% within several minutes),^[26, 134] which may be attributed to the presence of the organic charge transporting layers as has similarly plagued the traditional metal-chalcogenide nanocrystal LEDs.^[146] In this regard, Shi *et al.* exploited the n-type MgZnO and p-type MgNiO as the carrier injectors to replace the organic carrier-transporting layers used in previous reports.^[145] The stacking fully-inorganic multilayer LED structure is shown in Figure 18d, together with the simplified energy band alignment of the multilayered heterostructure illustrating the conduction and valence band energy levels with respect to the vacuum level (Figure 18e). By engineering the energy bands of carrier injectors and optimizing the layer thickness to facilitate the carrier injection, they realized the CsPbBr₃ nanocrystal-based LEDs with greatly improved efficiency of maximum luminance of 3809 cdm⁻², luminous efficiency of 2.25 cdA⁻¹, and EQE of 2.39%. Importantly, thanks to the absence of the labile organic charge-transporting layers, the fully-inorganic perovskite nanocrystal LEDs manifested significantly enhanced stability in air and free from encapsulation as compared to those with organic carrier injectors (Figure 18f).^[145] **Table 3** summarizes the development of

CsPbX₃ nanocrystal-based LEDs. It is notable that the inorganic perovskite nanocrystal LEDs are moving forward in such a remarkable speed, whereas the performance achieved by the CsPbX₃ nanocrystal-LEDs within two years is comparable to that of metal-chalcogenide nanocrystal-LEDs with development of two decades.^[147] Therefore, we can envisage the prospects of the industrial applications of CsPbX₃ nanocrystal-based LEDs in the near future.

5. Issues and challenges

5.1 Stability

Despite that the CsPbX₃ nanocrystals have demonstrated much better stability than the organic-inorganic hybrid perovskites, they still suffer from the poor stability against the moisture, light irradiation and heat.^[148-150] Due to the intrinsic ionic crystal nature, these CsPbX₃ nanocrystals are extremely sensitive to polar solvents, such as water, which seriously limits their application potential, especially for biological applications.^[25, 151] The photo- and thermo-induced degradation of the CsPbX₃ nanocrystals is found to be closely related to the damage of labile surface ligands and the ensuing particle aggregation or fusion at elevated temperatures, which leads to the loss of quantum confinement and formation of carrier traps.^[149-154] To address the above issues, a few attempts have been made, such as the surface coating, ligand treatment and introduction of host matrix.^[25, 27, 149, 151-153, 155-158] Loiudice *et al.* reported that the uniform surface coating of amorphous alumina by atomic layer deposition could greatly enhance the resistance exposed to air (for more than 45 days), to simulated solar spectrum irradiation (for more than 8h), and to thermal treatment (up to 200 °C).^[149] Palazon *et al.* applied a low flux X-ray irradiation on the CsPbX₃ nanocrystals to induce the intermolecular C=C bonding of the organic ligands. The carbonized surface ligands after X-ray treatment serve as the effective protection layer coating on the surface of the nanocrystals, such that the nanocrystal films exhibit great robustness against air, moisture and even

water.^[155] Besides, several groups have demonstrated the decoration of CsPbX₃ nanocrystals onto silica spheres and incorporation of the CsPbX₃ nanocrystals into polymer and semiconductor with larger bandgap matrix could block the aggregation of the nanocrystals, hence improving the stability.^[27, 151-153] Despite that impressive progress has been made in the past two years, it is still challenging to achieve high stability of the CsPbX₃ nanocrystals while preserve the superior optoelectronic properties. Moreover, most of the efforts are only focused on the green-emitting CsPbBr₃ nanocrystals, there is less progress on enhancing the stability of other halide perovskite nanocrystals. Especially, the red-emitting CsPbI₃ nanocrystals are found to be metastable, known as the “red wall”.^[16, 59] However, until now, robust red-emitting pure iodine all-inorganic perovskite nanocrystals are still unavailable.

5.2 Toxicity

The toxicity of lead is a concern for the environment and health safety, which may stifle the application commercialization of CsPbX₃ nanocrystals. In this regard, several recent works have been devoted to developing lead-free and less toxic metal halide perovskite nanocrystals as well as their derivatives, exemplified by the so-called double perovskites.^[159-163] Enlightened by the previous achievements in exploring bulk lead-free all-inorganic perovskites, colloidal metal halide nanocrystals containing tin (Sn), bismuth (Bi), germanium (Ge), indium (In), and antimony (Sb) may be favorable alternative candidates.^[159, 161-163] Jellicoe *et al.* fabricated the CsSnX₃ nanocrystals by the hot injection method and demonstrated the emission color tunability by both quantum confinement effects and composition control.^[153] Notably, due to the lower bandgap of bulk CsSnI₃, the emission spectra of the CsSnI₃ nanocrystals could be extended to as long as ~950 nm, which holds promise for applications in the near-infrared region. However, these CsSnX₃ nanocrystals show much lower PLQY (< 1%) than the lead analogues, indicating the poor optical

properties. Even worse, it turns out that the CsSnX_3 nanocrystals are more sensitive to the air due to the easy oxidation of Sn^{2+} , which may partially explain the low PLQY. Later, Wang *et al.* directly made use of the tetravalent tin (Sn^{4+}) instead of Sn^{2+} to produce the Cs_2SnI_6 nanocrystals.^[156] Although the synthesized Cs_2SnI_6 nanocrystals show much improved stability, they still suffer from the poor PLQY (<1%), making them unfavorable for light-emitting applications. Until now, although several reports have demonstrated the success in fabrication of the lead-free perovskite nanocrystals, none of nanocrystals could show satisfactory optical properties that matches those of the lead halide perovskites.^[159] Hence, there is a plenty of space in pursuing the high-quality lead-free perovskite nanocrystals, which requires the combined efforts from the theory, fabrication and characterization.

6. Summary and Outlook

We present the major contributions on the development of all-inorganic metal halide perovskite nanostructures. We have briefly introduced the representative fabrication methods of the perovskite nanocrystals including the size, composition and morphology control. The review is mainly focused on the photophysics and corresponding light-emitting applications of the perovskite nanocrystals, which include the electronic structures, steady-state and transient absorption and emission, nonlinear optical properties, exotic photophysical phenomena, lasers and LEDs. In spite of the remarkable achievements obtained in the research of the perovskite nanostructures, there is still a wealthy of open questions to be answered and challenges to be addressed.

Noticeably, so far, the overwhelmingly employed core-shell structure in metal-chalcogenide quantum dots is not applicable for halide perovskite nanocrystals due to the spontaneous anion-exchange reactions. The infeasibility of the core-shell heterostructure greatly limits the

optimization of the perovskite nanocrystals. On one hand, the coating of inorganic protective shell will significantly enhance the stability of the colloidal nanocrystals, which shall be the ideal approach toward robust perovskite nanocrystals as has been validated in metal-chalcogenide quantum dots.^[164] On the other hand, the core-shell heterostructure engineering has been demonstrated to be an effective strategy to suppress the Auger loss in colloidal nanocrystals.^[120] Due to the strong biexciton Coulombic interaction in the perovskite nanocrystals, the Auger lifetime is determined to be as short as several ten of picoseconds. Therefore, the fast Auger recombination will inevitably plague the optical performance under high excitation intensities or electrical pumping. Inspired by the previous lessons, the adoption of core-shell heterostructure may radically circumvent the Auger disaster, which will definitely improve the performance of perovskite nanocrystals in high flux applications such as lasing and solid state lighting.^[164]

Moreover, several other kinds of ternary lead halide compound are found to frequently co-exist with CsPbBr_3 , such as the CsPb_2Br_5 and Cs_4PbBr_6 , which has led to false understanding of the luminescent source in the composites.^[23, 165-170] For example, some works reported the bright green emission from Cs_4PbBr_6 ,^[165-167] however, Cs_4PbBr_6 is a wide bandgap semiconductor (~ 3.8 eV) or insulator whose emission locates at UV range (~ 375 nm).^[171] Similarly, CsPb_2Br_5 have been claimed to be highly luminescent material by several groups, while some other reports demonstrated it to be an indirect semiconductor with poor emission.^[23, 169, 170] Without doubt, further comprehensive studies are required to reach a consensus. Moreover, one may make full use of the flexibility of the perovskite phases to optimize the optical and device performance relying on the insightful understanding of the perovskite compound.^[154, 168]

Besides, the toxicity issue has to be solved before commercialization application of the perovskite nanostructures. Although lead-free alternatives have been explored, as a matter of fact,

none of them could exhibit comparable optical properties to those of the lead ones.^[159] As a result, further efforts should be devoted to figuring out the peculiarity of lead-based perovskites and, thereafter, rationally look for the substitutes.

In terms of the light-emitting applications, the performance of the inorganic perovskite nanocrystal LEDs including the EQE, current efficiency and device operational lifetime is far inferior to the state-of-the-art CdSe-based analogues at present.^[172] Advance of the perovskite LED is still an urgent issue, which relies on the optimization from both the device configuration and the active materials.^[173, 174] As for the inorganic perovskite nanocrystal lasers, although the pump thresholds reported are much lower than those of the CdSe ones, the demonstration of continuous wave (CW) operation is still lacking.^[28, 30] As a consequence, there is a long way to go toward the goal of electrically driven lasing.

In summary, we believe that the all-inorganic metal halide perovskite nanostructures are emerging as a new research paradigm and will compete favorably with both conventional metal-chalcogenide nanocrystals and organic-inorganic hybrid halide perovskites in light-emitting applications and hold new opportunity for optoelectronics in the near future.

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Figure 1. a) Photograph of CsPbX₃ nanocrystals with different emission colors under UV irradiation. b) Photograph of highly luminescent CsPbX₃ nanocrystals-PMMA polymer monoliths under UV irradiation. The inset shows the crystal structure of cubic CsPbX₃. Adapted with permission.^[16] Copyright 2015, American Chemical Society.

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Figure 3. a-c) Impact of reaction temperature on the morphology of CsPbBr₃ nanocrystals: (a) green emitting 8–10 nm nanocubes are formed at 150°C. (b) cyan emitting nanoplatelets with lateral dimensions of 20 nm are formed at 130 °C. (c) blue emitting thin nanoplatelets are observed at 90°C. Scale bar: 50 nm. Reproduced with permission.^[44] Copyright 2015, American Chemical Society. d) Sketch of the variation of the concentration of the perovskite nanostructures along with the reaction time. Reproduced with permission.^[48] Copyright 2015, American Chemical Society.

Figure 4. a) Calculated electronic band structures for cubic phase CsPbCl_3 , CsPbBr_3 , CsPbI_3 , respectively. Reproduced with permission.^[16] Copyright 2015, American Chemical Society. b) Density of states of the cubic CsPbBr_3 with elemental contributions to energy band. c) [Illustration of valence band maximum of \$\text{CsPbBr}_3\$](#) . d) [Illustration of conduction band minimum for \$\text{CsPbBr}_3\$](#) . Reproduced with permission.^[18] Copyright 2016, Wiley-VCH.

Figure 5. a) Simplified bonding (σ)/antibonding (σ^*) orbitals in CsPbX₃ showing the formation of the valence and conduction band. Reproduced with permission.^[71] Copyright 2016, American Chemical Society. b) Schematic depiction of surface self-passivation and the schematic diagram of the Br rich surface and assumed energy levels for CsPbBr₃ nanocrystals. Reproduced with permission.^[18] Copyright 2016, Wiley-VCH. c) Illustration that CsPbX₃ nanocrystals maintain high emission efficiency with removal of surface atoms and ligands. Reproduced with permission.^[69] Copyright 2015, American Chemical Society.

Figure 6. a) Size- and composition-tunable emission spectra from CsPbX₃ nanocrystals. Reproduced with permission.^[16] Copyright 2015, American Chemical Society. b) Comparison of emission line-widths of Rhodamine 6G, CdSe/CdS-ZnS nanocrystals, and CsPbBr₃ nanocrystals. Copyright 2015, Wiley-VCH. c) PL spectra of CdSe/CdS-ZnS nanocrystals in solution and film. d) PL spectra of CsPbBr₃ nanocrystals in solution and film. Reproduced with permission.^[17] Copyright 2015, Wiley-VCH. e) Composition dependent absorption and emission spectra of CsPbX₃ nanocrystals. Reproduced with permission.^[21] Copyright 2015, American Chemical Society. f) Size dependent absorption and emission spectra from 5–12 nm CsPbBr₃ nanocrystals manifesting the quantum confinement effect. Reproduced with permission.^[16] Copyright 2015, American Chemical Society. g) Plot of emission from CsPbX₃ nanocrystals (black data points) on CIE chromaticity coordinates, showing the wide color gamut. Reproduced with permission.^[16] Copyright 2015, American Chemical Society.

Figure 7. a) Room temperature confocal scanning PL image of individual CsPbBr₃ nanocrystals. b) Second-order autocorrelation $g^{(2)}(\tau)$ of individual CsPbBr₃ nanocrystal under pulsed excitation, indicating the achievement of single photon emission. Reproduced with permission.^[76] Copyright 2015, American Chemical Society. c) Low temperature PL spectra from single CsPbBr₃ nanocrystal, resolving three emission peaks. d) Polarization dependent PL intensity of the three resolved peaks, indicating that the fine structure of CsPbBr₃ nanocrystal is related to anisotropies or defect complexes. Reproduced with permission.^[77] Copyright 2016, American Chemical Society.

Figure 8. a) Composition dependent time-resolved PL decays of CsPbX₃ nanocrystals. Adapted with permission.^[16] Copyright 2015, American Chemical Society. b) Schematic illustration of thiocyanate salt treatment enabled trap-free CsPbBr₃ nanocrystals. The upper figure shows the PL decay of CsPbBr₃ nanocrystals before and after thiocyanate salt treatment. Reproduced with permission.^[20] Copyright 2017, American Chemical Society. c) Pump intensity dependent PL decay curves of CsPbBr₃ nanocrystals, showing the appearance of fast Auger recombination under high excitation intensities. Adapted with permission.^[28] Copyright 2015, Wiley-VCH. d-f) Temporally and spectrally resolved photoluminescence under high pump intensity (<N>=1.74) of CsPbBr₃ nanocrystals with different sizes or absorption cross-sections. g) Temporally and spectrally resolved photoluminescence under low pump intensity (<N>=0.07). h) Extraction of

biexciton binding energy according to the difference between the high and low excitation density at the short delay spectra (~ 100 ps) (shaded area). i) Size dependent biexciton binding energies of CsPbBr_3 and CsPbI_3 as well as conventional CdSe nanocrystals. Reproduced with permission.^[90] Copyright 2016, American Chemical Society.

Figure 9. a) Transient absorption spectra and kinetics of CsPbBr₃ nanocrystals under excitation

wavelength of 400 nm. b) Kinetics of photobleaching band and photoinduced absorption band as well as PL dynamics of the CsPbBr₃ nanocrystals. Adapted with permission.^[88] Copyright 2015, American Chemical Society. c) Transient absorption of CsPbBr₃ nanocrystals under excitation energy of the first excitonic band and (d) higher energy state ($\sim 1.26 E_g$) with short time delays (< 10 ps). e) and h) Schematic illustration of transient absorption evolution during the hot-carrier relaxation process. Reproduced with permission.^[93] Copyright 2017, Wiley-VCH.

Figure 10. a-c) Color-coded contour images of transient absorption signal (top) and early time transient absorption dynamics detected at the positions of photoinduced absorption and bleach features (bottom) for CsPbBr₃, CsPbBr_{1.5}I_{1.5}, and CsPbI₃ nanocrystals, respectively. Reproduced with permission.^[93] Copyright 2017, Wiley-VCH.

Figure 11. a) Transient absorption spectra and kinetics of CsPbBr₃ nanocrystals under excitation wavelength of 400 nm and varied excitation densities. With the increased of excitation density, photobleaching band shows a carrier-density-dependent blueshift and broadens, which originate from charge carrier accumulation at the band edges and can be explained on the basis of a dynamic Burstein–Moss shift. Reproduced with permission.^[88] Copyright 2015, American Chemical Society.

Figure 12. a) PL spectra from CsPbBr₃ nanocrystals under varied excitation intensities. The inset shows the photograph of solution of CsPbBr₃ nanocrystals under excitation wavelength of 800 nm. b) Plot of spectrally integrated PL intensity with respect to the excitation intensity. c) Open aperture Z-scan curves of CsPbBr₃ nanocrystal solution and the pure solvent. The red curve is the best-fitting curve on basis of Z-scan theory. d) PL dynamics of CsPbBr₃ nanocrystals induced by one-, two- and three-photon absorption. The inset depicts the absorption and emission process induced by one-, two- and three-photon excitation. Reproduced with permission.^[29] Copyright 2016, American Chemical Society.

Figure 13. a) Pressure-dependent PL spectra of CsPbBr₃ nanocrystals. The inset presents the schematic illustration of the diamond anvil cell in the measurements. Right panel displays the microimages of the sample under pressures of 1 atm, 0.49, 0.78, and 1.21 GPa. b) Schematic illustrations in polyhedral views of CsPbBr₃ crystal structures under high pressure. Reproduced with permission.^[107] Copyright 2017, American Chemical Society. c) PL spectra of 3-monolayer CsPbBr₃ nanoplatelet film under varied irradiation time (ML: monolayer). d) Schematic depiction of the photon driven transformation process. e) Bright field optical image taken by the confocal laser scanning microscopy. f) PL image recorded in range of 420-470 nm. g) PL image recorded in range of 500-560 nm. Scale bar: 50 μ m. Reproduced with permission.^[109] Copyright 2016, Wiley-VCH.

Figure 14. a) PL spectra from CsPbBr₃ nanocrystal thin film under varied pump intensities. The inset presents pump and detection configuration (excitation beam (EB), cylindrical lens (CL), collection lens set (CLS), charged coupled device (CCD)). Reproduced with permission.^[28] Copyright 2015, Wiley-VCH. b) Composition tunable stimulated emission spectra from CsPbX₃ nanocrystals. Reproduced with permission.^[30] Copyright 2015, Nature Publication Group. c) PL intensity and FWHM variation of CsPbBr₃ nanocrystal thin film under varied pump intensities. d) Plot of stimulated emission peak intensity with respect to laser shot. The inset shows the initial stimulated emission spectrum. Reproduced with permission.^[28] Copyright 2015, Wiley-VCH. e) Determination of modal net gain for the CsPbBr₃ nanocrystal film by variable stripe-length method. Reproduced with permission.^[30] Copyright 2015, Nature Publication Group.

Figure 15. a) Whispering gallery mode lasing emission by infiltrating CsPbBr₃ nanocrystals into a hollow capillary tube. Reproduced with permission.^[28] Copyright 2015, Wiley-VCH. b) Whispering gallery mode lasing emission by coating the CsPbBr₃ nanocrystals onto silica microsphere. Reproduced with permission.^[30] Copyright 2015, Nature Publication Group. c) Vertical cavity surface emitting lasers by sandwiching CsPbBr₃ nanocrystals between two distributed Bragg mirrors. Copyright 2017, Wiley-VCH. d) Photograph of blue and red vertical cavity surface emitting lasers from CsPbX₃ nanocrystals. Reproduced with permission.^[31] Copyright 2017, Wiley-VCH.

Figure 16. a) Dark-field image of a CsPbBr₃ nanowire. Reproduced with permission.^[50] Copyright 2016, National Academy of Sciences. b-d) PL images of the nanowire under increased excitation fluence. Scale bar: 2 μm . Reproduced with permission.^[50] Copyright 2016, National Academy of Sciences. e) Pump intensity dependent PL spectra from the CsPbBr₃ nanowire, indicating the development of lasing action. Reproduced with permission.^[50] Copyright 2016, National Academy of Sciences. f) Composition tunable broadband lasing spectra from single-crystal nanowires of CsPbX₃ (X = Cl, Br, and I). Reproduced with permission.^[127] Copyright 2016, American Chemical Society.

Figure 17. a) Schematic illustration of multilayer stacking perovskite nanocrystal-LED device. Reproduced with permission.^[26] Copyright 2015, Wiley-VCH. b) Cross-sectional view showing the multiple layers of material with detectable contrast. Scale bar: 50 nm. c) Plot of current density and luminance as a function of applied voltage for the LED device. d) EL spectrum under driving voltage of 5.5 V. The inset shows the photograph of a LED in operation. Reproduced with permission.^[26] Copyright 2015, Wiley-VCH. e-f) Photographs of perovskite nanocrystal-LED devices with the Nanjing University of Science and Technology (NUST) logo. Reproduced with permission.^[26] Copyright 2015, Wiley-VCH.

Figure 18. a) Schematic depiction of the surface ligand density control of CsPbBr₃ nanocrystals and the corresponding changes of ink stability, PLQY and carrier injection. Reproduced with permission.^[144] Copyright 2017, Wiley-VCH. b) Normalized EL spectra and PL spectra of CsPbBr₃ nanocrystal-LED. The inset displays a photograph of the LED device in operation. c) External quantum efficiency of CsPbBr₃ nanocrystal-LEDs under a number of ligand purification cycles (one-three) with respect to luminance. Reproduced with permission.^[144] Copyright 2017, Wiley-VCH. d) Cross-sectional SEM image of the p-MgNiO/CsPbBr₃/n-MgZnO/n⁺-GaN LED. (d) Simplified energy band alignment of the multilayered LED manifesting the conduction and valence band energy levels with respect to the vacuum level. Reproduced with permission.^[145] Copyright 2016, American Chemical Society.

Table 1. Phase transition points of CsPbX₃ bulk crystals^[33]

Materials	Cubic phase	Tetragonal phase	Orthorhombic phase
CsPbI ₃	>578 K		
CsPbBr ₃	>403 K	373-403 K	<373 K
CsPbCl ₃	>320 K	315-320 K	<315 K

Table 2. Stimulated emission (SE) and lasing characteristics from CsPbX₃ nanostructures

Materials	Pump mechanism	SE Threshold	SE Wavelength	Temperature	Laser configuration	Lasing threshold	Pump Source
CsPbBr ₃ microcrystalline film ^[116]	One-photon	100 kW cm ⁻²	~537	295 K			337.1 nm, 0.5 ns
CsPbX ₃ nanocrystal film ^[28, 31]	One-photon	22 μJ cm ⁻²	~470-620 nm	293 K	VCSEL, WGM	9 μJ cm ⁻²	400 nm, 100 fs
CsPbX ₃ nanocrystal film ^[30]	One-photon	5-22 μJ cm ⁻²	~470-640 nm	293 K	WGM, Random lasing		400 nm, 100 fs
CsPbX ₃ nanocrystal film ^[29, 32]	Two-photon	2.5 mJ cm ⁻²	~470-530 nm	293 K	WGM	0.8 mJ cm ⁻²	800 nm, 100 fs
CsPbBr ₃ nanowire ^[50]	One-photon		~532 nm	299 K	Fabry–Pérot	5 μJ cm ⁻²	400 nm, 150 fs
CsPbBr ₃ nanorod ^[126]	Two-photon		~539 nm	295 K	Fabry–Pérot	0.63 mJ cm ⁻²	800 nm, 80 fs
CsPbX ₃ nanoplatelet ^[66]	One-photon		~470-700 nm	293 K	WGM	~2 μJ cm ⁻²	400 nm, 50 fs
CsPbBr ₂ Cl nanoplatelet ^[133]	Two-photon		~510 nm	77 K	WGM	~4.8 μJ cm ⁻²	780 nm, 100 fs

Table 3. Development of CsPbX₃ nanocrystal based LEDs

All-inorganic perovskite nanocrystals	EQE (%)	Current Efficiency (CdA ⁻¹)	Power Efficiency (lmW ⁻¹)	Luminance (Cdm ⁻²)	EL Wavelength (nm)	Turn-on Voltage (V)
CsPbBr ₃ ^[26]	0.12	0.43	0.18	946	516	4.2
CsPb(Br/Cl) ₃ ^[26]	0.07	0.14	0.07	742	455	5.1
CsPb(Br/I) ₃ ^[26]	0.09	0.08	0.06	528	586	4.6
CsPbBr ₃ ^[134]	0.06	0.19		1377	516	3.5
CsPbBr ₃ ^[135]	0.19			206	523	2.8
CsPbI ₃ ^[135]	5.7			2335	698	2.2
CsPbBr _{1.5} Cl _{1.5} ^[135]	0.0074			8.7	480	3.5
CsPbBr ₃ ^[23]	2.21	8.89	3.4	3853	527	4.6
CsPbBr ₃ ^[144]	6.27	13.3	5.24	1585	512	3.4

Author biographies

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The table of contents entry: All-inorganic metal halide perovskite nanostructures (IHPNs) have sparked intense research interest by virtue of the superior optical properties and the cost-effective synthesis. Since the first report in 2015, striking development of the IHPNs has been witnessed in terms of fabrication strategy, optical characterization, and optoelectronic applications, which will be discussed in this review article.

*Yue Wang, Handong Sun**

All-inorganic metal halide perovskite nanostructures from photophysics to light-emitting applications

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