Study of energy transfer in mixed system of two different sized quantum dots

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It is important to investigate resonance energy transfer (RET) between quantum dots (QDs) because RET provides an approach for communication and coupling between QDs which is one of the central themes in numerous scientific efforts of present physical and technological interest [1, 2].

In this work, we characterized energy transfer process in mixed system of two different sized CdSe QDs by steady-state and time-resolved photoluminescence (PL) spectroscopy. Two kinds of CdSe QDs (2.6nm and 4nm in diameters) were used in this work. The concentration of small dots (donor) was 18.4nmol/ml and that of large dots (acceptor) was 10.4nmol/ml. The mixed sample was dispersed in toluene solvent (the concentration of small dots is 13.2nmol/ml and that of large dots is 3nmol/ml). The mixed solution was put in a quartz cell. The steady-state and time-resolved PL spectra were observed during evaporation of the solvent.

Figure.1 shows PL band variation of mixed sample with the increasing evaporation time. The PL emission band is composed of small QD emission (peak around 556nm) and large QD emission (peak around 586nm). It should be noticed that the both emission intensities of small and large dots drop monotonously with increasing evaporation time. This is called concentration quenching [3] originated from energy transfer between the same sized QDs. With increasing evaporation time, the PL intensity ratio of small to large QDs in PL band gradually decreases. The emission of small QDs is finally depressed by that of large QDs, which can be explained by RET process from small to large QDs [4, 5]. In order to confirm RET from small to large dots in mixed system, time-resolved PL spectra of small and large QDs are investigated in pure and mixed systems. The results show that the lifetimes of both small and large dots in mixed solution are the same as that of QDs in pure solution. However, compared with lifetimes of small and large QDs in pure solid films, in the mixed solid film the lifetime of the small QDs decreased while that of the large QDs increased, which can not be explained by concentration quenching. The observations strongly indicate that the transfer of excitation energy from small to large QDs is consistent with RET mechanism [4, 5]. We modeled our system using a proposed equation system. The RET rate from small to large dots and nonradiative recombination rate originated from concentration quenching are quantitatively estimated to be $0.13ns^{-1}$ and $0.08ns^{-1}$, respectively. The variation of integrated intensity ratio of small to large dots during the evaporation is shown in Fig.2. Three stages can be distinguished during the evaporation: initial stage (before 12.5min), transition stage (12.5-22.5min), and final stage (after 22.5min). In transition stage, since the ratio drastically decreases accompanied by the emission depression of small dots by that of large dots, we suggest that RET rate from small to large dots begins to increase in this stage. The effect of distance between small and large dots on RET in the three stages is evaluated.

References:

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Figures:

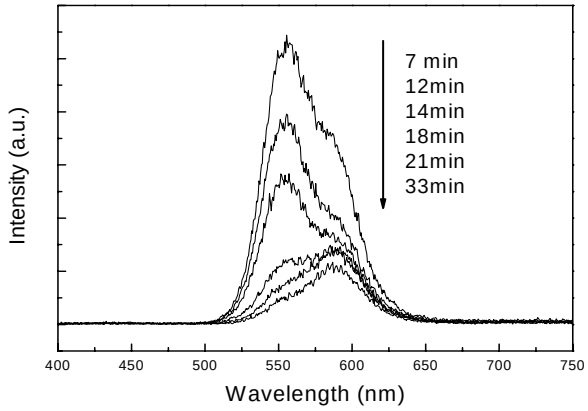

Fig.1: The PL band variation of mixed sample with the increasing evaporation time.

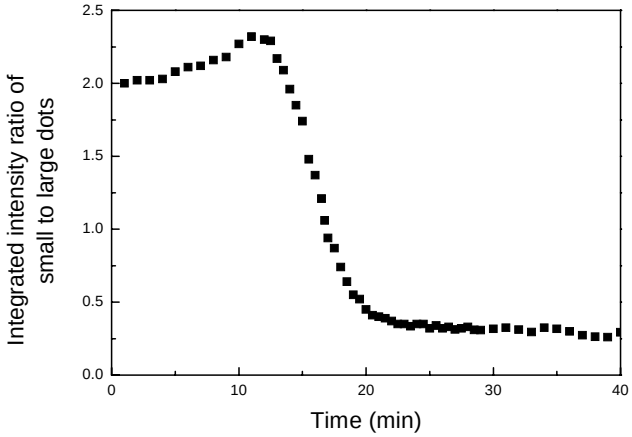

Fig.2: The variation of integrated intensity ratio of small to large dots with the increasing evaporation time.