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Enhanced optical performance of $FAPbBr_3$-MOF composite films

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Abstract. Metal halide perovskite nanocrystals have emerged as excellent candidate materials for various optoelectronic applications due to their distinguished optoelectronic properties. However, suffering from instability under environmental conditions such as humidity, temperature and ultraviolet radiation restricts its further development and larger-scale application. In this study, we present a one-step method for fabricating composite thin films of formamidinium lead bromide and a lead-based metal-organic framework with enhanced stability. Meanwhile, the obtained composite films exhibited a low amplified spontaneous emission threshold of 12.3 $\mu J \cdot cm^{-2}$ enhancing their potential for efficient optoelectronic applications, including light-emitting devices and lasers.

Keywords: perovskite, metal-organic framework, composites, enhanced spontaneous emission

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Улучшенные оптические характеристики композитных пленок $FAPbBr_3$-MOF

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Аннотация. Нанокристаллы металлогалогенидных перовскитов стали отличными материалами-кандидатами для различных применений в оптоэлектронике благодаря своим выдающимся оптоэлектронным свойствам. Однако нестабильность в условиях

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окружающей среды, таких как влажность, температура и ультрафиолетовое излучение, ограничивает их дальнейшую разработку и широкомасштабное применение. В этом исследовании мы представляем композитные тонкие пленки перовскита FAPbBr3 и металлоорганического каркаса на основе свинца с повышенной стабильностью, полученные одноэтапным методом. При этом полученные композитные пленки продемонстрировали низкий порог усиленного спонтанного излучения с пороговой плотностью энергии возбуждения 12,3 мкДж·см$^{-2}$, что свидетельствует об их высоком потенциале для эффективного применения в оптоэлектронике, включая светоизлучающие устройства и лазеры.

Ключевые слова: перовскит, металлорганический каркас, композитные материалы, усиленное спонтанное излучение

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Introduction

Metal-halide perovskite nanocrystals (MPNCs) based on lead halides exhibit exceptional optoelectronic properties, such as high absorption coefficients, high photoluminescence quantum yields, tunable band gaps, and defect tolerance [1]. These characteristics make them promising candidates for various optoelectronic applications, including light-emitting diodes and lasers. However, due to the ionic structure, halides perovskites are unstable under the influence of external influences such as high temperature, air and ultraviolet radiation.

To address the stability issue, researchers vary the composition of the elements included in the ABX_3 perovskite structure (A is an organic or inorganic cation, B is a divalent metal, X is a halide anion) or use ligands, polymer materials, or metal-organic frameworks [2–4]. Metal-organic frameworks are a class of compounds in which metals and organic ligands form a crystalline structure called a framework [3, 4].

In this work, we focused on the recombination dynamics and amplified spontaneous emission (ASE) characteristics of $FAPbBr_3$ (FA- $CH(NH_2)_2$, formamidinium) by incorporating it into a lead based metal-organic framework constructed with trimesic acid (Pb-MOF). Previous studies have demonstrated improved perovskite stability using Pb-MOF and a two-step method for film fabrication [3, 4]. In contrast, we developed a one-step fabrication approach for $FAPbBr_3$-MOF composite thin films, enabling a simplified and faster preparation process. This approach yielded a record-low ASE threshold for $FAPbBr_3$ (12.3 μJ·cm^{-2}), which we were able to rationalize by examining the underlying recombination mechanisms. A Low ASE threshold is important for photonic applications, as it enables the development of devices with higher efficiency and improved operational stability.

Materials and Methods

Materials. The following materials were used in experiments: Lead Nitrate ($Pb(NO_3)_2$, 99.7%, LenReaktiv), trimesic acid (1,3,5-H_3BTC, 95%, Sigma-Aldrich), Acetic acid (CH_3COOH, 93%, ECOS), dimethyl sulfoxide (DMSO, 99%, ECOS), formamidinium bromide (FABr, >99.99%, Dyesol).

Methods. Pb-MOF was synthesized using a sonochemical method [5]. The obtained Pb-MOF and FABr were dissolved in anhydrous dimethylsulfoxide (DMSO) with the addition of methylammonium acetate (MAAc). The solution was stirred for 5 min (3500 rpm) using vortex MSV-3500 BioSan. The resulting solution was used to fabricate $FAPbBr_3$-MOF composite

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thin films via spin-coating: first at 3,000 rpm for 90 seconds, then at 5,000 rpm for 10 seconds, followed by annealing at 80 °C for 10 minutes.

The morphology of the synthesized films was analyzed using a Zeiss LIBRA 200FE transmission electron microscope (TEM) and a scanning probe microscope AIST-NT SPM SmartSPM^{tm}–1000.

Optical measurements were performed using an Antaus Ytterbium femtosecond fiber laser operating at a repetition rate of 10 kHz and a wavelength of 343 nm. Time-resolved photoluminescence (TRPL) was measured using a single-photon avalanche diode detector (SPAD PDM, Micro Photon Devices). Photoluminescence spectra were recorded with a QE Pro fiber-optic spectrometer.

The ABC model (equation 1) was used to approximate the decay curves [6].

$$\frac{dn}{dt} = -An - Bn^2 - Cn^3. \tag{1}$$

The term A is responsible for trap-assisted recombination, B for bimolecular recombination, and C for Auger recombination. At low fluence, Auger recombination does not contribute to charge carrier recombination [6]. Thus, the normalized PL (equation 2) decay curve exhibits bimolecular behavior and can be approximated by the following equation:

$$n(t) = \frac{An \cdot \exp(-At)}{A + Bn(1 - \exp(-At))} + \text{const}, \tag{2}$$

where n is the initial charge carrier density. The initial charge carrier density was calculated using the following equation:

$$n = F \cdot Abs, \tag{3}$$

where F is the fluence and Abs is the fraction of incident photons that are absorbed. Abs is calculated as:

$$Abs = (1 - R) \cdot \left[1 - \exp(-ad)\right], \tag{4}$$

d is the thickness of the film and a is the attenuation coefficient.

Results and Discussion

The resulting $FAPbBr_3$-MOF composite film has a low surface roughness of 0.99 nanometers and a thickness of 116 nanometers (Fig. 1, *a*). The composition of $FAPbBr_3$-MOF composite films was confirmed by X-ray diffraction analysis (XRD). The presence of the cubic Pm–3m phase is indicated by strong peaks at 14.8 degrees (100) and 29.8 (200) (Fig. 1, *b*), along with weaker peaks at 20.9 (110) and 33.8 (210) [7]. Peaks corresponding to Pb-MOF also confirm the composite nature of the films [5]. Transmission electron microscopy (TEM) revealed encapsulation of perovskite within the Pb-MOF matrix and showed an average $FAPbBr_3$ nanoparticle size of 11.6 nm (Fig. 1, *c*).

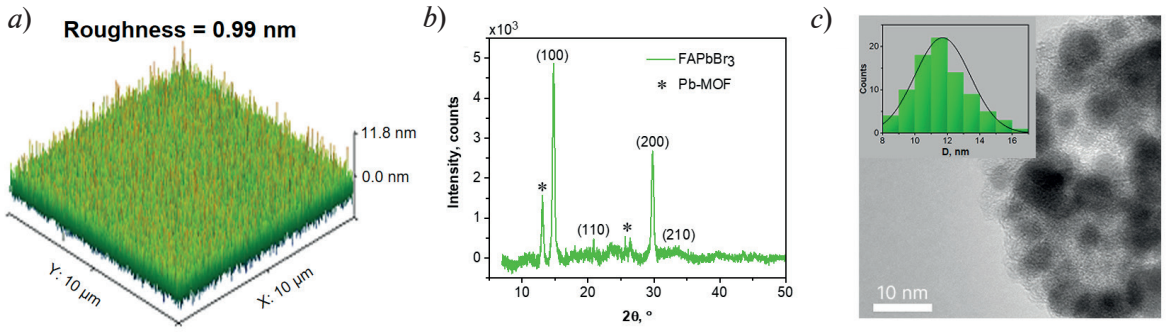

Fig. 1. AFM image of $FAPbBr_3$-MOF film (*a*), XRD analysis of $FAPbBr_3$-MOF (*b*), TEM analysis of $FAPbBr_3$-MOF (*c*). Inset in (*c*) is a histogram of the NCs size distribution

The photoluminescence (PL) peak of the film is at 530.1 nm with a full width at half maximum (FWHM) is 24.3 nm (Fig. 2, *a*) also a similar peak position was confirmed by studying the absorption spectrum of $FAPbBr_3$-MOF (532.5 nm). For the subsequent analysis of the optical properties of our film, we investigated the dependence of the photoluminescence quantum yield on the increasing power density (Fig. 2, *c*). The best PLQY is 43% at a power density of 993 mW·cm^{-2}, and the growing dependence of PLQY at an increasing power density indicates high film quality, as well as a greater contribution of radiative processes relative to non-radiative [8]. To study the origin of the recombination process in more detail, we performed time-resolved photoluminescence (TRPL) measurements, as shown in Fig. 2, *d*. TRPL measurements showed that the monomolecular (*A*, non-radiative) and bimolecular (*B*, radiative) recombination coefficients increased from $A = 2.4 \cdot 10^6$ s^{-1} and $Bn = 9.4 \cdot 10^6$ s^{-1} at 1.3 μJ·cm^{-2} to $A = 18.7 \cdot 10^6$ s^{-1} and $Bn = 6.5 \cdot 10^8$ s^{-1} at 10.4 μJ·cm^{-2} (Table). This enhancement of radiative recombination and acceleration of carrier dynamics at higher fluence are beneficial for ASE, as these effects support faster population inversion and gain buildup. As a result, PL spectra evolution under increasing pump fluence revealed an ASE threshold of 12.3 μJ·cm^{-2} (Fig. 2, *e*, *f*), which is lower than previously reported [9], demonstrating the suitability of $FAPbBr_3$-MOF films for integration into next-generation low-threshold laser systems and other light-emitting applications.

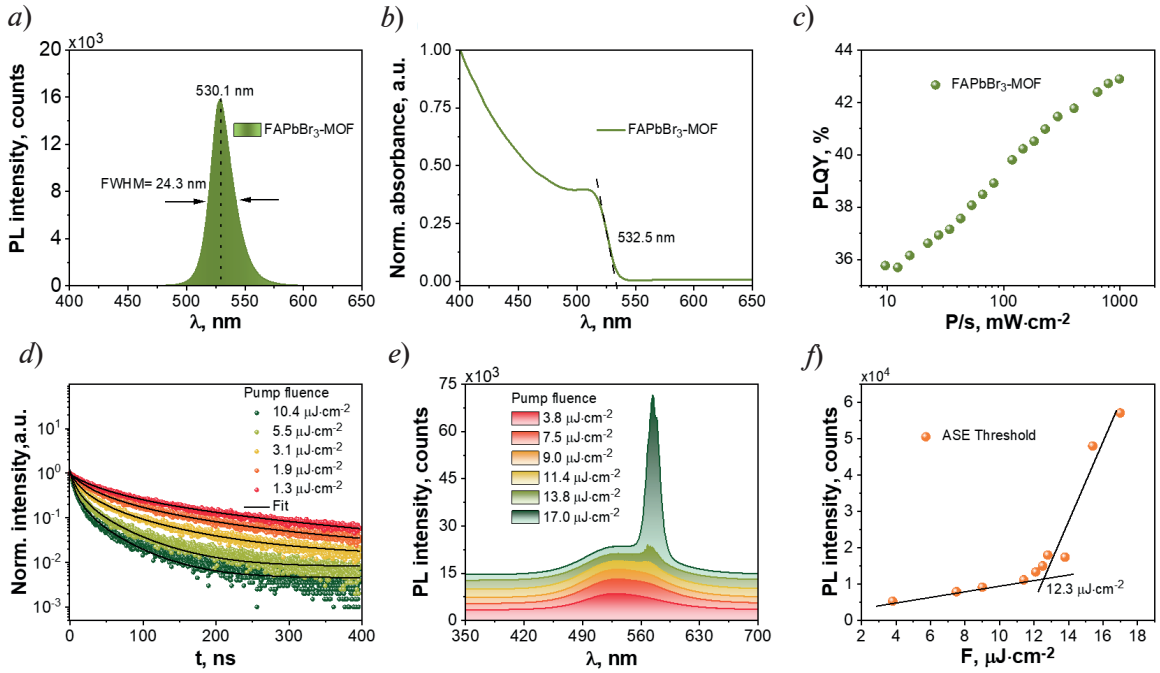

Fig. 2. Photoluminescence spectrum of $FAPbBr_3$-MOF (*a*), Normalized absorbance spectrum of $FAPbBr_3$-MOF (*b*), Dependence of PLQY on the power density (*c*), decay curves of $FAPbBr_3$-MOF under increasing pump fluence (*d*), evolution of the PL spectra of the $FAPbBr_3$-MOF films under increasing pump fluence (*e*), ASE threshold of $FAPbBr_3$-MOF (*f*)

Table

Analysis of decay curves of $FAPbBr_3$-MOF film

Pump fluence, μJ·cm^{-2}	*n*, cm^{-3}	A, s^{-1}	Bn, s^{-1}
10.4	$3.3 \cdot 10^{18}$	$18.7 \cdot 10^6$	$6.5 \cdot 10^8$
5.5	$1.7 \cdot 10^{18}$	$16.0 \cdot 10^6$	$2.0 \cdot 10^8$
3.1	$9.6 \cdot 10^{17}$	$8.1 \cdot 10^6$	$6.5 \cdot 10^7$
1.9	$5.9 \cdot 10^{17}$	$4.7 \cdot 10^6$	$2.2 \cdot 10^7$
1.3	$4.0 \cdot 10^{17}$	$2.4 \cdot 10^6$	$9.4 \cdot 10^6$

Conclusion

We developed a simple one-step method for fabricating $FAPbBr_3$-MOF composite thin films with improved structural stability and promising optical properties. The encapsulation of Pb-MOF ensured the formation of a stable composite matrix, while the films demonstrated efficient radiative recombination and a low ASE threshold (12.3 $\mu J \cdot cm^{-2}$), making them suitable for photonic applications.

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