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TOPICAL REVIEW

Recent progress in all-inorganic metal halide nanostructured perovskites: Materials design, optical properties, and application

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Low-dimensional all-inorganic metal halide perovskite (AIMHP) materials, as a new class of nanomaterials, hold great promise for various optoelectronic devices. In the past few years, tremendous progress has been achieved in the development of efficient and stable AIMHP nanomaterials for optical property studies and related applications. Here, we offer a critical overview on the unique merits and the state-of-the-art design of AIMHP using different composition strategies. Then, the effects of material compositions, dimensionality, morphologies and structures on optical properties are summarized. We also comprehensively present recent advances in the development AIMHP nanomaterials for practical applications including solar cells, light-emitting diodes, lasers and photodetectors. Lastly, the critical challenges and future opportunities in this emerging field are highlighted.

Keywords inorganic perovskite, nano-structure, materials design, optical properties, applications

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1 Introduction

In the last decade or so, metal halide perovskites have received substantial attention in all over the world due to their applications in solar cells (SCs) [1–3], light emitting diodes (LEDs) [4–6], lasers [7–9], photodetectors (PDs) [10–12], etc. However, there are some concerns remaining during their practical applications, especially the instability of organo metallic halide perovskites (OMHP) and hybrid organic-inorganic metallic halide perovskites (HOIMHP). Recent studies indicated that the organic groups play a vital role for enhancing their photo and thermal stability because metal halide perovskites are extremely sensitive to both oxygen and water [13, 14]. Thus, most of the attention has turned to the enhancement of environmental stability by hybridizing with other cations such as Cs according to a tolerance factor [15], by surface modification with moisture-tolerant molecules [16], by integrating OMHP with polymers [17] and by replacing organic carrier transport layers with inorganic oxides [18].

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Although various methods have been used to improve the stability of materials, the inherent instability of organic groups still hinders their practical applications, as shown in Fig. 1.

To overcome the challenges, the all inorganic halide perovskite $CsPbX_3$ ($X=Cl^+$, Br^+, I^+) nanocrystals were successfully synthesized in 2015 based on the concept that replacing the organic groups with inorganic cations [19]. After that, the $CsPbX_3$ are rapidly emerging as promising alternatives because of their excellent stability and comparable optical properties and the prepared all inorganic halide perovskite $CsPbX_3$ nanocrystals have been successfully applied in the SCs [20–22]. Subsequently, many strategies have been developed for $CsPbX_3$ fabrication, including colloidal synthesis, anti-solvent, substrate evaporation, and substrate vapor transport [23]. By controlling the rich reaction chemistry in the preparation of inorganic halide perovskites, high quality and well-defined morphologies of zero-dimensional (0D) quantum dots (QDs) [19], one-dimensional (1D) nanowires (NWs) [24–27], and two-dimensional (2D) nanoplates (NPLs) or nanosheets (NSs) [28–30] have been achieved. Besides, recently reported works suggested that compared to the OHP and HOIHP, AIMHP with general formula ABX_3 ($A=Cs^+$; $B=Pb^{2+}$, Sn^{2+}, Ge^{2+}; $X=Cl^+$, Br^+, I^+) are a promising alternative with attractive properties, such as high quantum efficiency [19–31], high carrier mobility [32, 33], narrow emission peak [34, 35], and exhibit higher stability and have many potential in various optoelectronics, including SCs [36, 37], lasers [38, 39], LEDs [40, 41], and PDs [42, 43].

Obviously, inorganic halide perovskite nanomaterials provide controllable morphology and tunable optoelectronic properties and can effectively improve the stability and performance of materials and devices [44, 45], which offer excellent platforms for distinct fundamental research and practical applications in the near future. Thus, a timely review on such a rapidly growing field of great significance is highly desirable. Following this line of thought, in this review, we summarize the synthesis, optical properties, and applications of AIMHP materials from their discovery to date. At last, we also discuss the challenges facing the field of AIMHP and some possible solutions based on the available literature are suggested.

2 Synthesis of all-inorganic metal halide perovskite nanomaterials

AIMHP nanomaterials have at least one dimension at the nanoscale (<100 nm), such as QDs (or NCs: nanoparticles), NWs (or nanorods), and NSs (or thin films). These materials have demonstrated excellent optical and electrical properties different from their bulk materials due to quantum confinement effects (QCE) and strong anisotropy [46–50]. In this part, we will review the progress of AIMHP nanomaterials in recent years, based on the differences of dimensions. Different synthesis approaches and growth conditions will be discussed along with their novel characteristics especially optical and stability properties.

2.1 All-inorganic metal halide perovskite 0D nanomaterials

In recent years, all-inorganic cesium lead halide perovskite $CsPbX_3$ (X=Cl, Br, and I) NCs have attracted a lot of attention due to their excellent properties, such as high photoluminescence quantum yield (PLQY), broad wavelength coverage, narrow PL emission bandwidth, and low trap state density [51–54]. These outstanding properties of $CsPbX_3$ NCs distinguish them from traditional semiconductor NCs and make them promising candidates for various optoelectronic applications.

As mentioned many times before, Protesescu *et al.* synthesized $CsPbX_3$ colloidal NPs for the first time by hot-injection method [19]. The $CsPbX_3$ colloidal NPs exhibited a cubic shape with a size of 4–15 nm and cubic perovskite crystal structure. Through compositional modulations and QCEs, the emission spectra are adjustable from 410 to 700 nm. Since then, many methods have been developed for the synthesis of $CsPbX_3$ NCs, such as solvothermal synthesis [55], post-treatment [56], ultrasonication [57], and mechanochemistry [58]. However, researchers still face challenges to achieve highly stable and photoluminescent $CsPbX_3$ NCs at room temperature by the direct-synthesis method [59].

Nedelcu *et al.* and Akkerman *et al.* further realized fast anion exchange in $CsPbX_3$ (X=Cl, Br, I) perovskite NCs at low temperature [60, 61]. By adjusting the halide ratios in the colloidal nanocrystal solution, the bright PL could be tuned over 410–700 nm while maintaining high quantum yields (QY) of 20%–80% and narrow emission line widths of 10–40 nm. Furthermore, the fast inter-nanocrystal anion exchange was demonstrated between the $CsPbCl_3$, $CsPbBr_3$, and $CsPbI_3$ NCs, leading to uniform $CsPb(Cl/Br)_3$ or $CsPb(Br/I)_3$ compositions. After that, there were many reports on $CsPbX_3$ colloidal NCs synthesized based on this method, but with different crystal phases. For example, Swarnkar *et al.* synthesized $CsPbBr_3$ colloidal NCs following the above methods, but obtained orthorhombic phase $CsPbBr_3$ [54]. Jellicoe *et al.* first synthesized lead-free $CsSnX_3$ (X=Cl, $Cl_{0.5}Br_{0.5}$, Br, $Br_{0.5}I_{0.5}$, I) perovskite nanocrystals and exhibited the tunability of the spectrum through both QCE and control of the anionic composition [62].

Inspired by the methodology for synthesis of perovskite $MAPbX_3$ colloidal NCs, Li *et al.* extended the supersaturated recrystallization method to synthesize inorganic perovskite $CsPbX_3$ [63]. These colloidal NCs have superior optical properties to those formed at high temperature, including PLQYs of 80%, 95%, and 70% for red, green,

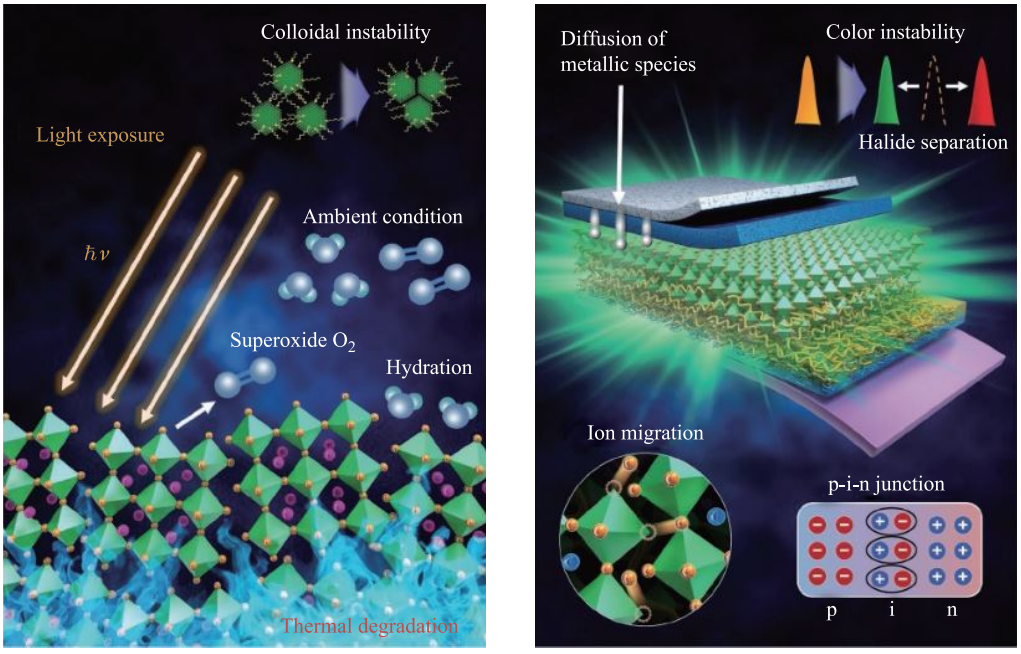

Fig. 1 A schematic illustration describing the intrinsic instability of OMHP and HOIMHP materials and corresponding devices. Reproduced with permission from Ref. [6].

and blue PLs, and a 90% retention rate after aging for 30 days under ambient conditions. Deng *et al.* used a reprecipitation strategy to systematically manipulate the shape of colloidal $CsPbX_3$ NCs, such as spherical colloidal NCs and nanocubes, by using different ligands [64]. De Roo *et al.* studied the dynamic ligand of the inorganic perovskite $CsPbX_3$ [65]. They found that compared to classical chalcogenide QDS, $CsPbX_3$ is more ionic in nature and the interactions with capping ligands are also more ionic and labile. Most recently, Swarnkar *et al.* [66] reported the synthesis of ambient stable cubic–phase QDs of $CsPbI_3$, a phase that was previously known to be stable only at high temperature. They have developed an improved synthetic route and purification approach that prevents the $CsPbI_3$ QDs from transforming their as-synthesized cubic phase to orthorhombic. Accurate control of emission bands and NC morphologies were vital prerequisites for most $CsPbX_3$ NCs practical applications. A facile method of synthesizing $CsPbX_3$ NCs in the nonpolar solvent octane was developed by Ye *et al.* [67]. The process was conducted in air at about 90°C and the prepared high-quality $CsPbX_3$ NCs showing 12–44 nm wide emission with high PL quantum yield, exceeding 90% [67].

Since last year, core-shell structures have been used to further improve the optical properties and stability of AIMHP nanomaterials. Lu *et al.* prepared the $CsPbBr_3$/Cs_4PbBr_6 core-shell structure NCs by means of a microchannel reactor. Without any protection, the outstanding stable NCs retain more than 90% of the PL intensity after 84 days [68]. Tang *et al.* were successfully synthesized $CsPbBr_3$/CdS core-shell structure QDs using a facile hot-injection method [69]. Chen *et al.* adopted an effective ion-doping strategy to improve the stability and efficiency of all-inorganic $CsPbI_2Br$ and related SC devices [70]. Wang *et al.* got the mixed $CsPbI_{2.85}Br_{0.15}$ by adding 5% Br ions into the $CsPbI_3$ [71]. The optimized PSCs based on this mixed $CsPbI_{2.85}Br_{0.15}$ obtain a record reverse scan PCE of 17.17% and stabilized PCE of 16.83%, and exhibit excellent thermal and compositional stability. Zhao *et al.* synthesized $CsPbI_3$ NCs with different size distributions and studied the lattice structure and confinement properties based on this perovskite nanocrystals, as shown in Fig. 2 [72]. Their findings suggest that adopt the perovskite nanocrystal structure can effectively improve the stability over thin-film perovskite materials. As summarized above, intensive efforts have been made in the development of AIMHP 0D nanomaterials since the first study in 2015. Significant progress has been achieved in the controlled preparations and optimizations of AIMHP NCs properties over the past five years. Their promising applications in different areas have also been partially demonstrated. Despite these great successes, the research in AIMHP 0D nanomaterials is still in its early infancy objectively. In particular, the aforementioned issues of stability and toxicity of lead must be addressed before the widespread practical applications of AIMHP 0D nanomaterials become possible.

2.2 All-inorganic metal halide perovskite 1D nanomaterials

Among differently shaped AILHP nanostructures, the 1D AILHP NWs have demonstrated outstanding performance when used in lasing and PD devices [73,

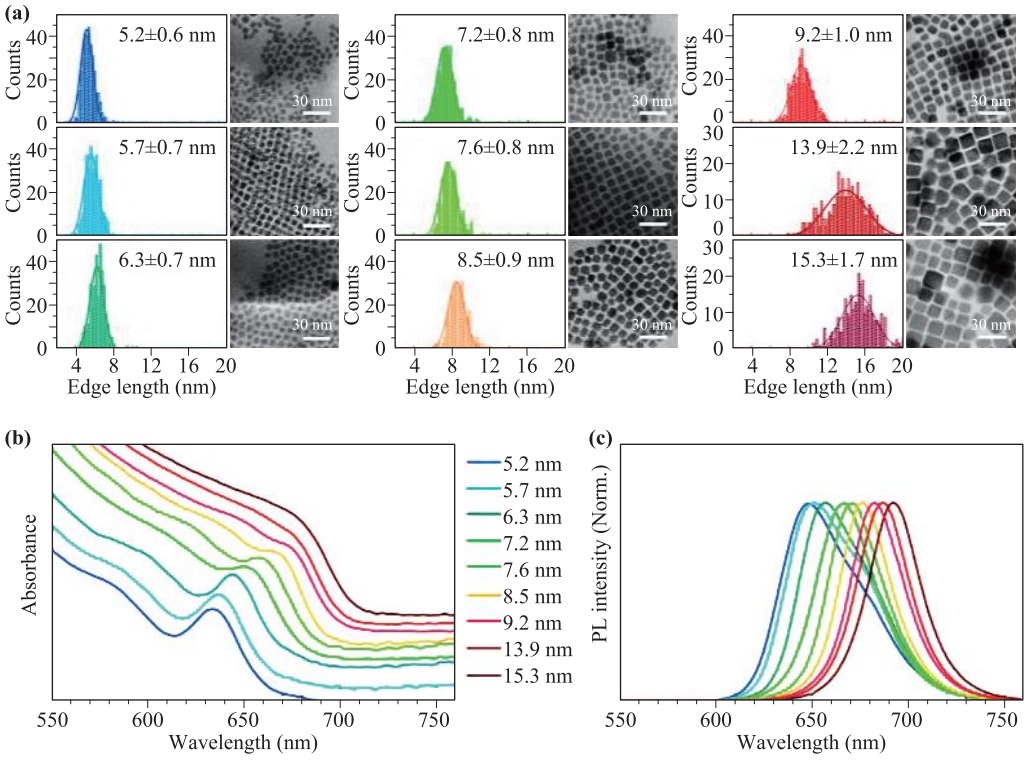

Fig. 2 The distributions, TEM images, absorption spectra and PL spectra of $CsPbI_3$ NCs with different sizes. **(a)** Size distribution and TEM images; **(b)** Optical absorption spectra; **(c)** Normalized PL spectra. Reproduced with permission from Ref. [72].

74]. Various methods have been developed to prepare the AILHP NWs [75, 76], including oriented assembly of nanocubes [77, 78], solution-phase methods [79–81], ultrasonication-assisted synthesis [57], self-assembly process [82], vapor-phase growth techniques [83–87] and catalyst-free colloidal synthesis method followed by stepwise purification strategy [40]. A solution-processed method with surfactant ligand was reported to control the synthesis of all-inorganic perovskite $CsPbX_3$ (X=Cl, Br, I or mixture of halides) NWs [39]. This method was widely used for previous metal and inorganic semiconductor NWs, using a surfactant ligand as the capping ligand to initiate one-dimensional crystal growth. The $CsPbX_3$ NWs were synthesized under an air-free environment by reacting cesium oleate with lead halide in the presence of oleic acid and oleylamine in octadecene at 150–250°C for 5–10 min. The as-synthesized $CsPbX_3$ perovskite NWs exhibit diameters less than 12 nm and lengths up to 5 mm and the yielding up to 90% after simple purification. Moreover, by an anion exchange reaction, $CsPbCl_3$ and $CsPbI_3$ NWs can be transformed from $CsPbBr_3$ NWs by reacting with other halide precursors. The NWs transformed from $CsPbBr_3$ NWs exhibit high quantum efficiency up to 83% and 30% for $CsPbI_3$ and $CsPbCl_3$, respectively [80].

Chen *et al.* reported a simple but efficient solvothermal approach to prepare ultrathin $CsPbX_3$ (X=Cl/Br, Br, and Br/I) NWs with almost 100% morphological yield. A strong QEC was observed in the ultrathin NWs, in which both the absorption and emission peaks shift to shorter wavelength range compared to their bulk bandgap [79]. The single-step ligand-mediated synthesis of single-crystalline $CsPbBr_3$ perovskite NWs directly from the precursor powders [88]. Studies of the reaction process and the morphological evolution revealed that the initially formed $CsPbBr_3$ nanocubes were transformed into NWs through an oriented-attachment mechanism. The optical properties of the NWs can be tuned across the entire visible range by varying the halide (Cl, Br, and I) composition through subsequent halide ion exchange. Single-particle studies showed that these NWs exhibit strongly polarized emission with a polarization anisotropy of 0.36. More importantly, the NWs can self-assemble in a quasi-oriented fashion at an air/ liquid interface. This process should also be easily applicable to perovskite NCs of different morphologies for their integration into nanoscale optoelectronic devices, as shown in Fig. 3 [89]. Shoaib demonstrates the directly controlled method to grow in-plane directional perovskite $CsPbBr_3$ NWs. The NWs have a very strong band-edge PL with a long PL lifetime of $\sim$ 25 ns and can realize high-quality optical waveguides [90]. Then, the direct synthesis of composition-graded $CsPbBr_xI_{3-x}$ NWs is reported by Huang *et al.* through vapor-phase epitaxial growth on mica [91]. The graded composition along the NW, with an increased Br/I from the center to the ends, comes

from desynchronized deposition of cesium lead halides and temperature-controlled anion-exchange reaction. The graded composition results in varied bandgaps along the NW, which induce a blue-shifted emission from the center to the ends. As an efficient gain media, the NW exerts position-dependent lasing performance, with a different color at the ends and center respectively above the threshold.

Last year, Liu *et al.* generated the visible-light-induced template-free synthesis of $CsPbBr_3$ NWs through self-assembly cubic $CsPbBr_3$ NCs [92]. Then, a post-treatment method for the preparation of ultrathin $CsPbX_3$ NWs by treating $CsPbBr_3$ nanocubes with thiourea solution was realized by Li *et al.* [93]. The ultrathin $CsPbBr_3$ NWs exhibited high PLQY (up to 60%) and high resistance to water treatment. Monodisperse $CsPbBr_3$ nanorods with tunable aspect ratio and a clean surface have been prepared through an interfacial conversion from Cs_4PbBr_6 nanocrystals. The nanorods showed excellent photophysical properties, including a PLQY up to 87% and a PL lifetime of 44 ns [94]. Zhang *et al.* developed a self-assembled nanotemplate-confined growth concept to prepare ultrathin $CsPbBr_3$ NWs [95]. He *et al.* reported a robust polar-solvent-assisted route to 1D $CsPbBr_3$ NWs via room temperature supersaturated recrystallization. $CsPbBr_3$ NWs were rapidly yielded, experiencing a cubic-to-orthorhombic phase transformation and a blue-shifted emission [96]. Chen *et al.* adopted a vapor-phase growth method and successfully prepared aligned cesium tin tolerant $CsSnX_3$ (X=Br, I) NWs [97]. And, Meng *et al.* directly synthesized all-inorganic perovskite NWs by the method of vapor-liquid-solid [98]. A facile synthesis of quantum-confined $CsPbBr_3$ NWs by using a continuous-flow microfluidic reactor was reported by Zhang *et al.* [99]. Recently, Liu *et al.* introduced an amphiphilic block copolymer to chemically modify the surface of colloidal $CsPbBr_3$ NWs and a modified Langmuir-Blodgett technique to enhance the stability [100]. The resulting core-shell NWs show enhanced PL emission and good colloidal stability against water. As discussed in this section, the anisotropi-cally structured AILHP NWs and nanorods have been prepared using a variety of methods and have demonstrated outstanding performance when used in lasing and photodetector devices. But the biggest obstacle to practical application of 1D AILHP nanomaterials is still their component stability against chemical, thermal, and photo disturbances. At the same time, the preparation of ultra-long and ultra-fine one-dimensional nanomaterials with good optical and structural properties is also a huge challenge.

2.3 Nanomaterials

The stable $CsPbX_3$ NPLs were first reported in 2015 by Bekenstein and collaborators using the hot-injection method [28]. Since then, the emerging 2D forms of AIMHP are attracting more interest due to the long charge carrier lifetime, high PLQY, and great defect tolerance [101–103]. In 2016, Akkerman *et al.* have demonstrated the synthesis of the $CsPbX_3$ NPLs also by hot-injection method at room temperature [104]. They have shown that anisotropic growth leads to the formation of NPLs through the injection of acetone in a mixture of precursor. The low growth temperature facilitates the control of the plate thickness down to three monolayers. Wang *et al.* reported a facile fast precipitation synthesis of highly luminescent perovskite-related $CsPb_2Br_5$ NPLs and their fast anion-exchange capabilities to tune optical properties [105]. Their results also indicated that the $CsPb_2Br_5$ layered structure has the best stability during the exchange processes when the thickness of the ion-exchange nanosheet is about 3 nm.

In addition to the solution-based approaches, CVD was another important way to grow high-quality 2D materials [106, 107]. Their results suggested that selecting the ideal encapsulation and protection materials for halide perovskites during the processing or fabrication is important. $CsPbX_3$ perovskite NPLs were grown on muscovite mica substrates using the vapor transport CVD method [106]. Shamsi *et al.* improved the synthesis of $CsPbBr_3$ NSs by introducing shorter alkyl chain ammonium ligands to the reaction [108]. The results showed that the lateral size of the NSs is tunable by varying the ratio of shorter ligands over longer ligands. The perovskite multiple quantum wells (MQWs) with red light emission were prepared using the solution-processed self-organized method [109, 110]. Their results demonstrated that the perovskite MQWs are a promising platform that allows achieving high-performance visible-range EL emissions through high-throughput manufacturing processes, which is attractive for low-cost lighting and display applications. In addition, due to the broad tunability of halide perovskites by controlling dimensionality and composition, it is possible to build heterojunctions between different halide perovskites. An example of periodic lateral $CsPbCl_3/CsPbBr_3$ heterojunctions in single NPLs by the cation exchange method and nanofabrication technique was shown by Du *et al.* [111]. 2D lead-free $CsSnI_3$ NPLs have been synthesized for the first time by Weidman [112, 113]. These $CsSnI_3$ NPLs (with thicknesses of less than 4 nm) exhibit significant QEC with PL at 1.59 eV compared to 1.3 eV in the bulk. The results also showed that Sn-rich conditions in $CsSnI_3$ can reduce defect density and increase stability. $Cs_3Bi_2Br_9$ perovskite NPLs were synthesized for the first time via the hot injection method [114]. The $Cs_3Bi_2Br_9$ NPLs were parallelogram-shaped and exhibit an extremely sharp first exciton peak which blue-shifts substantially with respect to their bulk bandgap.

Tin-based inorganic halide perovskites, such as $CsSnX_3$ (X=Cl, Br, I), have been studied as promising candidates that avoid toxic lead halide compositions [115].

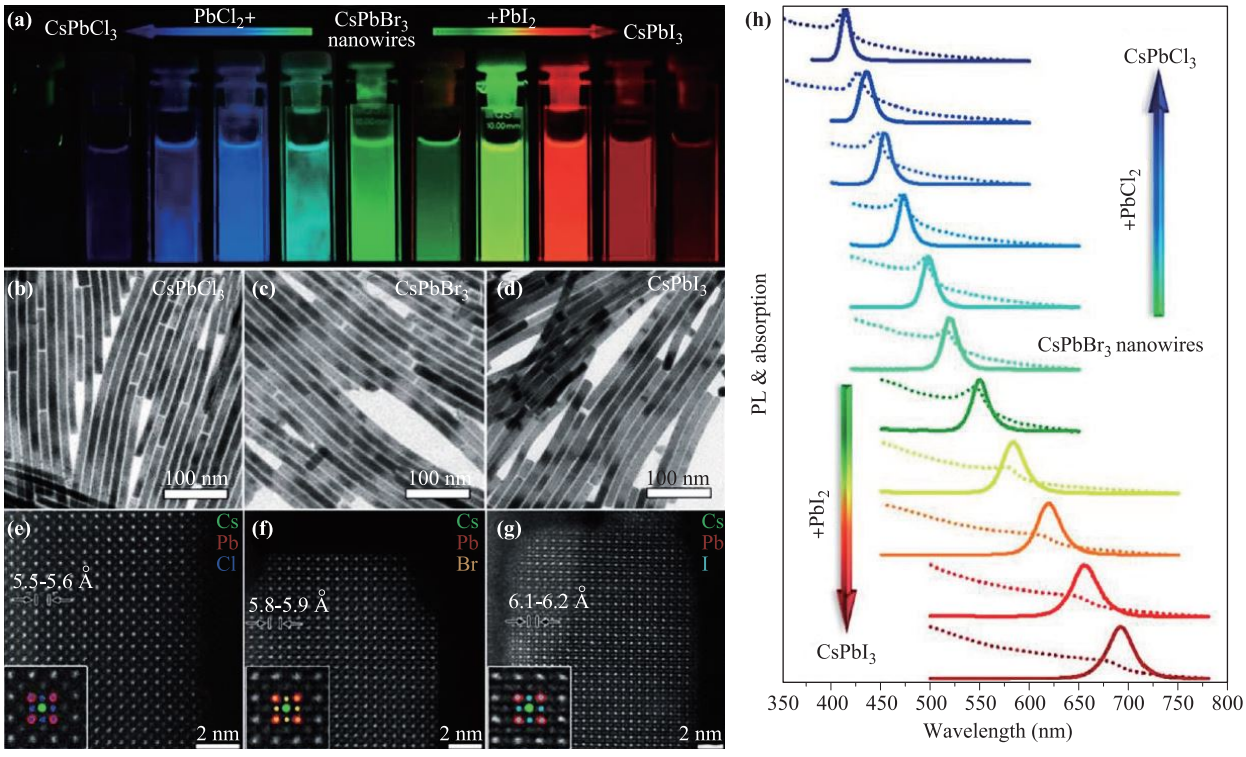

Fig. 3 The photograph, TEM images, UV/Vis absorption and PL spectra of colloidal $CsPbX_3$ (X=Cl, Br, and I) NWs. **(a)** Photograph of colloidal dispersions of $CsPbX_3$ NWs. **(b–g)** Bright-field TEM and high-resolution TEM images of $CsPbX_3$ NWs. **(h)** UV/Vis absorption and PL spectra. Reproduced with permission from Ref. [89].

The single-domain epitaxial growth of $CsSnI_3$ on closely lattice-matched single-crystal potassium chloride substrates was demonstrated [116]. Liu *et al.* introduced a monolayer polystyrene sphere confined growth method for obtaining $CsPbBr_3$ nanonet films (NFs) with ordered nanostructures grown in the preferred (110) orientation, which is beneficial for the charge carrier transport and the light-harvesting efficiency [117]. Recently, high-quality $CsPbBr_3$ film was deposited using the sequential thermal evaporation by Hua *et al.* [118], and they found a new epitaxially-stabilized tetragonal phase for controlling the electronic properties using in situ real-time diffraction techniques. Huo *et al.* prepared single $CsPbBr_3$ NPLs and single-photon emission without blinking is observed using these NPLs [119]. Gao *et al.* proposed a facile colloidal method to obtain $CsPbBr_3$ nanoribbons and deep-blue light emission based on this material is studied [120].

As shown in this section, AIMHP 2D nanomaterials represent an exciting new class of semiconductor nanomaterials. Their excellent absorption and emission properties and facile synthesis make them a promising material for many applications. However, there is still a lot of work to be done on how to obtain and utilize the strong QCE in 2D nanomaterials compared to 0D and 1D AIMHP nanomaterials. In Table 1, we summarized the compositions, shapes and optical properties of $CsPbX_3$ nanostructured material.

3 Optical properties of all-inorganic metal halide perovskite nanomaterials

In the second part, we summarized the advances in the synthesis of AIMHP nanomaterials based on their different dimensions. At the same time, we also introduced in detail the material preparation method, structure, morphology, size and related optimizati the material properties, especially the photoelectric properties. In this part, we will further summarize the excellent optical properties of AIMHP nanomaterials relative to bulk materials, mainly including the following three aspects:

3.1 Strong quantum confinement effect

As is known to all, one of the biggest characteristics of nanomaterials is that the QCE in the optical properties of materials can be observed when the size of an AIMHP is too small to be comparable to the Bohr radius of excitons. Excitons are confined in all three spatial dimensions, which results in a transition from continuous to discrete energy levels. Theoretical calculation shows that the exciton Bohr radius is about 7 nm of all-inorganic $CsPbBr_3$ PNCs; the QCE is quite prominent in $CsPbBr_3$ perovskite NCs when its size becomes comparable with the exciton Bohr radius. For instance, the emission of

Table 1 Comparison of the photophysical properties of IMH perovskite nanocrystals with different compositions and shapes.

Materials	Shapes	PL peaks (nm)	PLQY (%)	Reference
$CsPbX_3$	nanocrystals	410–700	90	[19]
$CsPbBr_3$	nanocrystals	480–560	90	[54]
$CsPbX_3$	quantum dots	410–700	72	[55]
$CsPbX_3$	nanocrystals	410–700	90	[57]
$CsPbBr_3$-$CsPb(Br/I)_3$	nanocrystals	520–580	75	[59]
$CsPbX_3$	nanocrystals	410–700	20–80	[60]
$CsPbBr_3$	nanocrystals	390–660	1–78	[61]
$CsSnX_3$	nanocrystals	607–696	~	[62]
$CsPbX_3$	quantum dots	blue-red	70–95	[63]
$CsPbX_3$	quantum dots	505	80	[64]
$CsPbX_3$	nanocrystals	408.7–691.3	15.93–91.56	[67]
$CsPbBr_3$-Cs_4PbBr_3	nanocrystals	425–575	~	[68]
$CsPbBr_3$/CdS	quantum dots	514	88	[69]
$CsPbI_3$	nanocrystals	648–692	~	[72]
$CsPbBr_3$	nanowires	442	30	[25]
$CsPbX_3$	nanorods	515	~	[64]
$CsPbX_3$	nanorods	505	34	[74]
$CsPbBr_3$	nanowires	475	~	[78]
$CsPbX_3$	nanowires	full visible spectrum	80	[79]
$CsPbX_3$	nanowires	entire visible spectrum	20–80	[80]
$CsPbCl_3$	nanowires	410–460	~	[81]
$CsPbX_3$	nanowires	420–710	~	[83]
$CsPbBr_3$	nanoplates	405–488	84	[28]
$CsPbBr_3$	nanoplates	452	33	[29]

$CsPbBr_3$ perovskite NCs can be truly tuned from around 2.7 eV to 2.4 eV with the size changing from 4 nm to 12 nm which is in good agreement with the theoretical calculation [61, 121, 122]. A large number of experimental results show that the QCE is also observed in 1D [123] and 2D [104, 113] AIMHP materials. The QCE provides a way to tune the emission of semiconductors, which results in potential for various light-emitting applications.

3.2 Wide range of adjustable optical properties

Carry on the discussion in the previous section, the bandgap and optical absorption (emission) properties could be tuned by changing the size of the AIMHP nanomaterials with the decrease of the crystal size. Kovalenko and co-workers first fabricated the $CsPbX_3$ structure with exceptionally tunable optical properties [38]. Compared to other low dimensional nanostructured materials, an attractive feature of AIMHP is facile tunable emission throughout the whole visible range, achieved through controlling the material composition, dimensions and structure. The emission of all inorganic-perovskite $CsPbX_3$ (X=Cl, Br, I, or their mixture), including QDs, QWs and NPLs, can be well-tuned from 400 nm (blue) to 700 nm (red), which covers the whole visible region [23, 124, 125]. Even, the emission of perovskite can be further tuned to the near-infrared or ultraviolet region through particle replacement method and new device structure [126].

3.3 High quantum yield efficiency

Quantum efficiency, defined as the ratio of the number of converted photons to absorbed photons, is one of the most crucial properties for light emitters [127–136]. High quantum efficiency usually signifies that most of the absorbed photons were converted through radiative recombination processes rather than non-radiative recombination processes. Perovskites are regarded as excellent light emitters due to their large absorption coefficient and high quantum efficiency [137]. High quantum efficiencies up to 96.9% have been reported in both all-inorganic $CsPbX_3$ and organic-inorganic methylammonium lead halide perovskite nanocrystals without any further surface treatment [138]. The high quantum efficiency in perovskite is the result of a clear bandgap with negligible charge-trapping states, which greatly promote the exciton radiative recombination efficiency. With their high quantum efficiency, perovskites are promising alternatives for light-emitting applications [139, 140].

4 Applications of all-inorganic metal halide perovskite nanomaterials

AIMHP nanomaterials have attracted great interest in recent years due to their good device performance with higher thermal stability than that of OMHP and HOIMHP. These $CsPbBr_3$ nanowires can be potentially used for nanoscale photonic, electronic, and optoelectronic devices, including SCs, LEDs, lasers, PDs, single-photon quantum sources (SPQS), and so on.

4.1 Solar cells

From the first report in 2015 to now, the PCE of $CsPbX_3$-based perovskite SCs (PSCs) has abruptly increased from 2.9% to 23.7% with much-enhanced stability [141–143]. High-efficiency PSCs were realized by tuning the bandgap and stabilizing the black perovskite phase of $CsPbI_2Br$ at lower temperatures. The device can be realized 9.8% power conversion efficiency (PCE) and over 5% stabilized power output [144]. Zhou *et al.* made inverse opal perovskite SCs based on the slow-photon effect of carbon QD sensitized $CsPbBr_3$ [145]. Compared with planar $CsPbBr_3$, the perovskite SCs PCE can be up to 8.29%

and incident photon-to-electron conversion efficiency can be up to 76.9%. It is worth mentioning that QD modified bifacial PSCs were reported by Li *et al.* [146]. The SC without encapsulation retains around 95% of the initial efficiency at 60% room temperature over a period of 1000 h. Tang *et al.* studied the effect of annealing temperature on $CsPbBr_3$ SCs, which show excellent stability in ambient air with the humidity up to 70% [147].

On the basis of the previous research, the scientists further adopt the material and device optimization techniques to improve the characteristics of SCs. For instance, interface engineering (self-passivation and electron transporting layer) and defect-engineering were introduced to the stability and QCE of the PSCs [148–152]. Scientists adopt a new precursor pair (cesium acetate/hydrogen lead trihalide and DMAI/DMAPbIx) or single-source thermal evaporation to fabricate high-quality $CsPbX_3$ perovskite film and to improve the stability of the device [153–155]. Nanocrystalline controlled synthesis methods were widely used methods to improve the properties of materials and devices [156–158]. Besides, the study found that designing new inverted structure was critical to realize efficient and stable PSCs. The results showed that the devices based on new structure exhibit improved optoelectronic properties, more favorable interfacial energetics, and reduced recombination due to an improved trap passivation effect [159, 160].

4.2 Light emitting diodes

In 2015, Song *et al.* fabricated tunable LEDs based on all-inorganic perovskite $CsPbX_3$ (X=Cl, Br, I) QDs for the first time [161]. The external quantum efficiencies (EQE) of the blue, orange, and green LEDs were 0.07%, 0.09%, and 0.12%, respectively. This study showed that fully inorganic perovskite can be used as the emitter of LEDs devices, although the EQE is still relatively low. Since then, scientists have taken various measures to improve the stability [54, 162–164], quantum efficiency [165–167] and color purity [168] of materials and devices. In the past five years, the performances (EQE, current efficiency, power efficiency, luminance or brightness, lifetime and voltage) of LEDs based on AIMHP have been greatly enhanced [169–173]. Currently, the highest EQE of all-inorganic LEDs is above 20% [174], which is comparable to the state-of-the-art organic LEDs. The results also showed that the operational stability of the light-emitting devices based on $CsPbBr_3$ QDs was 36 times higher because the surface ligand density of the corresponding QDs was lower.

So, the scientists further adopt the material and device optimization techniques to improve the characteristics of LEDs [175–180]. For example, interface engineering (interfacial passivation strategy) was introduced to prompt the efficiency and stability of perovskite QD LEDs [175–178]. Song *et al.* employed a tetrabutylammonium bromide to reduce nonradiative defects of $CsPbBr_3$ and it results in further promotion of electroluminescence performance of LED [175]. A two-step ligand-exchange strategy was developed by Shan et al. to fabricate all-inorganic perovskite $CsPbX_3$ (X=Br, Cl) QDs with halide-ion-pair ligands. Green and blue LEDs made from the halide-ion-pair-capped QDs exhibit high EQE compared with the untreated QDs [176]. Xu *et al.* proposed a bilateral passivation strategy through passivating both top and bottom interfaces of $CsPbBr_3$ QD film, which has drastically enhanced the efficiency and stability of perovskite LEDs with a maximum EQE of 18.7% and current efficiency of 75 cd A^{-1} [178]. Besides, device engineering technology was also used to improve LED efficiency and performance [177–180]. Some strategies such as improving the hole injection, enhancing the electron injection, increasing the charge balance and decreasing the charge leakage, and so on have been adopted to promoting efficiency and stability of perovskite LEDs.

4.3 Lasing

The emission wavelength tunability demonstrated in perovskite materials was an attractive property for nanoscale lasers. Cesium lead halides offer a robust alternative without sacrificing emission tunability or ease of synthesis. Eaton *et al.* first reported all-inorganic $CsPbX_3$ NW lasers in 2016, as shown in Fig. 4 [81]. The Fabry–Perot (F–P) perovskite NWs lasing displayed an excellent performance with a threshold of ~5 μJ/cm^2 and maximum quality factor (MQF) of ~1009±5 when pumped by 400 nm pulsed source. Remarkably, the lasing can be maintained for over 1 h when persist upon exposure to ambient atmosphere, showing much better performance than the hybrid perovskite NW lasers. Fu *et al.* reported a single-crystal $CsPbX_3$ NWs for lasing [181]. They synthesized $CsPbBr_3$ NWs with different lengths ranging from 5–21 μm and realized either single or multiple lasing with thresholds ranging from 2.8 μJ/cm^2 to 9 μJ/cm^2. The NW laser can output stable lasing emissions for longer than 8 h using continuous pumping. Park *et al.* synthesized the $CsPbX_3$ NWs with lengths of about 2–15 μm by using the CVD method and realized the lasers with low thresholds of 6, 3, and 7 μJ/cm^2, with MQFs of 1200, 1300, and 1400 for $CsPbI_3$, $CsPbBr_3$ and $CsPbCl_3$, respectively [182]. Zhang *et al.* comprehensively investigated the strong exciton-photon coupling in micro/nanowires system with low-threshold polariton lasing at room temperature [183]. Large-area $CsPbX_3$ NW laser arrays with lengths of ~10–20 μm were achieved by vapor growth method with a low threshold of ~4 μJ/cm^2 and high MQF of ~2256 [184]. Zhu *et al.* further realized the CW lasing in $CsPbBr_3$ NWs with a threshold of ~6 kW/cm^2 [185]. Wang *et al.* reported the multiphoton-pumped lasing from $CsPbX_3$ nanorods with thresholds of ~0.6 and 1.7 mJ/cm^2 under excitations of 800 and 1200 nm with 80 fs and 1 kHz [186].

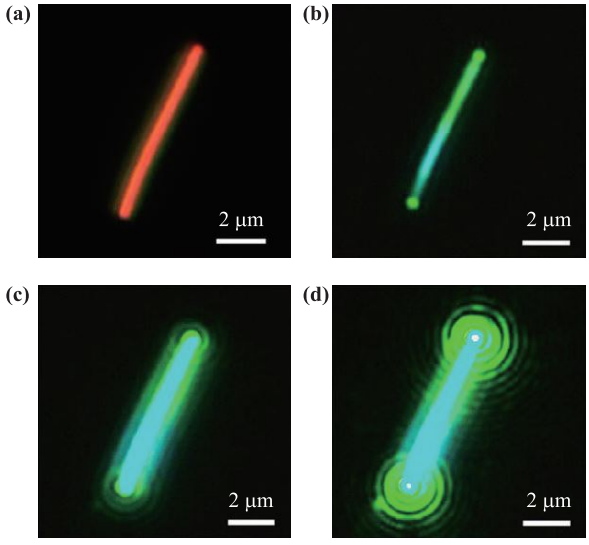

Fig. 4 Lasing in single-crystal $CsPbBr_3$ NW. **(a)** Dark-field image of a $CsPbBr_3$ NW; **(b–d)** the NW from (a) under excitation from a femtosecond pulsed laser with increasing excitation fluence. Reproduced with permission from Ref. [81].

Meanwhile, Hu *et al.* demonstrated F-P lasing in $CsPbBr_3$ microcubes with high MQF of 1150 under optical excitation (800 nm, 35 fs, 1 kHz) [187].

Temperature-dependent lasers based on AILHP have been studied in depth, because nanostructured lasers are very sensitive to temperature [188–191]. At the same time, scientists using new material system and material structure or device engineering have been used to improve the detector's properties [192–200]. Pushkarev *et al.* reported a novel approach to fabricate high-quality $CsPbBr_3$ nanolasers obtained by rapid precipitation from dimethyl sulfoxide solution sprayed onto hydrophobic substrates at ambient conditions [193]. In-plane self-assembly $CsPbBr_3$ NWs and $CsPbBr_3$ QDs embedded in silica sphere were used to improve the stability and performance of $CsPbBr_3$ nanolasers [194, 195]. In addition, inorganic and hybrid perovskites have been exploited to boost the external luminescence efficiency of lasers [197–199]. Recently, Polushkin *et al.* provided a comprehensive analysis of single-particle perovskite lasers with various compositions and shapes [200].

4.4 Photodetectors

PDs have many important applications including imaging, optical communication, remote control, chemical/biological sensing and so on. For an ideal PD, it is necessary to have high spectral selectivity, high signal-to-noise ratio, high sensitivity, high stability, and high speed. Dong *et al.* for the first time demonstrated the visible light PDs with synergetic effect of preferred-orientation and plasmonic effect based on the all-inorganic perovskite $CsPbBr_3$ NCs [201]. And, the photocurrentwas increased more than 200% after introducing the Au NCs into the device, which was contributed to the near field enhancement effect between $CsPbBr_3$ NCs and Au NCs. Their results suggested that all-inorganic perovskites are promising semiconductors for high-performance solution-processed PDs, as shown in Fig. 5. Next year, low-voltage PDs with high responsivity based on all-inorganic scattered $CsPbBr_3$ nanoplatelets are fabricated [202]. The photoresponsivity of PDs based on these nanoplatelets is as high as 34 $A{\cdot}W^{-1}$, with an external driven voltage of 1.5 V under illumination using a 442 nm laser. To improve the responsivity, a planar structure perovskite PDs based on the $CsPbBr_3$ nanosheet/CNT composite films are constructed and exhibit both high light-harvesting and high conductivity [203]. A high EQE of 7488% and high responsivity of 31.1 $A{\cdot}W^{-1}$ under a bias of 10 V are achieved.

In addition to the 0D and 2D $CsPbBr_3$ nanomaterials, 1D AILHP was widely used to prepare optical detectors with high stability and high detection rate [204–210]. Then, superior PDs based on a 1D $CsPbI_3$ nanorods were constructed, with a responsivity of 2.92×10^3 $A{\cdot}W^{-1}$, an ultrafast response time of 0.05 ms and a detectivity approached up to 5.17×10^{13} Jones [204]. Stabilized Sn-doped $CsPbI_3$ nanobelts with satisfied stability for up to 15 days when maintained under air condition were successfully synthesized. The as-constructed PDs based on the $CsPb_{0.922}Sn_{0.078}I_3$ nanobelts displayed an ultrahigh detectivity of up to 6.43×10^{13} Jones [205]. A sensitive deep ultraviolet (DUV) light PD based $Cs_3Cu_2I_5$ was reported [200]. The PD exhibited pronounced sensitivity to both DUV and UV light illumination with response speeds of 26.2/49.9 ms for rise/fall time.

Self-powered photodetectors (SPPDs) with no external power sources, high-sensitivity and fast response speed is an important type of PDs. Self-powered all-perovskite PDs with high detectivity, high-sensitivity and fast response speed were reported by different research groups [211–215]. At the same time, the scientists further adopted the material and device optimization techniques such as interfacial engineering, inverse structure, core-shell technique, vertical Schottky junction, and fast interfacial charge transfer layers to improve the detector's properties [216–220]. The experimental data showed that the enhanced device performance is attributed to the improved crystallinity and less surface defects of AIMHP nanomaterials.

4.5 Applications in other optoelectronic devices

In addition to the nanoscale photonic and optoelectronic devices, including SCs, LEDs, lasers and PDs, AIMHP nanomaterials were also widely used in other devices. A facile design of hybrid phototransistors with an outstanding responsivity of 4.9×10^6 $A{\cdot}W^{-1}$, fast response of 0.45 s/0.55 s, and long-term stability of 200 h in ambi-

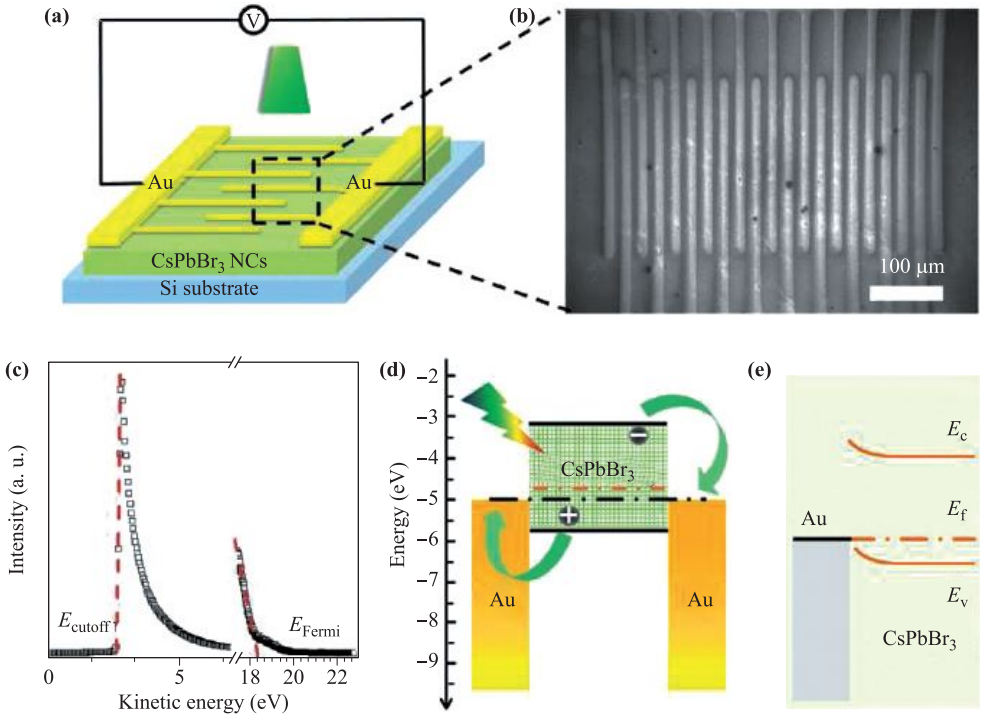

Fig. 5 The PD based on $CsPbBr_3$ NCs. **(a)** Schematic demonstration of device configuration; **(b)** The microscope image of interdigitated electrodes; **(c)** Ultraviolet photoelectron spectroscopy curve of $CsPbBr_3$ NCs; **(d)** Band profile of the $CsPbBr_3$ PD. **(e)** Schottky barrier between Au and $CsPbBr_3$ NCs. Reproduced with permission from Ref. [201].

ent based on the $CsPbBr_3$ thin film is reported [221]. Polarization-sensitive light sources based on well-aligned $CsPbBr_3$ and $CsPbCl_3$ NWs were reported [222]. Recently, a field-effect transistor (FET) has been fabricated using the all-inorganic $CsPbBr_3$ NCs [223, 224]. The $CsPbBr_3$-NC transistors exhibit a clean unipolar transport characteristic in a p-type mode featuring well-defined linear and saturation regimes.

Except for the conventional optoelectronic device described above, AIMHP has been employed as next-generation light sources, such as SPQS [224, 225]. The $CsPbX_3$ (X=Cl/Br) NCs exhibit stable, narrow-band emission with suppressed blinking and small spectral diffusion at low-temperature. Photon demonstrates unambiguously nonclassical single-photon emission with radiative decay on the order of 250 ps, representing a significant acceleration compared to other common quantum emitters.

5 Conclusions and outlook

In summary, taking all-inorganic perovskite $CsPbX_3$ (X=Cl, Br, I or mixture of halides) nanomaterials as representative, we provide an overview of the up-to-date developments in synthesis, attractive optical properties and related emerging applications of AIMHP nanomaterials. First of all, we can see that the material compositions, dimensionality, morphologies and structures of AIMHP nanomaterials are an important factor affecting the optical properties and performance of the device. Besides that, as shown in the paper, through adopting the material and device optimization techniques such as interface engineering (self-passivation and electron transporting layer), defect engineering, and device engineering (inverting device structure, enhancing the hole and the electron injection, increasing the charge balance, decreasing the charge leakage and so on) others in constructing optoelectric devices, the performance could be improved remarkably. Therefore, continuing to study the effects of material and device optimization techniques on nanostructured light-emitting devices from the microscopic perspective and understanding the correlation between the properties of nanostructured devices in depth is crucial. Even though this progress in the improvement of the stabilities and performances of AIMHP nanomaterials and devices is very encouraging, there are several shortcomings and challenges that stand in the way of commercialization. The following aspects are critical to improving the performance of devices.

1) Understanding of the physics in low-dimensional perovskites

   Since tremendous optoelectronic applications have been demonstrated, the investigations focusing on fundamental properties of low-dimensional perovskites

have not attracted much attention. With the reduced dimensions, some inherent natures like band structure and photon transport will behave differently. However, the study on this aspect is limited and more efforts should be devoted to further exploration. In addition, the low dimensionality could also bring about confined charge transport in the selected directions, leading to unique electrical properties such as 2D electron gas, which is the basis for field-effect devices. However, these fundamental investigations lag far behind the application. We have drawn more attention in the near future [226–228].

2) Precise control of the size and dimensionality

As one of the most valuable merits with reduced dimensionality, size and dimensional control for various bandgaps attract tremendous attention. Nowadays, although some groups can manipulate the perovskites with size and shape control to some degree, most synthesis methods for low-dimensional perovskites are trial-and-error, without a precise regulation. We are inquisitive whether this approach can be utilized as the guidelines for the synthesis of perovskites with accurate structures and dimensions we desire. Based on these, more explorations of in-depth physical mechanism and wide applications will be duly concomitant [229, 230].

3) Pb-free metal halide NCs

Concerning the environmental issues, lead-free IHPs are necessary and more appealing. As we have shown in part in this article, colloidal metal halide NCs containing tin (Sn), germanium (Ge), indium (In), bismuth (Bi), and antimony (Sb) can be explored as a replacement for Pb halides. The development of environmentally benign lead-free halide perovskites with comparable excellent features and exploration of their fundamental optical and electronic properties and environmental stability are interesting, such as Cs_2AgBiX_6, $Cs_3Bi_2X_9$, $Cs_3Sb_2X_9$, $CsEuCl_3$ and other analogues [231, 232].

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