APPLIED PHYSICS LETTERS **87**, 232110 (2005)

Experimental determination of electron and hole sublevels in modulation-doped InAs/GaAs quantum dots

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(Received 22 March 2005; accepted 13 October 2005; published online 1 December 2005)

Electron and hole sublevels in quantum dots (QDs) are experimentally determined using the excitation-power dependence of photoluminescence spectrum for a modulation-doped QD structure. The sublevel spacing between $n=1$ and $n=2$ electron states can be obtained from the photoluminescence (PL) spectrum under very low excitation and the spacing between $n=1$ and $n=2$ hole states can be obtained by comparing the PL spectrum under high excitation with the one under low excitation. The proposed method should give useful information in the design of QD devices, as well as for the verification of theoretical calculations of QD energy levels. © *2005 American Institute of Physics*. [DOI: 10.1063/1.2140882]

Quantum dots (QD) in which carriers are confined in all three directions have attracted great interest in both fundamental and applied research. Defect-free high-quality QDs have been grown by the Stranski–Krastanov growth mechanism; these high-quality QDs have improved our understanding of QDs and made it possible to fabricate high performance QD devices.[1] It is important to know the sublevel of each electron and hole in a QD because the sublevel is an important parameter both for fundamental issues such as carrier relaxation[2] and for applications, such as QD infrared detectors.[3] Carrier relaxation may depend on the sublevel spacing and is related to the device speed. The detection wavelength of a QD infrared detector also depends on the sublevel spacing, since the detector utilizes intersublevel transitions. It is difficult, however, to determine the sublevel of each electron and hole in QDs because only $\Delta n=0$ transitions are allowed in conventional photoluminescence (PL) measurements, and the PL peak spacing gives information only on the sum of the electron and hole sublevel spacings.

Several groups have calculated sublevels in QDs.[4–6] However, these calculations are complicated because the three-dimensional potential is sensitive to the shape and size of the QD, the strain distribution in the QD, and band offsets. The strain distribution in self-organized QDs is very complex because such QDs are grown using a large strain induced by a lattice mismatch between the QD and the barrier. In addition, for pyramid- or lens-shaped QDs, it is difficult to simplify the calculation due to the low symmetry of these structures.[5] Thus, to test the calculation results, and to determine the model calculation parameters without ambiguity, the sublevel of each electron and hole should be obtained by experiment. In this letter, we present a simple experimental method to determine the sublevel of each electron and hole in QDs.

The QD sample was grown on an undoped GaAs(100) substrate by molecular-beam epitaxy. The structure consists of a 0.6 μm thick undoped GaAs buffer layer, a 500 Å undoped $Al_{0.33}Ga_{0.67}As$ barrier, a 200 Å undoped GaAs quantum well (QW), a 400 Å undoped $Al_{0.33}Ga_{0.67}As$ barrier, a 400 Å n-doped (Si: 1×10^{18} cm^{-3}) $Al_{0.33}Ga_{0.67}As$ layer, and a 170 Å GaAs capping layer. A 2.15 monolayer InAs QD layer was grown at the center of the 200Å GaAs QW. Details are given in Ref. 7. Due to the inclusion of an n-doped layer in this system, electrons are present at the QDs even without photoexcitation.

Figure 1 shows the excitation power dependence of the PL spectrum at 10 K. The PL spectra were measured using a cooled InGaAs array detector. The full width at half maximum of the ground-state PL peak is about 35 meV, indicating rather uniform growth of the QDs. The PL intensity was very strong and was maintained more than 22% of the intensity at 10 K, in contrast to those of typical InAs/GaAs QDs.[8] Two

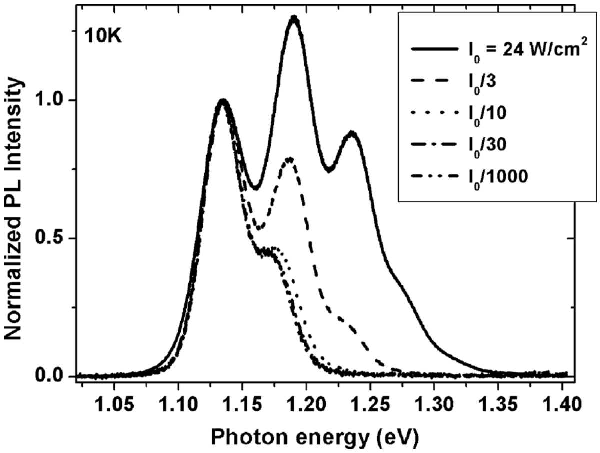

FIG. 1. Excitation power dependence of the PL spectrum from an n-modulation-doped self-organized InAs/GaAs QD at 10 K.

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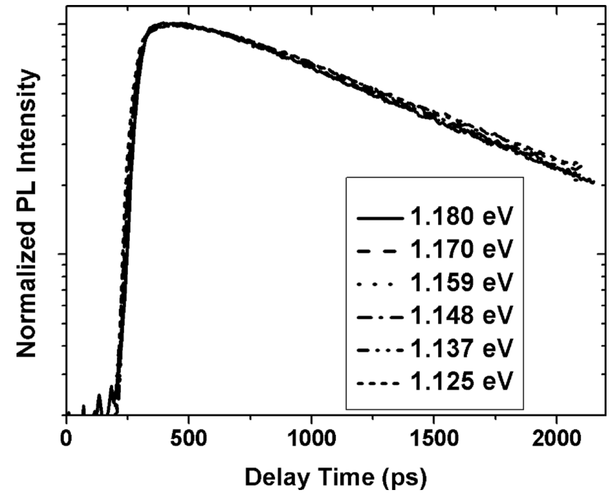

FIG. 2. PL decay curves at various energy positions. The numbers indicate the center wavelength of the employed bandpass filter of 10 nm. The carrier lifetimes are almost the same independent of energy position.

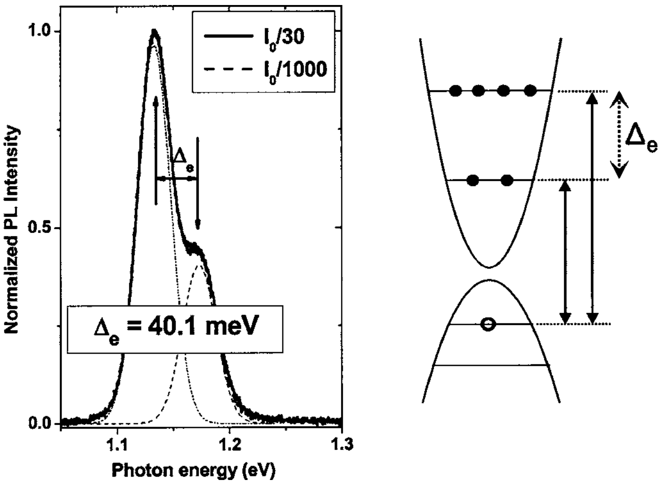

FIG. 3. PL spectra and a level diagram of the QD at an extremely low excitation. Filled circles indicate electrons and empty circles indicate holes.

excited state peaks are well resolved in the PL spectrum under the excitation intensity of 24 W/cm^2. The first excited state peak is stronger than that of the ground state since the degeneracy increases with energy level.[9]

In Fig. 1, it is interesting to note that the small peak at the high-energy side of the ground state peak does not disappear even at the lowest excitation. The position of this small peak was independent of the excitation intensity in the low excitation regime. Moreover, the small peak cannot be attributed to the transition between the $n=2$ electron sublevel and $n=2$ hole sublevel because it appears even at extremely low excitations, at which only a small part of the $n=1$ hole level will be occupied. Further evidence that the small peak cannot be from the transition between the $n=2$ electron sublevel and $n=2$ hole sublevel is provided by time-resolved PL measurements (Fig. 2), which show that the decay of the small peak is the same as that of the ground state under a low excitation of 0.8 W/cm^2. A picosecond streak camera and femtosecond Ti: Sapphire laser were used for the measurements. If the small peak had originated from the transition between the $n=2$ electron sublevel and $n=2$ hole sublevel, it would be expected to decay faster than the ground state.[10] The small PL peak is not related to an exciton complex because the energy difference between the ground state PL peak and the small PL peak is more than 40 meV, as shown in Fig. 1. In general, the energy differences between an exciton state and its complex states reported for In(Ga)As QDs are less than 10 meV.[11]

Previous studies have found that for a QD sample of very low height, size quantization is observed due to the discrete QD height.[12,13] In these systems, the emission wavelength was very short ($<1.0\ \mu$m) since the height contribution to the energy level is significant at a low dot height. In our system, however, the peak separation was not large and the emission wavelength was not short. Even if the small PL peak is related to the size quantization effect, the excited states of each small peak and the ground state peak should have appeared; however, no such features were observed in this experiment. In addition, a similar excitation power dependence of the PL spectrum was observed for another sample with the same structure but a different height.[7] Hence, the small PL peak cannot be related to size quantization effects.

Even without photoexcitation, electrons generated by the n-doped layer will be collected in QDs. Our experimental results can be adequately explained only if we assume that the doping causes electrons to fill up to the sublevel of $n=2$. At a low excitation, the electrons generated by doping and photoexcitation will fill up to the $n\geqslant 2$ electron sublevels, as shown in Fig. 3. On the other hand, the holes will occupy only the $n=1$ hole sublevel because holes are generated only by photoexcitation, which is low. Consequently, there will be no holes in $n\geqslant 2$ hole sublevels at 10 K.

Among the possible transitions, two would be considered the most likely to occur in our system: $n=1$ electron sublevel to $n=1$ hole sublevel and $n=2$ electron sublevel to $n=1$ hole sublevel. In general, only $\Delta n=0$ transitions are allowed in lens-shaped QDs, such as those used in the present study. In our sample, however, $\Delta n\neq 0$ transitions can occur because a symmetry toward a growth direction is destroyed by a built-in field caused by the n-modulation-doped layer. The relatively low intensity of the small peak is likely due to the dominant forbidden characteristic. Therefore, we can infer that the small peak originates from the transition between the $n=2$ electron sublevel and $n=1$ hole sublevel, while the ground state peak originates from the transition between the $n=1$ electron sublevel and $n=1$ hole sublevel. As a result, the energy difference between the two peaks corresponds to the electron sublevel spacing between the $n=1$ and $n=2$ states, as indicated in Fig. 3. From the fitting of the PL spectrum at low excitation, the electron sublevel spacing between the $n=1$ and $n=2$ levels was found to be 40.1 meV.

The above interpretation is also consistent with the results of the time-resolved PL measurements shown in Fig. 2. The decay time at low excitation in our QD sample is equal to the lifetime of minority holes in the $n=1$ hole sublevel because, in n-doped QDs, the temporal evolution of the PL is determined by the dynamics of minority holes. Hence, the decay time at low excitation is independent of the electron sublevel in our QD sample.

In the case of strong excitation, as shown in Fig. 4, electrons generated by the doping and photoexcitation will fill up to the $n>2$ electron sublevels and, at the same time, holes

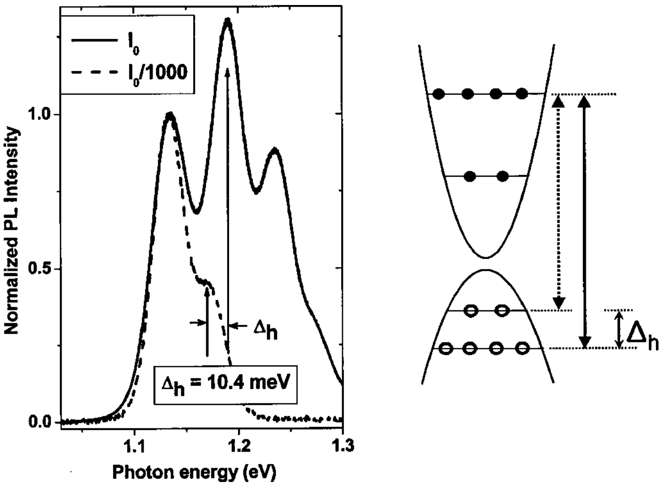

FIG. 4. PL spectra and a level diagram of the QD at a high excitation. Filled circles indicate electrons and empty circles indicate holes.

will also fill up to the $n > 2$ hole sublevels because the excitation intensity is high. In this case, $\Delta n = 0$ transitions rather than $\Delta \mathrm{n} \neq 0$ transitions are dominant. Hence, in the PL spectrum at high excitation, the first excited state peak originates from the transition between the $n=2$ electron sublevel and $n=2$ hole sublevel. The peak of the first excited state was independent of the excitation intensity in the high excitation regime. From the fitting of the PL spectra, the energy difference between the first excited state peak and the small peak was 10.4 meV, which corresponds to the hole sublevel spacing between $n=1$ and $n=2$. The present method may be very useful to determine whether the $n=2$ electron level is filled or not in an n doped QD sample, by observing a PL spectrum at a very low excitation, whereas it is difficult to calculate the number of electrons in a QD.

In conclusion, our method provides a simple and direct approach for determining sublevels without the need for simulations. Our method can be applied to both n and p doped QD samples, provided the sizes of the QDs are sufficiently uniform to allow the PL peaks to be resolved. The results obtained using our method should prove useful in the design of devices, such as QD infrared detectors utilizing sublevel transitions; in elucidating fundamental issues, such as carrier relaxation via the quantized energy levels; and in determining parameters for use in model calculations.

The authors appreciate the significant technical contribution of the late U. H. Lee. This work is supported by the National Research Laboratory program (Grant No. 2004-02403).

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