Research article

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Remarkable photoluminescence enhancement of CsPbBr₃ perovskite quantum dots assisted by metallic thin films

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Abstract: All-inorganic cesium lead halide perovskite quantum dots have recently received much attention as promising optoelectronic materials with great luminescent properties and bright application prospect in lighting, lasing, and photodetection. Although notable progress has been achieved in lighting applications based on such media, the performance could still be improved. Here, we demonstrate that the light emission from the perovskite QDs that possess high intrinsic luminous efficiency can be greatly enhanced by using metallic thin films, a technique that was usually considered only useful for improving the emission of materials with low intrinsic quantum efficiency. Eleven-fold

maximal PL enhancement is observed with respect to the emission of perovskite QDs on the bare dielectric substrate. We explore this remarkable enhancement of the light emission originating from the joint effects of enhancing the incident photonic absorption of QDs at the excitation wavelength by means of the zero-order optical asymmetric Fabry–Perot-like thin film interference and increasing the radiative rate and quantum efficiency at the emission wavelength mediated by surface plasmon polaritons. We believe that our approach is also potentially valuable for the enhancement of light emission of other fluorescent media with high intrinsic quantum efficiency.

Keywords: asymmetric interference; CsPbBr₃ quantum dots; metallic thin films; photoluminescence enhancement; surface plasmon polaritons.

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1 Introduction

In recent years, semiconducting halide perovskites have emerged as a new class of materials with promising application prospects in fields of photonics and optoelectronics [1–8]. Being the typical representative of this family, all-inorganic cesium lead halide perovskite (CsPbX_3 , $\text{X}=\text{Cl}$, Br , and I or hybrid composites Cl/Br and Br/I) quantum dots (QDs) are particularly striking because of their great luminescent properties, including tunable photoluminescence (PL) wavelength, narrow emission bandwidth, and high luminous efficiency [9–16], and have been demonstrated for many applications from solar cells [9], lasers [10, 11], light emitting diodes (LEDs) [12–14] to photodetectors [15, 16]. Although remarkable advances have been made in the development of novel devices based on such as perovskite QDs, the performance in terms of brightness and efficiency could still be improved. To address this issue, many approaches, such as interface doping [17], shape decorating [18], and surface ligand modification [19], have been proposed to enhance the luminescent properties of the perovskites based upon the material chemical engineering perspective.

It has been known for a long time that nanophotonic structures can be used physically to amplify the emission of luminescent materials by changing their local electromagnetic environment [20–26]. Some types of them have also been recently introduced for enhancing PL of the perovskites. For example, Hou et al. designed and fabricated photonic crystals directly on perovskite thin film, and experimentally obtained an order of magnitude enhancement of light emission compared to the unpatterned area in the same sample [23]. Caligiuri et al. created a double epsilon-near-zero (ENZ) metamaterial, and demonstrated a four-fold enhancement of PL of CsPbBr_3 perovskite as that was placed on top of the specific ENZ metamaterials [24]. However, although notable PL enhancement in perovskites has been demonstrated by using these optical engineering structures, the realizations of such nanostructures usually demand not only elaborate structure design, but also highly precise processing techniques; this would hamper them for real applications. In the past decades, as the easily fabricated and perhaps the simplest plasmonic structure, metallic thin films have been demonstrated for the enhancement of spontaneous emission of a number of different light-emitting media, including quantum wells [27], dye molecules [28], organic emitters [29], and semiconductor nanocrystals [30], while, which has yet to be explored for enhancing the efficiency of light emission in perovskites.

In the work, we demonstrate that the light emission from perovskite QDs can be greatly enhanced by using metallic thin films, a technique that was usually considered useful for improving the emission of materials with low intrinsic quantum efficiency, rather than for high efficiency materials [30, 31]. Experimental measurements were performed of the PL intensity versus wavelength and as a function of both the thickness of the metallic thin film and the dielectric spacer. A maximum PL enhancement of 11 is observed with respect to the emission of perovskite QDs on the bare dielectric substrate. We explore this remarkable enhancement of the light emission originating from the joint efforts of enhancing the incident photonic absorption of QDs at the excitation wavelength by means of the strong optical asymmetric Fabry–Perot-like (F–P-like) thin film interference effects and increasing the radiative rate and quantum efficiency at the emission wavelength assisted by surface plasmon polaritons. To better understand the physical mechanism of the PL enhancement effect [32–35], energy band diagrams and time-resolved PL spectrum have been applied to illustrate the energy transfer process and the PL decay kinetics. We believe that the proposed methods in this work could open up a new route toward the design and realization of spontaneous emission with improved functionalities that have potential applications in many technologically important fields, ranging from light emitting diodes and plasmonic lasers to biological sensors.

2 Results and discussion

Cesium lead bromide (CsPbBr_3 , CPB) perovskite QDs were chosen as the light emitters in our experiments (details of the CPB QDs synthesis see Supplementary Material Note 1). Figure 1(A) shows a transmission electron microscopy (TEM) image of as-synthesized CPB QDs, indicating the presence of uniformly distributed cubic-shaped QDs with an edge length of about 11 nm. The inset of a high resolution TEM (HRTEM) image in Figure 1(A) shows that the QDs are well-crystallized. The structural characteristics of the QDs were further confirmed by the X-ray diffraction (XRD) analysis (see Figure 1(B)). These QDs typically exhibit bright green emission with the peak wavelength (λ_{peak}) of around 520 nm (see Figure S1 in Supplementary Material). The quantum efficiency of the colloidal QDs solution was measured to be about 0.4 [36].

As schematically shown in Figure 1(C), our experimental structure basically consists of a perovskite QDs nanolayer and continuous silver (Ag) thin film, separated by a SiO_2 dielectric spacer. The bottom layer, Ag film with

Figure 1: Structural characterizations of CsPbBr₃ perovskite QDs and experimental setup for PL measurements. (A) Typical transmission electron microscopy (TEM) and high resolution TEM (inset) images of CsPbBr₃ QDs. (B) X-ray diffraction (XRD) spectrum for CsPbBr₃ QDs (black line) and standard XRD profile of JCPDS cards No. 84-0464 (red line). (C) Schematic of sample structure. (D) Experimental setup for PL measurements.

thickness (denoted by d_{Ag}) varied from 20 to 150 nm, was first deposited on polished quartz substrate by using radio-frequency (rf) magnetron sputtering. SiO₂ spacer was then deposited by the same sputtering system. The thickness of the spacer (denoted by d_{spacer}) was changed from 5 to 100 nm. Afterwards, we spin-coated 20 μL colloidal CPB QDs solution on the top of each structure at 2500 rpm for 50 s to achieve about 35 nm thick CPB QDs layer (determined by ellipsometry data analysis). The QDs was also prepared by the same method onto the bare polished quartz substrate for reference purpose. Figure 1(D) shows the experimental setup for the PL intensity measurements at room temperature. A 405 nm continuous wave laser was focused onto the sample using an objective lens (N.A. = 0.95). The fluorescence light was collected by the same objective lens, and then passed through a long-pass filter to filter the excitation signal before coupled into a spectrometer.

Figure 2(A) presents the measured PL spectra for CPB QDs on seven different thickness Ag films ($d_{\text{Ag}} = 0, 20, 40, 60, 80, 100,$ and 150 nm) separated by 10 nm thick SiO₂ spacers. It is seen that the emission from the samples containing Ag thin films shows large enhancement in comparison to that of the control sample ($d_{\text{Ag}} = 0$ nm), while the impact on the emission peak position of the QDs can be negligible for all the cases. To further clearly illustrate the emission enhancement effect, we define an enhancement factor as $EF = I/I_0$, where I and I_0 are the PL signals from the samples with Ag thin films and the control sample, and plot the PL EF obtained at the emission peak

wavelength in Figure 2(C) as a function of Ag film thickness. It is clear from the figure that as the Ag film thickness increases, the enhancement factor increases first until reaching a maximum, then decreases slightly thereafter, and it eventually saturates at thickness of 100 nm. The maximum PL enhancement factor was measured to be about 11 at the Ag film thickness of 60 nm. In previous works, such metallic thin-film-based fluorescence enhancement was commonly understood via near-field coupling to surface plasmon polaritons (SPPs) and thus increasing the spontaneous emission rate and the internal quantum efficiency [33, 37]. However, for the CPB QDs emitters of the control sample that have a QE of $Q_0 = 0.1$, even which is much lower than the one of the colloidal QDs solution due to QDs aggregation effects and reaction with oxygen and water without the protection of solution, the maximum achievable enhancement factor in internal QE should be less than 10, which is smaller than the observed magnitude of the large enhancement in PL. Thus, it is obviously inappropriate just simply to use the mediation by SPPs to explain the PL enhancement of the present case [38–40].

We identify that this remarkable PL enhancement is mainly attributed to a combination of process involving enhanced absorption of the CPB QDs at the excitation wavelength and the enhanced PL quantum efficiency at the emission wavelength. To properly study the absorption of the CPB QDs, the optical constants of the QDs were extracted by using spectroscopic ellipsometry. Figure 3(A) displays the measured amplitude ratio (Ψ) and the phase

Figure 2: PL spectra and enhancement factors.

(A) Measured PL spectra for CPB QDs on seven different thickness Ag films ($d_{Ag} = 0, 20, 40, 60, 80, 100$, and 150 nm) separated by 10 nm thick SiO_2 spacers. (B) Measured PL spectra of CsPbBr_3 QDs on 60 nm thickness Ag NP film separated by dielectric spacer with thickness changed from $d_{\text{spacer}} = 5$ to 100 nm. (C) PL enhancement factors as a function of Ag film thickness. The inset shows a schematic of a structure with Ag film thickness changed. (D) PL enhancement factors as a function of dielectric spacer thickness. The inset shows a schematic of a structure with spacer thickness changed. (E) The enhancement factors resulting from the enhanced absorption (denoted by $F_a = A/A_0$) and from the improved quantum efficiency (denoted by $F_q = Q/Q_0$) are plotted against the Ag film thickness. (F) The enhancement factors resulting from the enhanced absorption and from the improved quantum efficiency are plotted against the spacer thickness. Blue symbols denote the enhancement factors resulting from the measured quantum efficiency data.

Figure 3: Ellipsometry analysis and optical absorption spectra.

(A) Experimental measured Ψ and Δ (solid curves) for CsPbBr_3 QDs layer at angles of incidence $\theta = 45$ and 55° , and the fitted results from the multiple Lorentz oscillators model (symbol curves). (B) Real and imaginary parts of refractive index for the CsPbBr_3 QDs obtained from the ellipsometry data analysis. (C) Calculated and (D) experimental optical absorption spectra for the samples studied in Figure 2(A). (E) The absorption spectra for the total and each component (QDs and Ag film) of the sample with Ag thickness of 60 nm. (F) The normalized electric and magnetic field amplitudes and time-averaged power dissipation density of the structure studied in Figure 3(E) at the wavelength of 405 nm.

difference (Δ) for the QDs nanolayer in the wavelength range from 380 to 800 nm at the angles of incidence 45 and 50°. The complex refractive index (n and k) derived from measured spectroscopic ellipsometric data are shown in Figure 3(B). It is noted that as expected the extinction coefficient k of the QDs decreases sharply as increasing wavelength from about the emission peak wavelength λ_{peak} . Figure 3(C) shows the calculated absorbance spectra for the structures studied in Figure 2(A) at normal incidence. The absorbance was obtained by using the equation $A = 1 - R - T$, where R and T are the reflectance and transmittance, which were directly calculated based on transfer matrix method. The corresponding experimental absorbance spectra of these structures are presented in Figure 3(D). Good agreements are noted between the theoretical and experimental results. We infer that the difference between the experimental and calculated optical absorption spectra mainly results from the imperfections of the layered structure, including random thickness fluctuations and the diffusive intermixing at the interfaces [41]. From Figure 3(C) and (D), one can see that for the sample involving Ag thin film, the absorption is much larger than the one of control sample, and which increases with Ag thickness, finally saturates at thickness of 60 nm for the wavelength range less than λ_{peak} . Figure 3(E) shows the absorption spectra for the total and each component of the sample with Ag thickness of 60 nm. It is found that at the excitation wavelength 405 nm, more than 51% energy of incident light is absorbed by the QDs. Compared to the control sample that the absorption is only around 12% at this wavelength; the absorption of QDs is enhanced by a factor of 4.25.

To figure out the nature of this absorption enhancement effect, near field responses were investigated. Figure 3(F) shows the normalized electric and magnetic field amplitudes and time-averaged power dissipation density of the structure studied in Figure 3(E), as it is illuminated by a normally incident plane wave with the wavelength of 405 nm [42]. As can be observed, the distribution of electromagnetic field exhibits asymmetric characteristic and the most energy of incident light is concentrated and dissipated by the QDs. These results indicate that such high absorption enhancement is attributed to zero-order optical asymmetric Fabry–Perot-like (FP-like) thin-film interference effect [43, 44]. In addition, the dependence of the absorption enhancement on the perovskite QDs film thickness is presented in Figure S2 (Supplementary Material).

The dashed-dotted red line of Figure 2(E) shows the enhancement factor (denoted by $F_a = A/A_0$) resulting from the enhanced absorption of QDs as a function of the

thickness of Ag films. However, to understand the observed Ag film thickness dependence of the PL enhancement shown in Figure 2(C), the improvement of fluorescence efficiency because of SPP coupling should also be figured out. To address this issue, we used a semiclassical approach reported in previous literatures to model the modification of the emission of the QDs due to the presence of the metallic thin film [25, 45, 46]. In the approach, the QD emitter was treated as an oscillating electric dipole, our structure can thus be considered as a situation where the dipoles are placed on top of an Ag film (see Supplementary Material Note 2). The calculated Ag film thickness dependence of fluorescence efficiency enhancement at the QD emission energy based on this approach is shown in Figure 2(E) as the dashed-dotted olive line (denoted by $F_q = Q/Q_0$). The product of F_a and F_q is plotted in Figure 2(C) as solid blue line, which is consistent with the experimental results, indicating that the overall PL enhancements are indeed originating from the joint effects of the enhanced absorption at the excitation process and the enhanced radiation efficiency at the emission process. Moreover, it is worth pointing out that the radiation patterns of PL from the QDs on the metallic thin films are non directive and quite similar to the one of the control sample as determined by the angle-resolved fluorescence measurements (see Figure 4). This is in sharp contrast to previous works that used plasmonic nanostructures to enhance spontaneous emission [47, 48], where the increased directionality of emission also plays an important role in the enhancement of radiative properties.

The mechanism of PL enhancement of QDs assisted by Ag thin films can be readily described by classical simplified energy diagrams as presented in Figure 5(A) and (B). Compared to the QDs on the bare substrate (Figure 5(A)), the photon excitation rate is highly increased based on the

Figure 4: The angle-resolved fluorescence measurements.

The measured radiation patterns for the sample with the parameters $d_{\text{Ag}} = 60$ nm and $d_{\text{spacer}} = 10$ nm and the control sample. Shaded regions denote angular regions that PL signals cannot be collected in our experimental setup.

Figure 5: The energy diagrams and time resolved PL spectra for CPB QDs. Schematic of energy diagram of the absorption and emission process for CPB QDs without (A) and with (B) metallic thin film. (C) Time resolved PL spectra for QDs on different thickness Ag films ($d_{\text{Ag}} = 0, 20, 40, 60, 80, 100$, and 150 nm), with the other parameter $d_{\text{spacer}} = 10$ nm. (D) Time resolved PL spectra for QDs on a series of planar plasmonic structures with different thickness dielectric spacers ($d_{\text{spacer}} = 10, 20, 40, 60, 80$, and 100 nm), with the Ag film thickness taken as $d_{\text{Ag}} = 60$ nm.

optical asymmetric FP-like thin-film interference effect when the QDs are located atop Ag thin films (Figure 5(B)), at the meantime, a new emission channel emerges through the SPP coupling, which can significantly accelerate the decay rate. To experimentally investigate influence of QD–SPP coupling on the spontaneous emission rate, time resolved PL decay measurements of QDs on Ag thin-films and a bare quartz substrate (control sample) were performed (see Figure S3 in Supplementary Material). Figure 5(C) shows the results of PL lifetime measured by time-correlated single photon counting system [49]. It is observed that the delay times for QDs on Ag thin films are obviously decreased compared to those of control sample, and the trend of Ag film thickness dependence of PL decay is similar to that of fluorescence efficiency enhancement factor F_q . We use a biexponential function to fit the decay curves [13, 36], resulting in the fitted average decay times for QDs on Ag thin films of 2.13 ns ($d_{\text{Ag}} = 20$ nm), 1.89 ns (40 nm), 1.74 ns (60 nm), 2.02 ns (80 nm), 3.20 ns (100 nm), 3.58 ns (150 nm), which are all much shorter than the one (7.56 ns) of bare QDs. The PL lifetime shortened as the presence of Ag thin films show evidence of that SPP provides an additional high-rate emission channel invoked by the strong QD–SPP interaction.

We further investigate the dependence of PL enhancement on the thickness of spacer layer. Figure 2(B) shows the measured of CsPbBr₃ QDs on a 60 nm thickness Ag NP film separated by dielectric spacer with thickness changed from $d_{\text{spacer}} = 5$ to 100 nm. Correspondingly, the PL EF obtained at the emission peak wavelength is also

plotted in Figure 2(D) as a function of the spacer thickness. It notes that as the spacer thickness increases, the EF increases first and then decreases gradually producing a non exponential variation curve. This spacer-layer dependence of the EF is quite different from previous studies [47, 48], in which the EF generally decreases as exponential function accompanying with increasing the spacer thickness, as their enhancement in PL is mainly attributed to the coupling of SPP, that exponentially decays with distance from the metal surface. To understand the observed spacer thickness dependence of the PL enhancement, the enhancements of absorption and fluorescence efficiency for such cases were investigated and are plotted in Figure 2(F) as the dashed-dotted red line and the dashed-dotted olive line, respectively. The product of these two enhancement factors is shown in Figure 2(D) as solid blue line. Good agreements are found between measured values of the PL enhancement and the theoretical calculated results. Furthermore, to examine the mechanism and demonstrate the enhancement of the spontaneous emission rate, time-resolved fluorescence measurements were also carried out. Figure 5(D) presents the time-resolved emission from the QDs for each of spacer thicknesses. As expected, the decay times for the series of samples were clearly shorter than those of control sample. In particular, the stronger the QDs coupled to the Ag film, the shorter the PL lifetime is. All these results suggest that the PL enhancements are really attributable to joint effects including enhanced absorption at the excitation wavelength and improving the emissive properties of the QDs at the emission wavelength.

3 Conclusion

In summary, we have demonstrated that the light emission from native high-performance fluorophores, perovskite QDs, can be remarkably enhanced by using metallic thin films that were usually used to improve the emission of materials with low intrinsic quantum efficiency, rather than for high efficiency materials. PL intensity measurements versus wavelength and as a function of both the thickness of the metallic thin film and the dielectric spacer were performed. An impressive 11-fold maximal PL enhancement factor is obtained with respect to the emission of perovskite QDs on the bare dielectric substrate. The basic physics behind the large enhancement involves two aspects: First, the absorption of QDs at the excitation wavelength is greatly enhanced as a result of the strong optical asymmetric F-P-like thin film interference effects, which even plays a dominant role in the overall PL enhancement when the spacer thickness is less than 50 nm. Second, the radiative rate and quantum efficiency of QDs at the emission wavelength is also improved mediated by SPPs. Good agreements between the experimental and theoretical results confirmed our predictions. We believe that this research can open up new avenues to expand the practical applications of high-performance perovskites optoelectronic device, such as light emitting diodes, biological sensors, and plasmonic lasers.

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