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Progress in the preparation and application of  $\text{CsPbX}_3$  perovskites

In recent years, perovskite has become a popular material of increasing interest. All-inorganic perovskites have better stability compared to organic-inorganic metal halide perovskites. Therefore, all-inorganic  $\text{CsPbX}_3$  ( $X = \text{Br}, \text{I}, \text{Cl}$ ) perovskites are becoming popular and potential materials suitable for optoelectronic devices.  $\text{CsPbX}_3$  perovskites exhibited many excellent physical and optical properties, such as high carrier mobility, high fluorescence, etc. In this review, the recent progress on the preparation and application of  $\text{CsPbX}_3$  perovskites is outlined. A brief summary of the full paper is presented and the challenges facing the all-inorganic  $\text{CsPbX}_3$  perovskites field are discussed.

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# Progress in the preparation and application of CsPbX<sub>3</sub> perovskites

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In recent years, organic–inorganic metal halide (OIH) perovskites have become a popular material of increasing interest. Due to the presence of organic cations, OIH perovskites suffer from chemical instability and easy decomposition in harsh environments with high temperature and humidity. All-inorganic perovskites have better stability compared to OIH perovskites. Due to the intensive research and continuous optimization of the preparation process, the power conversion efficiency (PCE) of all-inorganic perovskite solar cells (PSCs) has exceeded 20%. Therefore, all-inorganic CsPbX<sub>3</sub> (X = Br, I, Cl) perovskites are becoming popular and are potential materials suitable for optoelectronic devices. CsPbX<sub>3</sub> perovskites exhibit many excellent physical and optical properties, such as high carrier mobility and high fluorescence. In this review, recent progress on the preparation and application of CsPbX<sub>3</sub> perovskites is outlined. Firstly, the physical structure and excellent optical properties of CsPbX<sub>3</sub> perovskites are briefly described. Next, the preparation of different morphologies of CsPbX<sub>3</sub> perovskites is described. Furthermore, this paper summarizes the application of CsPbX<sub>3</sub> in optoelectronic devices, including PSCs, light-emitting diodes (LEDs), and photodetectors (PDs). The performance parameters of CsPbX<sub>3</sub> devices are summarized. Finally, a brief summary of the full paper is presented and the challenges facing the field of all-inorganic CsPbX<sub>3</sub> perovskites are discussed.

## 1. Introduction

OIH perovskites are popular and promising materials that have aroused tremendous public attention due to their excellent photoelectric properties, such as high absorptivity, high carrier mobility, smaller exciton binding energy, and adjustable bandgap.<sup>9–13</sup> Perovskites were originally discovered by a German mineralogist Gustav Rose in 1839; the first discovered perovskite-structure substance was CaTiO<sub>3</sub>.<sup>14,15</sup> OIH perovskites possess the general molecular formula ABX<sub>3</sub>, which is composed of three ions occupying different positions of the crystal lattice. The A site is a monovalent organic cation that has a larger ionic radius, where A = methylammonium (CH<sub>3</sub>NH<sub>3</sub><sup>+</sup>, MA<sup>+</sup>) or formamidinium (CH<sub>3</sub>(NH<sub>2</sub>)<sub>2</sub><sup>+</sup>, FA<sup>+</sup>).<sup>22</sup> The B site is a metal cation whose ionic radius is smaller than that of A, where B = Pb<sup>2+</sup>, Sn<sup>2+</sup> or Ge<sup>2+</sup>.<sup>24</sup> The X site is a halide anion, where X = Cl<sup>-</sup>, Br<sup>-</sup> or I<sup>-</sup>.

Fig. 1a shows the crystal structure of the perovskite CaTiO<sub>3</sub> and Fig. 1b shows the crystal structure of OIH perovskites. The A ion occupies the octahedral void and is located in the centre of the cubic cell. The B metal cations are located at the apex of the cubic cell and are surrounded by six halogen anions. Thus, the perovskite has excellent photoelectric properties because of its unique physical structure.

Although OIH perovskites have many outstanding properties, their stability issues greatly limit their application. For instance, they are very sensitive to harsh environments. Due to the

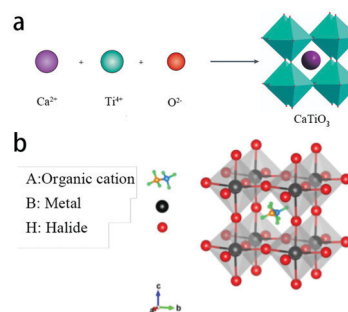


Fig. 1 (a) The crystal structure of the perovskite CaTiO<sub>3</sub>.<sup>34</sup> Copyright 2017, Springer Nature. (b) The crystal structure of OIH perovskites.<sup>36</sup> Copyright 2020, Walter de Gruyter.

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instability of organic cations in OIH perovskites, they decompose easily at high humidity and temperature.<sup>19,24</sup> Xue *et al.* studied the structural properties during the decomposition of MAPbI<sub>3</sub> (MA = CH<sub>3</sub>NH<sub>3</sub><sup>+</sup>) by first-principles calculations.<sup>30</sup> The intrinsically poor structural stability of MAPbI<sub>3</sub> can cause it to decompose into PbI<sub>2</sub>, whether in high humidity or not. In addition, high temperature will cause the perovskite phase transition, which results in instability of the perovskite. For instance, the black perovskite-type ( $\alpha$ -phase) of FAPbI<sub>3</sub> is stable at high temperatures above 160 °C, but it turns into the yellow  $\delta$ -phase when exposed to low temperatures and moisture.<sup>40</sup> Moreover, Schwenzer *et al.* investigated the effects of different cation compositions on the thermal stability of perovskite thin films; the surface material composition of FACsPbI<sub>3</sub> films was less affected than that of FAPbI<sub>3</sub> films after 500 h at 85 °C.<sup>41</sup> The poor thermal and moisture stability of organic–inorganic hybrid perovskite materials is caused by the facile phase transitions and the formation of hydrated phases near room temperature.<sup>43</sup> Kulbak *et al.* explored the thermal stability of MAPbBr<sub>3</sub> and CsPbBr<sub>3</sub> by thermogravimetric analysis.<sup>44</sup> MAPbBr<sub>3</sub> began to decompose at 220 °C, while CsPbBr<sub>3</sub> began to decompose at 580 °C; this indicates that all-inorganic perovskites have higher thermal stability than OIH perovskites. Therefore, it is known that all-inorganic perovskites are more stable than OIH perovskites.<sup>45</sup> All-inorganic perovskite nanocrystals (PNCs) have advantages of high photoluminescence quantum yield (PLQY), narrow band emission, and adjustable fluorescence wavelength.<sup>46</sup>

In recent years, increasing numbers of scholars have focused on all-inorganic perovskite materials owing to their unique advantages. The A site in the perovskite general formula can be replaced by an inorganic cation. In a perovskite structure, its structure and stability are determined by the tolerance factor  $t$  and octahedral factor  $\mu$ .<sup>47</sup> In DFT calculations at 0 K, the  $\gamma$  phase is always the most stable, and the most ideal perovskite structure is the  $\alpha$  phase, which has a tolerance factor of  $t = 1$ ; however, it is unstable.<sup>48</sup> When  $0.81 < t < 1.11$  and  $0.44 < \mu < 0.90$ , the crystal structure of perovskite is stable.<sup>11</sup> When  $t < 0.89$ , perovskite is a tetragonal ( $\beta$ -phase) or orthogonal ( $\gamma$ -phase) structure with poor symmetry. The Cs<sup>+</sup> ion can match the requirements of  $t$ , and it has replaced organic cations to improve the stability of perovskite in applications.

According to the citation report of American Chemical Society (ACS) Publications, we can conclude that the publications of CsPbX<sub>3</sub> have shown an upward trend in the last 10 years. Moreover, the hot injection method was the first to emerge, and in recent years, the new electrospinning method has gradually arisen (Fig. 2). In addition, based on the hot injection method, the PCE of CsPbX<sub>3</sub>-based PSCs has improved from 5.8% (CsPbBr<sub>3</sub>) and 2.9% (CsPbI<sub>3</sub>) to 17.1% (CsPbI<sub>3</sub>) from the first paper in 2015 to date, and the stability has been continuously improved.<sup>49</sup> Tong *et al.* improved the hot injection method, such as adding zeolite-encapsulated CsPbX<sub>3</sub> NCs, doping with Mn<sup>2+</sup>, and *in situ* growth of CsPbX<sub>3</sub> NCs in metal-organic framework crystals, which improved the luminescence efficiency of the materials.<sup>7,17,28,50–57</sup> Zhang *et al.* prepared dense and uniform CsPbX<sub>3</sub> thin films by thermal evaporation and applied them in lasers, LEDs, PSCs, and other photoelectric fields.<sup>1,16,35,58–64</sup> In recent years, electrospinning has

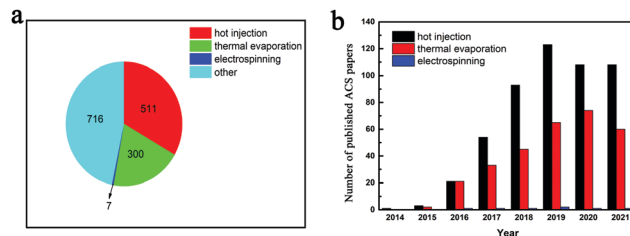


Fig. 2 (a) Number of articles on the three preparation methods of CsPbX<sub>3</sub> as percentages of core publications. (b) Statistics of core publications based on CsPbX<sub>3</sub> preparation methods.

emerged as a new method to prepare CsPbX<sub>3</sub>. Huang *et al.* encapsulated CsPbX<sub>3</sub> NCs in different polymers and prepared CsPbX<sub>3</sub> nanofibers by electrospinning.<sup>5,19,46,65–70</sup>

Here, in this review, we will describe the latest progress of CsPbX<sub>3</sub> perovskites and their applications in optoelectronic devices. The synthesis of CsPbX<sub>3</sub> perovskites in different morphologies, including single crystals, thin films and nanocrystals, are first described. Then, the optoelectronic applications of CsPbX<sub>3</sub> perovskite are reviewed, including PSCs, LEDs, and PDs. Finally, the challenges and prospects of the CsPbX<sub>3</sub> perovskites are discussed.

## 2. Synthesis of CsPbX<sub>3</sub> perovskites

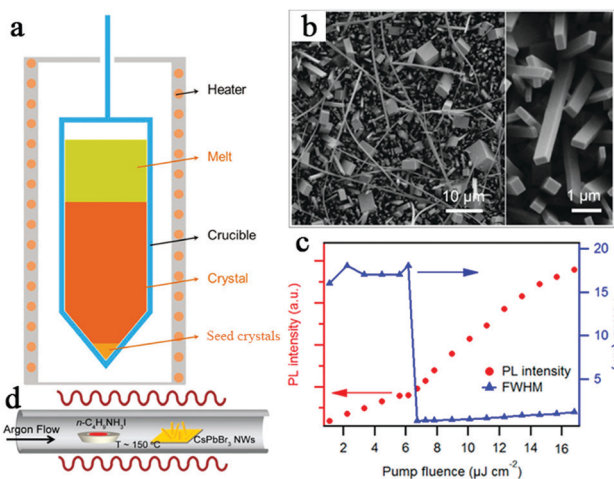
To explore the characteristics of CsPbX<sub>3</sub> perovskites and the application of different optoelectronic devices, various synthesis strategies of CsPbX<sub>3</sub> perovskites in different morphologies were summarized, such as single crystals, thin films and nanocrystals.

### 2.1 Synthesis of single crystals

Single crystals of all-inorganic perovskites are typically prepared by the Bridgman growth method. Song *et al.* obtained ultralarge 25 cm<sup>3</sup>-volume CsPbBr<sub>3</sub> SC ingot through an improved Bridgman growth method using a four-zone furnace.<sup>6</sup> The CsPbBr<sub>3</sub> SC was prepared by the reaction of equimolar amounts of CsBr (42.6 g) and PbBr<sub>2</sub> (73.4 g) in fused silica ampoules. A schematic of the growth is shown in Fig. 3a.

In addition to the traditional Bridgman growth method, the solution growth method has been developed to obtain high-quality perovskite single crystals. Fu *et al.* prepared solution-grown CsPbBr<sub>3</sub>, CsPbCl<sub>3</sub> and their halide alloys of CsPb(Br,Cl)<sub>3</sub> with high-quality single crystal nanostructures.<sup>18</sup> Nanoplates and nanocrystals were prepared by reacting PbX<sub>2</sub> and lead acetate films with CsBr (CsCl) solutions with methanol at room temperature. Fig. 3b displays the SEM image of CsPbBr<sub>3</sub> NWs grown at room temperature for approximately 15 hours. As shown in Fig. 3c, the photoluminescence (PL) spectrum of CsPbBr<sub>3</sub> NW peaked at 530 nm, with a FWHM of 17 nm. Since the direct synthesis of CsPb(Br,I)<sub>3</sub> NWs by the solution method is challenging, the low temperature vapor-phase method was developed. As shown in Fig. 3d, CsPbBr<sub>3</sub> NWs were successfully converted to CsPbI<sub>3</sub> by a low temperature vapor-phase halide exchange process, maintaining the NW morphology and perovskite lattice.





**Fig. 3** (a) Schematic diagram of the growth of a crystal by the Bridgman method.<sup>6</sup> Copyright 2017, John Wiley and Sons Inc. (b) Scanning electron microscope (SEM) image of the CsPbBr<sub>3</sub> nanostructure. The inset on the right is a magnified SEM image.<sup>18</sup> (c) Integrated emission intensity and full width at half maximum (FWHM) of the dominant emission peak plotted as a function of P, which shows that the lasing threshold is 6.2 μJ cm<sup>-2</sup>.<sup>18</sup> (d) Schematic diagram of the vapour conversion of CsPbBr<sub>3</sub> nanostructures into CsPb(Br,I)<sub>3</sub>.<sup>18</sup> Copyright 2016, American Chemical Society.

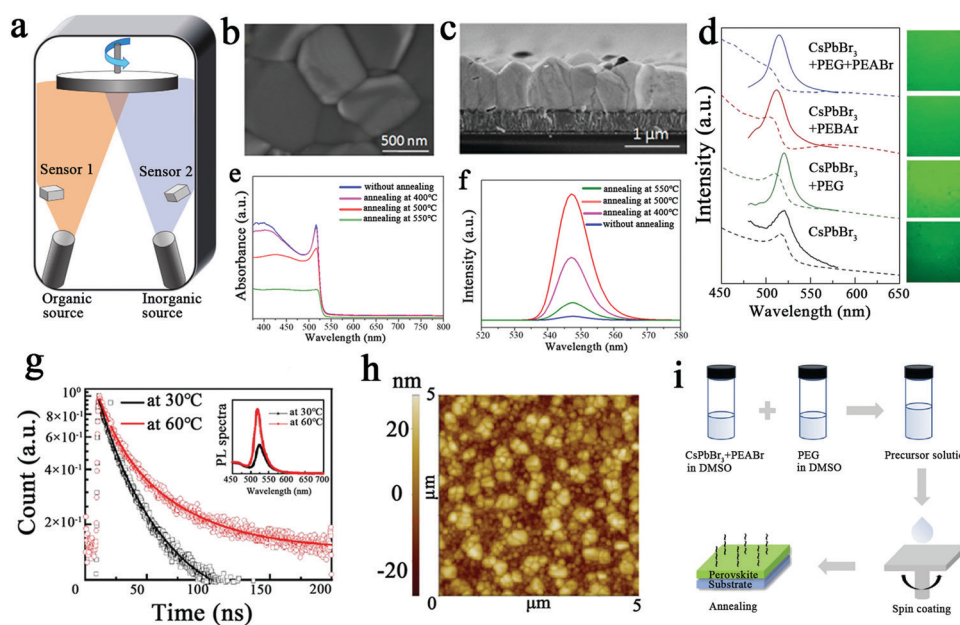
## 2.2 Preparation of thin films

Typically, perovskite films can be prepared by the solution method or by thermal evaporation. The thin films prepared by traditional thermal evaporation are compact, uniform and

reproducible, and this method can be used to prepare large-area uniform perovskite thin films.<sup>1,60</sup> Thermal evaporation mainly refers to vapor deposition; H. J. Snaith's group at the University of Oxford prepared planar heterojunction perovskite solar cells with 15% PCE using the vapor deposition method for the first time.<sup>1</sup> Fig. 4a illustrates the process of the preparation of CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3-x</sub>Cl<sub>x</sub> thin films by dual-source vapor deposition, in which organic source methylammonium iodide and inorganic source PbI<sub>2</sub> were used for dual-source thermal evaporation.

Liu *et al.* prepared CsPbBr<sub>3</sub> perovskite films by dual-source co-evaporation of CsBr and PbBr<sub>2</sub> under vacuum conditions.<sup>16</sup> When the substrate temperature was 300 °C and the evaporation rate ratio was 0.7 : 1, the best crystallinity and optimum stoichiometry of the CsPbBr<sub>3</sub> films were displayed. In addition, the CsPbBr<sub>3</sub> film films annealed at 500 °C had the largest grains, and they were tightly connected, as shown in Fig. 4b and c. This shows that photocarriers can travel to opposite electrodes without crossing through grain boundaries, which yields excellent photovoltaic performance. Fig. 4e and f shows the UV-Vis absorbance spectra of the deposited CsPbBr<sub>3</sub> films under different annealing temperatures, and the PL peak of all the films was 520 nm. On this basis, stable planar CsPbBr<sub>3</sub> solar cells were prepared with an efficiency of up to 6.95%. This finding suggests that dual-source thermal co-evaporation is an effective method for preparing CsPbBr<sub>3</sub> films with excellent properties.

Yang *et al.* prepared CsPbBr<sub>3</sub> perovskite films by thermal co-evaporation combined with continuous low-temperature thermal annealing and studied their morphology and crystallization



**Fig. 4** (a) Schematic diagram of the dual-source thermal evaporation system: the organic source is methylammonium iodide and the inorganic source is PbI<sub>2</sub>.<sup>1</sup> Copyright 2013, Springer Nature. (b) Surface SEM images of deposited CsPbBr<sub>3</sub> films under annealing at 500 °C.<sup>16</sup> (c) Cross-sectional SEM images of deposited CsPbBr<sub>3</sub> films under annealing at 500 °C.<sup>16</sup> (d) Absorption (dashed line) and PL (solid line) spectra of CsPbBr<sub>3</sub> films on a quartz substrate.<sup>26</sup> (e) UV-Vis absorption spectra of deposited CsPbBr<sub>3</sub> films under different annealing temperatures.<sup>16</sup> (f) PL spectra of deposited CsPbBr<sub>3</sub> films under different annealing temperatures.<sup>16</sup> Copyright 2018, Elsevier BV. (g) Time-resolved photoluminescence (TRPL) and steady-state photoluminescence spectroscopy (inset).<sup>35</sup> (h) AFM images of 100 nm thick CsPbBr<sub>3</sub> films prepared at 60 °C with deposition rates of 1.0 Å s<sup>-1</sup>.<sup>35</sup> Copyright 2020, American Chemical Society. (i) Preparation process of CsPbBr<sub>3</sub> perovskite thin films.<sup>26</sup> Copyright 2019, John Wiley and Sons Inc.



processes.<sup>35</sup> Dense and uniform CsPbBr<sub>3</sub> grains with an average size of 150 ± 74 nm were formed at a standard deposition rate of 1.0 Å s<sup>-1</sup> at 60 °C. As shown in the inset of Fig. 4g, the steady-state photoluminescence spectra of CsPbBr<sub>3</sub> perovskite films prepared at 60 °C is superior to that of films prepared at 30 °C. When the deposition rate is 1.0 Å s<sup>-1</sup>, a CsPbBr<sub>3</sub> film with a smoother surface can be seen under atomic force microscopy (AFM) in Fig. 4h. This article revealed for the first time the regulation of grain size and densification of CsPbBr<sub>3</sub> films during thermal co-evaporation fabrication by *in situ* thermal dynamic crystallization.

The general solution method for the preparation of perovskite films is a one-step or two-step spin coating method. Due to the morphological defects and serious non-radiative recombination of all-inorganic perovskite, the efficiency of the device is seriously affected. Tang *et al.* doped two additives, PEABr and polyethylene glycol (PEG), to prepare CsPbBr<sub>3</sub> perovskite films using the spin-coating method to passivate defects at the grain boundaries and thus reduce non-radiative recombination.<sup>26</sup> Fig. 4i shows the preparation process of the CsPbBr<sub>3</sub> perovskite films with the PEG and PEABr additives. Fig. 4d shows the PL spectra of various CsPbBr<sub>3</sub> perovskite films on PEDOT:PSS peaking at 520 nm. This method provides an effective strategy for preparing perovskite thin films with high density and few defects.

### 2.3 Synthesis of nanocrystals

All-inorganic perovskite nanocrystals can be synthesised by a variety of methods, the most mainstream being hot injection. The hot injection method has been widely used in the synthesis of traditional metal chalcogenide (PbS, CdS, CdSe) nanocrystals, and it is currently widely used for the preparation of quantum dots.<sup>71</sup> Based on the traditional quantum dot synthesis method, efficient luminescent CsPbX<sub>3</sub> nanocrystals (NCs) were synthesized by hot injection.<sup>72</sup> In the hot injection method, the

precursor solution was injected rapidly into the remaining solution of the synthetic compound under an inert atmosphere and a certain temperature, as can be seen in Fig. 5a.<sup>2,73</sup> For the synthesis of CsPbX<sub>3</sub> NCs, the Cs-containing precursor was first injected rapidly into the PbX<sub>2</sub> precursor solution containing the organic ligand at a certain temperature.<sup>74</sup> The precursor solution reacted in a high boiling point solvent with ligand-controlled growth to induce nucleation and obtain uniformly sized CsPbX<sub>3</sub> nanocrystals.

In 2015, Kovalenko *et al.* first prepared CsPbX<sub>3</sub> (X = Cl, Br, I and halogen mixtures) perovskite nanocrystals using the hot injection method.<sup>17</sup> They prepared CsPbX<sub>3</sub> NCs that possess 4–15 nm cubic perovskite crystals. Fig. 5b shows transmission electron microscopy (TEM) images of the CsPbBr<sub>3</sub> NCs; the size of the grains is uniformly distributed around 8 nm. As shown in Fig. 5c, the emission spectrum of the CsPbX<sub>3</sub> NCs is tunable in the entire visible spectral region of 410–700 nm by varying the ratio of the halides, and the corresponding FWHMs can be measured as 12–42 nm. CsPbX<sub>3</sub> NCs have high quantum yields of 50–90% and short radiation lifetimes of 1–29 ns, as can be seen in Fig. 5d. Fig. 5e shows the absorption and emission spectra of CsPbX<sub>3</sub>.

In recent years, CsPbX<sub>3</sub> perovskite quantum dots (PeQDs) have been embedded in intermediate structure metal-organic framework crystals (MOFs) to improve the stability of the PeQDs. In 2018, Wang's group prepared green-emitting CsPbBr<sub>3</sub> and red-emitting CsPbBr<sub>0.6</sub>I<sub>2.4</sub> PeQDs by hot injection and embedded CsPbX<sub>3</sub> PeQDs into intermediate structured zinc-based metal organic-framework (MOF-5) crystals to improve the stability of the PeQDs.<sup>7</sup> The PLQYs of the CsPbBr<sub>3</sub>/MOF-5 and CsPbBr<sub>0.6</sub>I<sub>2.4</sub>/MOF-5 composites were 52% and 56%, respectively; these were much higher than the PLQY of pure CsPbX<sub>3</sub> powder, indicating that MOF-5 improved the performance of the CsPbX<sub>3</sub> PeQDs. The average composite lifetimes of the CsPbBr<sub>3</sub>/MOF-5 and CsPbBr<sub>0.6</sub>I<sub>2.4</sub>/MOF-5

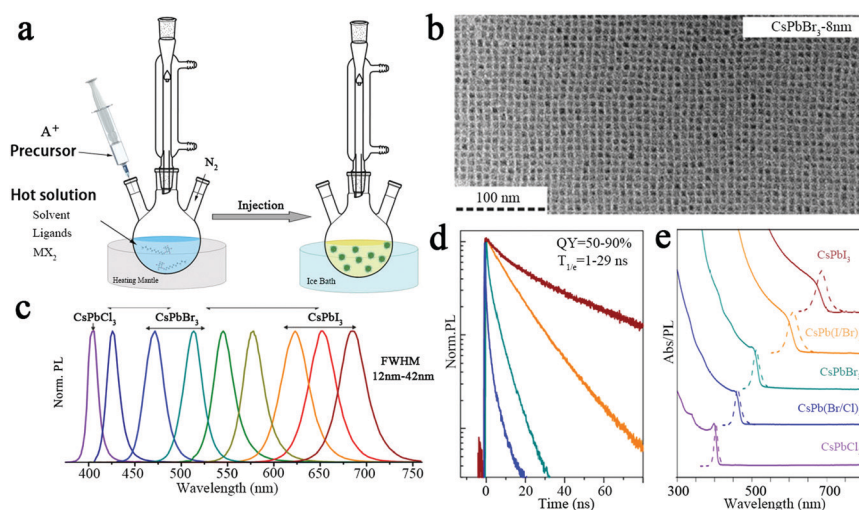
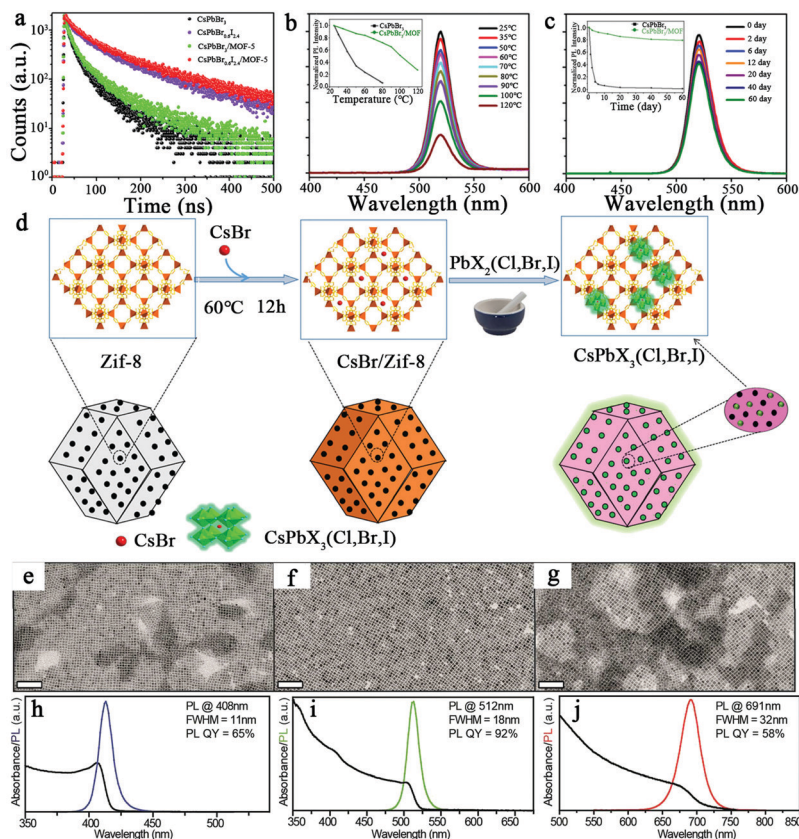


Fig. 5 (a) Schematic diagram of the hot injection method.<sup>2</sup> Copyright 2019, American Chemical Society. (b) TEM images of the CsPbBr<sub>3</sub> NCs.<sup>17</sup> (c) PL spectrum of the CsPbX<sub>3</sub> NCs.<sup>17</sup> (d) Time-resolved PL decay for all samples shown in (c) except CsPbCl<sub>3</sub>.<sup>17</sup> (e) Typical optical absorption and PL spectra.<sup>17</sup> Copyright 2015, American Chemical Society.





**Fig. 6** (a) Time-resolved photoluminescence decay curves of the CsPbBr<sub>3</sub> powder, CsPbBr<sub>0.6</sub>I<sub>2.4</sub>, CsPbBr<sub>3</sub>/MOF-5 and CsPbBr<sub>0.6</sub>I<sub>2.4</sub>/MOF-5 composites.<sup>7</sup> (b) Thermal stability test.<sup>7</sup> (c) Long-term storage stability test of CsPbBr<sub>3</sub>/MOF-5.<sup>7</sup> Copyright 2019, Elsevier. (d) Synthesis of CsPbX<sub>3</sub>/ZIF-8.<sup>25</sup> Copyright 2021, American Chemical Society. TEM images of (e) CsPbCl<sub>3</sub>, (f) CsPbBr<sub>3</sub>, and (g) CsPbI<sub>3</sub> NCs. Absorption and PL spectra of (h) CsPbCl<sub>3</sub>, (i) CsPbBr<sub>3</sub>, and (j) CsPbI<sub>3</sub> NCs dispersed in toluene.<sup>28</sup> Copyright 2018, American Chemical Society.

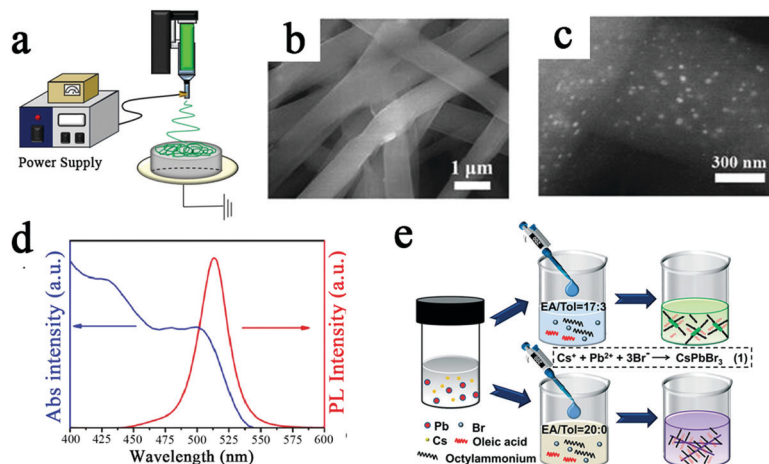
composites were longer than those of the pure CsPbX<sub>3</sub> PeQDs (Fig. 6a). CsPbBr<sub>3</sub>/MOF-5 maintained 72.8% of its initial PL intensity at 80 °C and could also emit light at 120 °C, which demonstrated the high thermal stability of CsPbBr<sub>3</sub>/MOF-5 (Fig. 6b). CsPbBr<sub>3</sub>/MOF-5 maintained 79.2% photoluminescence intensity after 60 days at room temperature, which demonstrated its good long-term storage stability (Fig. 6c). In addition, Zhao *et al.* grew CsPbX<sub>3</sub> (X = Cl, Br, and I) QDs *in situ* in zeolitic imidazolate framework-8 (ZIF-8), which improved the stability of the CsPbX<sub>3</sub> QDs.<sup>25</sup> The synthesis process of CsPbX<sub>3</sub>/ZIF-8 is shown in Fig. 6d. Furthermore, ZIF-8 can control the size of CsPbX<sub>3</sub> quantum dots. Among them, the PLQY of CsPbBr<sub>3</sub>/ZIF-8 is up to 72%.

Imran *et al.* replaced the halide precursor PbX<sub>2</sub> (X = Cl, Br or I) salt with benzoyl halide in the hot injection method, which facilitates the injection of the metal cation and the desired ligands into the solution.<sup>28</sup> This method also allows strict control of the size distribution, regulation of the CsPbX<sub>3</sub> NC components and high phase purity. Fig. 6e–g shows TEM images of CsPbX<sub>3</sub>. Fig. 6h–j shows the emission peaks of the PL spectra of CsPbX<sub>3</sub> NCs and the narrow FWHMs of the CsPbX<sub>3</sub> NCs, from 11 nm (X = Cl) to 18 nm (X = Br) to 32 nm (X = I). In addition, the PLQYs of the CsPbBr<sub>3</sub> NCs were up to 92%. The synthesized CsPbX<sub>3</sub> NCs showed high phase stability, uniform size distribution, and unprecedented optical properties.

Electrospinning has become a popular method for the preparation of nanofibers in recent years, and it has aroused public attention because of its low cost, easy fabrication process, and fine-tuning of the morphology of fibers, as well as the large specific surface area of the prepared fibers.<sup>5,75</sup> Electrospinning technology is used to prepare nanofibers by overcoming the surface tension of the jet and the internal viscous force by using electrostatic forces as the traction force.<sup>76</sup> A schematic of the electrospinning process is shown in Fig. 7a. The CsPbBr<sub>3</sub> NCs were coated with polymer by electrospinning, which improved their water and heat resistance, showing their promising application in optoelectronic devices.

Liao *et al.* investigated a general strategy that is based on the one-step electrospinning method to grow CsPbX<sub>3</sub> NC *in situ* in fibers.<sup>19</sup> The composition of CsPbX<sub>3</sub> can be finely tuned to be uniformly encapsulated in polymer (PS) fibers by reasonably adjusting the ratio of PbX<sub>2</sub> and CsX salts in the precursor solution. Fig. 7b displays a typical SEM image of the CsPbBr<sub>3</sub>@PS fiber film with 300 mg mL<sup>-1</sup> PS; the average diameter of the fibers is 800 nm. The average size of CsPbBr<sub>3</sub> NCs encapsulated in fibers is about 20 nm (Fig. 7c). Fig. 7d shows a clear absorption peak at around 500 nm and a PL emission peak at 513 nm with an FWHM of 24 nm. The PLQY values of CsPbBr<sub>3</sub>@PS remained above 70% of their original





**Fig. 7** (a) Schematic diagram of the CsPbX<sub>3</sub> (X = Cl, Br, I) nanofiber preparation process.<sup>5</sup> Copyright 2019, American Institute of Physics. (b) SEM images of the CsPbBr<sub>3</sub>@PS fibers.<sup>19</sup> (c) Typical backscattered electron SEM images of the CsPbBr<sub>3</sub>@PS fibers.<sup>19</sup> (d) UV-vis absorption and PL spectra of the CsPbBr<sub>3</sub>@PS fibers.<sup>19</sup> Copyright 2018, John Wiley and Sons Inc. (e) Schematic diagram of the process for synthesising CsPbBr<sub>3</sub> nanorods and nanowires.<sup>29</sup> Copyright 2021, Royal Society of Chemistry.

values after immersion in water for 192 h and above 50% after heating at 80 °C for 120 min. Eventually, the CsPbBr<sub>3</sub>@polymer fibers prepared by this method showed greatly improved water thermal stability and a PLQY of 48%.

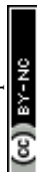
Ligand-assisted reprecipitation (LARP) is also a facile method for the synthesis of CsPbX<sub>3</sub> NCs at room temperature. Lai *et al.* applied the green solvent acetic acid instead of DMF to the LARP technique for the controlled synthesis of CsPbBr<sub>3</sub> NCs under

ambient conditions.<sup>29</sup> The workflow of LARP technology is shown in Fig. 7e. The length of the CsPbBr<sub>3</sub> NCs can be varied by changing the EA/Tol volume ratio to adjust the polarity of the antisolvent medium, enabling continuous conversion from nanorods to NWs. The prepared CsPbBr<sub>3</sub> NCs have broad photoluminescence (471–508 nm) and a high quantum yield of up to 78%.

To clearly summarize the preparation processes and optical properties of different CsPbX<sub>3</sub> materials, the recent

**Table 1** Comparison of the optical properties of CsPbX<sub>3</sub> perovskite nanocrystals with different preparation methods

Materials	Shapes	Preparation methods	PL peaks (nm)	PLQY	FWHMs (nm)	Ref.
CsPbBr <sub>3</sub>	Single crystal	Solution growth	530	—	17	18
CsPbBr <sub>3</sub>	Single crystal	Bridgman growth	527–538	—	—	6
CsPbBr <sub>3</sub>	Film	solution process	520	—	—	26
CsPbBr <sub>3</sub>	Film	Thermal evaporation	522.5	15.7%	18.5	58
CsPbBr <sub>3</sub>	Film	Thermal evaporation	525	12.2%	18	59
CsPbBr <sub>3</sub>	Film	Thermal evaporation	547	—	—	16
CsPbBr <sub>3</sub>	Film	Thermal evaporation	524	7.3%	18	62
CsPbCl <sub>3</sub>	Film	Thermal evaporation	410	—	—	64
CsPbX <sub>3</sub>	Film	Thermal evaporation	413.33–700.56	—	12.42–31.03	63
CsPbX <sub>3</sub>	Nanocrystals	Hot injection	410–700	50–90%	12–42	17
CsPbBr <sub>3</sub>	Nanocrystals	Hot injection	517	81% (in <i>n</i> -hexane)	17	7
CsPbBr <sub>0.6</sub> I <sub>2.4</sub>	Nanocrystals	Hot injection	653	85% (in <i>n</i> -hexane)	35	7
CsPbBr <sub>3</sub> /MOF-5	Nanocrystals	Hot injection	519	52%	21	7
CsPbBr <sub>0.6</sub> I <sub>2.4</sub> /MOF-5	Nanocrystals	Hot injection	655	56%	35	7
CsPbBr <sub>3</sub> /ZIF-8	Nanocrystals	Hot injection	520	72%	18	25
CsPbCl <sub>3</sub>	Nanocrystals	Hot injection	408	65%	11	28
CsPbBr <sub>3</sub>	Nanocrystals	Hot injection	512	92%	18	28
CsPbI <sub>3</sub>	Nanocrystals	Hot injection	691	58%	32	28
CsPbX <sub>3</sub> @zeolite	Nanocrystals	Hot injection	436–648	—	15.9–42	57
CsPbBr <sub>3</sub> @SBS	Nanofiber	Electrospinning	513	29%	21	5
CsPbBr <sub>3</sub> @PMMA	Nanofiber	Electrospinning	515	27%	22	5
CsPbBr <sub>3</sub> @PS	Nanofiber	Electrospinning	516	23%	23	5
CsPbX <sub>3</sub>	Nanocrystals	Electrospinning	450–660	—	18–35	46
CsPbX <sub>3</sub>	Nanofiber	Electrospinning	405–675	—	16–35	65
CsPbBr <sub>3</sub>	Nanofiber	Electrospinning	517	—	—	66
CNTs/CsPbBr <sub>3</sub> @PAN	Nanofiber	Electrospinning	511	—	20	67
CsPbBr <sub>3</sub> @PS	Fiber	Electrospinning	517	45.7%	—	68
CsPbBr <sub>3</sub> @PS	Fiber	Electrospinning	518	37%	22	70
CsPbBr <sub>3</sub>	Nanofiber	Electrospinning	521.26	—	20.19	70
CsPbBr <sub>3</sub>	Nanocrystals	LARP	471–508	78%	—	29
CsPbBr <sub>3</sub>	Nanocrystals	LARP	507	82.2%	26	81



developments of the different preparation methods and different compositions of  $\text{CsPbX}_3$  are detailed in Table 1.

### 3. Applications of $\text{CsPbX}_3$ perovskites

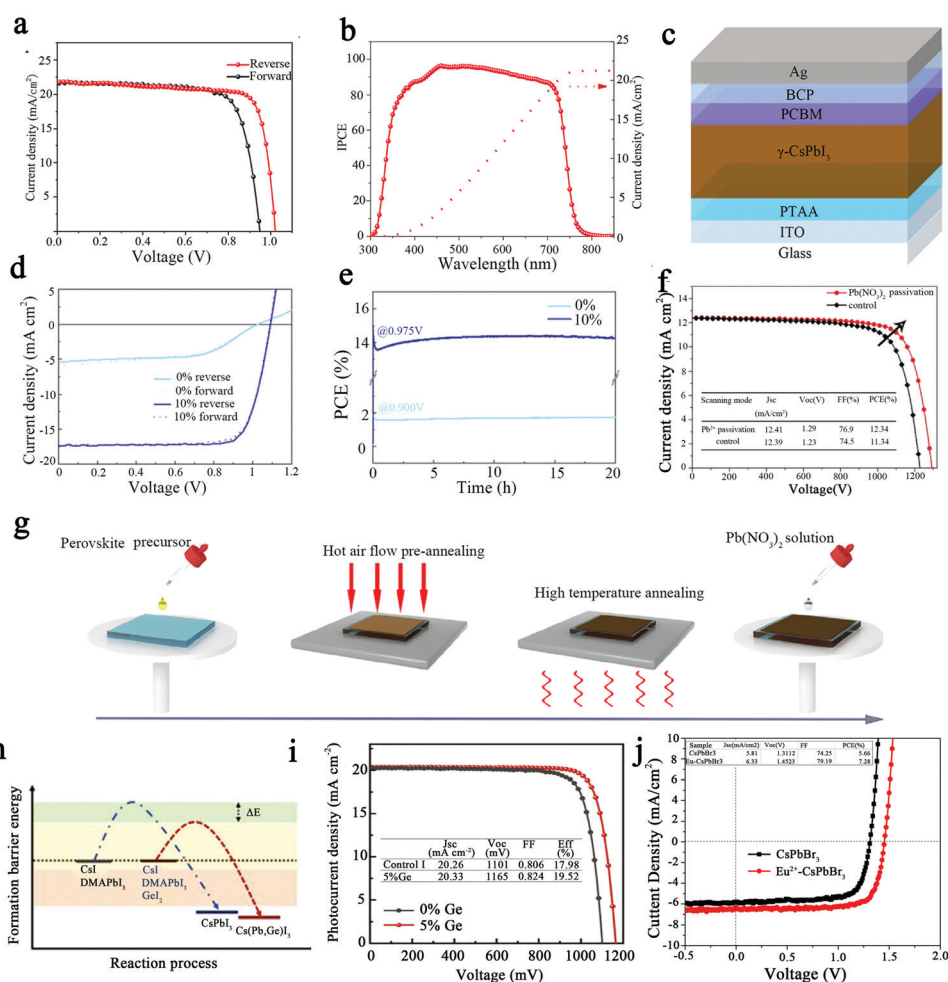
#### 3.1 Perovskite solar cells

OIH perovskites have been studied for decades and have received increasing attention in recent years for their application prospects in the photovoltaic field due to their excellent photovoltaic properties.<sup>77</sup> However, compared to OIH perovskites, all-inorganic perovskites have better thermal stability. Therefore, the application of all-inorganic chalcogenides in solar cells can yield excellent wet thermal stability.<sup>44,78</sup> Next, the application of all-inorganic  $\text{CsPbX}_3$  perovskites in the field of solar cells will be presented.

Some scholars have incorporated organic molecules into  $\text{CsPbX}_3$  PSCs to achieve efficient and stable inorganic  $\text{CsPbX}_3$  PSCs.<sup>79</sup> Recently, Zhou *et al.* incorporated inorganic 2D

Cl-terminated  $\text{Ti}_3\text{C}_2$  ( $\text{Ti}_3\text{C}_2\text{Cl}_x$ ) MXene into  $\text{CsPbBr}_3$  films; the all-inorganic  $\text{CsPbBr}_3$  PSC showed an efficiency of 11.08% and an open-circuit voltage of up to 1.702 V, which is the highest recorded in  $\text{CsPbBr}_3$  solar cells.<sup>80</sup> Shao *et al.* obtained Cs-rich  $\text{FA}_{0.15}\text{Cs}_{0.85}\text{PbI}_3$  perovskite by doping FA cations into  $\text{CsPbI}_3$  perovskite, which improved its thermal and moisture stability.<sup>4</sup> The  $\text{FA}_{0.15}\text{Cs}_{0.85}\text{PbI}_3$  films were used as the active layer to prepare solar cells with FTO/compact- $\text{TiO}_2$ /mesoporous- $\text{TiO}_2$ /perovskite/Spiro-OMeTAD/Au. As shown in Fig. 8a, the  $J$ - $V$  curve demonstrated a  $V_{\text{OC}}$  of 1.02 V,  $J_{\text{SC}}$  of 21.4  $\text{mA cm}^{-2}$ , and fill factor of 80.0%. The external quantum efficiency (EQE) of the  $\text{FA}_{0.15}\text{Cs}_{0.85}\text{PbI}_3$  solar cell is displayed in Fig. 8b. The  $\text{FA}_{0.15}\text{Cs}_{0.85}\text{PbI}_3$  perovskite solar cell achieved a high PCE of 17.5%.

In the second section of this paper, the preparation of  $\text{CsPbX}_3$  perovskite by thermal evaporation is presented, and the solution-processed devices in the field of photovoltaics usually are significantly better than those fabricated by thermal



**Fig. 8** (a)  $J$ - $V$  curve of  $\text{FA}_{0.15}\text{Cs}_{0.85}\text{PbI}_3$  solar cells.<sup>4</sup> (b) EQE curves of  $\text{FA}_{0.15}\text{Cs}_{0.85}\text{PbI}_3$  solar cells.<sup>4</sup> Copyright 2020, Wiley-Blackwell. (c) Structure of a  $\text{CsPbI}_3$  perovskite solar cell.<sup>21</sup> (d)  $J$ - $V$  characteristics of  $\text{CsPbI}_3$  (0%) and  $\text{CsPbI}_3$  with a 10% PEAI device.<sup>21</sup> (e) PCE of  $\text{CsPbI}_3$  (0%) and  $\text{CsPbI}_3$  with a 10% PEAI device.<sup>21</sup> Copyright 2021, Wiley-VCH Verlag. (f)  $J$ - $V$  curves for the passivated  $\text{CsPbI}_2\text{Br}$  devices and control devices.<sup>32</sup> (g) Schematic diagram of the fabrication of perovskite films and the passivation process using  $\text{Pb}(\text{NO}_3)_2$  ethyl acetate solution.<sup>32</sup> Copyright 2018, Wiley-Blackwell. (h) Schematic diagram of the crystallization kinetics of  $\text{CsPbI}_3$  and Ge-doped  $\text{CsPbI}_3$  thin films.<sup>38</sup> (i)  $J$ - $V$  curves of the champion  $\text{CsPb}_{0.95}\text{Ge}_{0.05}\text{I}_3$ -based PSC and control group.<sup>38</sup> Copyright 2021, Wiley-VCH Verlag. (j)  $J$ - $V$  curves of  $\text{CsPbBr}_3$  and  $\text{Eu}^{2+}$ -doped  $\text{CsPbBr}_3$ -based PSC.<sup>42</sup> Copyright 2020, Wiley-Blackwell.





evaporation. Nevertheless, Zhang *et al.* prepared high-quality  $\gamma$ -CsPbI<sub>3</sub> perovskite layers with PCEs of up to 15% by thermal evaporation of PbI<sub>2</sub> and CsI with a small amount of phenylethylammonium iodide (PEAI), exceeding the performance of most solution-processed inorganic CsPbX<sub>3</sub> (X = Cl, Br, I) PEA I perovskite solar cells.<sup>21</sup> Fig. 8c shows the structure of the CsPbI<sub>3</sub> solar cell. The addition of the appropriate amount of PEA I not only provided high-quality perovskite films but also ensured charge transport in the active layer of the perovskite films, which exhibited a longer charge recombination lifetime; this indicates that the charge recombination is strongly suppressed, thus improving the performance of the PSCs. Fig. 8d displays the current density–voltage ( $J$ – $V$ ) characteristics of the device with a  $V_{OC}$  of 1.09 V,  $J_{SC}$  of 17.33 mA cm<sup>−2</sup>, and fill factor of 79.41%. When PEA I was added at 10%, the device obtained 15% PCE, as shown in Fig. 8e.

Traps on the surface of perovskite can affect the conversion efficiency of perovskite solar cells. In recent years, some scholars have used post-treatment methods to reduce surface traps to further improve the PCE of inorganic PSCs. Yuan *et al.* introduced a Pb<sup>2+</sup> solution post-processing strategy to passivate the deep trap state of CsPbI<sub>2</sub>Br films and obtained PSCs with a PCE of 12.34%.<sup>32</sup> The dissociative Pb<sup>2+</sup> in the solution can combine with the excess halide ions on the surface of perovskite, thus reducing the deep trap density of states and charge recombination. The reduction of defect states results in doubling of the average lifetime of perovskite, increasing the  $V_{OC}$  and PCE of the solar cell device. The preparation and passivation process of CsPbI<sub>2</sub>Br perovskite films is shown in Fig. 8g. The structure of the planar heterojunction perovskite solar cells is in the form of FTO/SnO<sub>2</sub>/perovskite/spiro-OMeTAD/Ag. SnO<sub>2</sub> is used as an electron transport layer to reduce the hysteresis effect, resulting in a  $J_{SC}$  of 12.41 mA cm<sup>−2</sup>,  $V_{OC}$  of 1.29 V, and FF of 76.9% for perovskite solar cells, as shown in Fig. 8f. In addition, Li *et al.* synthesized *n*-methyl-2-pyrrolidinone iodide (NMPI) as a functional precursor additive to improve the phase stability and optical properties of CsPbI<sub>3</sub>.<sup>82</sup> As a result, the PCE of the quasi-2D ( $n = 20$ ) CsPbI<sub>3</sub> PSC was increased to 14.59%, and the stability was significantly enhanced.

Doping has been used to participate in the crystallization process, altering the energy state of the perovskite and promoting the stability of the PSCs. Meng *et al.* found that Ge doping was able to modify the crystalline growth of CsPb<sub>1− $x$</sub> Ge <sub>$x$</sub> I<sub>3</sub> films and reduce the annealing temperature and processing time by lowering the CsPbI<sub>3</sub> formation energy.<sup>38</sup> As shown in Fig. 8h, the first addition of Ge to the precursor of DMAPbI<sub>3</sub> accelerated the crystallization process by lowering the formation energy and improving the thermodynamic stability. The GeO<sub>2</sub> generated in situ on the surface of the perovskite film acts as a passivation layer to passivate the grain boundaries and surface defects, thereby improving the hydrothermal stability of the perovskite film and PSCs. The device structure of the solar cell is FTO/compact TiO<sub>2</sub>(c-TiO<sub>2</sub>)/PCBA/perovskite/spiro-OMeTAD/Au. As shown in Fig. 8i, the PCE of the best PSC is 19.52% (a certified PCE of 18.8%) with a  $J_{SC}$  of 20.33 mA cm<sup>−2</sup>,  $V_{OC}$  of

1165 mV, and FF of 0.824. Karunakaran *et al.* doped Eu<sup>2+</sup> into all-inorganic CsPbBr<sub>3</sub> PSCs, leading to an increase in PCE from 5.66% to 7.28%.<sup>42</sup> Owing to the addition of rare earth elements, the perovskite films displayed crystallinity and smooth morphology while optimizing the charge transfer kinetics within the perovskite films. As shown in Fig. 8j, the photovoltaic performance was greatly improved when Eu<sup>2+</sup> was introduced up to 60  $\mu$ L.

This section focuses on the enhancement of the performance of all-inorganic PSCs by doping, which passivates the surface defects of perovskite and improves the average lifetime, thus optimizing the performance of PSCs. At the end of this section, some of the advances in CsPbX<sub>3</sub> perovskite solar cells reported in the literature are summarized in Table 2.

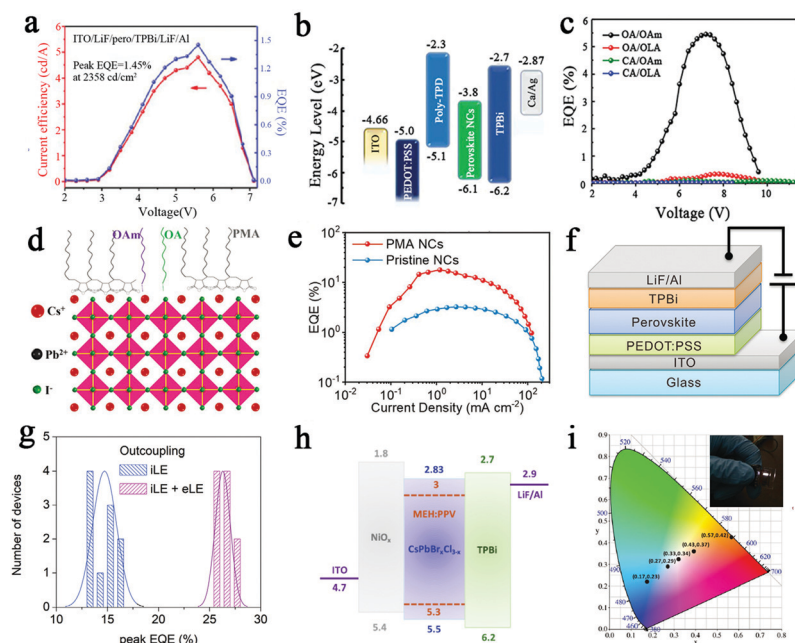
### 3.2 Light-emitting diodes

Recently, increasing numbers of scholars have applied all-inorganic perovskite as light-emitting materials in LEDs. Next, we describe the application of perovskite with different luminescent colors in LEDs. The PN junctions are the basis for LEDs, where electrons and holes will migrate directionally and recombine near the PN junction when a forward bias is applied, releasing heat or light.<sup>88</sup> Tang *et al.* prepared CsPbBr<sub>3</sub> green-emitting LEDs by all-vacuum processing, and high-throughput combination strategies were adopted to optimize the perovskite composition, annealing temperature and functional layer thickness.<sup>8</sup> The structure of CsPbBr<sub>3</sub> perovskite LED is indium-doped tin oxide (ITO)/LiF/perovskite/TPBi/LiF/Al, and the energy band diagram of the AVBD-processed device is shown in Fig. 8a. When the Cs/Pb ratio and the thickness of the perovskite film are 1.17:1 and 110.6 nm, the LED will achieve optimal performance. The appropriate functional layer thickness can balance electron and hole injection, thus enhancing the EQE of the LED performance. The best device showed a current efficiency (CE) of 4.8 cd A<sup>−1</sup> (EQE of 1.45%) at 2358 cd m<sup>−2</sup>, as shown in Fig. 9a. In addition, the CE improved to 10.15 cd A<sup>−1</sup> and the EQE improved to 3.26% for perovskite LEDs by improving the hole-injection layer to NiO <sub>$x$</sub> . CsPbX<sub>3</sub> perovskite is usually prepared in an inert environment under high temperature conditions, which hinders the large-scale production of LED devices, and a low-temperature preparation strategy is essential. Wang *et al.* synthesized CsPbBr<sub>3</sub> NCs in the

Table 2 Performance comparison of the PSCs based on CsPbX<sub>3</sub> perovskites

Materials	$J_{SC}$ (mA cm <sup>−2</sup> )	$V_{OC}$ (V)	FF (%)	PCE (%)	Ref.
FA <sub>0.15</sub> Cs <sub>0.85</sub> PbI <sub>3</sub>	21.4	1.02	80	17.5	4
CsPbX <sub>3</sub>	17.33	1.09	79.41	15	21
CsPbI <sub>2</sub> Br	12.41	1.29	76.90	12.34	32
CsPbX <sub>3</sub>	12.6	1.05	78.70	10.40	83
CsPbI <sub>3</sub>	15.68	1.20	64	12.04	84
CsPbI <sub>2</sub> Br	15.34	1.32	80	16.14	85
CsPbI <sub>2</sub> Br	15.23	1.14	64.51	11.20	86
CsPbI <sub>3</sub>	0.415	0.643	42.9	0.11	87
CsPbBr <sub>3</sub>	2.96	0.851	44.5	1.21	87
CsPb <sub>1−<math>x</math></sub> Ge <sub><math>x</math></sub> I <sub>3</sub>	20.33	1.165	82.4	18.8	38
CsPbBr <sub>3</sub>	6.33	1.4523	79.19	7.28	42





**Fig. 9** (a) Current efficiency and EQE curves of the best device.<sup>8</sup> Copyright 2019, Wiley-VCH Verlag. (b) Energy level diagram of LED devices.<sup>20</sup> (c) External quantum efficiency versus voltage of LED devices.<sup>20</sup> Copyright 2019, Wiley-Blackwell. (d) Schematic diagram of the PMA-incorporating  $\beta$ -CsPbI<sub>3</sub> NCs.<sup>27</sup> (e) Relationship between the current density and EQE curve of the CsPbI<sub>3</sub> LED.<sup>27</sup> Copyright 2020, Wiley-Blackwell. (f) Schematic diagram of the structure of the PSC devices.<sup>33</sup> (g) Histogram of the EQEs of the devices.<sup>33</sup> Copyright 2021, Wiley-VCH Verlag. (h) Schematic of the band structure.<sup>39</sup> (i) CIE coordinates of the white LED.<sup>39</sup> Copyright 2016, Wiley-Blackwell.

presence of the short-chain ligands propionic acid (PA) and butylamine (BLA) in a room temperature environment.<sup>20</sup> In addition, studies have shown that CsPbBr<sub>3</sub> NCs anchored by OA and OAm produce the best performance. Fig. 9b shows the energy level diagram of the CsPbBr<sub>3</sub> LED and the structural composition of each part of the device. Fig. 9c demonstrates that the LEDs with OA/OAm-anchored NCs exhibit the best performance over a wide range, with a luminance of 5033 Cd m<sup>-2</sup>, CE of 18.6 Cd A<sup>-1</sup>, turn-on voltage as low as 3.2 V, and EQE of 5.4%.

The stability of perovskite LEDs has been a great challenge, especially as red perovskite LEDs typically have a lifetime of only a few hours. Li *et al.* have prepared stable  $\beta$ -CsPbI<sub>3</sub> for red-emitting LED applications through incorporation of poly(maleicanhydride-*alt*-1-octadecene) (PMA) in synthesizing the nanocrystals.<sup>27</sup> As shown in Fig. 9d, the PMA can interact with PbI<sub>2</sub> in the precursor through the coupling between the O group in PMA and Pb<sup>2+</sup>, and the PMA greatly passivated Pb<sub>Cs</sub> anti-situ defects on the surface of  $\beta$ -CsPbI<sub>3</sub>, which improved the stability of the crystal. The photoluminescence quantum yield of the  $\beta$ -CsPbI<sub>3</sub> NCs films was significantly increased from 34% to 89% due to the addition of PMA. The structure of this device is indium tin oxide (ITO)/poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS)/poly-TPD/ $\beta$ -CsPbI<sub>3</sub> NCs/tris(1-phenyl-1*H*-benzimidazole) (TPBi)/lithium fluoride (LiF)/Al. Fig. 9e shows red-emitting LEDs obtaining an EQE of 17.8%. The LEDs were stable for 317 h at a constant current density of 30 mA cm<sup>-2</sup>. Miao *et al.* prepared  $\gamma$ -CsPbI<sub>3</sub> cuboid crystallites of deep red light perovskite LEDs based on a one-step method.<sup>89</sup> Due to the addition of diammonium iodide to passivate the trap state of the *in situ* formed  $\gamma$ -

CsPbI<sub>3</sub>, the high peak external quantum efficiency (EQE) value of the LED was 15.03%. Moreover, this one-step method offers a new strategy for the flow-line production of large-area perovskite LED modules.

Most of the current research on perovskite blue-emitting LEDs has been prepared by adjusting the band gap of a mixture of Br and I halogens. However, the film morphology of the mixed halide perovskite has not been well improved. Yao *et al.* prepared blue-emitting (470 nm) perovskite nanocrystals by mixing a 2 : 1 weight ratio of CsPbBr<sub>3</sub> and CsPbI<sub>3</sub> nanocrystals.<sup>39</sup> By controlling the size of the thin film grains, high quality blue-emitting perovskite LEDs were obtained. Shen *et al.* proposed a simple device architecture to achieve highly efficient blue-emitting perovskite LEDs.<sup>33</sup> Interfacial engineering is employed to manipulate the perovskite crystallization to achieve highly compact perovskite nanocrystals. The device structure of the blue perovskite LEDs is shown in Fig. 9f. On this basis, the EQE is further increased from 16.8% to 27.5% by integrating a lens-based structure for substrate mode light (Fig. 9g).

The preparation process of white perovskite LEDs is more complex than that of red, blue and green LEDs. Yao *et al.*, mentioned in the previous paragraph, prepared a white light perovskite LED by mixing an orange light polymer with blue perovskite as the active layer.<sup>39</sup> The device structure of white LEDs with a composite orange polymer material is shown in Fig. 9h. By adjusting the ratio between perovskite nanocrystals and polymers, white LEDs were achieved with CIE coordinates at (0.33, 0.34). Chen *et al.* first prepared red-emitting CsPbBr<sub>2.5</sub>I<sub>0.5</sub> perovskite films by a one-step spin-coating method using polyacrylonitrile



(PAN) polymer.<sup>90</sup> Then, white LEDs were prepared by combining *N,N'*-bis(naphthalen-1-yl)-*N,N'*-bis(phenyl)-benzidine (NPB), CsPbBr<sub>2.5</sub>I<sub>0.5</sub> perovskite films doped with PAN, and CsPbBr<sub>3</sub>. As shown in Fig. 9i, the LEDs displayed a maximum of 360 cd m<sup>-2</sup> with CIE coordinates of (0.31, 0.36) at 7 V.

For further study, some optical parameters of CsPbX<sub>3</sub> perovskite-based LEDs are summarized in Table 3. In recent years, many different methods of preparing perovskite LEDs have emerged, gradually improving their luminescence properties and stability. All-inorganic perovskite LEDs show great promise in the field of optical displays.

### 3.3 Photodetectors

CsPbX<sub>3</sub> perovskite is used in photodetectors due to its high light absorbance and long carrier diffusion length. Next, we describe the application of different structures of perovskite in photodetectors. Zeng *et al.* prepared CsPbBr<sub>3</sub> nanosheets by improving the hot injection method and applied them in photodetectors.<sup>3</sup> As shown in Fig. 10a, the edge length of the nanosheet could be greater than 1.5 μm. Large-area CsPbBr<sub>3</sub> nanosheet films were prepared by simple ink printing or the roll-to-roll method and applied to flexible photodetectors, as shown in Fig. 10b. In the presence of an external electric field, the electron-hole pairs generated in the perovskite are rapidly separated and collected by the copper electrode, and then the CsPbBr<sub>3</sub> nanosheet photodetector can work effectively. As shown in Fig. 10c, the maximum value of the responsivity was located at 517 nm, and the responsivity was 0.64 A W<sup>-1</sup>; the peak EQE value of the device at 515 nm was 53% at 10 V bias. A visible photodetector prepared using CsPbBr<sub>3</sub> nanosheets exhibited a high sensitivity with a light on/off ratio of > 10<sup>3</sup> under 442 nm light at a power of 0.35 mW cm<sup>-2</sup> as well as high stability and outstanding fluorescence excitability (> 10 000 cycles). In addition, high stability and outstanding fluorescence excitability were still demonstrated after 10 000 cycles.

Large perovskite single crystals have the potential to produce high performance photodetectors due to their higher carrier mobility, longer diffusion length and longer carrier

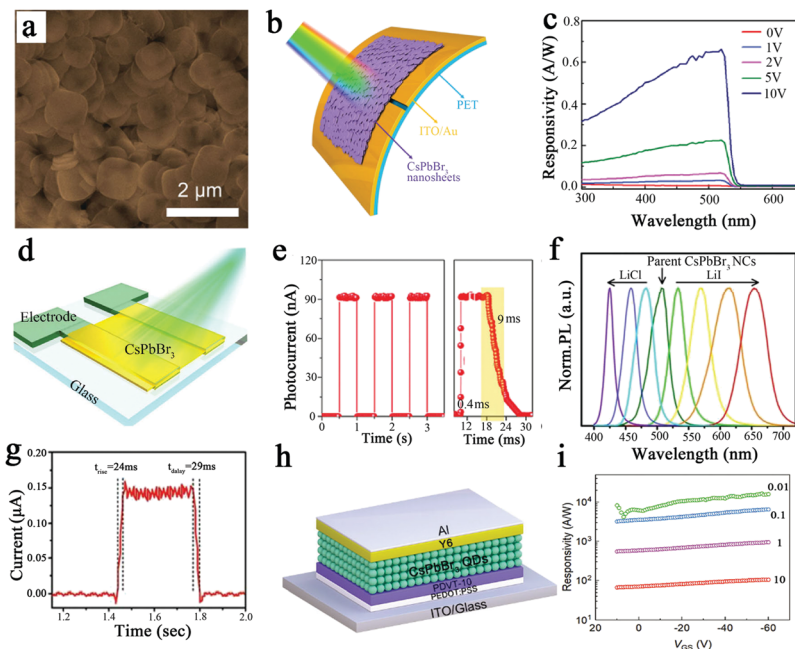
lifetime. Zheng *et al.* demonstrated a low-temperature and substrate-independent strategy to prepare millimeter-scale CsPbBr<sub>3</sub> perovskite single-crystal films.<sup>23</sup> Fig. 10d shows a schematic of the device structure. The superior carrier transport properties will improve the performance of the photodetector. The trap density, diffusion length and migration lifetime of CsPbBr<sub>3</sub> films and CsPbBr<sub>3</sub> bulk crystals are comparable, but the resistivity is superior to that of the CsPbBr<sub>3</sub> bulk crystals, which improves the performance of the photodetector. Fig. 10e shows the photodetector with a light on/off ratio > 10<sup>3</sup> and rise and decay times of 0.4 ms and 9 ms, respectively. The CsPbBr<sub>3</sub> films showed no significant photoluminescence loss after two months of storage, indicating that they exhibited excellent long-term stability to humidity and heat treatment. Ramasamy *et al.* prepared CsPbX<sub>3</sub> (X = Cl, Br, I) nanocrystals (NCs) using halide ion exchange reactions at room temperature, and the photoluminescence could be tuned throughout the visible region (425–655 nm).<sup>31</sup> In this report, CsPbBr<sub>3</sub> perovskite colloidal NCs prepared by halogen exchange reactions were applied to photodetectors for the first time. An adjustable luminescence range was achieved through the halide ion exchange reactions. The green emission from CsPbBr<sub>3</sub> NCs was tuned by lithium salts (LiX, X = I, Cl, Br) in the entire visible spectral region (425–655 nm) at room temperature. Fig. 10f demonstrates that different concentrations of lithium salts can tune the PL peak from 508 to 654 nm. The group selected CsPbI<sub>3</sub> NCs with relatively long radiation lifetimes for photodetector applications. The CsPbI<sub>3</sub> photodetector exhibits an excellent on/off photocurrent ratio of 10<sup>5</sup>. In addition, the CsPbI<sub>3</sub> photodetector showed a relatively fast rise time (*t*<sub>rise</sub> = 24 ms) and decay time (*t*<sub>decay</sub> = 29 ms), as shown in Fig. 10g.

Planar heterojunction (PHJ) structures consisting of different semiconductors also show great potential in photodetectors. Chen *et al.* prepared solution-processed perovskite quantum dots (PQDs)/organic semiconductors (OSC) planar heterojunctions and applied them to photodetectors.<sup>37</sup> CsPbBr<sub>3</sub> QDs were prepared by the hot-injection method, and the quantum dot films were prepared by spin coating. Fig. 10h shows a schematic of the phototransistor for CsPbBr<sub>3</sub>

Table 3 Performance comparison of the LEDs based on CsPbX<sub>3</sub> perovskites

Materials	Luminous colours	EL λ <sub>max</sub> (nm)	V <sub>on</sub> (V)	Max. EQE (%)	Max. CE (cd A <sup>-1</sup> )	Max. L (cd m <sup>-2</sup> )	Ref.
CsPbBr <sub>3</sub>	Green	523–524	3.92	—	1.07	1607	62
CsPbBr <sub>3</sub>	Green	516	—	1.45	4.8	2358	8
CsPbBr <sub>3</sub>	Green	516	3.2	5.4	18.6	5033	20
CsPbBr <sub>3</sub>	Green	514	—	13.14	37.14	45 990	26
CsPbBr <sub>3</sub>	Green	517	2.3	5.7	19.9	46 000	91
CsPbBr <sub>3</sub>	Green	516	4.2	0.12	0.43	946	50
CsPbI <sub>3</sub>	Green	586	4.6	0.09	0.08	528	50
CsPbCl <sub>3</sub>	Green	455	5.1	0.07	0.14	742	50
CsPbBr <sub>3</sub>	Green	—	—	0.25	0.96	637	92
CsPbBr <sub>3</sub>	Green	510	2–2.5	0.37	0.72	1661	93
CsPbI <sub>3</sub>	Red	689	5	17.8	—	618	27
CsPbI <sub>3</sub>	Red	699	2.8	15.3	—	1272	89
CsPbBr <sub>x</sub> Cl <sub>3-x</sub>	Blue	470	5	0.07	0.18	350	39
CsPbBr <sub>3-x</sub> Cl <sub>x</sub>	Blue	486	3.3	27.5	33.4	3651	33
CsPb(Br <sub>2.5</sub> I <sub>0.5</sub> ) + CsPbBr <sub>3</sub>	White	—	7	—	0.2	360	90
CsPbBr <sub>x</sub> Cl <sub>3-x</sub> + MEH:PPV	White	—	8	—	—	—	39





**Fig. 10** (a) SEM images of CsPbBr<sub>3</sub> nanosheets.<sup>3</sup> (b) Schematic diagram of PDs based on CsPbBr<sub>3</sub> nanosheets.<sup>3</sup> (c) Responsivity of CsPbBr<sub>3</sub> nanosheet photodetector devices under different forward biases.<sup>3</sup> Copyright 2016, Wiley-Blackwell. (d) Schematic of the photodetector device structure.<sup>23</sup> (e) Left: Time-dependent optical response of the photoelectric detector. Right: Rise time and decay time of the photodetector under 530 nm light (216 μW m<sup>-2</sup>) at a bias of 2 V.<sup>23</sup> Copyright 2018, Wiley-Blackwell. (f) PL spectra of CsPbBr<sub>3</sub> NCs exchanged with different concentrations of LiI and LiCl.<sup>31</sup> (g) Rise and decay times of the photodetector device.<sup>31</sup> Copyright 2016, Royal Society of Chemistry. (h) Schematic device structure of the CsPbBr<sub>3</sub> QDs/PDVT-10 phototransistor.<sup>37</sup> (i) Responsivity of CsPbBr<sub>3</sub> QDs/PDVT-10 phototransistors.<sup>37</sup> Copyright 2021, Wiley-VCH Verlag.

QDs/PDVT-10 PHJs. As shown in Fig. 10i, the *R*-value can reach  $1.64 \times 10^4$  A W<sup>-1</sup> when the illumination intensity is 0.01 mW cm<sup>-2</sup>. In addition, the double layer PQD/PDVT-10 PHJs phototransistor has a specific detectivity of  $3.17 \times 10^{12}$  Jones and a photosensitivity of  $5.33 \times 10^6$ . The presence of the PHJ structure allows the exciton dissociation process to become more efficient. This photosensitive effect is important for achieving high responsiveness or gain in PHJ-based phototransistors due to the extended excess carrier lifetime. For comparison, the properties of CsPbX<sub>3</sub> perovskite PDs are summarized in Table 4.

## 4. Conclusions

In summary, here, we reviewed some advances in OIH perovskites, mainly some different preparation methods of all-inorganic perovskites and their applications in optoelectronic fields. Recently, due to the special physical structure and excellent optical properties of CsPbX<sub>3</sub> perovskite, researchers have conducted extensive research on CsPbX<sub>3</sub>. However, there

are still some challenges in the practical application of perovskite remaining to be addressed. Therefore, in this section, we focus on a summary of the current challenges and opportunities for perovskite applications.

Firstly, the toxicity of lead halide perovskite needs to be considered. The heavy metal ion lead has severe consequences on human health and the environment. Appropriate encapsulation strategies are needed to reduce the toxicity of lead halide perovskite. When the lead halide perovskite encapsulated by the polymer is exposed to water, the lead concentration is comparable to that in tap water. Another alternative solution is the development of lead-free halide perovskite, a non-toxic element (such as Sn, Bi, Sb, Cu, and Ge) to replace the toxic lead.<sup>94</sup>

Secondly, the instability of the internal structure of perovskite is also a problem that limits its commercialization. The water stability of perovskite has been improved through different approaches, including surface passivation and the polymer encapsulation strategy. However, polymer encapsulation can affect the electrical properties of perovskites, reducing

**Table 4** Performance comparison of photodetectors based on CsPbX<sub>3</sub> perovskites

Materials	Shapes	Responsivity (A W <sup>-1</sup> )	On/off ratio	Rise time (ms)	Decay time (ms)	EQE (%)	Ref.
CsPbBr <sub>3</sub>	Nanosheet	0.64	> 10 <sup>4</sup>	0.019	0.24	54	3
CsPbBr <sub>3</sub> -CNTs	Nanosheet	31.1	> 832	0.016	0.38	—	95
CsPbBr <sub>3</sub>	Film	0.21	10 <sup>3</sup>	0.4	9	6.2	23
CsPbBr <sub>3</sub> /ZnO	Film	4.25	> 10 <sup>4</sup>	0.21	0.24	—	96
CsPbI <sub>3</sub>	Nanocrystal	—	10 <sup>5</sup>	24	29	—	31



the efficiency of their optoelectronic devices. Subsequent research needs to focus on surface modification, improvement of surface passivation strategies or improvement of chemical components.

Third, compared to OIH perovskites, the band gap of all-inorganic perovskites should be suitably adjusted to better improve the performance of optoelectronic devices. Therefore, the band gap of all-inorganic perovskites is tuned and their stability is further improved by doping, metal-organic framework crystal embedding and ligand-based studies.

Ultimately, we hope that theoretical calculations and experiment studies in CsPbX<sub>3</sub> will be of great benefit to the synthesis of crystals and device performance. All-inorganic perovskites still have a long way to go, but we believe that they will make great progress in a variety of applications. CsPbX<sub>3</sub> can play important roles in photovoltaics and improving human well-being.

## Conflicts of interest

There are no conflicts to declare.

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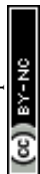
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