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Observation of OD Subband Structures in Single Quantum Dots by Microscopic Photoluminescence Excitation Spectroscopy

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Recently, observations of photoluminescence from single quantum dots by microscopic optical experiments were reported from several authors [1] These experiments are very valuable since one can exclude inhomogeneous broadening that is a major obstacle to investigating low dimensionality. Reported luminescence in these experiments was very sharp (with FWHM of 100-200 μeV). However, it seems difficult to conclude that this sharpening is a direct consequence of zero dimensionality because reported homogeneous width of 2D excitons is 200 μeV if inhomogeneous broadening is excluded. More unambiguous aspect of zero dimensionality is observation of discrete subband structures, which is the most prominent feature of 0D system. This paper reports a first observation of zero dimensional discrete subbands of single quantum dot by microscopic photoluminescence excitation (μ-PLE) spectroscopy.

$In_{0.4}Ga_{0.6}As/Al_{0.4}Ga_{0.6}As$ quantum dot samples were fabricated by MOVPE self-organized growth on (311)B GaAs substrate [2]. The averaged size is 30 Å in vertical and 400 Å in lateral direction. Figure 1 shows μ-PL spectrum (A) and normal macroscopic PL spectrum (B) at 5 K. The excitation light source is a cw-wavelength tunable Ti: sapphire laser. The spatial resolution is approximately 1 μm for μ-PL and PLE measurement. Number of illuminated dots are several tens for (A) and approximately 10^7 for (B). Very sharp luminescence lines observed in the μ-PL spectrum are thought to be emitted from individual quantum dots, as reported by other authors [1]. These sharp lines were observed up to 50 K. Figure 2 shows μ-PL spectra at 5 K and 50 K plotted with a narrower spectral region. The inset shows temperature dependence of FWHM, which clarifies that FWHM was unaffected by thermal broadening. This is strong indication of 0D density-of-states character.

By tuning the detection wavelength at the emission peak, we measured μ-PLE spectrum. The line PLE A in Fig. 3 was taken with the detection wavelength resonant to the emission peak and the line PLE B was taken with that off-resonant to the peak. The remarkable sharp absorption lines are observed only at the resonant case. This indicates that the sharp absorption lines are directly connected to the emission lines, and these thus originated from 0D subbands.

Since the energy position of the absorption peaks are different for different PL peaks, we can excite each line resonantly. Figure 4 (a) shows a PLE (shown in Fig. 4(b)), we measured PL spectra with resonantly exciting the sharp absorption line (labeled A). the results are shown as a line A in Fig. 4(b). The luminescence peak evolved dramatically. When we set the excitation wavelength slightly off-resonant to the absorption line, the PL peak intensity markedly decreased whereas the background was almost constant. The line B was taken at resonant to another subband. These results directly demonstrated the discrete nature of the observed absorption line which can be attributed to their zero-dimensional nature. Note that we selectively picked up single dot from the many illuminated dots within the laser beam by the resonant excitation.

We also confirmed that absorption peak positions can be explained by the energy level calculation using Fourier expansion method. Furthermore, we investigated homogeneous broadening by analyzing the half width of the absorption peaks. The result indicates that there is no threshold-like behavior at the LO phonon energy, which suggests that the LO phonon scattering is suppressed due to discreteness of 0D subbands.

In summary, we observed zero-dimensional subband structure of single quantum dots by μ-PLE spectroscopy for the first time. We confirmed discrete nature of the absorption spectra by resonant and off-resonant conditions. This nature enabled us to excite only single dot from the many illuminated dots by resonant excitation. The analysis of the half width of the absorption also gave us information about the intersubband relaxation phenomenon of 0D state.

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1 J.-Y. Martzin et al. Phys. Rev. Lett. **73**, 716 (1994); S. Fafard et al. Phys. Rev. B **50**, 8086 (1994); M. Grundmann et al. Phys. Rev. Lett. **74**, 4043 (1995).
2 R. Nötzel, J. Temmyo, and T. Tamamura, Nature **369**, 131 (1994).

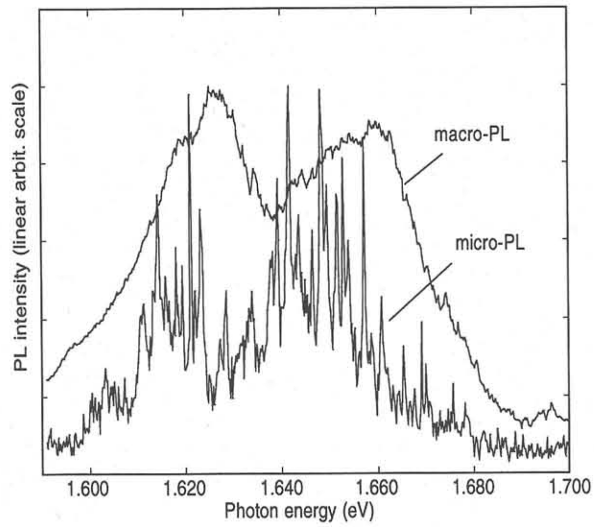

Fig. 1. PL spectra of $In_{0.4}Ga_{0.6}As$ quantum dots exited by macroscopic laser beam and microscopic beam.

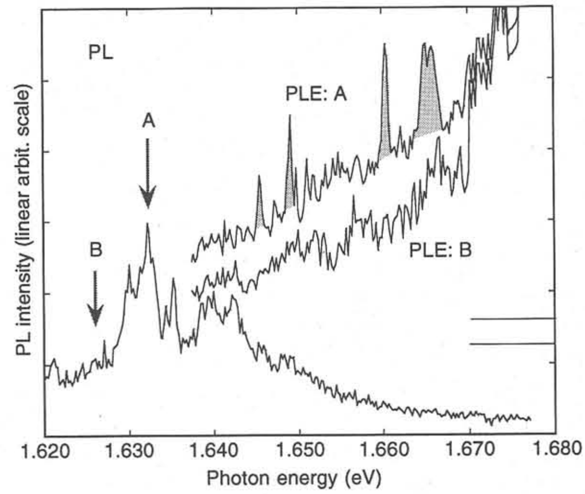

Fig. 3. μ-PLE and μ-PL spectra of InGaAs quantum dots. Detection photon energy for two PLE curves is indicated by arrows in the PL spectrum. Vertical axis of PLE curves is shifted and zero lines for both curves are represented by horizontal lines inside the figure.

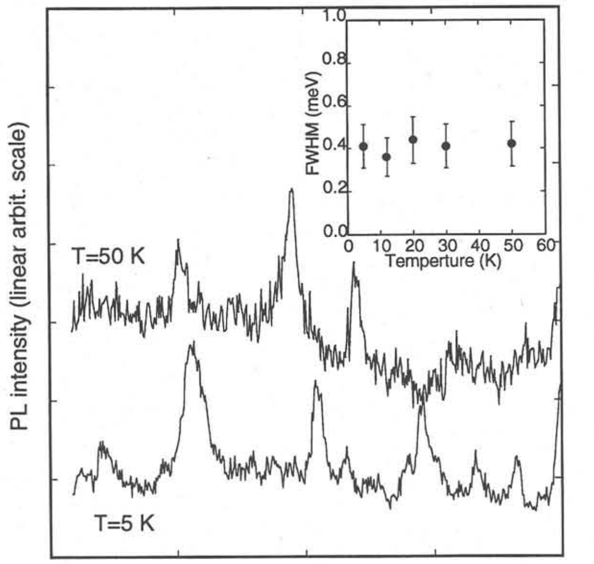

Fig. 2. μ-PL spectra at 5 K and 50 K. Temperature dependence of FWHM is shown in the inset.

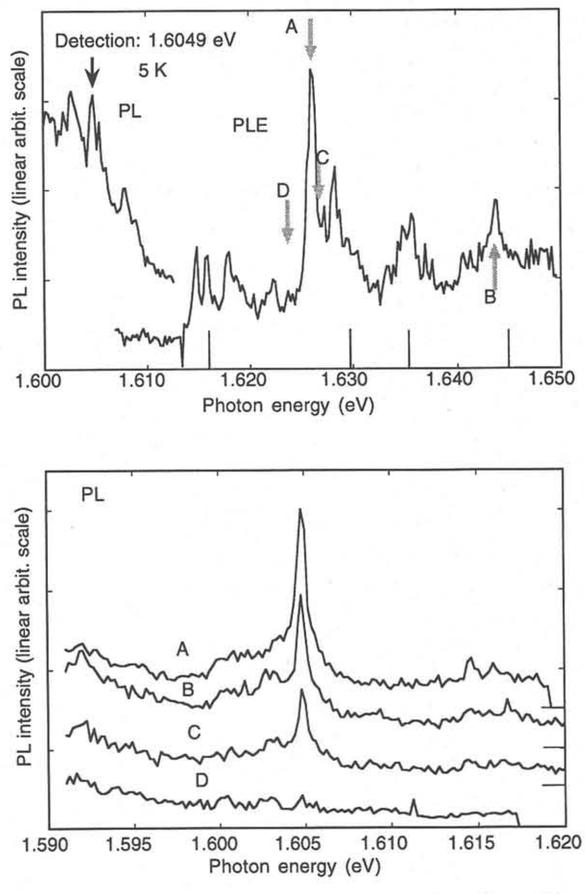

Fig. 4. (a) μ-PLE spectrum and corresponding PL spectrum excited at 1.72 eV. (b) Resonantly and non-resonantly excited PL spectra. The excitation photon energy for each PL curve in (b) is indicated by arrows in (a). Vertical axis of PL curves (A to D) in (b) is shifted and zero lines are shown by horizontal bars.