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Self-trapped Excitons in Metal Halides for Lighting Applications

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Abstract: Metal halides have attracted worldwide attention as exceptional optoelectronic materials. Over the past decade, research on metal halides has yielded remarkable progress, and their color-conversion applications have shown considerable promise for commercialization. With the reporting of self-trapped exciton (STE) emission in perovskites, the application of metal halides as broadband emitting materials in the lighting field has gained increasing interest. Herein, we provide a comprehensive review of metal halide STE emitters, especially for lighting applications. We begin with highlighting the ideal spectral characteristics and corresponding performance metrics for lighting. This is followed by a systematic summary of the mechanisms, optimization strategies, and recent advances of STE emission in metal halides. Finally, we outline the major challenges and prospective trends for metal halide STE emitters. This review aims to offer valuable insights into metal halide STE emitters and their lighting applications for facilitating the future commercialization.

Keywords: self-trapped exciton; metal halide; lighting; optoelectronic applications
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金属卤化物中的自陷态激子助力照明应用

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摘要：金属卤化物作为卓越的光电材料引起了全世界的关注。在过去的十年中，金属卤化物的研究取得了显著进展，其颜色转换应用展现出巨大的商业化前景。随着钙钛矿中自陷态激子(STE)发射被发现和报道，将金属卤化物作为宽光谱发光材料应用于照明领域的研究越来越受到关注。本文对金属卤化物自陷态激子发光体进行了全面的综述，特别是用于照明应用。我们首先强调了照明的理想光谱特征和相应的性能指标。随后系统总结了金属卤化物自陷态激子发光的机理、优化策略和最新进展。最后，概述了金属卤化物自陷态发光体的主要挑战和前景趋势。本文旨在为金属卤化物STE发射器及其照明应用提供宝贵的见解，以促进未来进一步的商业化。

关　键　词：自陷态激子；金属卤化物；照明；光电子应用

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1 Introduction

Benefiting from a comprehensive understanding of down-conversion materials and the underlying theories, color conversion has long been an indispensable technology in the field of lighting. Its excellent compatibility and spectral broadening capability support diverse applications from outdoor to indoor lighting. Conventional phosphors such as YAG∶Ce^{3+} with yellow emission under short-wavelength excitation can be blended with a blue component to generate white light emission[1-3]. However, such systems are limited by inherent drawbacks, including discontinuous broad spectra, insufficient absorption, and reduced luminescence efficiency[4-5].

Metal halides (MHs) have emerged as promising candidates for optoelectronic applications due to their high color purity, exceptional efficiency, and cost-effectiveness[6-8]. Initially, MHs were employed in advanced color-conversion displays owing to their efficient free-exciton (FE) emission with high color purity[9]. Subsequently, self-trapped exciton (STE) emission was discovered in metal halide perovskites (MHPs), particularly in double perovskites (Fig. 1 (a)), making them equally attractive for color-conversion lighting[10-11]. Over the past decade, continuous exploration and optimization have led to significant improvements in the efficiency and spectral breadth of STE emission in MHs[12-15]. Several lighting prototypes and advanced applications have already been demonstrated[16-18]. Undoubtedly, these progressive investigations on STE emission will substantially contribute to the commercialization of MH-based lighting in the near future. Nevertheless, it is important to recognize that both MHs and their STE emission still face several challenges[19-21]. A systematic summary of achieved progress and potential improvement strategies is essential to guide future research and development.

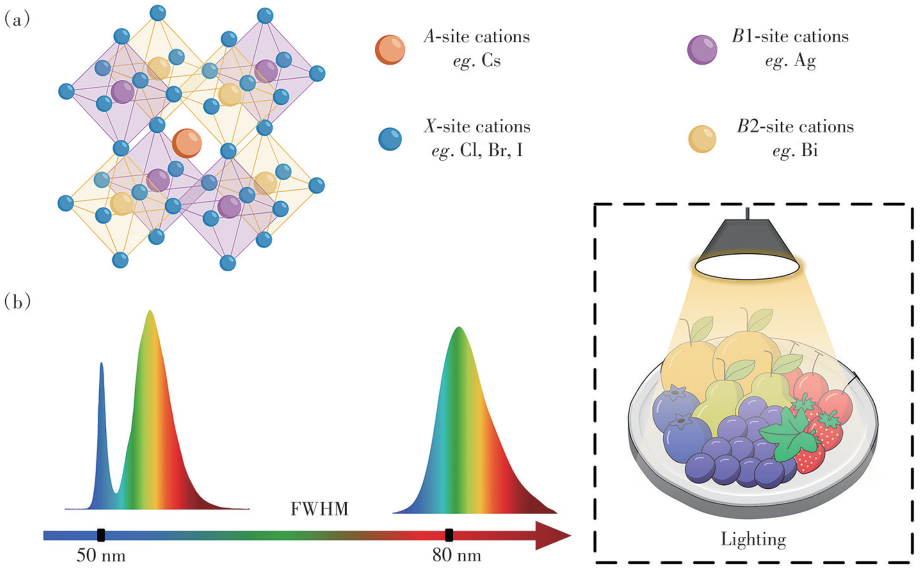

Fig.1 (a) The structure of metal halide double perovskites, which are known as efficient STE emitters. (b) The spectral trends in MHs for lighting applications

This review focuses on the origins, enhancement strategies, and potential development directions of STE emission in MHs for lighting applications. We begin by introducing the importance of broad and continuous visible spectra for illumination, which are the potential advantages of STE emission. The fundamental mechanisms and optimization pathways of STE emission in MHs are then summarized to guide subsequent studies. Based on these strategies, recent advances in highly efficient STE emitters are reviewed. Finally, key challenges toward the commercialization of MHs with STE emission are discussed. It is believed that this review will offer valuable insights and inspiration for the future optimization and application of STE emission in MHs.

2 Lighting

The majority of lighting applications require white light to accurately replicate the appearance of objects under sunlight[22]. However, mature GaN-based light-emitting diodes (LEDs) typically emit only monochromatic light, making color converters essential for achieving white light through color mixing[23-25]. Consequently, the overall performance of the lighting device is largely determined by the properties of the color converter. Specifically, the luminous efficacy(lm/W) of an individual lighting device is closely related to the light utilization efficiency (for blue or ultraviolet excitation) and the light conversion efficiency (LCE) of the color converter[26-27]. Here, we emphasize that high light utilization can be achieved by enhancing the absorbance of the color-conversion material, while the LCE is governed collectively by both the absorbance and the photoluminescence quantum yield (PLQY).

For most conventional materials, achieving uniform size distribution and high radiative recombination often requires complex and precise processing procedures. In contrast, MHs can attain near-unity PLQYs and desirable emission spectra without numerous treatments, owing to their inherent high defect tolerance and low aggregation-induced losses[28-29]. Furthermore, MHs exhibit notably high absorption coefficients, which is beneficial for short-wavelength light utilization[30-31]. These attributes underscore the significant potential of MHs in emerging optoelectronic devices. A large number of studies have demonstrated the suitability of MHs for lighting applications, especially those exhibiting STE emission[32-33]. Therefore, we begin with emphasizing the distinct requirements of lighting, which may serve as a foundational guide for the subsequent customization of materials and device architectures.

In lighting, correlated color temperature (CCT) and color rendering index (CRI) are used to assess the white color performance of devices and the color reduction of objects, respectively (Fig. 1(b)). The sun, as an ideal lighting source, shows a suitable CCT between 2 800-6 500 K and a CRI of 100[34-36]. It is found that both CCT and CRI are strongly related to the emission spectrum of the lighting source. That means the emission spectrum is expected to cover as wide a visible band as possible and be located on the CCT curve[37-38]. Traditional phosphors and nanocrystals (NCs) usually exhibit monochromatic emission, such as rare earth ion-doped ceramic phosphors (blue $BaMgAl_{10}O_{17}:Eu^{2+}$, green $MgAl_{11}O_{19}:(Ce^{3+}, Tb^{3+})$, and red $Y_2O_3:Eu^{3+}$), quantum dots (CdSe/ZnS, InP), and so on[39-42]. Therefore, it is common to use more than two phosphors or NCs for white color with a broader spectrum[43-44]. This usually leads to severe cross-absorption or conversion, which directly affects the LCE and spectral stability. Meanwhile, the non-uniform distribution of different materials will aggravate the color uniformity of the lighting device, due to the Lambertian distribution of LEDs[45]. These disadvantages will ultimately lead to lighting devices with high power consumption and undesirable colors. MHs with STE emission demonstrate high absorption coefficients and efficient color conversion, which makes them one of the strong contenders for the next generation of white light down-conversion materials[46].

3 STE Emission in Metal Halides

As color conversion in MHs advances, it becomes increasingly important and urgent to expand their applications as efficient and colorful emitters in various scenarios. As mentioned above, since the discovery of STE emissions in perovskites, they have been promising candidates for white lighting. However, the STE emission commonly exhibits undesirable performance due to the conflicts between spectra and efficiency. Additionally, the complicated mechanisms and formation processes of STE impede the development of efficient STE emitters in MHs. Therefore, a comprehensive overview and understanding of these mechanisms and improvement strategies are crucial for future progress in this field.

3.1 Mechanism of STE

Unlike FE emissions, STE emissions require lattice distortion-induced traps to capture carriers. This process, known as intrinsic self-trapping (The topmost schematic in Fig. 2(a)), refers to the simultaneous occurrence of transient lattice

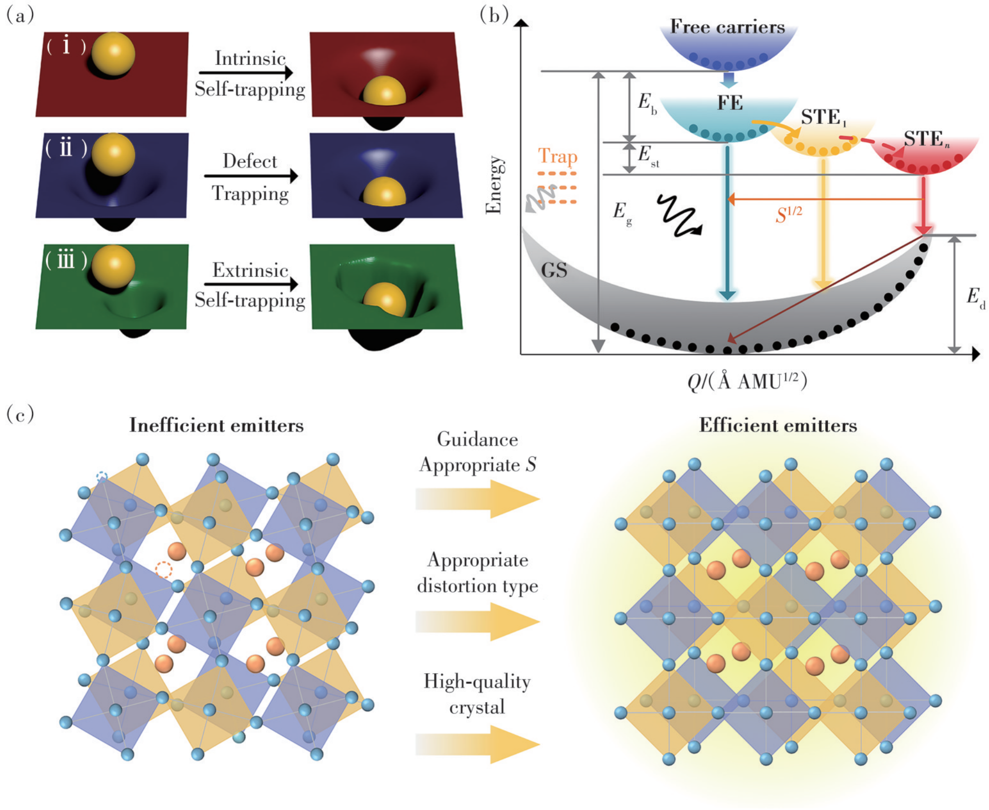

Fig.2 (a)Schematic illustration of three typical trapping modes by a ball interacting with a rubber sheet: (ⅰ)intrinsic self-trapping, (ⅱ)defect trapping, and (ⅲ)extrinsic self-trapping[11]. (b)Configuration coordinate diagrams illustrating the STEs process. (c)Guidance for efficient STE emission in MHs

distortions and carrier capture. Although the other two types of self-trappings caused by defects can also lead to a wide spectrum (Fig. 2(a)), the associated non-radiative recombination is typically considered unfavorable for luminescence performance. Therefore, there is greater emphasis on manipulating lattice distortion to generate STE emission.

The STE emission in MHs is usually attributed to the intrinsic soft lattice and strong exciton-phonon coupling. In MHs, the strength of exciton-phonon coupling is strongly influenced by composition and structural characteristics. The Huang-Rhys factor (S) is proposed to quantitatively evaluate the degree of exciton-phonon coupling, where a value of S significantly greater than 1 indicates strong exciton-phonon coupling[32-33]. Then, the lattice distortion will easily form to trap the exciton due to the strong exciton-phonon coupling. This energy valley, therefore called self-trapped states, will assist the red-shift emission and broadened spectrum.

Although the process of STE emission has been thoroughly discussed, it is still essential to reiterate the mechanisms to guide further improvement strategies[47-48]. From the perspective of energy states, strong exciton-phonon coupling leads to the formation of one or more self-trapped states with lower energy compared with the FE state[49-50]. That means excitons will be trapped by different excited states, and then both FE and STE will release energy to return to the ground state, resulting in emission with a broad spectrum. Taking the excited state of the free exciton as a reference, the position of STE_{1-n} can be given by E_{st} and S, which can be regarded as the reason for the Stokes shift (Fig. 2(b))[51]. Therefore, the wavelength (E_{PL}) and FWHM of emission can be derived according to the following equation[33]:

$$E_{\mathrm{PL}} = E_{\mathrm{g}} - E_{\mathrm{b}} - E_{\mathrm{st}} - E_{\mathrm{d}}, \qquad (1)$$

$$\mathrm{FWHM} = 2.36\sqrt{S}\,\hbar\omega_{\mathrm{phonon}}\sqrt{\coth\frac{\hbar\omega_{\mathrm{phonon}}}{2k_{\mathrm{b}}T}}, \qquad (2)$$

where E_{g} represents the band gap between the ground state and the free carriers, E_b represents the exciton binding energy, E_{st} represents the self-trapping energy (relative to FE), E_d represents the lattice distortion energy, $\hbar$ is Planck's constant, and ω is the phonon frequency.

3.2 Guidance for Efficient STE Emission

As aforementioned, STE in MHs can be enhanced by strengthening the exciton-phonon coupling with a larger S. However, this process involving phonons introduces non-radiative recombination, which is detrimental to light emission. Additionally, the distortions caused by strong exciton-phonon coupling may lead to more defects, potentially exacerbating non-radiative recombination. Therefore, achieving efficient STE emitters requires a trade-off between efficiency and spectral properties (Fig. 2(c)).

Firstly, we discuss the strength of exciton-phonon coupling, represented by the magnitude of S. Considering that phonons are temperature-dependent energy quanta, S can be obtained by directly fitting the temperature-dependent FWHM curve (Eq. (1), (2)). This allows us to determine whether S meets the prerequisite for STE emission. However, an excessively large value of S can lead to a significant decrease in PLQY due to the enhanced non-radiative recombination[52]. Therefore, achieving a well-balanced value of S for highly efficient STE emission is a great challenge. For example, even the highly promising $Cs_2NaInCl_6$ and $Cs_2AgInCl_6$ only exhibit PLQY values below 1%[10]. In addition, the parity-forbidden transitions are obvious in some perovskites with inversion symmetry, leading to low efficiency due to the Laporte rule. Breaking inversion symmetry and allowing transitions is an effective method to enhance the PLQY of emitters, such as increasing the Jahn-Teller distortions and asymmetric vibrations.

The trade-off between different types of distortions is another important consideration. For instance, Jahn-Teller distortion is more conducive to realizing high PLQY in STE emitters. This is because lattice distortion is intrinsically a type of defect, while other distortions will easily exacerbate non-radiative recombination, leading to significant efficiency losses[53]. Ensuring that Jahn-Teller distortion predominates is also beneficial for efficient STE emission, even though multiple distortions can contribute to spectral broadening[54]. Notably, the MHs with BX_6 octahedral exhibit great potential for the formation of Jahn-Teller distortions. Instead, some undesired distortions will introduce additional non-radiative recombination, thus affecting the luminescence. Considering the growth characteristics of MHs, controlling crystal growth perfection can be effective for realizing efficient STE emission, especially in eliminating the defects caused by the distortions.

The final consideration is the appropriate energy level of the FE, especially for white light emission in lighting applications. The FE generally determines the shortest wavelength of the light emission, as STE emission is red-shifted compared to FE emission. To achieve a broad coverage of the visible spectrum, it is crucial to rationally design the band gap between the excited state and ground state of the FE. Typically, a band gap corresponding to ≤450 nm ultraviolet or deep blue emission is preferred, such as $Cs_2AgInCl_6$ (3.02 eV) and $Cs_2NaInCl_6$ (4.55 eV)[10,50,55]. Certainly, the choice of band gap and S needs to be highly matched, since they both determine the redshift of STE relative to FE. This allows for long-wavelength emissions generated by STE to fall within the visible range, resulting in white light emission with a broad spectrum instead of infrared emissions.

The strategies for efficient STE emitters are required to not only address the conflict between broad spectrum and efficiency, but also to effectively eliminate unexpected defects. MHs, for their inherent ionic crystal characteristics, offer a convenient platform for implementing such strategies through component engineering. With ongoing advancements, MHs with STE emission have yielded outstanding achievements in both theoretical understanding and practical applications. In recent years, the primary development systems and performance parameters of STE emitters have been summarized in Tab. 1.

Tab. 1 Advances in STE emitters

Materials	Strategy	PLQY	FWHM/nm	CRI	CCT/K	Stability	Ref.
$Cs_2NaLuCl_6$	doping	91%	216	95	3 656	NA	[4]
					5 584	NA	
					8 568	NA	
$Cs_2AgInCl_6$	doping	91.00%	NA	NA	4 054	1 000 h, ≈97%	[10]
$Cs_2HfCl_6:Sb^{3+}/Ln^{3+}$	doping	≈80%	NA	≈98.1	4 843	50 h, ≈97%	[13]
$[AMP]_2Cu_2PbX_8$	new structure	48.44% (Br)	135(Br)	NA	NA	NA	[14]
X=Br/I		26.64% (I)	105(I)				
Cs_2KInCl_6	doping	86.98%	≈230	85.7	6 570	NA	[16]
$Cs_2AgScCl_6$	doping	> 60%	>280	≈90	4 100	NA	[25]
$Cs_2NaInCl_6$	doping	31.1%	NA	NA	NA	NA	[55]
$Cs_{4-x}A_xSn(Br,I)_6$	doping	(15±5)%	NA	NA	NA	NA	[56]
$Cs_2AgInCl_6$	doping	≈45%	188	NA	NA	NA	[57]
Cs_2KInCl_6	doping	95%(without Mn^{2+})	114(without Mn^{2+})	NA	NA	NA	[58]
		87% (with Mn^{2+})	NA	≈89.5 (with Mn^{2+})	4 252(with Mn^{2+})	NA	
$(DETA)_3InCl_6$	doping	nearly 100%	NA	NA	NA	NA	[59]
$Cs_2NaTbCl_6$	doping	96.07%	NA	NA	NA	NA	[60]

4 Improvement Strategy of the STE Emission

MHs with STE emission can be prepared either as phosphor powders or as colloidal NCs[61-62]. The composition of these MHs is entirely determined by the ionic types and ratios of the precursors, enabling the manipulation of their luminescent properties. As a representative metal halide semiconductor, perovskites demonstrate considerable potential for STE emission. In this section, they are regarded as an example to show typical strategies for enhancing STE. Based on the valence of the B-site, they can be classified into two types: single perovskite and double perovskite. The B-site of single perovskite is occupied by divalent metal cations like Pb^{2+} and Sn^{2+}. Instead, the double perovskite is commonly taken up by positively monovalent and trivalent metal cations like Ag^+, Cu^+, Na^+, Bi^{3+}, In^{3+}, Sb^{3+}, *etc*[63]. The single perovskite to realize stable STE emission is commonly low-dimensional structures such as $A_nB_nX_{3n+1}$ and A_4BX_6[56]. Therefore, early reports on single perovskite have focused on A-site component engineering to realize dimensionality reduction from 3D to 2D (Fig. 3(a)-(b)). This is primarily due to the presence of large A-site cations, which can induce octahedral distortion and generate STE emission. However, the large distortion and deep defects in 2D metal halides will lead to undesirable efficiency[11]. 0D metal halides are another well-established choice for excellent STE emission[56, 64-65]. These 0D nanocrystals show a high radiative recombination rate due to their strong quantum confinement effect (Fig. 3(c)). However, the mechanism of these 0D nanocrystals is more complicated and lacks deep exploration, which limits their further development.

Compared with the common MHs, the double perovskites are another STE system with the potential for non-toxicity (Fig. 3(d)). These double perovskites with different optoelectronic properties can be completely manipulated, especially the PLQY. Since the realization of Bi^{3+}-doped alloyed $Cs_2Na_xAg_{1-x}InCl_6$ with high PLQY by our group, the research on double perovskites with efficient emission has become a hot spot[10]. The origin of this efficient perovskite is that some alloyed double perovskite can form 3D structures with substantial 0D electronic behavior. Certainly, the optimization of double perovskite NCs or phosphors does not necessarily require intentional dimensional reduction to induce distortion. Instead, it can fully focus on achieving appropriate S values, desirable distortion

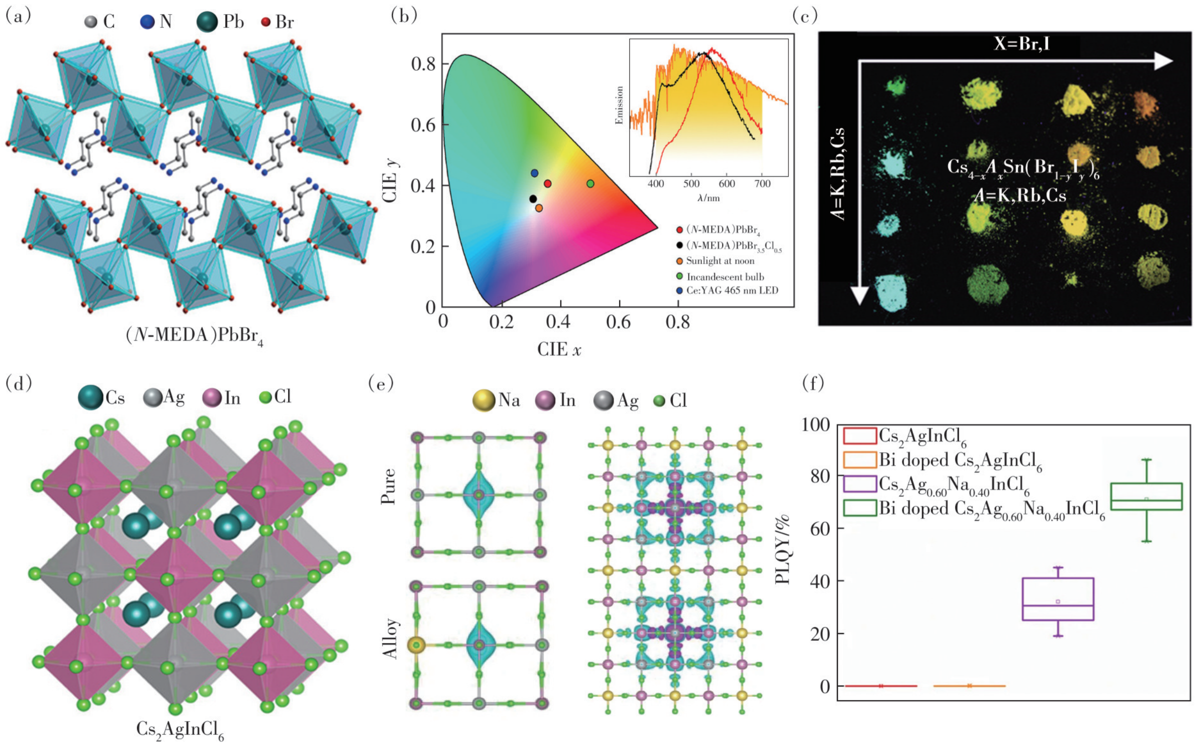

Fig.3 (a) Schematic of the (*N*-MEDA)PbBr$_4$ crystal structures[11]. (b) Chromaticity coordinate diagram for some 2D STE emitters with white light emission[11]. (c) The image of Sn-based perovskite powders with different components under 365 nm UV light[56]. (d) The structure of CsAgInCl$_6$ is regarded as a typical double perovskite[63]. (e) Parity-forbidden property of the electron wavefunction of the STE before and after Na incorporation[10]. (f) The PLQY results of Cs$_2$AgInCl$_6$ and Cs$_2$Ag$_{0.60}$NA$_{0.40}$InCl$_6$ with and without Bi doping[10]

types, and minimizing non-radiative recombination to obtain efficient STE emission. Therefore, the *B*-site doping strategy is widely employed in double perovskites to form alloyed structures. Notably, the rigorous design of component engineering is essential for efficient STE emitters with a double perovskite structure. Obviously, the original double perovskite needs to have a sufficiently high *S* to ensure high exciton-phonon coupling and thus stable broad-spectrum STE emission. Secondly, the ions involved in the alloying are usually required to be able to break the inversion symmetry and thus eliminate the parity-forbidden property, like our chosen Na$^+$ (Fig. 3(e))[66]. At the same time, the alloyed double perovskites need to have similar crystal structures and negligible lattice mismatch. When such components are confirmed, the alloyed double perovskites will show significantly enhanced emission without harmful phase separation and additional defects. On this basis, the trace introduction of some metal ions (like Bi^{3+} and Yb^{3+}) will assist in passivating the defects and improve the perfection of the crystal. These will boost the double perovskites to have a low non-radiative recombination rate and obtain an increased PLQY (Fig. 3(f))[10, 57, 67]. Some studies have shown that the effective component modulations can endow the double perovskite phosphors with a broad spectrum and more than 80% PLQY, which shows great potential for lighting applications. Surely, this strategy will be also effective in NCs synthesized by solution method due to the similar structural and emission principles[50, 68-69].

In addition, there are some studies combining efficient STE emitters and ion-emitting centers to achieve great white light emission[58]. From this perspective, some MHs with FE emission can also be used to compensate for some color gaps. For example, the MHs with FE emission can absorb the extra blue light and fill the deficiency of cyan emission to realize a natural daylight-like spectrum for better lighting[70]. In the future, it is worth rethinking how to realize a natural daylight-like spectrum with a single component.

In summary, the research on MHs with STE emission has gradually adjusted from dimensional engineering to a doping strategy[59]. Continuing to achieve white light emission with 100% PLQY and a natural daylight-like spectrum will be a major goal for MHs with STE emission. At the same time, conducting research to deepen the mechanism understanding of the STE in metal halide nanocrystals and achieving pixelated lighting will also be an important direction.

5 Color-conversion Application for Lighting

Unlike displays with the three-primary-color subpixels, lighting units generally emit white light directly. Similarly, lighting devices can be divided into unpixelated devices and pixelated devices. When the unpixelated device is switched on, it will light the space or object directly, which is convenient and advantageous for most everyday use scenarios (Fig. 4(a)). However, it does have significant drawbacks for the applications where illumination of the specific area is required. In contrast, pixelated lighting devices offer significantly enhanced functionality through local dimming or even independent pixel-level control. Pixelated lighting devices refer to an illumination system composed of mini-LEDs or micro-LEDs arranged in a specific order. A typical application is through the combination of Micro-LEDs as automotive headlights to reduce energy loss (Fig. 4(b)). At the same time, this pixelated lighting source can additionally be used as a simple projection engine to enhance interaction with people. Due to the differentiation between pixelated and unpixelated devices, different dimensions of down-conversion materials are also worth considering. For example, bulk phosphors are important in unpixelated lighting devices, while the low-dimensional NCs are more suitable for use as pixelated lighting devices.

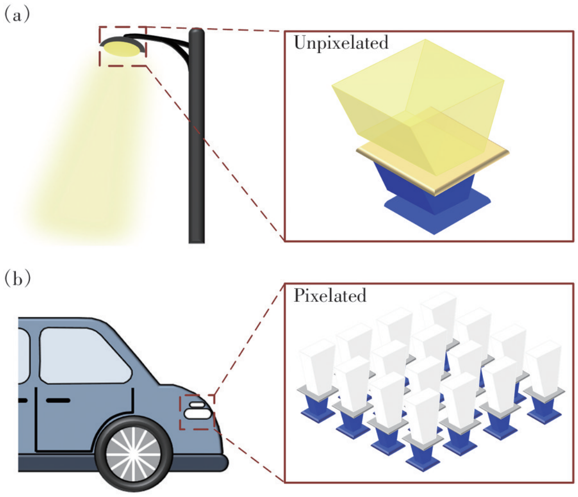

Fig.4 Schematic illustration of pixelated and unpixelated lighting source: (a) unpixelated device used in space and object illumination, (b) pixelated device used in automotive headlights

6 Future Development for STE Emitters

Firstly, current research efforts are largely focused on expanding the range of material systems exhibiting STE emission. The relationship between the specific characteristics of STEs and the underlying physics in different materials remains to be further elucidated. A deeper understanding of specific material systems is essential for optimizing the performance parameters of individual systems and assessing their potential for practical engineering applications.

Secondly, it is essential to emphasize the stability of color converters[9]. Lighting applications typically require high-power short-wavelength excitation under continuous operation, imposing extremely stringent stability requirements on the converters. Although most STE emitters are fabricated into bulky powder with improved stability, their intrinsic ionic crystal structure and soft lattice nature remain limiting factors[71]. Therefore, their stability should be enhanced by following two aspects: improving the intrinsic stability of the material and developing encapsulation strategies. Both approaches require further investigation into underlying degradation mechanisms (such as photo-induced and thermal-induced degradation) to guide the long-term stability strategies[72-73].

Thirdly, sustainability considerations must be addressed. Unlike the advantageous FE emission for displays, STE emission is often highlighted in lead-free systems, particularly in double perovskites[10]. Since double perovskites can achieve efficient STE emission without lead or cadmium, they hold great potential for next-generation green lighting technologies[10]. Such eco-friendly lighting systems would be well-suited for indoor human-centric healthy lighting applications[60]. Moreover, the fabrication process of color converters should minimize harm to humans and the environment. Green and healthy lifecycle management of STE emitters from synthesis to disposal will be an important focus for future research[74].

Furthermore, advanced fabrication methods deserve emphasis. Conventional powder or nanocrystal synthesis routes are often inefficient and incompatible with integrated devices. Vacuum deposition has emerged as a novel and efficient approach for fabricating monolithically integrated devices. Our research group has previously demonstrated its advantages in electroluminescent devices based on metal halide STE emitters[75]. Extending this method to the fabrication of STE emitters could potentially revolutionize application formats and scenarios[76]. Other advanced manufacturing techniques also offer routes to pattern bulk STE emitters into microscale structures or functional devices, such as nanoimprinting, inkjet printing, and direct laser writing[77-79]. These techniques could benefit emerging miniaturized lighting applications, including medical fiber illumination and automotive headlight projection[60].

Finally, we believe that the potential of metal halide STE emitters in other optoelectronic applications deserves emphasis, particularly as progress continues to be reported. To explore broader applications, our group recently demonstrated the capability of STEs in achieving efficient blue emission and exciton spin polarization. One study employed STE to assist the realization of the highly efficient deep-blue LEDs with color coordinates approaching the Rec. 2020 standard, supporting the development of wide-color-gamut electroluminescent displays[75]. Another work highlighted the potential of STEs for fabricating efficient and environmentally friendly tunable spintronic devices[80]. More efforts should be devoted to exploring the diverse characteristics of STE emission, which will benefit the development of a broader range of semiconductor devices.

Response Letter is available for this paper at: http://cjl.lightpublishing.cn/thesisDetails#10.37188/CJL.20250239

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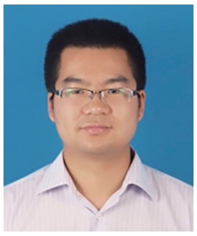

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