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# Luminescence lifetime enhanced by exciton-plasmon couple in hybrid CsPbBr<sub>3</sub> perovskite/Pt nanostructure

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#### Abstract

Photoluminescence (PL) and time-resolved spectroscopic studies on plasmonically coupled semiconductor nanoparticles (SNPs) have demonstrated the PL quenched and lifetime enhanced of SNPs in the presence of metal nanoparticles (MNPs). The hybrid colloidal CsPbBr<sub>3</sub> perovskite SNPs/Pt MNPs (S-M) structures exhibit novel optical properties due to the synergetic interaction between the individual components. In hybrid S-M nanostructures colloidal chemistry incorporates SNP and MNP into a single unit resulting in the formation of plexciton (or excimon) which has now been established in a series of hybrid structures. The experimental results of femtosecond transient absorption (TA) spectroscopy based on the time-resolved pump—probe confirm the transformation from excitons to plexcitons. It was found that the experimental data can't be well described by the theory based on conventional Föster resonance energy transfer (FRET). The differences between theory and experiment may be due to the missing some PbBr<sub>2</sub> PL peaks, the reason will be revealed further.

#### 1. Introduction

Metal nanostructures have attracted considerable attentions because of their ability to manipulate light at the nanometer scale. Free electrons within the metal nanostructures collectively oscillate at the frequency of incident radiation, a process known as localized surface plasmon resonance (LSPR), resulting in the confinement of the electromagnetic fields near the metal nanostructure and a significant enhancement of the local density of optical states [1-7]. The excitons, discrete quantum confined electronic states in the SNPs and plasmons, dielectricconfined electromagnetic modes in the MNPs are coupled each other through long-range Coulomb interactions in the hybrid nanostructure. The SNP is quantum emitter, whereas the MNP is amplifier or damper in this hybrid nanostructure. When SNPs are situated close to noble MNPs, their emitting behavior can be conveniently tuned because of the interaction between the excitons of the SNPs and the plasmons of the MNPs. This interaction gives rise to PL enhanced or quenched. However, the mechanism of how plasmonic structures cause the changes in the SNP emission remains unclear. The degree of exciton-plasmon coupling in S-M systems depends on many factors, such as interparticle distance, spectral overlap between S-M, orientation of the electric dipole of SNP relative to the MNP, as well as the topology of the metal surface. The presence of plasmon radiative field in metals, caused by resonant oscillations of low-energy conduction electrons, can strongly affect the dynamics of excitons in SNPs via two distinct interaction mechanisms, including exciton-plasmon energy transfer and modification of the local radiation field in SNP domains.

The conventional Föster resonance energy transfer (FRET) suffers from a limited length scale of approximately 8  $\sim$  10 nm. Plasmonic structures provide a route toward longer range energy transfer via nanometal surface energy transfer or LSPR-coupled FRET [5, 6]. The near-field of the plasmonic structure can modify the emission of a fluorophore [7–10]. The modified emission can be a consequence of many processes

such as LSPR energy transfer to the MNP, scattering and absorption by the MNP, and changes in the emitter's radiative and nonradiative decay rates.

The hybrid SNPs-MNPs nanosystem consisted of colloidal CsPbX3 perovskite and noble Pt nanoparticles. In particular, CsPbX3 perovskite nanoparticles with outstanding signatures of combined advantages of both QDs and halide perovskites have size-tunable emissions (410–700 nm), narrow emission line widths (12–42 nm) and high quantum yields (~90%), which enable their advantages over the Cd–chalcogenide NC [11].

The many studies on fluorescence quenching and lifetime shortening in S-M hybrid complexes were reported [1–7, 12–16], however, lifetime enhancement or radiative rate reductions were rarely reported [17–19]. In the present study, we found a pronounced effect on the PL lifetime enhanced by the couples between excitons of colloid CsPbBr3 perovskite SNPs and plasmons of Pt MNPs (S-M). The experimental data have been analyzed and discussed. It is suggested that the new theory beyond the conventional FRET is needed. The exciton-plasmon interaction in the hybrid S–M nanostructures alters the exciton generation and recombination dynamics of the QDs, which facilitates developments in the fields of optoelectronics, lasing, sensing, and solar cell [20–24]. The aim of this study is to investigate the underlying mechanism of both fluorescence quench and lifetime enhancement.

#### 2. Materials and methods

#### 2.1. Materials

CsPbBr<sub>3</sub> SNPs were synthesized at room temperature followed the supersaturated recrystallization strategy of Li and co-workers [25]. All the purchase reagents were used directly without further purification.

#### 2.2. Preparation and characterization

In a typical synthesis of CsPbBr<sub>3</sub>, PbBr<sub>2</sub> (0.0734 g) and CsBr (0.0426 g) were dissolved in dimethyl formamide (DMF) (5 ml). The surface ligands oleic acid (OA) (0.5 ml) and oleylamine (OAm) (0.25 ml) were added to stabilize the precursor solution. Then, 5.75 ml of the precursor solution was quickly added into toluene (57.5 ml) under vigorous stirring. Pt NPs were dispersed homogeneously into toluene solution, then which were added into CsPbBr<sub>3</sub> NPs toluene solution according to designed wt% amount. The procedure of synthesis of bulk CsPbBr<sub>3</sub> is similar just without the surface ligands. The crystals were dried under flowing nitrogen at room temperature.

Powder x-ray diffraction (XRD) measurements were carried out by using a Bruker D8 advanced powder diffractometer with Cu KR radiation ( $\lambda = 1.54056$  Å) as the incident beam. The morphology was investigated using field emission scanning electron microscopy (SEM) (Hitachi S-4800). UV–vis absorption spectra were taken with UV–vis–NIR scanning spectrophotometer (Shimadzu, UV-3101PC). Emission spectra were performed on a Hitachi F-7000 fluorescence spectrophotometer equipped with a 150 W xenon lamp as the excitation light source. Fluorescence decay measurements were experimented on grating spectrometer FL 980 equipped with the time-correlated single photon counting, the samples were excited at 405 nm using a picosecond diode laser (IBH Nanoled-07) in an IBH Fluorocube apparatus monitoring at 517 nm. The typical full width at half maximum (FWHM) of the system response using a liquid scatter is about 90 ps.

Ultrafast TA experiments were conducted using a Ti:sapphire laser (Spectra-Physics, 800 nm, 0.3 mJ/pulse, FWHM 120 fs, 1 kHz repetition rate). The white-light probe was generated by the fundamental laser output focused on a CaF2 window with 5 mm thickness. The probe beam at the sample had a diameter of 150  $\mu$ m. Pump pulses at 400 nm were generated by frequency doubling of the fundamental laser in a BBO crystal. The pump beam at the sample had a diameter of 660  $\mu$ m. The energy pump beam used for the measurement was controlled by a variable neutral-density filter wheel. The energy of the pump pulse were changed by a variable neutral density filter and chopped by a synchronized chopper to 500 Hz. After passing through the sample, the probe beam was focused into a fiber-coupled spectrometer (Avantes; AvaSpec-1650F-USB2,). The angle between pump and probe polarizations was set at magic angle (54.6°) to ensure the dynamics free from reorientation effects. The group velocity dispersion of the whole experimental system was compensated by a chirp program. During the data collection, samples were constantly stirred to avoid degradation. All experiments were performed at room temperature.

#### 3. Results and discussion

#### 3.1. XRD and SEM analysis

Powder x-ray diffraction (XRD) measurements were collected at room temperature with a  $2\theta$  range of  $20^{\circ}$ – $60^{\circ}$ , shown in figure 1(a). It is indicated that bulk CsPbBr<sub>3</sub> with a orthorhombic phase. The diameters of CsPbBr<sub>3</sub> and Pt nanoparticles are 11 nm [25] and 10 nm, respectively. The morphology of hybrid CsPbBr<sub>3</sub>/Pt nanostructure



was shown in figure 1(b), 'Pt MNPs (bright white points in dashed circle) were adhered to the surfaces of CsPbBr3 SNPs.'

#### 3.2. Spectroscopy studies

The absorption spectra were given in figure 2(a); (a)/20 of SNPs in toluene without Pt MNP (sample 0, molar concentration 7.47  $\mu$ M) and of hybrid system with Pt MNPs, (b) 1.5 wt%), (c) 1.8 wt%, (d) 2 wt% and (e)





2.2 wt%), (f) 2.5 wt%, (g) 5 wt% and (h) 8 wt%, respectively. It is indicated that there are not sharp peaks structures in the range of 300–700 nm for Pt MNPs.

The main emission about 517 nm is an exciton transition, the 450 nm emission is relating to PbBr<sub>2</sub> in figure 2(b) PL spectra. The narrow FWHM ~20 nm and obvious red-shift of emission profile were shown in the PL spectra. The ratios of integrated emissions spectra  $I^{S+M}/I^S$  of the hybrid nanostructure S-M ( $I^{S+M}$ ) and only

 ${\bf Table 1.} \ {\bf Experimental \ data \ and \ calculated \ parameters.}$ 

Samples wt%		$ au_1$		$ au_2$		$\langle \tau  angle$ ns	$\mathrm{I}_{PL}^{S+M}/\mathrm{I}_{PL}^{S}$	R <sub>S,M-M,S</sub> nm
	$A_1$	ns	$A_2$	ns	$\mathbb{R}^2$			
0	772	6.7	185.5	63.6	0.9793	17.7		
1.5	728.8	8.2	186.5	54	0.9223	17.5	0.95	7.10
1.8	672.4	12	146.4	96	0.9551	27.0	0.98	6.77
2	706.6	11.4	133.3	86.4	0.9508	23.3	0.91	6.68
2.2	729.2	7.8	208.5	62.9	0.9777	20.1	0.84	7.61
2.5	729.5	11.3	151.1	70.6	0.9619	21.5	0.94	6.57
5	608.3	11.8	167.7	97.3	0.9342	30.3	0.10	5.79
8	616.6	13.2	158.9	98.9	0.9280	30.8	0.11	5.37

SNPs (I<sup>S</sup>) were tabulated in table 1. As previously reported, most photoluminescence were quenched due to situating near the MNPs. The quenching mainly is due to modifying of both radiative and nonradiative rates by MNPs of the hybrid nanostructure S-M.

The radiative rate can be calculated from the integrated extinction coefficient [26].

$$k_r \approx 2.88 \times 10^{-9} n^2 \langle v_f^2 \rangle \int \varepsilon(v) dv \tag{1}$$

where n is refractive index of environmental medium and

$$\langle v_{\rm f}^2 \rangle = \int v^2 F(v) dv \Big/ \int F(v) dv$$
 (2)

F(v) is the photoluminescence profile measured experimentally,  $v_f$  is the fluorescence frequency (wavenumber),  $\varepsilon(v)$  is molar extinction coefficient. Then the radiative lifetime  $\tau_r = 29.8$  ns was obtained from equations (1) and (2). In addition, the PL lifetime observed experimentally can be expressed as

$$\tau_{\rm obs} = (k_r + k_{nr})^{-1} = (\tau_r^{-1} + \tau_{nr}^{-1})^{-1} = \tau_r \tau_{nr} / (\tau_r + \tau_{nr}), \tag{3}$$

Then the quantum yield QY was estimated by

$$QY = k_r / (k_r + k_{nr}) = \tau_{obs} / \tau_r \tag{4}$$

So  $QY_D = 59.4\%$  of CsPbBr3 SNPs was derived.

Time-resolved spectra of samples figure 3(a), (a) colloid CsPbBr<sub>3</sub> in toluene or sample 0, and hybrid system (b) with Pt MNPs 1.5 wt%, (c) 1.8%, (d) 2 wt%; (e) 2.2%, (f) 2.5 wt%; (g) 5 wt% and (g) 8 wt% respectively. The PL decay can be described by biexponential fitting,

$$I(t) = I_0 + \sum_{i=1,2} A_i \exp(-(t-t_0)/\tau_i),$$
(5)

where  $\tau_i$  and  $A_i$  are the decay time and fractional amplitude of the *i*th component, shown in table 1 respectively. Then amplitude-weighted lifetime was written as

$$\langle \tau \rangle = \sum A_i \tau_i / \sum A_i$$

The environment around the SNPs is modified or engineered to change the radiative and nonradiative decay rate of the SNPs. The PL lifetimes of colloid CsPbBr3 SNPs were extended from 17.7 ns to 30.8 ns, nearly 2 times, see figure 3(a). As a result of the energy redistribution, M-S energy transfer process suppresses the reduction of relaxation rate led to extending the lifetime of SNPs. This property should likewise favor the catalytic function of the semiconductor component, as it extends the exciton lifetime, increasing the probability of charge transfer into a surface-appended catalyst, also could be beneficial for solar cell applications since a longer lifetime allows more time for charge separation to occur [27].

The SNP PL is strongly modified by two different phenomena which are (1) FRET between SNPs-MNPs; (2) the enhanced electric field around MNPs that arises from surface plasmon oscillations. It is usually considered that energy transfer is essentially one way or S-M transfer which is due to the very short lifetimes of plasmons, typically <10 fs [15]. A model function [28] based on the rate equations of the system are developed by considering solely FRET, however it not only takes into account the S-M energy transfer, but also the M-S transfer. The integrated PL spectra ratios, I<sup>S+M</sup> of hybrid S-M complexes to I<sup>S</sup> of absence of Pt MNPs were given by [28]





$$\frac{I_{PL}^{SPL}}{I_{PL}^{S}} = (1 - A_{em}) \times \frac{\left[1 + \frac{4\pi\varepsilon_{M}\rho_{M}\rho_{S}R_{M-S}^{12}}{3\varepsilon_{S}\left(r_{\min}^{3} + \frac{4}{3}\pi R_{M-S}^{6}\rho_{S}\right)(R_{M-M}^{6}\rho_{M} + R_{M-S}^{6}\rho_{S}}\right]}{1 + \frac{4\pi R_{S-M}^{6}\rho_{M}}{3r_{\min}^{3}}}$$
(6)

Since the Pt MNP concentrations are too dilute ( $\sim \mu M$ ) the energy transfer among the MNPs (M-M process) to be ignored, or R<sub>M-M</sub> = 0, then equation (6) was simply rewritten as

$$\frac{I_{PL}^{S+M}}{I_{PL}^{S}} = (1 - A_{em}) \\ \times \frac{\left[1 + \frac{4\pi\varepsilon_{M}\rho_{M}R_{M-S}^{6}}{3\varepsilon_{S}\left(r_{\min}^{3} + \frac{4}{3}\pi\rho_{S}R_{M-S}^{6}\right)}\right]}{\left[1 + \frac{4\pi\rho_{M}R_{S-M}^{6}}{3r_{\min}^{3}}\right]}$$
(7)

where,

 $A_{em} = 10^{-\varepsilon(\lambda_{em})C_{AL}}$ 

 $\varepsilon(\lambda_{\rm em})$  and  $\varepsilon_{\rm S,M}$  are molar extinction coefficients,  $C_{\rm A}$  and L are concentrations of MNPs and a half of sample cell thick (~0.5 cm), respectively.  $\rho_{\rm S,M}$  are the particle number densities,  $r_{\rm min}$  is the minimum separation of the particles, i.e. the thickness of the ligand shells (~1 nm) and  $R_{\rm S,M-M,S}$  are the critical distances for the energy



Scheme 1. Schematic diagram of the exciton-plasmon interaction mechanism in hybrid colloid perovskite CsPbBr3 and metal Pt (SNP-MNP) nanostructure.

transfer [28],

$$R = 0.02108 [\kappa^2 Q Y_D n^{-4} J(\lambda)]^{1/6}, \tag{8}$$

 $\kappa^2 = 2$  in the case of randomly distributed, spherical and isotropic particles, n is the refractive index of the medium and

$$J(\lambda) = \int F_D(\lambda) \varepsilon_A(\lambda) \lambda^4 d\lambda / \int F_D(\lambda) d\lambda$$
(9)

is the overlap integral of donor emission spectrum  $F_D(\lambda)$  and acceptor absorption spectrum  $\varepsilon_A(\lambda)$ .  $R_{S,M-M,S}$ were calculated by equation (8), shown in table 1. If there is no energy transfer from MNP to SNP, then  $R_{M-S} = 0$ ,

$$\frac{I_{PL}^{S+M}}{I_{PL}^{S}} = \frac{1 - A_{em}}{1 + \frac{4\pi\rho_{M}R_{S-M}^{6}}{3r_{\min}^{3}}}$$
(10)

In figure 3(b) the black and red lines were calculated by equations (7) and (10), and blue spheres are the experimental data, respectively. Explicitly, the experimental data can be satisfactorily described by neither the black line nor the red line. Equations (7) and (10) derived from a model function for the interaction of S-M by considering solely FRET [28]. It is known that FRET is a well-defined distance-dependent dipole-dipole interaction. It has been widely used for measuring the distance between two fluorophores. However, as a spectroscopic ruler, conventional FRET suffers from a limited length scale of approximately 8–10 nm [29]. The energy transfer becomes too weak beyond this distance. The competition between these processes results in two opposite observations as reported in the literature: luminescence enhancement or quenching [30–33]. Quenched emission mainly comes from that MNPs could modify the fluorescence decay rates (both radiative and non-radiative), which could significantly change the fluorescence lifetime and quantum yield. The optical excitations of the SNPs are the excitons, with a sharp, discrete response. The excitons act as quantum emitters. The strong, local, plasmonic excitations of the MNPs provide a continuous spectrum of response. Enhanced local fields in the vicinity of the MNP provide strong coupling to neighboring SNPs. There is no direct tunneling between the MNP and SNP. However, due to the long-range Coulomb interaction, there is a dipole-dipole interaction that will allow them to couple and leads to excitation transfer. LSPR exists when the MNP is smaller than the incident wavelength, making the electron oscillations in phase. The collective oscillations lead to a large absorption and scattering cross-section, as well as an amplified local electromagnetic field [34]. The positions of energy levels were determined using vacuum level as potential energy reference point coming from electrons. The conductor and valence bands of CsPbBr<sub>3</sub> SNPs are -3.4 and -5.6 ev respectively [35]. The Fermi level  $E_F = -5.1 \text{ ev of Pt MNPs}$  which is depicted in scheme 1 [36]. Different parameters play a determinant role in the quenching and enhancement mechanisms but their influence is still debated [34, 37]. In particular, the excited energy in SNP and MNP domains can be efficiently shared though the mechanisms of FRET, Rabi oscillations, or radiative dipole-dipole interaction. Local surface energy transfer does not require a resonant electronic transition like Förster process and it originated from the interaction of the electromagnetic field of SNP dipole with the free conduction electrons of MNP. There are plenty of scopes for understanding the SNP-MNP actions. In hybrid S-M nanostructures colloidal chemistry incorporates SNP and MNP into a single unitplexciton [1, 16, 38-43]. The formation of such plexciton or excimon has now been established in a series of hybrid structures. The energy splitting between the coupled hybrid states has been analyzed, and Rabi splitting of



up to several 100 meV have been found. Such large splittings show that the local enhancement of the near-field in these structures can result in very strong coupling effects.

The femtosecond TA spectroscopy based on the time-resolved pump—probe method is a powerful tool to investigate the ultrafast dynamics in SNPs [44–46]. Figure 4 TA spectra of the colloid CsPbBr<sub>3</sub> perovskite in toluene solution (a) without Pt MNPs and (b) with 1.8 wt%. The photobleaching signals at 454 nm (PB1) is corresponding 441 nm absorption in the colloid CsPbBr<sub>3</sub> perovskite toluene solution related to PbBr<sub>2</sub>, and 520 nm (PB2) related to the exciton transition of the colloid CsPbBr<sub>3</sub> perovskite. Comparing figures 4(a) and (b), there are two differences: (i) the intensities of the former larger than the latter, (ii) the positions of PB2 of the latter have obvious blue-shift. The changes can be just caused by the coupling of Pt NPs to the CsPbBr3 SNPs exciton emitters, which result in hybrid plasmon—exciton mixtures. The interaction between the participating modes enables the significant modification of their properties. It is indicated that both the different bleaching behaviors. The Coulomb-interactions after excitation lead to modification for the exciton features.

The main reason for the differences between theory and experiment lies very likely in inappropriateness of the Förster mechanism for describing the energy transfer processes involving MNP. Therefore, to bring the theory presented to a more quantitative level, it is of primary importance to derive theoretical expressions for energy transfer processes involving MNP, in addition to conducting further PL measurements using SNP of precisely determined quantum yield.

#### 4. Conclusions

In this article we have synthesized colloidal CsPbBr<sub>3</sub> perovskite nanoparticles and prepared hybrid CsPbBr<sub>3</sub> perovskite/Pt nanostructures. PL spectra of the hybrid nanostructures show a narrowing (FWHM ~20 nm) and emission energy red-shift. Significant both PL quenching and lifetime enhancement by the interaction of

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exciton-plasmon couples in hybrid CsPbBr<sub>3</sub> perovskite/Pt nanostructure were observed. It is dedicated that the results can't well be explained only based on conventional FRET theory. The hybrid structures often exhibit unconventional optoelectronic properties due to the synergetic interaction between the individual components. A modifiation from the exciton feature to the exciton-plasmon couple has been demonstrated by the femtosecond TA experiment. To explore underlying mechanics of both PL quenching and lifetime enhancements, quantitative analyst for excitation energy near resonance of plexciton should be necessary.

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