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Enhanced photoluminescence of a CsPbBr$_3$/Al$_2$O$_3$ film heterojunction enabled by coupling of gold nanoparticles

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Abstract

We studied an enhanced photoluminescence (PL) effect of CsPbBr$_3$/Al$_2$O$_3$ heterojunction by using gold nanoparticles (NPs). The CsPbBr$_3$/Al$_2$O$_3$ heterojunction and CsPbBr$_3$/Au/Al$_2$O$_3$ heterojunction were respectively prepared on porous Al$_2$O$_3$ film by using a colloidal self-assembly method. The defect fluorescence of CsPbBr$_3$/Al$_2$O$_3$ heterojunction was enhanced up to 40 times due to the transfer of photo-generated carriers from the CsPbBr$_3$ quantum dots (QDs) to the porous Al$_2$O$_3$ film. To suppress the defect fluorescence of heterojunction, the CsPbBr$_3$/Au/Al$_2$O$_3$ heterojunction was prepared by loading the gold NPs between the CsPbBr$_3$ QDs film and the porous Al$_2$O$_3$ film. The results showed that the gold NPs facilitated the transfer of the photo-generated carriers from the CsPbBr$_3$ QDs to the surface of porous Al$_2$O$_3$ film. Compared with the porous Al$_2$O$_3$ film and CsPbBr$_3$/Al$_2$O$_3$ heterojunction, the peak fluorescence of CsPbBr$_3$/Au/Al$_2$O$_3$ heterojunction was enhanced up to 6.4 times and 2.8 times, respectively. This method of enhanced fluorescence of QD heterojunction provides a new pathway in the applications of photovoltaic, photodetector, LED, image sensor, and photocatalysis.

1. Introduction

Compared with the traditional organic or inorganic quantum dots (QDs), the perovskite quantum dots (PQDs) have a high photo-generated carrier density and quantum yield [1, 2]. Especially, all inorganic colloidal CsPbBr$_3$ QDs have a good optical stability and air stability among the organic or inorganic PQDs, which are also well semiconductor heterojunction materials and luminescent materials [3]. Usually, the PQDs act as optical active materials to form a heterojunction with other oxide semiconductor materials for increasing the photoelectric conversion efficiency (PCE) of the photovoltaic and LED device. Furthermore, the PQDs heterojunction is also the core of perovskite photovoltaic [4–6], LED [7, 8], photocatalysis [9, 10], and so on, which determines the optoelectronic performance of PQD solar cell. However, the defects and impurities of PQD heterojunction can capture the carriers of heterojunction interfaces, which reduces the carrier utilization and results in an inefficient optical radiation [11–13]. Besides, these defects and impurities can also result in a loss of open circuit voltage in PQD heterojunction device, which makes it difficult to improve the PCE. Therefore, it is a good solution to decrease the defects and impurities of PQD heterojunction for improving their PCE.

Gold nanoparticles (NPs) are one of the most widely used nanomaterials in the fields of nanophotonics and nano-optoelectronic devices, which have attracted a wide range of scientific interests [14, 15]. Moreover, the gold NPs and their coupled system can not only boost the light absorption [16, 17] and scattering [18, 19] to enhance the fluorescence [20, 21] and scattering signal [22, 23], but also promote the carrier transfer between different nanomaterials, which have been applied for photovoltaic, LED [24], biological detection [25] and photocatalysis [26].
In addition, the gold NPs can not only increase the light absorption and utilization efficiency, but also improve the physical and chemical properties of heterojunction interface to suppress the defects of PQD heterojunction. Therefore, they have also been applied in the electron transport layer (ETL) and hole transport layer (HTL) of PQD solar cells to improve the PCE of solar cells [27]. Recently, many studies have reported that Surface Plasmons (SPs) effect of gold NPs can increase the PCE of solar cell and LED in quantum dot heterojunctions [28, 29]. Snaith and co-workers make use of Au@SiO2 nanocomposites to passivate the surface of the PQDs, the PCE of solar cells is increased up to 11.4% [30]. Fan and co-workers have reported that the ZnO nanorods modified with the gold NPs can eliminate its defect emission to improve the band edge emission [31]. In addition, the gold NPs can also storage and transfer electrons of QD heterojunction to increase the carrier utilization. Thus, the gold NPs were an ideal carrier transport material of nano-heterojunction to enhance native emission and suppress the defect emission.

In this paper, we prepared self-assembly the CsPbBr3/Al2O3 heterojunction by using colloidal CsPbBr3 QDs and porous Al2O3 film. The defect fluorescence of CsPbBr3/Al2O3 heterojunction was enhanced up to 40 times. To suppress the defect fluorescence, we used the colloidal gold NPs acted as the medium of electron storage and transfer to enhance the fluorescence of CsPbBr3/Al2O3 heterojunction. It was found that the band edge emission of porous Al2O3 film was enhanced, while the defect emission was effectively suppressed due to the addition of gold NPs.

2. Materials and methods

The porous Al2O3 film (SINTI Ltd China) is a commercial porous film of aluminum substrate, which is fabricated by the electrochemical etching method. It has a distribution of cellular porous structure. The size and depth of single hole are respectively about 90 nm and 5 μm. Usually, the Al2O3 film is a wide band gap metallic oxide, which can’t radiate the fluorescence in the visible light range. However, the porous Al2O3 film has a high ratio surface area. The oxygen vacancies and Al interstitials defects are the predominant ionic defects in defect Al2O3 film [32]. The oxygen vacancies of the donor defects create an energy level below the conduction band, whereas the Al interstitials of acceptor defects create an energy level above the valence band. Thus, a defect band gap is formed between the conduction band and valence band inside Al2O3 band gap. The gap is formed between the conduction band and valence band inside Al2O3 band gap. The fluorescence in visible light range derives from the defect band gap.

The raw colloidal CsPbBr3 QDs (its number density is about 2.0 × 1018 ml⁻¹) were prepared by using a solvothermal synthesis method (See the supplementary materials is available online at stacks.iop.org/MRX/7/106201/mmedia). The size of single CsPbBr3 QDs was about 9 nm, which was packaged of oleic acid molecules. It was a cubic structure. Thus, it was stable in air. In addition, the colloidal gold NPs were a commercial colloidal nanoparticle solution (its number density is about 2.0 × 1017 ml⁻¹). The size of single gold NP was about 15 nm. Its surface was packaged with the citric acid molecules. The raw colloidal CsPbBr3 QD solution was diluted in n-hexane by a volume ratio of 1:10 to prepare the precursor solution of colloidal CsPbBr3 QDs. The solution of gold NPs was dispersed into ethanol by a volume ratio of 1:5 to prepare the gold NPs solution. All chemical reagents were not further purified.

A 20 mm × 20 mm porous Al2O3 film of aluminum substrate was averagely divided into four equal parts. Two sheets were used for preparing the CsPbBr3/Al2O3 heterojunction and CsPbBr3/Au/Al2O3 heterojunction. The others were used for measuring the fluorescence of porous Al2O3 film. Before used, they were washed three times by ethanol solution to get rid of surface dusts, and then were dried in vacuum drying oven for drying at 80 °C temperature.

The colloidal CsPbBr3 QDs were directly self-assembled on the surface of porous Al2O3 film to prepare CsPbBr3/Al2O3 heterojunction in a standard cuvette. To compare the enhancement effect of gold NPs, the colloidal gold NPs were self-assembled on the surface of porous Al2O3 film to prepare the Au/Al2O3 film in the standard cuvette, and then the colloidal CsPbBr3 QDs at same concentration were successively self-assembled on the surface of Au/Al2O3 film again to form CsPbBr3/Au/Al2O3 heterojunction. All film samples were not measured until they were completely dried in the vacuum drying oven.

The AFM surface morphologies and the photoluminescence (PL) spectra were in situ measured by the scanning near-field optical microscope (SNOM, Nanonics Ltd, Mv4000). A linear continuous laser with a central wavelength of 360 nm was guided into the multimode fibers and attained to the tip of a SNOM probe with a 100 nm aperture to form a 100 nm laser spot for exciting PL spectra of all film samples. In the tapping mode, the AFM picture and fluorescence spectra could be simultaneously in situ obtained in the process of scanning. The PL signals were collected by a ×50 objective lens (Nikon, plan SWLD NA = 0.40) to a multimode optical fiber, then attained to the optical fiber spectrometer (Ocean optics, QE6500). The morphology of peak optical spectra was measured in the process of AFM scanning. The integral time of single PL spectra was set as 1000 ms.
3. Results and discussions

3.1. Surface morphologies of CsPbBr3/Al2O3 and CsPbBr3/Au/Al2O3 heterojunctions

Figure 1(a) shows the AFM surface morphology of CsPbBr3/Al2O3 heterojunction. The CsPbBr3 QDs and their clusters were clearly observed in figure 1(a). The size of single CsPbBr3 QD was vertically about 9 nm, which was agreed with the raw material of colloidal CsPbBr3 QDs. Compared with the CsPbBr3/Al2O3 heterojunctions, the surface of CsPbBr3/Au/Al2O3 heterojunctions was smoother in figure 1(c). The lateral size of CsPbBr3 QD clusters were reduced from 159 nm to 139 nm. Furthermore, the single CsPbBr3 QD could be still observed, too. This also indicated that the gold NPs could modify the surface properties of porous Al2O3 film to reduce the aggregation of CsPbBr3 QDs on the surface of Au/Al2O3 for well-distributed CsPbBr3 QDs.

3.2. Enhanced PL spectra of CsPbBr3/Al2O3 and CsPbBr3/Au/Al2O3 heterojunctions

We measured respectively the PL spectra of porous Al2O3 film, colloidal CsPbBr3 QDs, CsPbBr3/Al2O3 heterojunction and CsPbBr3/Au/Al2O3 heterojunction at room temperature by a scanning near optical microscope in figure 2(a). The PL peaks of porous Al2O3 film and CsPbBr3 QDs were respectively at 450 nm and 500 nm, which provided a convenience for observing the electron transfer between the CsPbBr3 QDs and the porous Al2O3 film. The porous Al2O3 film had a relatively weak PL emission originated from its surface defects. The PL peak at 500 nm was the excitonic emission of CsPbBr3 QDs. After the CsPbBr3 QDs were self-assembled on the surface of porous Al2O3 film to form a CsPbBr3/Al2O3 heterojunction, the fluorescence of CsPbBr3 QDs was reduced, while that of porous Al2O3 film was dramatically increased. It showed that most energy of the colloidal CsPbBr3 QDs was transferred to the porous Al2O3 film for enhancing the fluorescence of porous Al2O3 film.

To clearly observe the enhanced fluorescence of differential heterojunctions, we drew the bar diagram at their peak fluorescence of two heterojunctions and the bar diagram at interfacial fluorescence of porous Al2O3 film, respectively, as shown in figure 2(b). Compared with the porous Al2O3 film, the fluorescence of two heterojunctions were both enhanced, furthermore, the level of CsPbBr3/Au/Al2O3 heterojunction was highest due to the induction of gold NPs. It showed that the photo-generated carriers of CsPbBr3 QDs were transferred...
to the porous Al₂O₃ film to increase the peak fluorescence and the interfacial fluorescence of two heterojunctions. The gold NPs played a more important role in the process of promoting the transfer of photo-generated carriers.

In order to illustrate defect enhancement effect, we divided the PL spectra of the CsPbBr₃/Al₂O₃ heterojunction to that of the porous Al₂O₃ film, as shown in figure 2(c). The fluorescence of CsPbBr₃/Al₂O₃ heterojunction was distinctly enhanced due to the photo-generated carrier transfer from CsPbBr₃ QDs to the porous Al₂O₃ film. The peak of enhanced fluorescence spectrum was located around 720 nm, which was resulted in by the interfacial defects of the porous Al₂O₃ film and the CsPbBr₃ QD film. The maximum enhancement factor was up to 40 times. However, the enhancement of interfacial fluorescence was relatively small. The photo-generated carriers had a large loss in the process of transport, which reduced the carrier utilization. In addition, there is a lot of noise behind the 650 nm range in figure 2(c). The reason was that there was almost no fluorescence radiation on the surface of porous Al₂O₃ film behind 650 nm range. The signal-to-noise ratio was very poor, and the fluctuation of fluorescence baseline was obvious. The fluctuation of the ratio between the fluorescence of CsPbBr₃/Al₂O₃ heterojunction and that of porous Al₂O₃ film in this range was also bigger. Thus, the bigger noise could be observed behind 650 nm range.

To suppress the defect fluorescence of CsPbBr₃/Al₂O₃ heterojunction, the gold NPs were added between the CsPbBr₃ QDs film and the porous Al₂O₃ film to act as the medium of carrier transfer and storage. Here, the gold NPs had two fold effects. (1) It could modify the surface of porous Al₂O₃ film to suppress the defects and reduce the surface energy. (2) It could storage and transfer the photo-generated carriers of CsPbBr₃/Al₂O₃ heterojunction to increase the carrier utilization. The PL spectra of CsPbBr₃/Au/Al₂O₃ heterojunction were further enhanced in comparison with the CsPbBr₃/Al₂O₃ heterojunction in figures 2(a) and (b). Moreover, the excitonic peak of CsPbBr₃ QDs was almost disappeared, while the fluorescence of porous Al₂O₃ film was also further increased in figure 2(a). It was further confirmed the gold NPs could boost the transfer efficiency of photo-generated carriers from the CsPbBr₃ QDs to the porous Al₂O₃ film. Besides, the surface of CsPbBr₃/Au/Al₂O₃ heterojunction was smoother than that of CsPbBr₃/Al₂O₃ heterojunction due to the modified of gold NPs. This result is consistent with the related literature reports [33].
The gold NPs could suppress the defect transfer rate of CsPbBr3 (shown in QDs lost the energy in the process of diffusion to the porous Al2O3 interface, they were trapped by the defect.

The CsPbBr3 heterojunctions.

The physical mechanism analysis of enhanced transfer effect of gold NPs in comparison with the CsPbBr3

Owing the electron transfer of CsPbBr3 QDs, the higher density of photogenic carriers would radiate more at their interfaces. And then the CB electrons were recombined with the holes to radiate the photo-generated electrons for transferring quickly them from the CsPbBr3 QDs to the porous Al2O3 defects via a diffusion effect and a built-in electric field.

The excited hot electrons and CB electrons were easily transported to the CB of porous Al2O3 and then were recombined with the holes at their defect centers. So, the defect fluorescence of CsPbBr3/Al2O3 heterojunction was also effectively suppressed and was also blue-shifted. Thus, the gold NPs could suppress the defect fluorescence of the heterojunction interface and could boost the electron transfer rate of CsPbBr3/Al2O3 heterojunction.

3.3. The physical mechanism analysis of enhanced fluorescence of CsPbBr3/Al2O3 and CsPbBr3/Au/Al2O3 heterojunctions

The CsPbBr3/Al2O3 heterojunction had a staggered energy band distribution, which could be described as shown in figure 3(a). The transfer processes of carriers could be explained as follows: (1) The electrons of CsPbBr3 QDs and porous Al2O3 films were simultaneously pumped by the 360 nm laser to their conduction band (CB), leaving the holes in valence band (VB). Because the CB of CsPbBr3 QDs was higher than that of porous Al2O3 defects, the excited hot electrons and CB electrons were easily transported to the CB of porous Al2O3 defects via a diffusion effect and a built-in electric field. (2) Accordingly, the holes were also accumulated at their interfaces. And then the CB electrons were recombined with the holes to radiate the fluorescence. The higher the density of photogenic carriers would radiate more fluorescence photons on the interface of porous Al2O3. Owing the electron transfer of CsPbBr3 QDs, the fluorescence of porous Al2O3 film was intensively enhanced, while that of CsPbBr3 QDs was decreased. (3) Some of the electrons that had not been transferred to the interfaces fell back to their VB for recombination with holes to radiate their intrinsic fluorescence. Thus, the excitonic fluorescence of CsPbBr3 QDs could also be observed. (4) As the photo-generated electrons of CsPbBr3 QDs lost the energy in the process of diffusion to the porous Al2O3 interface, they were trapped by the defect states of CsPbBr3 QDs, and then were recombined with the holes at their defect centers to radiate the defect fluorescence. Thus, the defect fluorescence could be enhanced up to 40 times.

The physical mechanism of enhanced fluorescence of CsPbBr3/Al2O3 heterojunction by gold NPs could be explained by the transfer of photo-generated carriers as shown in figure 3(b). The gold NPs played a lead role in the process of photo-generated electron transfer and storage, avoiding being captured by the defect state of the heterogeneous interface. It also increased the transport efficiency of photo-generated electrons from the CsPbBr3 QDs to the porous Al2O3 film for boosting the fluorescence of porous Al2O3 film, and for suppressing the defect fluorescence of heterojunction interface.

When the CsPbBr3/Au/Al2O3 heterojunction was pumped by 360 nm laser, the gold NPs acted as the bridge of photo-generated electrons for transferring quickly them from the CsPbBr3 QDs to the porous Al2O3 film. The fluorescence of porous Al2O3 film was respectively enhanced up to 2.8 times and 6.4 times, respectively. The peak of enhanced fluorescence at 450 nm was derived from the porous Al2O3 film interface. However, the defect fluorescence of CsPbBr3/Al2O3 heterojunction was also effectively suppressed and was also blue-shifted. Thus, the gold NPs could suppress the defect fluorescence of the heterojunction interface and could boost the electron transfer rate of CsPbBr3/Al2O3 heterojunction.
suppressed the defect states. And the tail of enhanced fluorescence of porous Al₂O₃ film was also blue-shifted. Thus, the gold NPs could facilitate the utilization of photo-generated carriers of CsPbBr₃/Al₂O₃ heterojunction, which transferred the photo-generated electrons from the CsPbBr₃ QDs to the porous Al₂O₃ film to enhance the fluorescence of heterojunction, and suppressed the defect fluorescence of heterojunction interface.

3.4. Comparison of peak fluorescence distribution on the surface of CsPbBr₃/Al₂O₃ heterojunction and CsPbBr₃/Au/Al₂O₃ heterojunction

In order to further verify the mechanism of enhanced fluorescence of CsPbBr₃/Au/Al₂O₃ heterojunction by gold NPs, we in situ measured the distribution of peak fluorescence spectra on the surface of CsPbBr₃/Au/Al₂O₃ heterojunction, as shown in figure 4. Figure 4(a) showed the distribution of peak fluorescence spectra of CsPbBr₃/Al₂O₃ heterojunction surface, and figure 4(b) was its projection plan picture. The distribution of peak intensity was not uniform, while the distribution of wavelength was uniform besides a disturbed area. It showed that the distribution of CsPbBr₃ QDs was not uniform on the surface of porous Al₂O₃ film, which depended on the self-assembly preparing process of CsPbBr₃/Al₂O₃ heterojunction. Furthermore, the peak fluorescence of CsPbBr₃ QDs played a lead role on the CsPbBr₃/Al₂O₃ heterojunction. The photo-generated carriers of CsPbBr₃ QDs hadn’t absolutely transferred to the porous Al₂O₃ film.

Compared with the CsPbBr₃/Al₂O₃ heterojunction, the distribution of peak fluorescence of CsPbBr₃/Au/Al₂O₃ heterojunction was well-distributed. The fluorescence peak was dominated from porous Al₂O₃ film instead of the CsPbBr₃ QDs. It demonstrated that the photo-generated carriers of CsPbBr₃ QDs were almost completely transferred to the porous Al₂O₃ film by the addition of gold NPs. These results were further confirmed our previous theoretical conclusions.

4. Conclusions

In conclusions, we had demonstrated an enhanced photoluminescence effect of CsPbBr₃/Au/Al₂O₃ heterojunction by using gold NPs. The heterojunctions of CsPbBr₃/Al₂O₃ and CsPbBr₃/Au/Al₂O₃ were fabricated by using a colloidal self-assembly method. Owing to the transfer of photo-generated carriers from the
CsPbBr3 QDs to the porous Al2O3 film, the defect fluorescence of CsPbBr3/Al2O3 heterojunction was enhanced to 40 times. To reduce the defect fluorescence, the gold NPs were self-assembled added into the surface of porous Al2O3 film to fabricate a CsPbBr3/Au/Al2O3 heterojunction. Compared with the CsPbBr3/Al2O3 heterojunction, the fluorescence of porous Al2O3 film was increased up to 2.8 times, suppressing effectively the defect fluorescence of heterojunction. These methods which enhanced fluorescence and suppressed defect fluorescence by gold NPs can be applied for photovoltaic, optical detection, LED and other devices to improve the photoelectric conversion efficiency of heterojunction.

See the supplementary materials for additional results for raw materials of CsPbBr3 QDs and prepared methods of CsPbBr3/Al2O3 heterojunction and CsPbBr3/Au/Al2O3 heterojunction.

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Data Availability

The main data that support the findings of this study are available with the article and its supplementary materials. Extra data are available from the corresponding author upon reasonable request.

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