Photoexcited Hole Cooling Dynamics in PbSe/CdSe Core/Shell Heterostructure Quantum Dots

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Introduction

Core/shell quantum dot (QD), in which the shell layer can not only improve the stability of core QD, but also result in totally new physical properties of the overall heterostructure. For instance, C. M. Cirloganu et al [1] found that the PbSe/CdSe quasi-type-II core/shell QD showed almost a fourfold increase in the multiple exciton generation (MEG) yield over core PbSe QD. However, photoexcited carrier dynamics in those quasi-type II QDs are not clear. In this study, we used transient absorption (TA) spectroscopy to reveal the photoexcited carrier dynamics in the PbSe/CdSe core/shell quasi-type-II QDs.

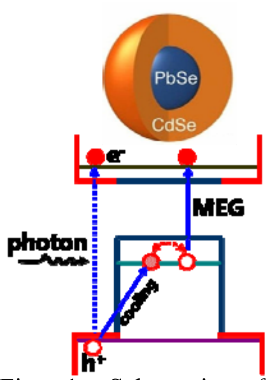

Fig. 1. Schematic of electronic state in PbSe/CdSe quasi type II core/shell QDs.

Experimental Method

The synthesis of colloidal PbSe and PbSe/CdSe QDs was performed via a reported method. [2] PbSe QDs and PbSe/CdSe QDs were dispersed in trichloroethylene (TCE) solution. In TA measurements, we used a titanium/sapphire laser (wavelength: 775 nm, repetition rate: 1 kHz, pulse width: 150 fs) as the laser source, the pump light wavelength was 470 nm and the probe beam wavelength was changed from 1140 nm to 1180 nm.

Results and Discussion

Fig. 2 shows the normalized TA responses (-ΔA) of PbSe and PbSe/CdSe QDs up to 600 ps for a probe wavelength of 1180 nm, which correspond to the electron-hole recombination of LUMO-HOMO in PbSe QDs. The photoexcited electron-hole pair lifetime of PbSe/CdSe QDs was longer than that of PbSe QDs, which suggested that the surface traps at PbSe/CdSe QDs are fewer than those of PbSe QDs. What's interesting is that, we found a new peak appeared around 200 ps in the TA response of PbSe/CdSe QDs. The 470 nm pump light can excite electron-hole pairs both in PbSe core and CdSe shell, we believe that the new TA peak means that there existed a hole cooling process from CdSe HOMO to PbSe HOMO (Fig. 1) and the hole cooling rate is as slow as 100 ps. This striking result indicates that the slow hole cooling process in the PbSe/CdSe QDs may cause the generation of MEG more efficiently.

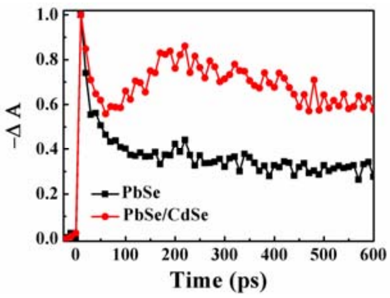

Fig. 2 Comparison of normalized TA responses of PbSe and PbSe/CdSe

References

1. C. M. Cirloganu, L. A. Padilha, Q. Lin, et al. Nat. Commun., 2014, 5, 4148.

2. G. Zaiats, D. Yanover, R. Vaxenburg, et al. Materials, 2014, 7, 7243-7275.