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PLE spectra analysis of the sub-structure in the absorption spectra of CdSeS quantum dots*

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The semiconductor CdSeS quantum dots (QDs) embedded in glass are analysed by means of absorption spectra, photoluminescence (PL) spectra and photoluminescence excitation (PLE) spectra. The peaks of absorption spectra shift to lower energies with the size of QD increasing, which obviously shows a quantum-size effect. Using the PLE spectra, the physical origin of the lowest absorption peak is analysed. In PLE spectra, the lowest absorption peak can be deconvoluted into two peaks that stem from the transitions of $1S_{3/2}$–$1S_e$ and $2S_{3/2}$–$1S_e$ respectively. The measured energy difference between the two peaks is found to decrease with the size of QD increasing, which agrees well with the theoretical calculation for the two transitions. The luminescence peak of defect states is also analysed by PLE spectra. Two transitions are present in the PLE, which indicates that the transitions of $1S_{3/2}$–$1S_e$ and $2S_{3/2}$–$1S_e$ are responsible for the defect states luminescence.

Keywords: CdSeS, PL spectra, PLE spectra, defect state
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1. Introduction

Since the discrete optical transitions in quantum dots (QDs) were observed, great efforts have been made to study the internal electronic structure of the bound electron–hole pair states.[1−3] These atomic-like discrete electronic energy states are expected to have some unique features. For example, quantum-dot lasers are predicted to have a very low threshold current, so quantum dots may offer possibilities of building low-power nonlinear devices. The energy levels of quasi-0-dimensional semiconductor systems have been widely discussed by means of absorption spectra[4−6] and photoluminescence (PL) spectra.[7−9] Using photoluminescence excitation (PLE) spectra, energy relaxation mechanisms have been discussed.[10−13]

We have obtained a series of glass of semiconductor QDs by use of the method of co-melting.[7] At a temperature of 10 K, the QDs are studied by means of PL and absorption spectra. The quantum-size effect and the temperature effect of energy levels are analysed.[14] At room temperature, QDs' electronic energy and the relaxation process of electronic transition luminescence are analysed by means of PLE spectra.[15]

In this paper, the lowest absorption feature and PL spectra are analysed by means of PLE at room temperature. There are two peaks in the PLE spectra, corresponding to the lowest absorption peak, and the energy difference between the two PLE peaks diminishes with the size of the QD increasing. It indicates that sub-structure exists in the lowest absorption peak.

2. The samples and the analysis of absorption spectra

A series of CdSeS QDs embedded in glass with different sizes is obtained by annealing for 8, 16, 24 and 39 hours at 900 K respectively. These samples are labelled as 64H, 65H, 66H and 67H. Their QD radii are 2.5, 2.7, 3.0 and 3.2 nm, respectively. We study these samples at room-temperature by means of absorption spectra, PL spectra and PLE spectra. The

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samples for PL and PLE measurements are polished to a thickness of about 0.8 mm. PL and PLE spectra are recorded using the Fluorolog-3 spectro-fluorometer. A wavelength of 400 nm from a xenon arc lamp is applied in PL measurement. The samples for absorption measurement are polished to a thickness of 0.1 mm. Absorption spectra are recorded using the TU-1221 spectrophotometer.

The absorption spectra and PL spectra for QDs are shown in Fig.1. The first absorption peaks are at about 2.329 eV, 2.317 eV, 2.214 eV and 2.163 eV for samples 64H, 65H, 66H and 67H, respectively. The absorptions for bound states at higher energies are clearly resolved. We mark the two lower absorption peaks as A_1 and A_2. The spectral positions of A_1 and A_2 shift to higher energies with the size of the QD decreasing, which obviously shows a quantum-size effect.

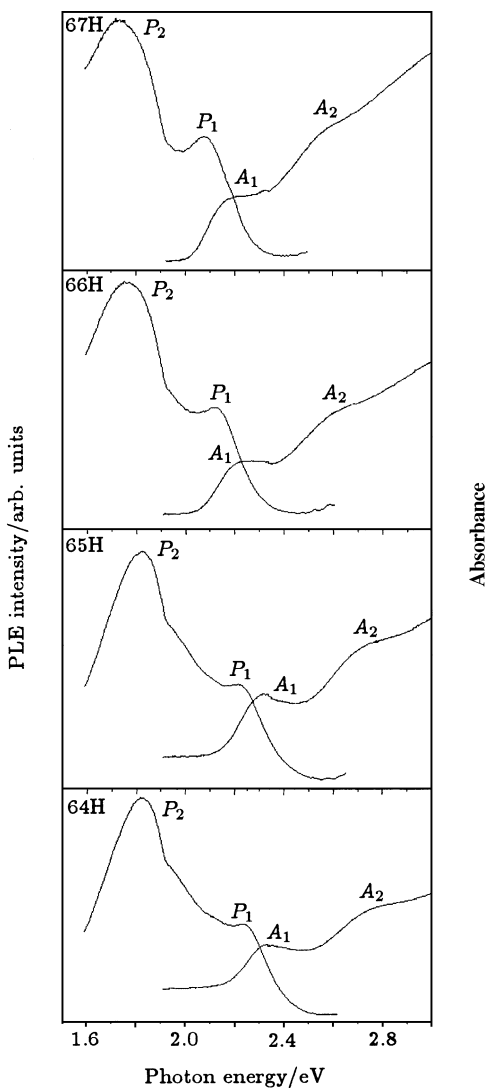


Fig.1. PL and absorption spectra of QD samples with different QD average sizes. The left is for PL spectrum, and the right is for absorption spectrum.

There are two prominent peaks in PL spectra in Fig.1, and they are labelled as P_1 and P_2. The spectral position of P_1 shifts to lower energies with the size of the QD increasing. This also shows the quantum-size effect. P_2 is caused by the defect states of surface of QDs.[14,16]

3. PLE spectra analysis of the sub-structure in the absorption spectra

In PLE, a narrow-band detection has been used in the low-energy tail of the absorption spectrum. Tuning the excitation energy through the whole spectrum and assuming fast relaxation, the PLE can probe the density of states. It is a probe for discrete, absorbing electron–hole-pair states. The prerequisite for this experiment is that the relevant luminescence of QDs can be effectively observed.

PLE spectra of samples 64H∼67H are shown in Fig.2. The detected energies are 2.066 eV, 2.066 eV 2.032 eV and 1.984 eV, respectively. The relative absorption spectra are shown in Fig.2, too. In the photon energy range corresponding to the extent of the spectral line profile with a peak A_1 in absorption spectra, there are two peaks, L_1 and L_2 in PLE spectra. The positions of the peaks are marked out by vertical bars in Fig.2, and their energies are shown in Table 1. L_1 and L_2 shift to lower energies with the size of QD increasing, which also shows the quantum-size effect.

The theoretical calculation[4] indicates that the lowest absorption peak A_1 is contributed from $1S_{3/2}$–$1S_e$ and $2S_{3/2}$–$1S_e$ transitions, and the energy difference between those two transitions decreases with the size of QD increasing. From Table 1, the difference in energy between L_1 and L_2 decreases as the size of QD increases. It is obviously shown that L_1 and L_2 originate from $1S_{3/2}$–$1S_e$ and $2S_{3/2}$–$1S_e$ transitions respectively, and the absorption feature A_1 results from the superposition of the two transitions.

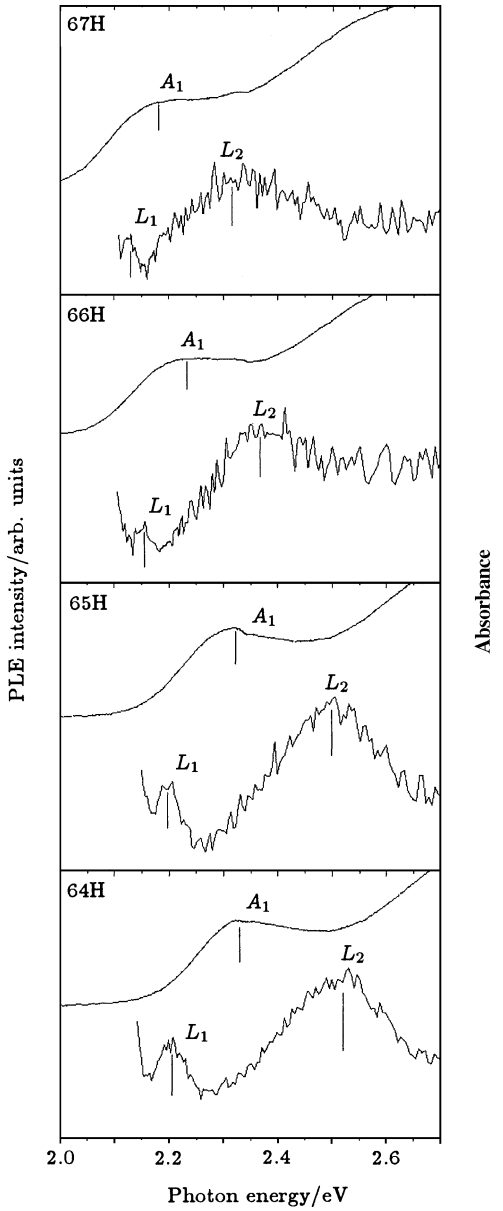


Fig.2. PLE spectra and absorption spectra of samples 64H~67H.

Table 1. The energies and energy differences between the two peaks L_1 and L_2.

Sample	A_1/eV	L_1/eV	L_2/eV	L_2–L_1/eV
64H	2.329	2.202	2.518	0.316
65H	2.317	2.195	2.495	0.300
66H	2.214	2.155	2.363	0.208
67H	2.163	2.129	2.317	0.188

4. PLE spectral analysis of defect state photoluminescence

The intensity of P_2 in PL spectra in Fig.1 is stronger. We preliminarily confirm P_2 to be responsible for the defect state luminescence.[14,16] In order to analyse the luminescence mechanism of P_2, PLE spectra of the samples are detected at the energy of P_2. Figure 3 shows the PLE spectrum of Sample 64H, detected at 1.83 eV (the spectral position of P_2). The dash line is for the PL spectrum of the sample.

At about 2.2 eV and 2.5 eV, PLE shows structures with a shoulder and a peak. Using Gauss-Fit, two peaks are present as shown in Fig.3. The two peaks corresponding to L_1 and L_2 in Fig.2, stem from the transitions $1S_{3/2}$–$1S_e$ and $2S_{3/2}$–$1S_e$, respectively. The PLE spectrum in Fig.3 indicates that the transitions of $1S_{3/2}$–$1S_e$ and $2S_{3/2}$–$1S_e$ are the main source of the defect state luminescence P_2.

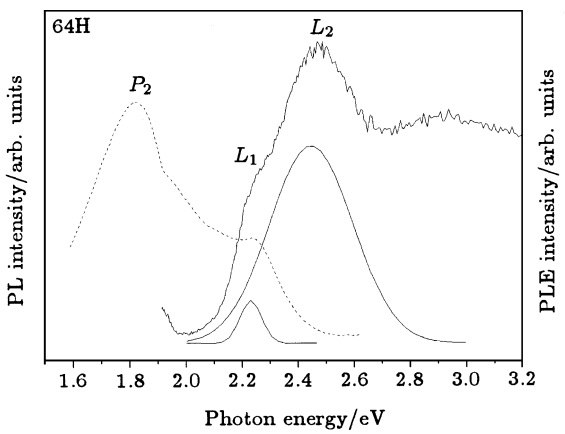


Fig.3. The PLE spectrum of sample 64H, detected at 1.83 eV of P_2. The dash line is for the PL spectrum of the sample. The two Gauss-Fit peaks of PLE spectrum are at the positions of L_1 and L_2.

5. Conclusions and discussion

The semiconductor CdSeS QDs are studied by means of absorption spectra, PL and PLE spectra. The spectral positions of A_1 and A_2 in absorption spectra shift to lower energies with the size of QD increasing, which obviously shows a quantum-size effect.

Using PLE spectra, the physical origin of the lowest absorption peak A_1 is analysed. In PLE spectra, A_1 is deconvoluted into two peaks, L_1 and L_2, which originate from the transitions of $1S_{3/2}$–$1S_e$ and $2S_{3/2}$–$1S_e$ respectively. When measuring the spectral positions of L_1 and L_2, we can find that the energy difference between the two peaks decreases with the size of QD increasing, which agrees well with the theoretical calculation for the two transitions.[4] The PLE spectra for A_1 show the sub-structure of A_1 obviously. The absorption peak A_1 is the superposition of two peaks from $1S_{3/2}$–$1S_e$ and $2S_{3/2}$–$1S_e$ transitions re-

spectively.

The luminescence peak P_2 of defect states is also analysed by PLE spectra. The two transitions of L_1 and L_2 are present in the PLE, which indicates that the transitions of $1S_{3/2}$–$1S_e$ and $2S_{3/2}$–$1S_e$ are responsible for the defect states luminescence.

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