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The Origin of Multi-Peak Structures Observed in Photoluminescence Spectra of InAs/GaAs Quantum Dots*

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Abstract: Multi-peak structures in photoluminescence spectra of InAs/GaAs quantum dots are investigated. Excitation power-dependent photoluminescence spectra are used to identify the nature of different peaks. By combining experimental results and an energy-level structure analysis, origins of the multi-peaks are identified. Furthermore, inter-subband spacing of electrons and holes are deduced.

Key words: quantum dots; multi-peak structure; energy-level structure; photoluminescence
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1 Introduction

Self-assembled quantum dots (QDs) with atomic-like discrete energy states are important for applications in optical and electronic devices[1,2] and fundamental physics[3] due to the three dimensional confinement on the carriers[4]. Correspondingly, a number of well resolved peaks due to the discrete electronic shells have often been observed in state-filling spectroscopy[5~8]. Multiple peak structures due to different transition processes in the photoluminescence (PL) spectra of InAs QDs have been identified to be transitions between excited electron and hole states with the same quantum number[9], from bimodal or multimodal size distribution[10], or transitions between the ground electron state and excited hole levels in the dots[11,12] etc. Information of energy levels in a QD can be directly obtained according to the origins of the PL peaks, which is the basic parameter for such investigations as carrier relaxation[13] in fundamental researches and QD infrared detectors[14,15] in device applications.

Besides the transitions mentioned above, Yang *et al*.[16] reported the observation of a small PL peak at the high-energy side of the ground peak at low excitation power P_{ex}. Usually under a low P_{ex}, only the ground peak is observed. Yang *et al*. confirmed that the small peak cannot be attributed to the transition between the first excited electron and hole states with $n = 2$ in a QD by time-resolved PL measurements. They explained the appearance of the small peak in terms of the transition between the first excited electron state with $n = 2$ and the ground hole state with $n = 1$. Such a transition was usually thought to be forbidden for a QD symmetrical in geometrical shape. Therefore, they thought the appearance of such peaks was related to an n-type doping and an asymmetric built-in electric field[16].

In this work, excitation power dependent PL spectra of InAs/GaAs QDs with a multi-peak structure are investigated. Besides the peak due to the transition between the first excited electron state with $n = 2$ and the ground hole state with $n = 1$ reported by Yang *et al*., more peaks belonging to the forbidden transitions are observed. The excitation power dependence of the position of these peaks is systematically analyzed and the detailed inter-subband spacing for both electron and hole is deduced accordingly.

2 Experiment

The sample was fabricated on GaAs (001) substrate using molecular beam epitaxy (MBE). After the growth of the GaAs buffer layer at a substrate temperature of 600℃, an InAs QD layer and a 10 nm GaAs layer were deposited at ~535℃. Finally, a GaAs cap layer grown at 600℃ completed the epitaxial structure. For PL measurement, the sample was mounted on the cold finger in a closed cycle cryostat and excited by a double-frequency diode-pumped Nd:YAG laser (532nm). The PL emission was detected by a Fourier transform infrared spectrometer (IFS 120HR) with an InGaAs detector.

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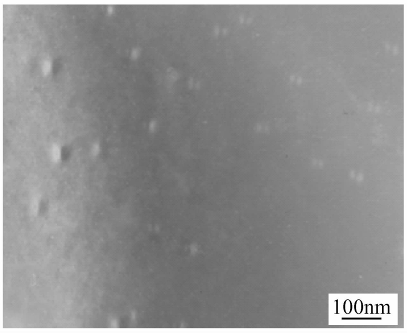

Fig. 1 Plane-view TEM image of the QD sample

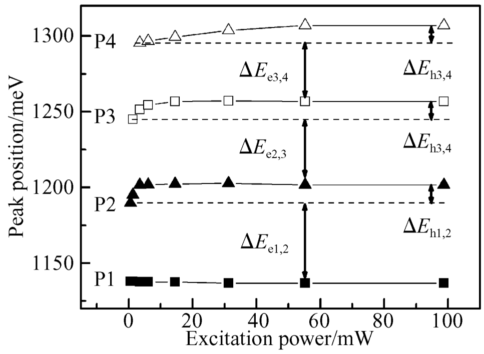

Fig. 3 Shifts in peaks P2, P3, and P4 as excitation power increases

3 Results and discussion

Figure 1 is the plane-view transmission electron microscopy (TEM) image of the QD sample. The sample shows a relatively low sheet density of $2.89\times 10^{9}cm^{-2}$. Figure 2 shows the PL spectra of the QD sample under different excitation powers at 80K. At the lowest excitation power of 0.5mW, only two peaks, P1 and P2, were observed, at 1137.9 and 1189.7meV, respectively. As P_{ex} increases, more peaks appear at the high energy side of the PL spectra. At 3.5mW, the third peak P3 occurs at 1245.0meV; at 14.5mW, the fourth peak P4 occurs at 1295.5meV. A peak at 1438.8meV was observed under all excitations, which is identified as the emission from the wetting layer (WL). The P2, P3, and P4 peaks shift towards high energy as the excitation power increases, as shown in Fig. 3. P2 reaches 1201.7meV at 3.5mW and does not increase any more as P_{ex} increases further. P3 increases from 1245.0 to 1256.8meV when P_{ex} increases from 3.5 to 14.5mW, and no more shifting can be observed as P_{ex} continues to increase. P4, originally occurring at 1295.5meV at 14.5mW, moves to 1307.0meV at 55mW and remains there to 98mW, the highest excitation power. Such multiple-peak structures correspond to the low sheet density and uniformity of QDs observed in the TEM image shown in Fig. 1.

The feature of the PL peaks changing with the increase of P_{ex} indicates that P1 under various P_{ex} and other peaks under high P_{ex} can be interpreted as excited state transitions of electrons and holes with the same quantum number. However, with P1, unchanged in position as P_{ex} increases, being identified with the ground-state transition, the small shift in P2, P3, and P4 may be explained in a similar way to Yang *et al*.[16]. At relatively low P_{ex}, there are more electrons than holes due to n-type background doping, and the former can fill up to the sublevels with $n+1$, whereas the holes fill at least up to the sublevels with n. Therefore, at the respective low excitation powers, these peaks should mainly originate from the transitions of electron sublevel $n+1$ to the hole sublevel n. As P_{ex} increases, more holes and electrons are photoexcited with the hole sublevels of $n+1$ being filled up, and transitions between the excited sublevels with the same quantum number mostly occur.

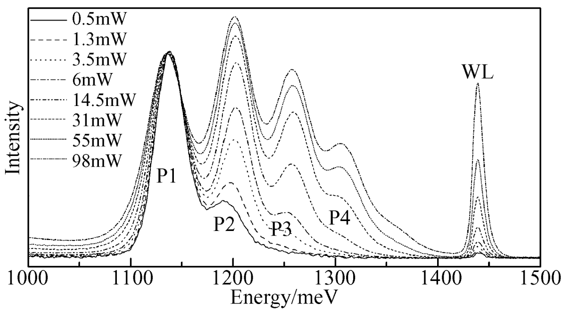

Fig. 2 Normalized excitation power dependent PL spectra of the QD sample at 80K

To demonstrate the situation more clearly, we take peak P2 as an example. At 0.5mW excitation, P2 originates mainly from the transition between the first excited electron sublevel with $n=2$ and the ground hole state with $n=1$ because the number of holes at the sublevel with $n=2$ is significantly smaller than that of the electrons. At relatively high excitation power ($P_{ex}\geqslant 3.5mW$), more holes were excited and the sublevel with $n=2$ is mostly filled up, and the transition mostly occurs between the sublevels with the same quantum number. At the intermediate excitation range with $0.5mW < P_{ex} < 3.5mW$, the $n=2$ hole levels can be filled in some QDs but remain empty in others due to the a relatively low P_{ex} and QDs size fluctuation. Under such conditions, the peak positions were determined by different weights in the transitions between the electrons and holes of the same quantum number and between the $n+1$ electron sublevel to the n hole sublevel. Therefore, P2 apparently shifts from 1189.7meV at 0.5mW to 1201.7meV at 3.5mW.

According to the shifts in peaks P2, P3, and P4 as

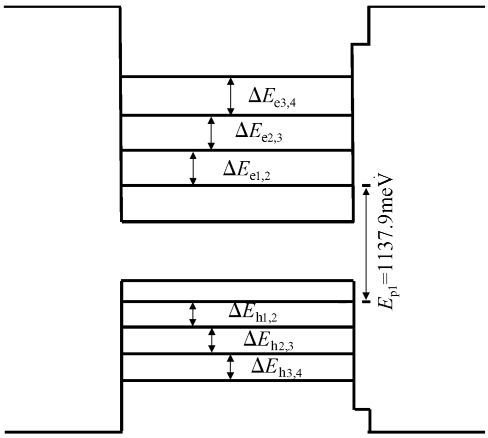

Fig. 4　Energy level structures in an InAs QD deduced from Fig. 3

excitation power increases, the electron sublevel spacings between the levels with $n=1$ and $n=2$; between $n=2$ and $n=3$; and between $n=3$ and $n=4$ can achieve $\Delta E_{e1,2}=51.8$meV, $\Delta E_{e2,3}=43.3$meV, and $\Delta E_{e3,4}=38.7$meV respectively by comparisons of peaks due to the $n+1$ electron to the n hole transition and the n electron and hole transition (as indicated in Fig. 3). The energy difference between the peaks due to the $n+1$ electron and hole transition and the $n+1$ electron to the n hole transition corresponds to the hole sublevel spacings between the levels with $n=1$ and $n=2$; between $n=2$ and $n=3$; and between $n=3$ and $n=4$, as indicated in Fig. 3. So, $\Delta E_{h1,2}=12.0$meV, $\Delta E_{h2,3}=11.8$meV, and $\Delta E_{h3,4}=11.5$meV can be achieved. The sublevels of electrons and holes in a QD were schematically shown in Fig. 4.

4　Summary

In summary, multi-peak structures in PL spectra of InAs/GaAs quantum dots were investigated. Excitation power-dependent PL measurements were used to identify the origins of different peaks. The positions of several peaks were observed shifting to higher positions as excitation powers increased. The shifts were explained in terms of the different weights in the transitions between the electrons and holes of the same quantum number and between the $n+1$ electron sublevel to the n hole sublevel. From the amount of the shifts in peak position, the electron and hole sublevel spacings were estimated to be $\Delta E_{e1,2}=51.8$meV, $\Delta E_{e2,3}=43.3$meV, $\Delta E_{e3,4}=38.7$meV, $\Delta E_{h1,2}=12.0$meV, $\Delta E_{h2,3}=11.8$meV, and $\Delta E_{h3,4}=11.5$meV, respectively.

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InAs/GaAs 量子点光致发光光谱多峰结构发光本质*

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摘要： 研究了 InAs/GaAs 量子点光致发光光谱中出现的多峰结构.观察到随着激发功率的增加光谱中发光峰的数目逐渐增多并且部分发光峰的峰位随激发功率的增加向高能量方向移动.解释了各发光峰的来源并结合量子点能级结构的特点,计算了量子点中电子和空穴各子带间的能级间距.

关键词： 量子点；多峰结构；能级结构；光致发光

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