Photoluminescence spectroscopy of self-assembled InAs quantum dots in strong magnetic field and under high pressure

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We have investigated the photoluminescence spectrum of self-assembled InAs quantum dots embedded in a GaAs matrix in magnetic field B up to 23 T and under hydrostatic pressure up to 8 kbar. A strong anisotropy in the diamagnetic shift is found depending on whether B is applied parallel or perpendicular to the growth direction. In the former case, the spatial extent of the carrier wave function in the dot is estimated to be 60 Å. The pressure coefficient for the dot emission line is (9.1 ± 0.2) meV/kbar, about 20% smaller than for the Γ-point band gap in bulk GaAs. © *1997 American Institute of Physics.* [S0003-6951(97)01804-4]

Self-assembled InAs quantum dots (QDs), grown in the self-organized Stranski–Krastanov mode on the lattice-mismatched GaAs surface, are presently the subject of intense interest.[1–5] The electron and hole energies in the dots are strongly influenced by both the large strain in the region of the dot and by size quantization. The dot ensemble has a fairly wide distribution of eigenenergies due to variations in size, shape, and strain. Typically, an optical emission line from the dot ensemble has energy $\hbar\omega\approx1.1$–1.3 eV and a full width at half-maximum (FWHM) linewidth of $\Delta\approx40$–60 meV.[2,5–7]

In this letter we report low-temperature photoluminescence (PL) studies of self-assembled InAs QDs in magnetic field B and under high quasihydrostatic pressure P. Due to the small size ($\approx$10 nm) and large quantization energies (of order 100 meV) of the QD, a magnetic field of $\approx$10–20 T can be treated as a perturbation, and the diamagnetic shift of the PL line provides an estimate of the spatial extent of carrier wave functions in the dot.[8] The main effect of the applied hydrostatic pressure on the energy levels of the QDs is expected to arise from the change in the Γ-point band gaps in the dot and in the GaAs matrix. In the QD the pressure can also affect the quantization energies of electrons and holes, due to nonparabolicity of the bands. This contributes to a shift of the PL line with pressure which can differ from that in bulk material.

Our sample was prepared by molecular-beam epitaxy on a $\langle 100\rangle$ GaAs substrate. A GaAs buffer layer was grown at 600 °C, and 1.8 monolayers of InAs were deposited at 450 °C after a growth interrupt. The dots were then capped by a 25-nm-thick GaAs layer at 450 °C. Magneto-optical measurements were performed using a resistive magnet, with the sample in a continuous-flow cryostat at temperature $T=10$ K. High-pressure experiments were performed at $T=4.2$ K using a low-temperature clamp cell with a sapphire window. An oil–gasoline mixture was used as a pressure transmission medium, and the pressure was measured by a calibrated InSb manometer. An optical fiber was used both to transfer the Ar^+-laser excitation to the sample and to collect the PL signal, which was dispersed by a monochromator (1200 lines/mm grating and 10 Å/mm dispersion), and detected by a cooled Ge diode.

The PL spectra of the sample show an emission line, typical of self-assembled InAs QDs,[2,5–7] as well as bulk GaAs exciton- and acceptor-related lines. A representative set of QD spectra for B normal to the growth plane is shown in Fig. 1(a). A relatively small FWHM linewidth $\Delta=40$ meV indicates a reasonably high homogeneity of dot sizes. For B increasing to 23 T the line shifts to higher energies with little change in shape. The solid squares in Fig. 2 show the field dependence of the weighted line center (its first moment). The dependence is quadratic in B, typical of a diamagnetic shift. A least-squares fit gives $\Delta\hbar\omega=\alpha B^2$ with $\alpha=(1.1\pm0.1)\times 10^{-5}$ eV/T^2. The error in α is relatively small, because the weighted line center can be determined rather precisely.

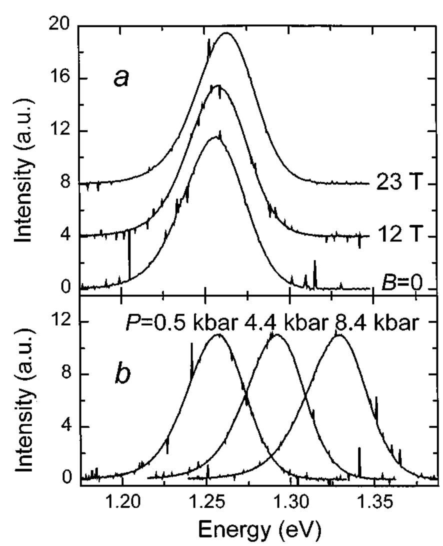

FIG. 1. Representative sets of PL spectra (a) in magnetic field normal to the plane of the sample, spectra are offset for clarity, and (b) under high pressure.

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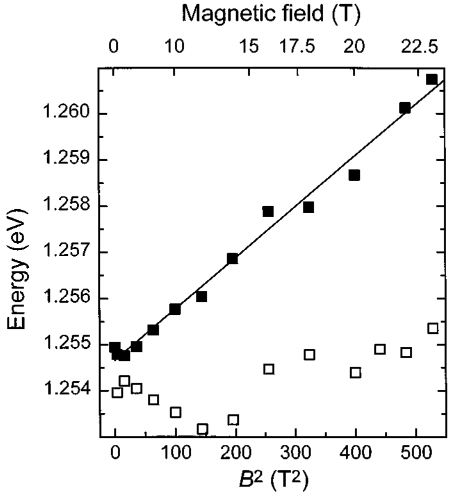

FIG. 2. Energy position of the weighted PL line center in magnetic field B normal (solid squares) and parallel (open squares) to the sample plane. Solid line: least-squares fit for the former case.

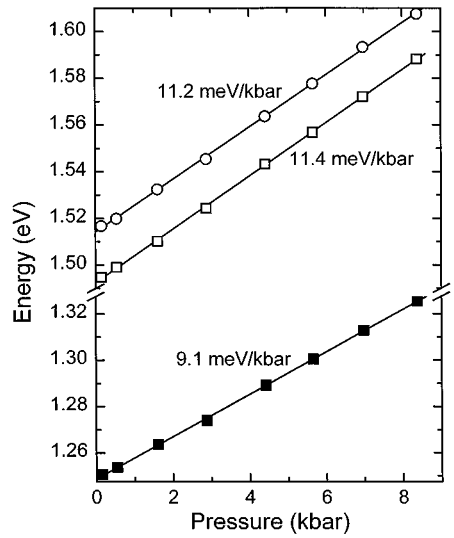

FIG. 3. PL line energies under high pressure: solid squares: QDs, weighted center; open squares and circles: maxima positions, acceptor-related and free exciton lines, respectively, in bulk GaAs; solid lines: least-squares fits.

The diamagnetic shift of the electron and hole levels in the QD is given by

$$\Delta E_{e(h)} = \gamma e^2 B^2 \langle x^2_{e(h)} \rangle / 2m^*_{e(h)},$$

where m^* is the effective mass, $2\sqrt{\langle x^2 \rangle}$ is a measure of the spatial extent of the carrier wave function, and γ is a geometrical factor. The shift of the PL line is $\Delta \hbar\omega = \Delta E_e + \Delta E_h$. To estimate the size of the QDs we use $\gamma = 0.5$, which is exact for QDs with circular symmetry in the plane, and we assume the same spatial extent for electrons and holes. As the dot height is typically smaller than the in-plane size, we expect heavy-hole character in the growth direction for the ground hole state, with light-hole character for in-plane motion,[7,9] similar to the case of the heavy-hole subband in quantum wells.[10] Therefore, for B normal to the sample plane we should use the light-hole effective mass. The crucial question is: What values of m^*_e and m^*_{lh} should be taken? Grundmann and co-workers[9] used the electron and light-hole masses of bulk InAs in their calculations, although the InAs in a QD is strongly strained, and corrections due to nonparabolicity should be important. We believe that an estimate within a simple $\mathbf{k}\cdot\mathbf{p}$ model provides more realistic values,[10] taking the PL line position of 1.26 eV as the ''effective'' band-gap energy. This gives $m^*_e \approx 0.055 m_e$ and $m^*_{\mathrm{lh}} \approx 0.1 m_e$ for the InAs dots. We then obtain $\Delta x = 2\sqrt{\langle x^2 \rangle} = 60$ Å for the in-plane spatial extent of the wave function in the QD. Despite the uncertainty in the effective masses, the accuracy of the estimate is probably reasonable since $\Delta x \sim (m^*)^{-1/2}$. It is also consistent with typically reported values of the geometrical dot size of around 100 Å,[3,5] and the estimate of Δx obtained in magnetotunneling experiments.[8]

The diamagnetic shift of the line is strongly anisotropic. The open squares in Fig. 2 show the weighted PL line center for B applied normal to the growth direction (i.e., parallel to the sample surface). The shift of the line is much smaller. Moreover, between $B = 0$ and 12 T the line shows evidence of a small shift to lower energies. There are at least two reasons for expecting a smaller diamagnetic shift in this orientation: the strong confinement Δz of the carriers in the growth direction and the smaller value of ΔE_h due to contribution of the heavy-hole effective mass for motion in this direction. We cannot account for the decrease in line energy at low B. The shift is less than 2 meV, small compared with the linewidth of 40 meV. Nevertheless, it is reproducible and it is not accompanied by any noticeable change in the line shape.

Figure 1(b) shows a representative set of QD emission lines under high pressure up to 8.4 kbar. The line shifts to higher energies with pressure, as expected for Γ-valley-related states, with no change in shape. Figure 3 shows the pressure dependence of the energy of the weighted line center. Also shown for comparison are the positions of the maxima of the band-edge exciton and residual acceptor emission lines from the bulk-GaAs buffer layers in our sample. All lines shift linearly with pressure, but the shift for the QD line is smaller.

A least-squares fit gives the pressure coefficient $\kappa = d(\hbar\omega)/dP$ for the GaAs exciton line as $\kappa_{\mathrm{EX}} = (11.2 \pm 0.2)$ meV/kbar. It is in good agreement with the band-edge shift in GaAs, $dE_g/dP = (11.6 \pm 0.2)$ meV/kbar.[11] This confirms the accuracy of the pressure calibration. For the acceptor-related line we obtain a very similar value, $\kappa_A = (11.4 \pm 0.2)$ meV/kbar, but for the QD line the coefficient is $\kappa_{\mathrm{QD}} = (9.1 \pm 0.2)$ meV/kbar, about 20% smaller. This is a significant difference. For example, in high-pressure investigations of InAs quantum dots grown on a terraced surface on a slightly misoriented GaAs substrate,[12] a difference of only 7% between the QD and bulk lines was reported [but see contribution (2) below].

Although a comprehensive quantitative analysis is difficult, we now indicate some possible reasons for the difference in the pressure coefficients. The following contributions to the PL line shift with increasing pressure are typically expected:[11]

(1) *The change in the energy gap*. The pressure coefficient for the bulk InAs band edge is known with much less accuracy than for GaAs.[13] In addition, one should take into account the huge internal strain of the InAs dot.[9] Typical

reported data indicate a value of dE_g/dP for InAs close to that for GaAs,[12,14] but a lower value is possible.[13]

(2) *The lowering of the quantization energy of electrons*. This is due to the effective mass increase in the QD with pressure, which results from conduction band nonparabolicity. For a deep confining potential, as in our QDs, this effect is about 0.6% per kbar,[15] which would require a size quantization energy of about 340 meV to explain the observed difference if other factors were neglected. This effect should be smaller for shallower levels, as in Ref. 12, when the QD levels are very close to the GaAs band edges.

(3) *The change of the quantization energy for holes*. The main cause of ΔE_h is the change in the light-hole effective mass with P. Even the sign of the change is uncertain. For example, the simple $\mathbf{k}\cdot\mathbf{p}$ model predicts an increase of $m^*_{\rm lh}$ with P, while in $In_{0.2}Ga_{0.8}As$/GaAs quantum wells a decrease of $m^*_{\rm lh}$ with P has been reported recently.[16]

Only minor contributions to the line energy are expected from the change in Coulomb interaction energy in the QD and from the small decrease of the dot size under compression.

Finally, we note that for the strongly confined states of the QD it may be necessary to consider also the contribution of the X and L valleys to the wave functions of the confined electron states. Any such contributions would decrease significantly the pressure coefficient. A more detailed theoretical analysis of the electronic states of QDs is clearly required to explain our pressure data.

To conclude, we have measured the diamagnetic shift and the pressure coefficient for the PL recombination line of self-assembled InAs QDs in a GaAs matrix. Large anisotropy of the diamagnetic shift has been observed. The in-plane spatial extent of the carrier wave function in the dot is estimated as 60 Å. The pressure coefficient is found to be (9.1 ± 0.2) meV/kbar, 20% less than for bulk GaAs.

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