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Ultrafast Charge Transfer Dynamics and Auger Recombination of CdTe/CdS Core/Shell Quantum Dots

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【Introduction】

Core/shell semiconductor quantum dots (QDs) have potential applications in solar cell, bio-imaging and QD-based laser because of their unique optical properties.[1] For type I core/shell QDs, their quantum yields are much higher than the core because of the confinement of the charge carriers in the cores and the surface passivation of the shells. For type II, the band edges can be tailored from blue to near-IR wavelengths and the lifetime of charge carriers becomes longer due to the indirect recombination and the charge separation of the core/shell QDs. The exciton dynamics of CdTe/CdS core/shell QDs have been discussed in terms of the change of energy structure from type-I to type-II.[2] However, little has been reported on Auger recombination of CdTe/CdS core/shell QDs. In the present study, Auger recombination and the change of energy structure of CdTe/CdS core/shell QDs with different size of CdTe core are discussed according to the experimental results of time-resolved luminescence and transient absorption (TA) spectra.

【Experimental】

Aqueous soluble CdTe/CdS core/shell QDs were hydrothermally synthesized with successive ionic layer adsorption and reaction method. Steady-state absorption, luminescence, XRD spectra and TEM were used to confirm the formation of CdS shells on CdTe cores. Time-resolved luminescence dynamics were measured by a time-correlated single-photon counting system. TA spectra were performed with different excitation wavelengths at the first exciton states of core and core/shell QDs.

【Results and discussion】

Steady-state absorption and luminescence spectra of CdTe and CdTe/CdS core/shell QDs are

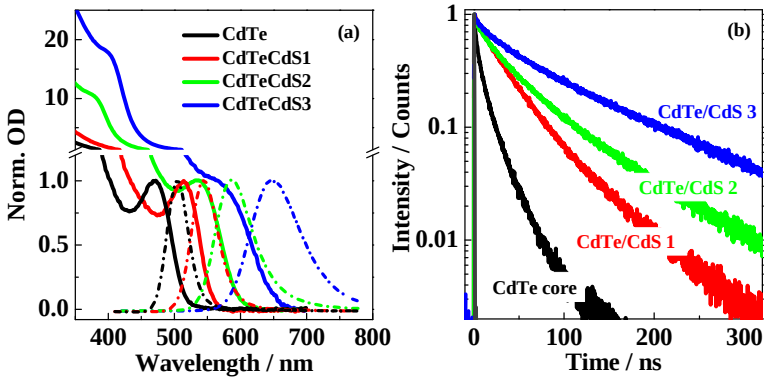

Fig. 1 Steady-state absorption and luminescence spectra of CdTe core and CdTe/CdS core/shell QDs (a) and the luminescence decays of CdTe core (2.2 nm) and CdTe/CdS core/shell QDs (b).

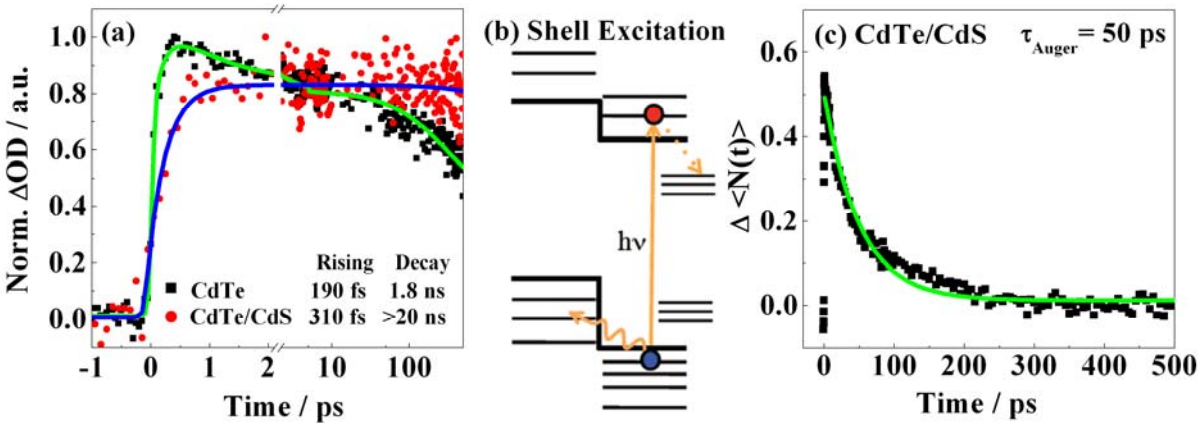

Fig. 2 The decay profiles corresponding to 1S states of CdTe core and CdTe/CdS core/shell QDs (a); the charge separation scheme of CdTe/CdS core/shell QDs with 400 nm shell excitation (b); Auger recombination dynamics of CdTe/CdS core/shell QDs (c).

shown in Fig. 1a. Additional peaks around 400 nm for core/shell QDs represent the formation of CdS shells. Moreover, the 1S states were red-shifted and finally became much featureless with the increasing of shell thickness. The luminescence spectra were also red-shifted and broadened. In Fig. 1b, the luminescence lifetime of core/shell QDs increased with the increase of the CdS shell thickness, which was explained in terms of the modification of surface trapping and the delocalization of electrons in type-II core/shell QDs. With 400 nm excitation, the decay profiles at 1S states of CdTe core and CdTe/CdS core/shell QDs were plotted in Fig. 2a. The exciton lifetime, fitted with two exponential components, of core/shell QDs became longer than that of CdTe QDs, due to the surface passivation and charge separation. Under the shell excitation of CdTe/CdS QDs with thicker shells, the charge separation dynamics are schematically illustrated in the Fig. 2b. Type-I CdTe QDs converted to type-II CdTe/CdS heterostructures, which was characterized by spatial separation of charge carriers, elongated luminescence lifetimes and significant red-shifted spectra. In addition, Auger recombination lifetime of CdTe/CdS core/shell QDs (~50 ps) became much longer as compared with that of CdTe core (~4 ps), as shown in Fig. 2c. These results are in a good agreement with the previous reports in our group.[3,4]

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