Size dependent energy level positions of InAs quantum dots determined by selective excitation photoluminescence

H. S. Ling [a), S. D. Lin, S. Y. Wang, M. C. Luo, C. P. Lee
Department of Electronic Engineering, National Chiao Tung University,
1001 Ta Hsueh Road, Hsinchu 300, Taiwan
a) karshe.ee92g@nctu.edu.tw, Tel: 886-3-5712121-54248, Fax: 886-3-5733722

Abstract:

Self-assembled quantum dots (QDs) grown on lattice mismatched material systems using Stransky-Krastanov growth mode has been widely studied in recent years. Because of the randomness of the island growth, the QDs usually exhibit a wide size distribution. In conventional photoluminescence (PL) measurement, where the excitation energy is larger than the bandgap energy of the matrix material, the emission spectrum from the QDs usually reveals a broad Gaussian shape with a FWHM around 50 meV as a result of this size fluctuation. However, if we lower the energy of the pumping photons to the range where only the QDs can absorb, i.e., an energy lower than the absorption edge of the wetting layer, we enter the resonant excitation regime [1]. Because of the property of δ-function like density of states, only certain size QDs with transition energies identical to the excitation energy can be resonantly excited. In this work, the selective excitation technique was used to probe QD samples. ***Despite the wide size distribution, we were able to obtain detailed information pertaining to individual dots. The size dependent hole level positions can be accurately deduced.***

The single InAs QD layer used in this study were grown by molecular beam epitaxy on (001) semi-insulating GaAs substrates in a Varian GEN Ⅱ reactor. The selective excitation PL spectra of the QDs were obtained with a Ti-sapphire laser pumped by an Argon ion laser. The AFM image shows that the density of the QDs is about $1x10^{11}$ cm^{-2} and the diameter is around 8 nm. The PL peak can be fitted perfectly by a Gaussian function, suggesting natural size distribution of QDs with one predominant size. With a sufficient pumping power, the peak position of this Gaussian-like PL spectrum, measured at 1.269eV, represents the transition energy of the QDs with the highest population density. Such high transition energy, due to the ultra-small size of the dots, tells us there is only one electron bound state in the QDs [2]. The selective excitation PL spectra with three different excitation energies are shown in Fig. 1. When the excitation energy E_{ex} is higher than the wetting layer energy (1.43eV determined from the PLE measurement), there is only one broad peak, the same as conventional PL. However, when E_{ex} is lower than the wetting layer energy, the PL reveals two major peaks, which correspond to the ground state transitions of two QD subgroups with different sizes [3].

The reason for the appearance of the two emission peaks when the excitation energy is low can be explained using the size dependent QD band diagram shown in Fig. 2. If there is a continuous size distribution, the energy levels of the electrons in the conduction band and the hole levels in the valence band are continuously varied. Since there is only one electron level in the conduction band, we use it as a reference. The various hole levels are shown as sloped lines. As the QDs are radiated by a pumping source, only the dots with transition energies identical to the excitation energy are excited. For example, when E_{ex} is 1.3778eV, there are two groups of QDs can be resonant-excited: one (smaller dots) with C-2Hh excitation and the other (larger dots) with C-3Hh excitation. Once the electron-hole pairs are generated in the dots, the emission, however, is all from C-1Hh ground state transition. Because the two groups have different sizes, two emission peaks are observed. When the excitation energy is changed, different dot groups with different sizes can be selectively excited. So we can basically scan the whole population of the QDs and obtain the spectral information of individual QDs. Since we know the ground state transition energy of the dots with the highest population, it is convenient to use this energy as a reference to obtain the hole energy level information for this group of dots (let's call them group A). As shown in Fig.2, photons with two excitation energies can resonantly excite group A dots. One with an excitation energy of 1.3778eV and the other with an energy of 1.3464eV. The 1.3778eV photons excite the group A dots with the C-3Hh transition and the 1.3464eV photons excite them with the C-2Hh transition. Besides the group A dots, two other groups of QDs are excited by these photons as shown in Fig.2. The emission spectra, shown in Fig.1, show the emission peaks of the two groups of dots that are excited. For the 1.3778eV excitation, besides the emission from the group A dots, we also see a peak at 1.300eV corresponding to a smaller size group. For the 1.3464eV excitation, the additional emission peak is at 1.252eV, which corresponds to a larger size group. From the diagram shown in Fig.2, we can easily deduce the separation of the hole energy levels for the group A QDs. The difference between 1.3464eV and the ground state transition energy, which is 1.269eV, gives the energy separation between 1Hh and 2Hh. The difference between 1.3778eV and 1.3464eV gives the energy separation between the 2Hh and 3Hh levels. The obtained ΔE (1Hh-2Hh) and ΔE (2Hh-3Hh) are 77.4meV and 31.4meV, respectively.

In the same way, the hole sub-level information can be obtained for QDs with other sizes. Fig.3(b) shows the deduced size dependent energy positions of three hole sub-levels. The corresponding size distribution is shown in Fig.3(c). Our experimental result, is in good agreement with the theoretical prediction given by Grundmann *et al*[2]. For the dominant size dots (group A), the deduced two excited-state hole level positions are confirmed by the result of PLE measurement, as shown in Fig.3(a)(b).

In conclusion, we have developed a selective excitation photoluminescence spectroscopy technique to probe QDs with a large size distribution and, for the first time, we are able to obtain detailed spectral information for individual QDs. The size dependent energy level positions for different hole sub-levels can be deduced.

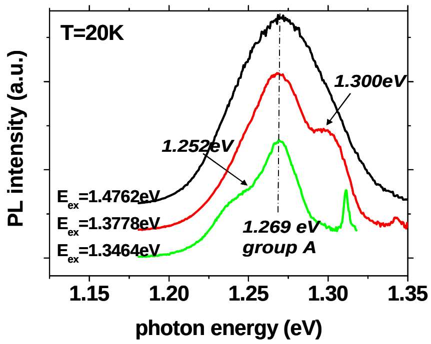

Fig. 1. Selective excitation PL spectra of InAs-GaAs QDs at 20K. Excitation energy of 1.4762eV is above InAs wetting layer, while that of 1.3778eV is below InAs wetting layer.

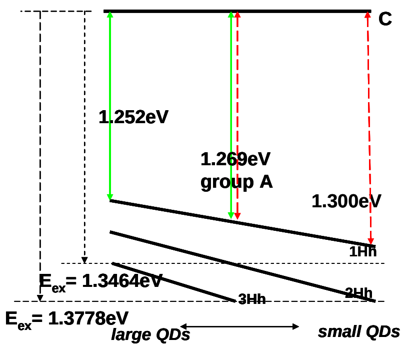

Fig. 2. Schematic size-dependent QD energy levels. The electron sub-level of different size QDs is leveled for reference.

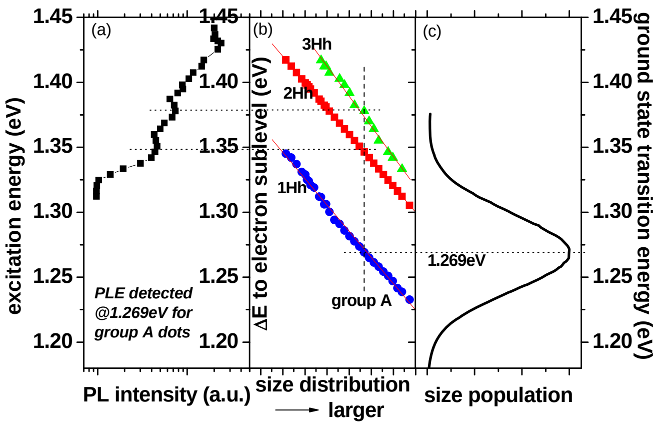

Fig. 3. Extracted hole sub-level information of individual QDs from selective excitation PL technique. (a) The PLE of dominant size QDs (group A), of which the detection energy is 1.269eV. (b) The energy difference to electron sub-level of three hole sub-levels. (c) The Gaussian-shape size population reflected by the high power PL spectrum of QD ensemble.

Reference:

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