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Magneto-photoluminescence and electroluminescence spectroscopy of self-assembled (InGa)As quantum dots on high index planes

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Abstract

We have investigated a wide range of structures containing InAs and $In_{0.5}Ga_{0.5}As$ self-organised quantum dots grown on (1 0 0), (3 1 1)A and (3 1 1)B GaAs substrates. The photoluminescence line width for quantum dots grown on the high-index planes is significantly narrower than those typically observed for (1 0 0) quantum dots. Bright room temperature electroluminescence is observed from a p–i–n device with the intrinsic region containing $In_{0.5}Ga_{0.5}As$ quantum dots grown at the (3 1 1)B orientation. Photoluminescence spectra were measured in magnetic fields up to 43 T for both quantum dot and wetting layer samples. From the diamagnetic shift of the photoluminescence lines, we have estimated the spatial extent of the carrier wave functions and compare these with the geometrical dot size obtained from microscopic investigations. This comparison suggests a qualitative model to explain the narrow photoluminescence of the quantum dots grown on (3 1 1)A and B planes. © 1998 Elsevier Science B.V. All rights reserved.

Keywords: Quantum dot; Magneto-photoluminescence

Semiconductor quantum dots (QDs) of a lower band gap material buried in a higher band gap matrix have attracted intense interest both for potential device applications and for studies of mesoscopic physics. In the case of lattice-mismatched semiconductor heterostructures, the self-assembly of nanometer-sized QDs is driven by the elastic forces exerted at the interface between the two lattices [1,2]. Most studies have been devoted to $In_xGa_{1-x}As/GaAs$ strained heterostructures. Both microscopic [3] and optical measurements [4,5] have shown the possibility of obtaining defect-free QDs with good size uniformity. Although the photoluminescence (PL) spectrum of an individual dot is a discrete sharp line [4,5], the PL of an *ensemble* of dots is characterised by a full-width at half-maximum (Δ) typically around 40 meV, due to the size distribution of dots.

This paper reports the optical properties in high magnetic fields of InAs and $In_{0.5}Ga_{0.5}As$ QDs and

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Table 1
Details of quantum dot and wetting layer samples. The PL line width was measured at 5 K in zero magnetic field

Sample type	QD	QD	QD	WL	WL
GaAs substrate	(1 0 0)	(3 1 1)A	(3 1 1)B	(3 1 1)A	(3 1 1)B
L	0.59 nm	1.13 nm	1.13 nm	0.68 nm	0.68 nm
Composition	InAs	$In_{0.5}Ga_{0.5}As$	$In_{0.5}Ga_{0.5}As$	$In_{0.5}Ga_{0.5}As$	$In_{0.5}Ga_{0.5}As$
PL line width Δ	40 meV	13 meV	11.4 meV	8 meV	8.2 meV

wetting layers (WLs) grown by molecular beam epitaxy on GaAs substrates having (1 0 0), (3 1 1)A and (3 1 1)B crystal orientations. The growth details are as follows: after heating to remove surface oxide, a 0.7 μm-thick buffer layer is grown, the first 0.2 μm at 580°C and the remaining 0.5 μm at 600°C. The substrate temperature is reduced to 450°C before the InAs or $In_{0.5}Ga_{0.5}As$ strained layer is deposited. The structure is completed with a 25 nm GaAs cap, also grown at 450°C. The strained layer compositions (i.e. indium fraction x) and thicknesses L are given in Table 1, together with the substrate orientations and PL line widths Δ. For each type of substrate, QDs grown at 450°C have the narrowest PL line width. During growth of the strained layer, the reflection high-energy electron-diffraction (RHEED) pattern is monitored and, if QDs are required, deposition is stopped when the RHEED pattern shows a transition from two-dimensional (2D) to three-dimensional (3D) growth.

PL measurements were performed at 4.2 K in static magnetic fields up to 11 T and pulsed fields up to 43 T. In static fields the optical excitation was provided by a He–Ne laser ($\lambda_{exc} = 633$ nm) and the PL was dispersed by a $\frac{3}{4}$ m monochromator and detected by a cooled Ge diode detector. For the pulsed fields, the laser beam ($\lambda_{exc} = 488$ nm) was chopped and the sample illuminated for 1 ms at the top of the field pulse, with the PL emission detected by a two-dimensional CCD array. The magnetic field was applied in two different orientations with respect to the growth axis (defined as z) of the samples: parallel and perpendicular to z.

Recently it has been shown that the photoluminescence (PL) properties of $In_{0.5}Ga_{0.5}As$ QDs are improved by growing on (n 1 1) substrates [6,7]. Specifically in Ref. [6], a remarkable narrowing of the PL spectrum line width is observed for InAs and $In_{0.5}Ga_{0.5}As$ QDs grown on (3 1 1)B with respect to the same structures grown on (1 0 0). This observation has been confirmed by our measurements (as shown in Table 1) with a value of the QD PL line width Δ narrower than the best reported in similar systems [8]. The good quality of the $In_{0.5}Ga_{0.5}As$ QD samples grown on (3 1 1) and their potential use for applications is demonstrated by the room temperature electroluminescence spectrum obtained from a p–i–n diode in which the $In_{0.5}Ga_{0.5}As$ QD layer is embedded in the intrinsic region (see Fig. 1). In Fig. 1 the room temperature PL spectrum measured on the same sample is also shown.

Consider now the low-temperature PL measurements. The WL PL is narrow as expected for a quantum well, while the 40 meV line width of the InAs QDs is typical for this system due to the size distribution of the dots [4]. The microstructure of the dot samples has been investigated by both transmission electron microscopy (TEM) and atomic force microscopy (AFM). TEM has been performed on the same capped (3 1 1)A sample studied by PL, but from a different part of the wafer. The measured dot size is between 5 and 8 nm with a dot density of 4×10^{11} cm^{-2}. For AFM imaging, samples were grown as described above, but the final cap layer was omitted, leaving the dots exposed. For $In_{0.5}Ga_{0.5}As$ QDs grown on (3 1 1)B a dot size ranging between 12 and 20 nm with a dot density of about 2×10^{11} cm^{-2} was obtained by AFM (in agreement with the values reported in Ref. [8]). The discrepancy in QD dimensions can probably be attributed to the presence of the GaAs cap layer which could quench the dot evolution, rather than to a size difference between dots grown on differently oriented substrates. This can be inferred

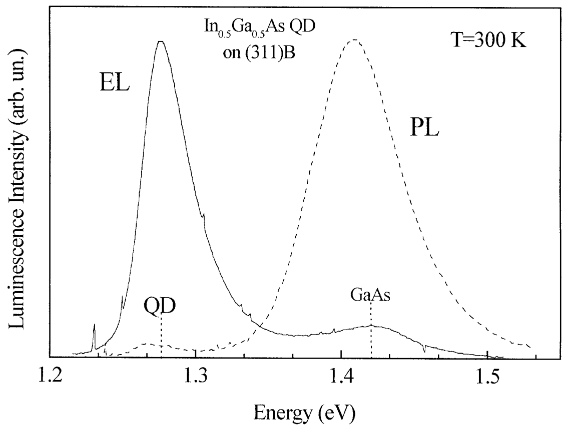

Fig. 1. Room temperature electroluminescence (EL) (continuous line) and photoluminescence (PL) (dashed line) spectra of a p–i–n diode having $In_{0.5}Ga_{0.5}As$ QDs embedded in the intrinsic region. The substrate orientation is (3 1 1)B. The EL spectrum was recorded at a device current of 14 mA. For the PL spectrum, the excitation wavelength was 515 nm. The lower-energy peak arises from the QDs, while the higher-energy peak is due to carrier recombination in the GaAs barrier.

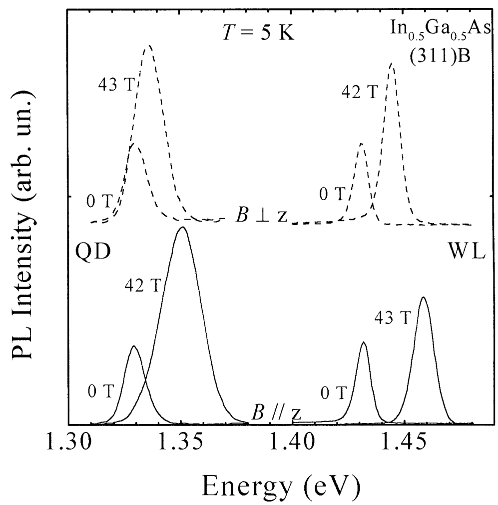

Fig. 2. Low-temperature PL spectra at zero field and ~ 43 T for $In_{0.5}Ga_{0.5}As$ QDs (left) and WL (right) samples grown on (3 1 1)B GaAs. For each sample, spectra are shown for magnetic field B parallel (continuous line) and perpendicular (dashed line) to the growth axis z. Note the higher emission energy of the WL sample.

by nearly the same PL peak energy and line width of QD samples grown on (3 1 1)A and B substrates. Because the TEM sample came from the same wafer studied by PL, we take the TEM dimensions to be the most reliable.

Fig. 2 shows the PL spectra of the $In_{0.5}Ga_{0.5}As$ QDs and WL grown on (3 1 1)B recorded for different orientations and magnitudes of the magnetic field. For ease of comparison, the zero-field PL spectra of the QD and WL samples have been normalised for both field orientations. At zero field the integrated PL intensity was found to be nearly the same for QD and WL structures. By measuring the diamagnetic shift of the PL line in a magnetic field (B) it is possible to obtain a measure of the wave function extent of the excitons confined in the QDs. Moreover, by changing the orientation of the field, useful insights into the QD potential geometry can also be derived [9,10]. Qualitatively, for B both parallel and perpendicular to the growth axis, a blue shift of the PL peak and an increase of the integrated intensity can be observed. Similar results have been found for QD and WL samples grown on (3 1 1)A.

The increase in PL intensity is due to exciton squeezing in the plane perpendicular to the field direction, which increases the transition oscillator strength and so its relative weight with respect to non-radiative channels [11,12]. The magnetic field acts as an additional confining potential which enhances the electron–hole pair recombination probability. In Fig. 3 the integrated PL intensity is plotted as a function of the magnetic field for two different field orientations. For $B \| z$ the enhancement of the PL is stronger than for B in the orthogonal direction. This is in agreement with a stronger confinement of the exciton along the growth axis which prevents the electron and hole from moving freely along z when the field is applied in the (x, y) plane. Microscopic measurements have shown [8] that the QD shape has an axial symmetry (e.g. pyramidal) and this is consistent with the difference observed in the increase of the luminescence intensity for the two orientations of the field.

Fig. 4 shows the PL peak shift ΔE as a function of the magnetic field for InAs QDs grown on (1 0 0) and $In_{0.5}Ga_{0.5}As$ QDs and WL grown on (3 1 1)B. The magnetic field ranged from 0 to ~ 43 T and was applied parallel and perpendicular to the growth axis. Results for the $In_{0.5}Ga_{0.5}As$ QDs and

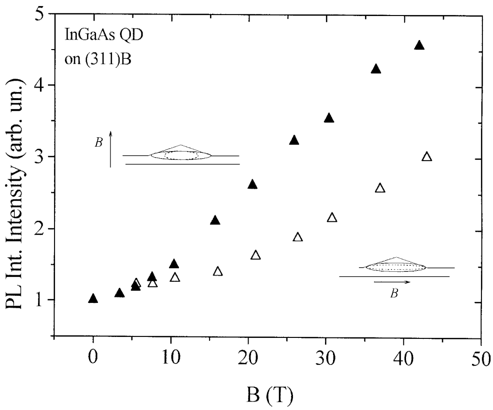

Fig. 3. Integrated PL intensity as a function of magnetic field for $In_{0.5}Ga_{0.5}As$ QDs grown on (3 1 1)B GaAs for two magnetic field orientations (B parallel to the growth axis: full triangles; B perpendicular to the growth axis: open triangles). The PL spectra were recorded at 5 K. The insets show schematically the magnetic field direction and the effect on the exciton wave function.

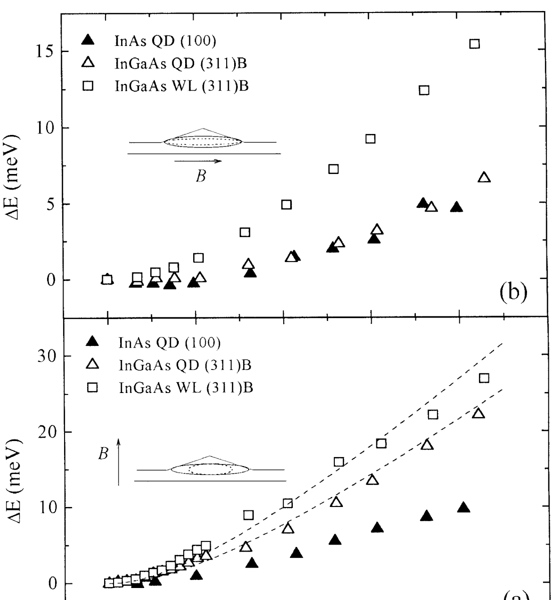

Fig. 4. Shift in energy ΔE of the PL line with magnetic field applied (a) parallel and perpendicular (b) to the growth axis. The different samples are indicated in the figure. In (a), the dashed lines are a fit with only one parameter (the exciton effective mass, μ) following Ref. [13]. The values obtained are: for $In_{0.5}Ga_{0.5}As$ L, $\mu = 0.045m_e$ and for $In_{0.5}Ga_{0.5}As$ QDs, $\mu = 0.051m_e$ where m_e is the bare electron mass.

WL on (3 1 1)A substrates are very similar to the (3 1 1)B layers and so are not included.

It is possible to distinguish two different limits of weak ($\gamma \ll 1$) and strong magnetic field ($\gamma \gg 1$) where $\gamma = a_0/l_B$ is the ratio of the Bohr radius a_0 ($=\hbar^2\varepsilon/e^2\mu$) of an exciton to the magnetic length l_B ($=\sqrt{\hbar/(eB)}$). (μ is the in-plane exciton effective mass). In the limit of weak ($\gamma \ll 1$) magnetic field we expect a diamagnetic shift of the PL energy given by

$$\Delta E = \frac{e^2\langle x^2\rangle}{4\mu}B^2, \tag{1}$$

where $\langle x^2\rangle$ ($=\langle y^2\rangle$) is the mean-square expectation value for the exciton wave-function in the plane perpendicular to the applied magnetic field. We take $\Delta x = 2\sqrt{\langle x^2\rangle}$ to be a measure of the extent of the wave function. At very high magnetic fields ($\gamma \gg 1$), confining QD or Coulomb potentials become less important and ΔE increases more linearly [11,12].

Considering the results for $B\|z$ in Fig. 4a, we find that the diamagnetic shift for the (1 0 0) InAs QDs is very similar to that previously measured in static fields up to 23 T [10]. For the present measurements we find a slightly smaller diamagnetic shift, and using the same effective masses as in [10], Δx is found to be 5.5 nm. The magnetic length at 40 T is 4 nm, so we do not expect the high field limit to be achieved for the QD in this sample. The InAs QDs show a good match between the wave function extent and the geometrical dot size of about 10 nm as reported previously [10].

For the (3 1 1)B QD and WL samples a quantitatively different dependence on B is found. MacDonald and Ritchie [13] have derived a numerical method for calculating the energy of a 2D Coulombic system such as an exciton in arbitrary magnetic fields. Using a single fitting parameter, the reduced effective mass of the exciton μ, this provides a good fit to the diamagnetic shift of the (3 1 1)B WL PL shown in Fig. 4a (dashed line), as might be expected since the WL acts as a narrow quantum well with approximately 2D confinement for excitons. Perhaps surprisingly, a good fit is also obtained for the PL of the (3 1 1)B QD. The reduced effective masses used in these fits are given in the caption of Fig. 4a and are reasonable values for μ in this type of $In_{0.5}Ga_{0.5}As$ heterostructure. If these values of μ are used in Eq. (1), together with the data in the quadratic low field limit, we find that Δx is equal to 13 and 14 nm for the (3 1 1)B QD and WL, respectively. An alternative approach is to use the same values of μ in the formula given in Ref. [13] for a 2D hydrogen atom:

$$\langle x^2 \rangle = \frac{\langle r^2 \rangle}{2} = \frac{3}{16} a_0^2.$$

This approach gives the same values for Δx for both the (3 1 1)B QD and WL. Note that the value of Δx for the (3 1 1)B QD is considerably larger than the QD size estimated from TEM measurements (5–8 nm) in the same sample. This suggests that the electrons and holes in $In_{0.5}Ga_{0.5}As$ QDs are less strongly confined by the dot potential, with their wave functions possibly spreading out into the wetting layer. It may be reasonable to consider the excitons in $In_{0.5}Ga_{0.5}As$ QDs as wetting layer excitons localised by the potential fluctuations of the QDs. Such a picture would account for the narrow PL of the (3 1 1) QDs compared to those grown on (1 0 0): although the (3 1 1) QDs have a similar geometrical size distribution, the excitonic recombination of (3 1 1) QDs is less strongly determined by the precise QD dimensions.

In Fig. 4b where the magnetic field is perpendicular to the growth direction, the diamagnetic shifts are much smaller, reflecting the stronger confinement along this direction. For magnetic fields up to 10 T, the PL peak moves to slightly lower energy. This is not expected for the diamagnetic shift and could be due to an increase in the exciton binding energy. Both (1 0 0) and (3 1 1)B QD samples show very similar diamagnetic shifts in this field orientation, suggesting a similar degree of confinement by the QD potential in the growth direction. This contrasts with the much larger shift shown by the (3 1 1)B WL, and confirms the localised nature of the excitons giving rise to the PL spectra in the (3 1 1) $In_{0.5}Ga_{0.5}As$ QDs.

In conclusion, we have performed magneto-photoluminescence spectroscopy on InAs QDs on (1 0 0) and $In_{0.5}Ga_{0.5}As$ QDs and WLs on (3 1 1)A and (3 1 1)B substrates. The PL line width for QDs on the high-index planes is significantly narrower than that of the (1 0 0) InAs QDs. Analysis of the PL intensity and diamagnetic shift as a function of the magnetic field yields information about the spatial extent of the carrier wave function in the QDs. We propose a model to explain qualitatively the narrow PL of the QDs grown on (3 1 1)A and B planes.

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