Time-Resolved Photoluminescence Spectroscopy: A Novel Technique for Determination of Luminescence of Quantum Dots *

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The time-resolved photoluminescence (PL) spectroscopy measured by the gradually increasing start delay time is utilized as a tool for the determination of the luminescence of quantum dots (QDs). The luminescence evolution of self-assembled CdSe QDs during the luminescence decay is fully revealed in terms of the experiment technique. The characteristic narrow luminescence lines of self-assembled CdSe QDs are obtained with increasing start delay time.

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High quality semiconductor quantum dots (QDs) have been grown by molecular beam epitaxy (MBE) in terms of Stranski–Krastanow (S-K) grown modes.[1] In comparison with quantum wires fabricated by etching techniques, the self-assembled grown QDs can be dislocation-free and less defects, hence it has attracted much attention in opto-electronic field.[2−5] However, it is very difficulty to obtain the luminescence information with the separated or same-size QDs because of the nanometre size distribution and the size fluctuating of QDs. Usually in the measurement of PL for QDs, the radiative transition from QDs shows a broad luminescence band because the excited region involves many QDs with different sizes. Thus, the origin of the luminescence is unclear because the broad luminescence band can be caused by defects or by other recombination processes.

In this Letter, we propose a novel technique based on the time-resolved PL spectroscopy obtained by the gradually increasing start delay time and demonstrate it in self-assembled CdSe QDs. This technique is used to determine that the CdSe QDs can show the characteristic narrow luminescence lines. Usually, the luminescence signal is integrated during a time interval in the process of luminescence decay for the measurement of the conventional time-resolved PL spectroscopy. Here the time-resolved PL spectroscopy of the gradually increasing start delay time also involves the signal integrating over a time interval, the start time is gradually delayed while the stop time is fixed for all such time-resolved PL spectra.

The experiments were performed in a sample structure consisted of MBE grown 100 nm ZnSe cladding layer on a GaAs(110) substrate, followed by a 5-monolayer (ML) CdSe layer, and finally, a 50 nm ZnSe top cladding layer. The PL of the sample was excited by the 325 nm line of a continuous-wave (cw) He-Cd laser and detected by a cooled charge coupled device. The time-resolved PL measurements were carried out using a frequency-doubled beam of a mode-locked Al_2O_3:Ti laser pumped by an Ar^+ laser, and a photon counting method with a synchro-scan streak camera in conjunction with a single grating monochromator. The wavelength, the pulse width and the repetition rate of the excitation beam were 3800 Å, about 2 ps and 82 MHz, respectively.

The PL spectra at different temperatures are shown in Fig. 1(a). When the temperature is raised to 220 K, a broad luminescent band due to the QDs appears at the lower-energy side of the W band, and a shoulder appears at the higher-energy side. In order to understand the evolution of the bands clearly, the enlarged luminescence spectrum at 21 K is displayed in Fig. 1(b). The 2 ML and W bands originate from the radiative recombination of wetting quantum wells with CdSe layer thickness of 2 and 4 ML in the wetting layer,[6] respectively. The QDs band located at the lower-energy side in Fig. 1(b) can be ascribed to the excitonic luminescence of CdSe QDs.[6] It is noticed that the luminescence quenching from the wetting quantum well is quicker than that from the QDs with increasing temperature, it thereby results in the relative increasing luminescence intensity of QDs compared with wetting quantum well. In order to prove that the broad QDs luminescence band is composed of the luminescence of many quantum dots with different sizes, the time-resolved PL spectroscopy with the gradual start delay time is utilized to study the luminescence evolution of QDs in the process of luminescence decay.

Figure 2(a) shows the time-resolved PL spectra with the gradual start delay time at 250 K. It is found that the luminescence of the wetting layer gradually disappears with gradually increasing start delay time,

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whereas the emission processes continuously change. As the delay time increases further, the luminescence evolution of the QDs becomes clearer. The dominating luminescence of CdSe QDs evolves into a number of narrow lines around 547.2 nm from the broad band with increasing delay time. When the start delay time increases to 2002 ps, the dominating sharp lines become completely separate lines. In order to see clearly the emission lines, Fig. 2(b) displays the enlarged time-resolved PL spectrum in the time interval between 2002 and 2093 ps. Because the measured results are repeatedly carried out, it is found that the obtained dominated sharp luminescence lines keep invariable in the time interval between 2002 and 2093 ps. Thus, noise effect on sharp lines is fully excluded. These sharp lines are the contribution of the corresponding CdSe QDs to the luminescence. Why can the time-resolved PL spectra with the gradual start delay time clearly presents the emissions from different QDs? This is because the recombination lifetime of excitons in the CdSe wetting layer is shorter than that of CdSe QDs, and the lifetimes for different QDs are dependent on the size.

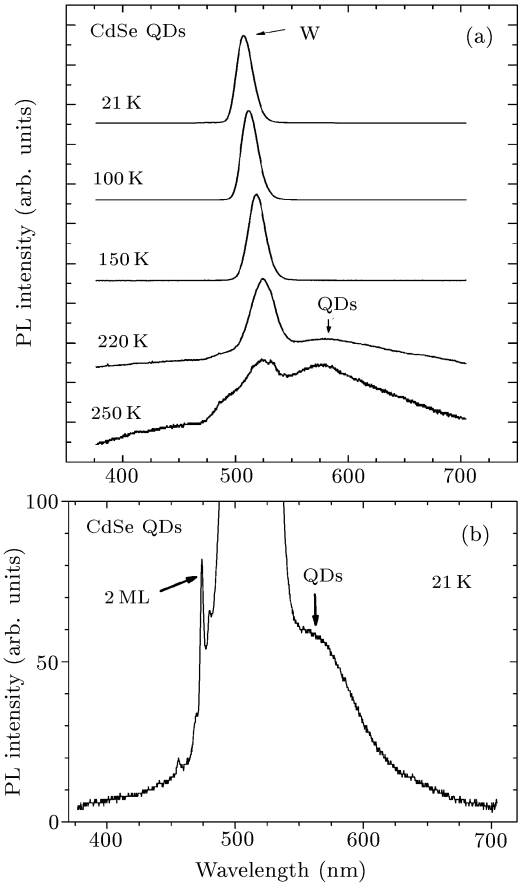

Fig. 1. (a) PL spectra of the self-assembled CdSe QD sample at different temperatures under the excitation of the 325 nm line from a cw He-Cd laser. (b) Enlarged PL spectrum of the self-assembled CdSe QDs at 21 K.

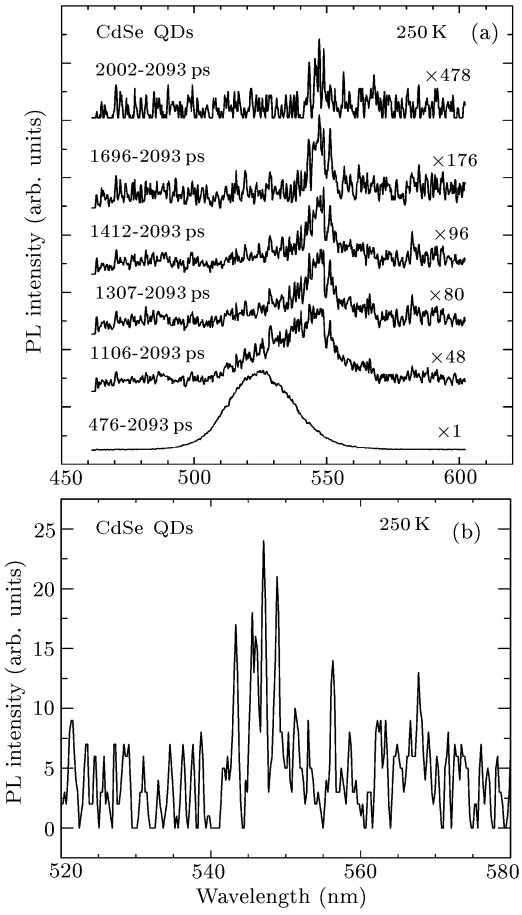

Fig. 2. (a) Time-resolved PL spectra of the self-assembled CdSe QDs measured by the gradual start delay time at 250 K. (b) Enlarged luminescence spectrum of Fig. 2(a) in the time interval between 2002 and 2093 ps.

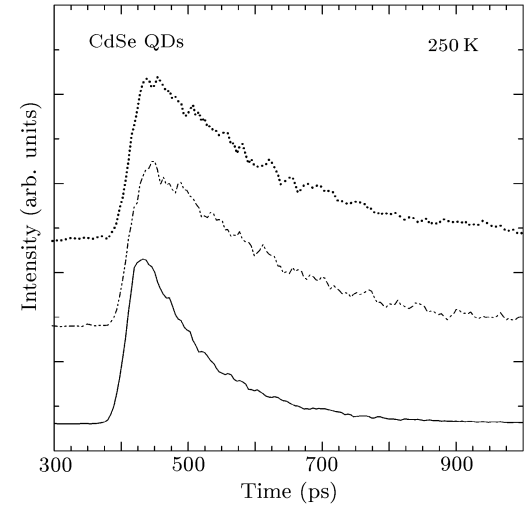

Fig. 3. PL decay curves of the wetting layer in the spectral range between 521.5 and 523.3 nm (solid line) and the QDs in the spectral ranges from 543.1 to 545.5 nm (dashed line) and from 545.9 to 551.2 nm (dotted line).

Figure 3 displays the PL decay curves of the wet-

ting layer and QDs in different luminescence regions. Their recombination lifetimes are 106 ps for the wetting layer, and 192 and 205 ps for QDs in the spectral ranges from 543.1 to 545.5 nm and from 545.9 to 551.2 nm, respectively. The measurement principle for the narrow luminescence lines of QDs is based on the differences of excitonic recombination lifetimes between wetting layer and QDs. As the start delay time increases, the luminescence of wetting layer and the luminescence of those QDs with shorter recombination lifetime have been excluded by the gradual increasing delay time. These residual sharp lines mainly result from the CdSe QDs with a longer recombination lifetime.

In summary, we have demonstrated the time-resolved PL spectroscopy measured by the gradually increasing start delay time as a tool for the determination of the luminescence of QDs. According to the difference of the recombination lifetimes, the unresolved broad luminescence band, which includes the recombination processes from the wetting layer and different QDs, evolves the sharp luminescence lines of different QDs with increasing delay time. Thus, the sharp luminescence lines from different QDs are clearly revealed in terms of the experiment technique.

References

[1] Leonard D, Krishnamurthy M, Reaves C M, Denbars S P and Petroff P M 1993 *Appl. Phys. Lett.* **63** 3203

[2] Kirstaedter N, Ledentsov N N, Grundmann M, Bimberg D, Ustinov V. M, Ruvimov S S, Maximov M V, Kopev P S, Alferov Z I, Richter U, Werner P, Gosele U and Heydenreich J 1994 *Electron. Lett.* **30** 1416

[3] Shernyakov Yu M, Egorov A Y, Zhukov A E, Zaisev S V, Kovsh A R, Krestnikov I L, Lunev A V, Ledentsov N N, Maximov M V, Sakharov A V, Ustinov V M, Chen C, Kopev P S, Alferov Z I and Bimberg D 1997 *Tech. Phys. Lett.* **23** 149

[4] Pan D and Towe E 1998 *Electron. Lett.* **34** 1883

[5] Chu L, Zrenner A, Böhm G and Abstreiter G 1999 *Appl. Phys. Lett.* **75** 3599

[6] Hu X B, Zheng Z H, Zheng J J, Gong W W and Gao W 2007 *Thin Solid Films* (to be published)