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Photoluminescence Studies on Self-Organized InAlAs/AlGaAs Quantum Dots under Pressure

J. D. PHILLIPS (a), P. K. BHATTACHARYA (a), and U. D. VENKATESWARAN[1]) (b)

(a) Department of Electrical Engineering and Computer Science, University of Michigan, Ann Arbor, MI 48109, U.S.A.

(b) Department of Physics, Oakland University, Rochester, MI 48309, U.S.A.

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The pressure dependence of the low temperature photoluminescence (PL) in self-organized $In_{0.5}Al_{0.5}As/Al_{0.25}Ga_{0.75}As$ quantum dots (QD) has been investigated up to 8 GPa. Interesting features of the QD PL observed in our study are: (i) a decrease in the linewidth up to 1.8 GPa, (ii) no significant shift in the PL energy between 0.8 and 2.2 GPa, (iii) anticrossing behavior in the region of 2.2 to 2.6 GPa, and (iv) complete quenching of PL beyond 2.6 GPa. The observed pressure behavior is explained on the basis of the crossing between Γ and X conduction bands.

Quantum dots (QD) of fairly uniform size and very high optical quality have been grown by self-organization during strained layer molecular beam epitaxy of InAs on GaAs and related ternary compounds [1]. In this method, known as the Stranski–Krastanow growth mode, growth starts two-dimensionally and after a few monolayers of growth, dislocation-free pyramidal islands are formed spontaneously. Efficient emission in the near-infrared (≈1.2 eV from InGaAs/GaAs QD) [2] or in the visible (≈1.8 eV from InAlAs/AlGaAs QD) [3] regions and for room-temperature laser operation [4] have been reported. In this paper, we report the temperature and pressure dependence of the PL in self-organized $In_{0.5}Al_{0.5}As$ QD of high density embedded in $Al_{0.25}Ga_{0.75}As$ barriers. Interesting pressure-induced energy shifts and narrowing of the QD PL as well as sharp PL peaks in the temperature range of 275 to 150 K are reported. We discuss this anomalous pressure shift of the QD PL in terms of the effects of strain on the energy band structure and crossover between Γ and X_{xy}, X_z conduction bands.

Two heterostructures, an $In_{0.5}Al_{0.5}As/Al_{0.25}Ga_{0.75}As$ QD sample and a reference sample without QD, were grown on semi-insulating GaAs substrates by molecular beam epitaxy. The QD structure consisted of a 0.2 μm GaAs buffer layer followed by a 0.2 μm thick $Al_{0.25}Ga_{0.75}As$ barrier layer. The $In_{0.5}Al_{0.5}As$ QD were then grown and capped with a 0.2 μm thick $Al_{0.25}Ga_{0.75}As$ layer. An additional surface layer of $In_{0.5}Al_{0.5}As$ QD was deposited under the same conditions of growth as for the buried QD layer. The reference sample was similar to the QD sample except for the absence of the embedded and surface QD.

Atomic force microscopy measurements show an areal density of 1×10^{11} dots/cm^2. PL was excited with 30 mW (or less) of 488 nm laser focused to a spot diameter of

[1]) Corresponding author; Tel.: (248) 370-3423, Fax: (248) 370-3408, e-mail: venkat@oakland.edu

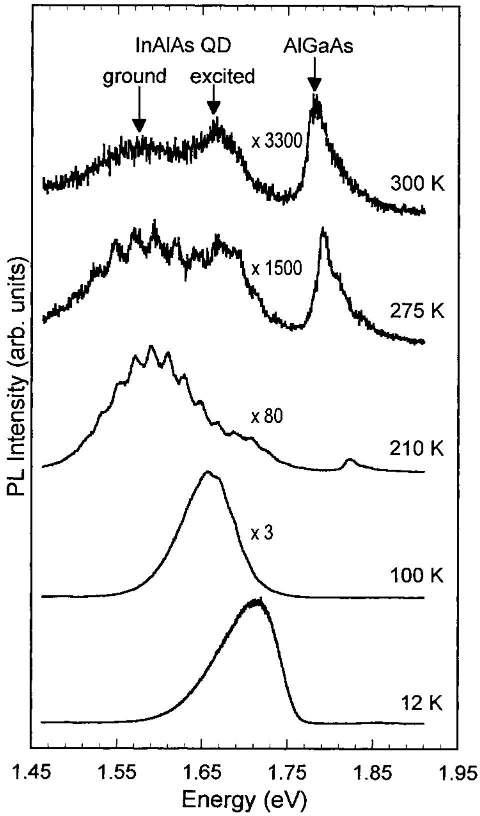

Fig. 1. Temperature dependence of the PL in $In_{0.5}Al_{0.5}As/Al_{0.25}Ga_{0.75}As$ QD sample. The sharp modulations in the PL intensity in the 275 and 210 K spectra are not due to Fabry-Perot fringes or LO phonon replicas

≈ 50 μm. A standard grating spectrometer (resolution of $\approx 4\ cm^{-1}$) and a photomultiplier with photon counting electronics were used for recording the PL spectra. A closed cycle helium refrigerator with a temperature controller (accuracy better than $\pm 1°$) and a Merrill–Bassett-type diamond anvil cell were used for the low-temperature and high-pressure measurements. The pressure transmitting medium was a 4:1 methanol–ethanol mixture and the pressure calibration was done using the standard ruby R-line fluorescence. Pressure was always adjusted at room temperature before making PL measurements at low temperature.

Fig. 1 shows the PL from the $In_{0.5}Al_{0.5}As/Al_{0.25}Ga_{0.75}As$ QD structure at a few representative temperatures. A series of sharp peaks, as shown in the spectra at 275 and 210 K, is observed between 275 and 150 K. From a sum of several Gaussians fit to the data, the energy separation between the sharp peaks is found to be between 24 and 19 meV in this temperature range. This energy separation is significantly small for LO phonon replicas or Fabry-Perot fringes to be the likely causes of the sharp peaks.[2]) The observed temperature-dependent characteristics of the PL spectra may be understood in terms of the distribution of carrier amongst the dots. At temperatures below 100 K, carriers are frozen in the dots in a non-equilibrium distribution and the single inhomogeneously broadened PL peak observed corresponds to the ground state transitions from this group of dots. At higher measurement temperatures, carriers are distributed more widely amongst the entire dot population and the observed sharp peaks may be attributed to emissions from discrete groups of dots with slightly different size and shape.

The low-temperature PL spectra of the QD sample at several pressures are shown in Fig. 2. In the atmospheric pressure (100 kPa) spectrum, three peaks at 1.5, 1.705, and 1.845 eV, corresponding to the emissions from the GaAs buffer/substrate, $In_{0.5}Al_{0.5}As$ QD, and $Al_{0.25}Ga_{0.75}As$ barriers, respectively, can be identified. The pre-

[2]) The LO phonon energies in AlAs, GaAs, and InAs are 50, 35, and 29.6 meV, respectively. The cavity thickness corresponding to a fringe separation of 20 meV is about 5 μm, much larger than the layer thickness in our sample.

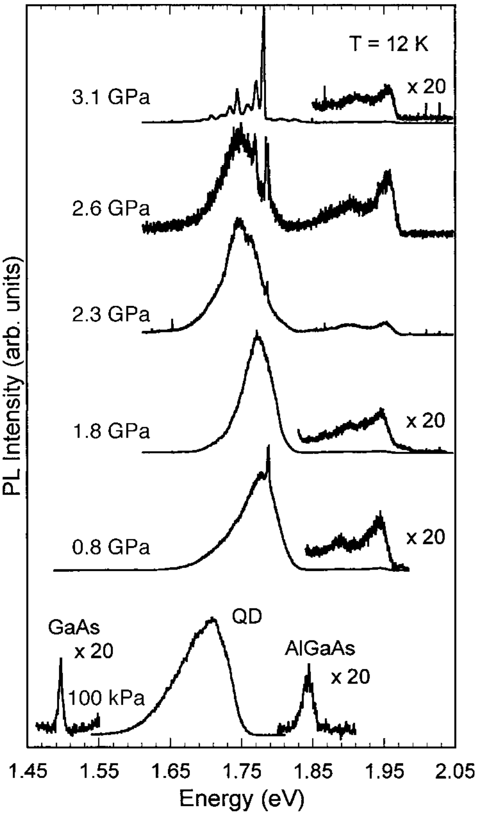

Fig. 2. Pressure dependence of the PL in $In_{0.5}Al_{0.5}As/Al_{0.25}Ga_{0.75}As$ QD sample. QD PL quenches beyond 2.6 GPa and the sharp peaks in the 3.1 GPa spectrum are due to deep levels in the barrier

sence of GaAs PL, although weak (not shown at all pressures in Fig. 2), serves as an internal reference for ascertaining the pressure shift of other observed peaks. With increasing pressure up to 0.8 GPa, all three PL peaks shift to higher energy. Between 0.8 and 1.8 GPa, there is a gradual narrowing in the linewidth of the QD PL and almost no shift in its energy position. For further increase in pressure, the QD PL is red-shifted as shown in the spectrum at 2.3 GPa. Above 2 GPa, there is a significant decrease in the QD PL peak intensity and it is quenched beyond detection above 2.6 GPa. Between 2.8 and 8 GPa, a series of sharp peaks, as shown in the 3.1 GPa spectrum, is observed. These peaks are attributed to deep levels in AlGaAs and GaAs, in comparison to similar peaks seen in the reference sample.

The variation in the peak intensity and full width at half maximum (FWHM) of the QD PL with pressure are depicted in Fig. 3. The QD PL intensity remains the same within experimental uncertainties up to 1.8 GPa and drops by more than two orders of magnitude between 1.8 and 2.6 GPa. There is a gradual decrease in the FWHM (≈50%) up to 1.8 GPa, and an increase (≈30%) thereafter.[3]) If the emission from larger size dots has a smaller pressure shift than those of smaller dots, inhomogeneous broadening will decrease with pressure resulting in reduced linewidths at elevated pressure. The decreasing intensity and increasing FWHM observed beyond 1.8 GPa, suggest a crossover between energy levels around this pressure.

A plot of the energy positions of all the PL peaks as a function of pressure is given in Fig. 4a. The GaAs PL shows a shift of 106 meV/GPa as expected. The QD PL shows an initial shift of 90 meV/GPa up to 0.8 GPa, no significant shift (or a small negative shift) between 0.8 and 2.2 GPa, and a decrease in the PL peak energy reminiscent of anti-crossing of energy levels until the PL completely quenches at 2.6 GPa. This pressure behavior is quite interesting and different from that reported for InAs/GaAs QD [5], or InGaAs/GaAs [6, 7] and InGaAs/AlGaAs [8, 9] strained layer quantum wells.

[3]) The GaAs PL overlaps with the QD PL for $P > 2$ GPa, but its linewidth is quite narrow and hence the error in the FWHM of the QD PL is <5% above 2 GPa.

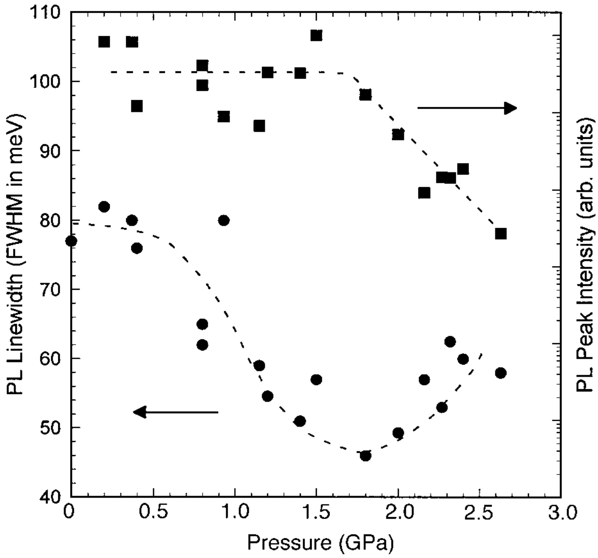

Fig. 3. Variation in the linewidth and peak intensity of the QD PL as a function of pressure. Lines through the data points are guides to the eye

A possible explanation of the observed pressure behavior is given below. Of the many factors that could influence the pressure shift of the QD PL, the dominant contributions come from energy level crossings under pressure and the built-in strain in the heterostructure. From the known composition dependence of the Γ and X CB energies in ternary alloys [10], it can be seen that the Γ CB has type-I and X CB has type-II

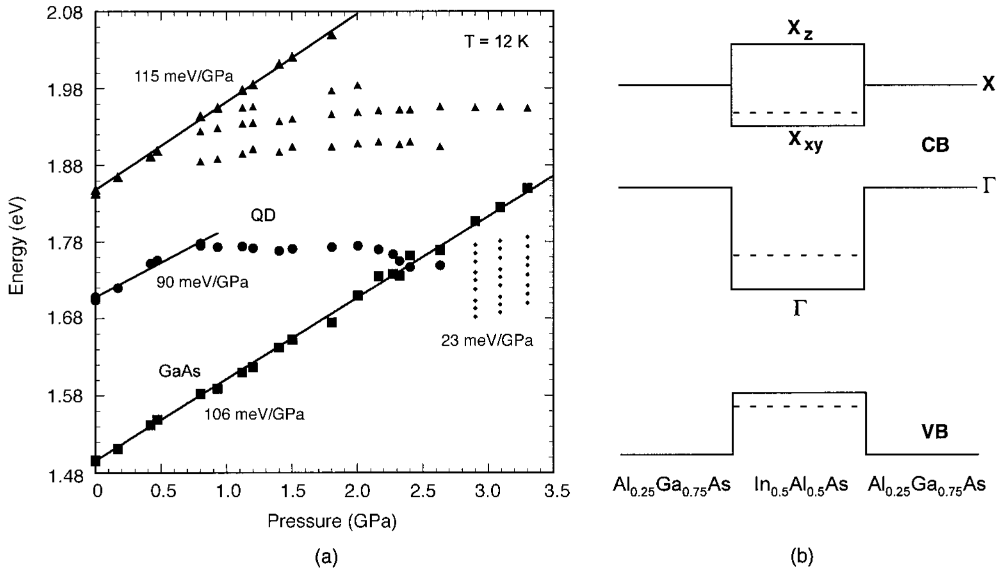

Fig. 4. a) Energy of the PL peaks in the $In_{0.5}Al_{0.5}As/Al_{0.25}Ga_{0.75}As$ QD structure plotted as a function of pressure. b) Sketch of the energy band alignments in InAlAs/AlGaAs heterostructure. Γ and X_{xy} CB have type-I structures whereas X_z CB has type-II (staggered) alignment. Dotted lines represent confined energy levels in various bands

band alignments in InAlAs/AlGaAs heterostructure for all values of the valence band offset. The biaxial component of the built-in strain in the InAlAs layers lifts the degeneracy between the X_z and X_{xy} bands, with the X_{xy} valley being lower in energy than the X_z minimum. Thus, it is possible to have a type-I alignment for X_{xy} and type-II alignment for X_z, as shown in Fig. 4b. We believe therefore, that the change in the pressure shift of the QD PL at 0.8 GPa (see Fig. 4a) is due to the crossover between the quantized levels in the Γ and X_{xy} bands of $In_{0.5}Al_{0.5}As$. After this crossover, absorption of the incident laser generates photoexcited carriers in the Γ level, which are efficiently scattered to the X_{xy} level before recombining with the $n = 1$ heavy holes. QD emission is still type-I and strong but the pressure shift is small because of its origin in the indirect X_{xy} level. As pressure increases further, there is yet another crossover between the Γ confined level and X_z or the barrier X band around 2.6 GPa. The mixing of these bands causes an anticrossing behavior in the vicinity of the crossover pressure similar to that reported in the InAs/GaAs QD grown on misoriented substrate [7]. After the crossover, the type-II QD PL is undetectable because the transition is indirect in both real and *k*-space.

It should be noted that in bulk $In_{0.5}Al_{0.5}As$, the Γ–X crossover occurs around 4 GPa. Because of the built-in strain (3.8%) and quantum confinement of energy levels, the crossover pressure in the QD structure would be lower than that in bulk. The observed pressure behaviour of the PL in InAlAs QD is explained on the basis of successive crossing of the Γ confined level with X_{xy} and X_z or barrier X bands.

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