Electron and Hole g Factors and Exchange Interaction from Studies of the Exciton Fine Structure in $In_{0.60}Ga_{0.40}As$ Quantum Dots

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Self-assembled $In_{0.60}Ga_{0.40}As$ quantum dots (QD's) have been studied by single dot magnetophotoluminescence spectroscopy ($B \leq 8$ T). At $B = 0$ a splitting of the exciton (X) emission is observed which we ascribe to an asymmetry of the confinement potential. With increasing B the emission splits into a quadruplet corresponding to the $m = \pm 2$ and ± 1 X states, which originates from a reduction of the cubic symmetry of the QD's. From the spectroscopic data we obtain values for the electron (e) and hole (h) g factors. We also determine the X singlet-triplet splitting which is found to be enhanced over bulk values by about an order of magnitude due to the increase of the e-h overlap in the QD's. [S0031-9007(99)08567-1]

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During the past several years spectroscopic studies of electronic confinement effects in quantum dots (QD's) have evolved into a very active research area ranging from basic physics regarding, e.g., properties of zero dimensional X's to technological applications like QD lasers. Semiconductor QD's fabricated by self-assembled growth have attracted particular interest due to their high optical quality [1–3]. However, a major obstacle in accessing fully detailed information on the electronic properties has been the inhomogeneous broadening of spectral lines due to size and composition variations over ensembles of dots. Recently several experimental techniques have been developed which permit the study of individual QD's for which inhomogeneous broadening effects are suppressed [1–9].

Because of the possibility of performing sensitive spectroscopy of single QD's, experiments giving clear insight into their electronic states may be realized. The electronic fine structure of the QD levels, which arises from the coupling of the spin and the single particle electronic states, gives particularly detailed information. For example, in studies of single QD's formed at $GaAs/Al_xGa_{1-x}As$ interfaces a splitting of the X states has been reported [6]. Very recently studies of the Coulomb interactions between carriers forming multi-X complexes have been performed in self-assembled QD's [7,8].

From the Zeeman splitting of X's in QD's formed by laser induced diffusion [10] the X g-factor has been obtained. However, to date it has not been possible to determine the underlying e and h g-factors separately in QD systems. These g factors are very sensitive to band mixing and furnish detailed insight into the confined electronic structure. To our knowledge, at present there is no detailed understanding of g factors in quantum wires and dots, either experimentally or theoretically.

In the present work we have used small mesa structures to study single $In_{0.40}Ga_{0.60}As$ QD's formed by self-assembled growth. We observe at high B emission from heavy hole X angular momentum states $m = \pm 1$ and $m = \pm 2$ (bright and dark X's). The latter emission results from a reduced symmetry of the QD's which we attribute to a shape asymmetry. From the splitting of the X lines in magnetic field we determine the e and the h g-factors. In addition, we find that the X singlet-triplet splitting is 200 μeV, which is drastically increased over the bulk value.

The QD's were grown on GaAs [001] substrates at a temperature of 470 °C by depositing the equivalent of 4.8 monolayers of $In_{0.60}Ga_{0.40}As$. Stransky-Krastanow growth [11] starts after deposition of about three monolayers. The QD's were capped without growth interruption. An average dot density of 1×10^{10} cm^{-2} is determined from scanning electron microscopy of a sample without cap layer. The QD's have an average lateral size of about 20 nm. In low excitation photoluminescence (PL) the emission from QD arrays has a spectral width of about 25 meV. Lithography was used to fabricate small mesa structures with lateral sizes down to 100 nm, for which we obtain an average number of one dot per mesa. For optical studies only those mesas were selected whose low excitation spectra consist of a single emission line. From this observation we conclude that only one QD is contained in such a mesa which is to be contrasted with other structures, whose spectra consist of a few lines.

The samples were held at a temperature of 1.5 K in the helium insert of an optical split-coil magnetocryostat ($B \leq 8$ T). The magnetic field was aligned normal to the heterostructure (z direction). The samples were optically excited by a cw Ar^+ laser using very low excitation densities (~ 0.1 W/cm^{-2}). The emission was dispersed by a double monochromator and detected by a charge-coupled-device camera. Its polarization could be analyzed by a quarter-wave-retarder and a linear polarizer.

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PL spectra from different single QD's can be divided into two classes with different behavior in magnetic field: The solid lines in Fig. 1 show typical PL spectra from a single QD belonging to one class. The spectra of these QD's show a splitting into four components with increasing B. Because of the weak excitation, all lines are associated with the recombination of single e-h pairs in the dot. The two outer lines have strong intensities, whereas the two inner ones have comparatively weak intensities. The spectra shown by dashed lines for $B = 0$ and 8 T belong to a dot from the second class and exhibit the well-known behavior of X transitions in magnetic field, namely a splitting of the emission line into a doublet due to the Zeeman effect.

Figure 2 shows polarized PL spectra of a QD which gives the four transitions in the unpolarized spectra of Fig. 1. At zero magnetic field (lower panel) the luminescence is linearly polarized parallel either to the [110] or the $[1\bar{1}0]$ direction [6]. The emission lines are split from one another by about 150 μeV. With increasing B the polarization of the emission changes from a linear into a circular one, as seen from the spectra at $B = 8$ T (upper panel). In contrast, the emission spectra of dots with a doublet splitting at high B show no significant linear polarization at $B = 0$.

Figure 3 gives the observed transition energies for a single QD with a quadruplet splitting versus B. The magnetic field dependences of the transition energies result from the diamagnetic shift of the X plus the Zeeman energy due to the interaction of the X spin with B, which depends on the spin orientation. In strongly confined QD's the X diamagnetic shift is determined by the single particle wave functions and is independent of the X spin orientations. The different B dependences of the emission lines therefore result from their Zeeman splittings. Both the splitting between the spectral lines with strong emission intensities and that between the weak lines increase linearly with B at high fields (inset of Fig. 3). The splitting between the two strong spectral lines is 1.4 meV at $B = 8$ T, whereas for the weak lines the splitting is only 0.7 meV. At low fields, however, a deviation from the linear dependence is observed for the strong lines. There the behavior can be well described by a square dependence on B.

The heavy hole X is fourfold degenerate at $B = 0$ neglecting the e-h exchange interaction [12]. The X states are characterized by the total angular momentum m along the z direction, $m = S_{e,z} + J_{h,z} = \pm 1, \pm 2$ with the e spin $S_{e,z} = \pm 1/2$ and the h spin $J_{h,z} = \pm 3/2$. The spin Hamiltonian for an X in magnetic field is given by [13–16]

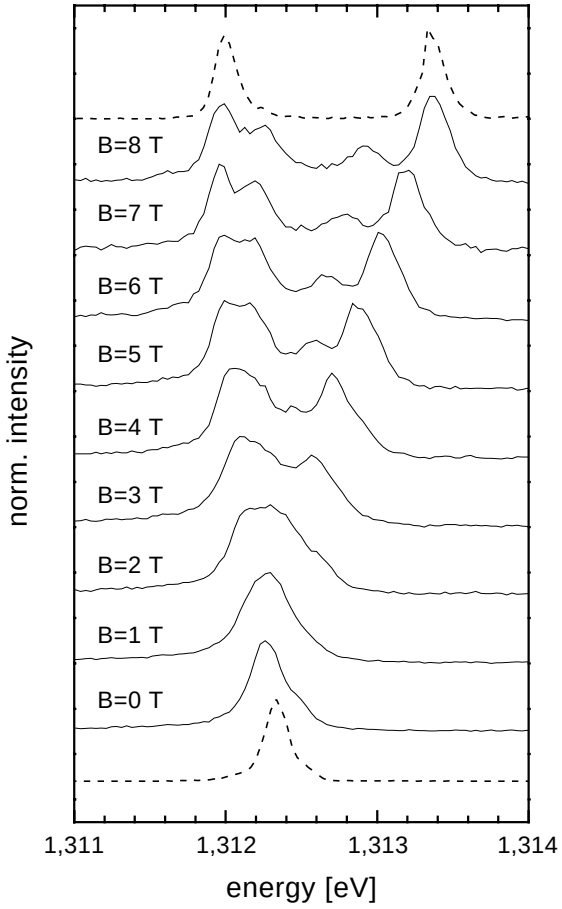

FIG. 1. Solid traces: PL spectra of an asymmetric QD for varying B. Dashed lines at $B = 0$ and 8 T give spectra of a symmetric QD. To facilitate comparison of magnetic field effects the emission of the symmetric QD which occurs at 1.3195 eV at $B = 0$ is shifted by 7.2 meV to lower energies.

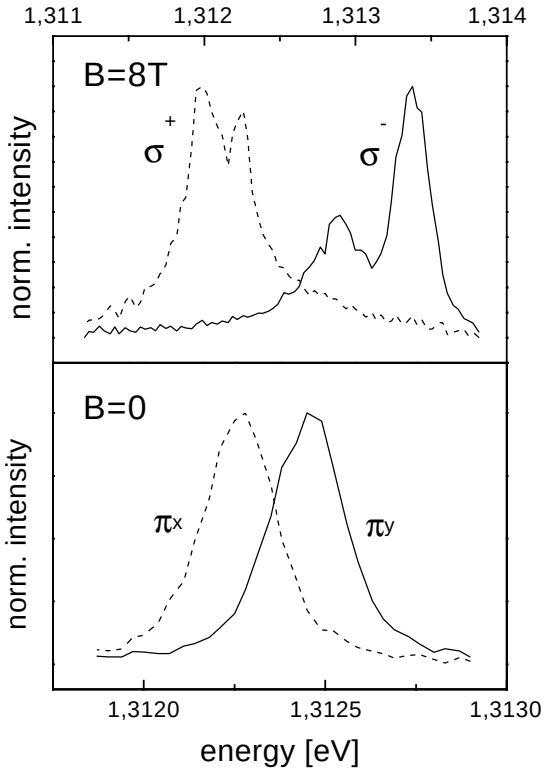

FIG. 2. Spin-polarized PL spectra of a QD with reduced symmetry.

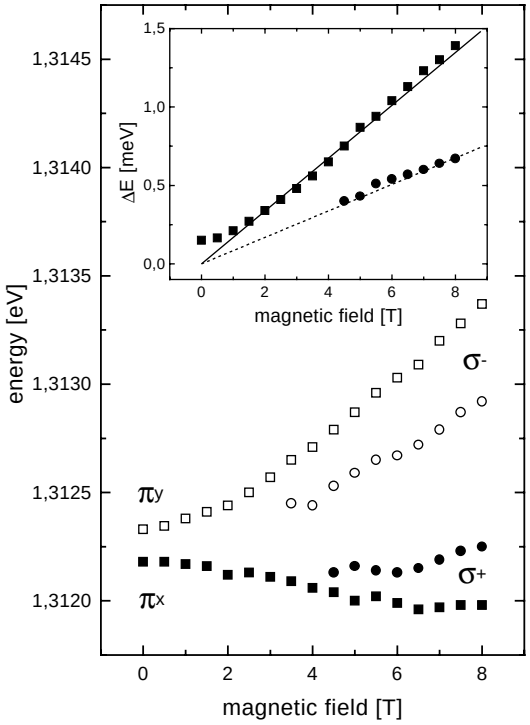

FIG. 3. Magnetic field dependence of the spin-polarized X transition energies of $In_{0.60}Ga_{0.40}As$ QD's. The inset shows the spin-splittings of the bright and dark X's versus B.

$$\mathcal{H} = a_z J_{h,z} \times S_{e,z} + \sum_{i=x,y,z} b_i J_{h,i}^3 \times S_{e,i} + \mu_B \left(g_e S_{e,z} - \frac{g_{h,z}}{3} J_{h,z} \right) B , \quad (1)$$

where the first two terms give the e-h exchange energy and the third one the interaction of the X spin with B. At $B = 0$, the radiative states $m = \pm 1$ and the nonradiative ones, $m = \pm 2$, are split by $E_{ex} = 1.5a_z + 3.375b_z$ due to exchange interaction (singlet-triplet splitting) [17].

An asymmetry of the confinement potential in the QD plane ($b_x \neq b_y$) results in an exchange energy splitting of the $|m| = 1$ X states (besides modifying the $|m| = 2$ splitting). The split states are linear combinations of the $m = \pm 1$ X's, $|L_{1/2}\rangle = 1/\sqrt{2}\,(|+1\rangle \pm |-1\rangle)$. The mixing results in a linear polarization of the QD emission, as observed for the QD's in Fig. 2 at $B = 0$. From the splitting of the two polarized spectral lines we obtain an asymmetry splitting $E_{as,\pm 1} = 0.75|b_x - b_y|$ of 150 μeV.

In a magnetic field the X eigenstates with $|m| = 1$ are given by

$$|L_{1/2}\rangle = \frac{(|+1\rangle + (r \pm \sqrt{1 + r^2})|-1\rangle)}{\sqrt{2(1 + r^2 \pm r\sqrt{1 + r^2})}} \quad (2)$$

where r gives the ratio of the spin-splitting $(g_e + g_h)\mu_B B$ to the asymmetry energy $E_{as,\pm 1}$. At high fields (at large r) $E_{as,\pm 1}$ can be neglected and the two states transform into circularly polarized states. The B dependence of the splitting between these states is given by $\Delta E_{\pm 1} = \sqrt{(g_e + g_h)^2 \mu_B^2 B^2 + E_{as,\pm 1}^2}$. Similarly, the $|m| = 2$ X's are split by $\Delta E_{\pm 2} = \sqrt{(g_e - g_h)^2 \mu_B^2 B^2 + E_{as,\pm 2}^2}$. At low B the splitting increases quadratically with magnetic field, and then transforms into a linear dependence. The changeover occurs at rather small magnetic fields of about 2 T, because the Zeeman energy then becomes considerably larger than the asymmetry splitting.

For the present QD's we observe a splitting of the X emission into a quadruplet at high B. In magnetoPL experiments on crystals with cubic symmetry only the heavy hole X's with angular momenta $m = \pm 1$ can be observed when $B \parallel [001]$, while the X's with $m = \pm 2$ are dark. Therefore we attribute the observation of a quadruplet splitting to a symmetry reduction of the QD's, that is the orientation of B effectively deviates from [001] [18]. In that case the dark X's mix with the optically active ones and gain oscillator strength. This mixing is, however, not very strong, as seen from the comparatively weak emission intensities of the inner spectral lines.

The different dependences of the Zeeman splittings of the $m = \pm 1$ and $m = \pm 2$ X's permit a separate determination of the e and h g factors. Neglecting the exchange energies at high B the splitting of these doublets is given by $\Delta E_{\pm 1} = |g_e + g_h|\mu_B B$ and $\Delta E_{\pm 2} = |g_e - g_h|\mu_B B$. From the splitting of the $m = \pm 2$ X's we obtain a g factor $g_{\pm 2} = g_h - g_e = -1.39 \pm 0.05$, whereas for the $m = \pm 1$ X's $g_{\pm 1} = g_h + g_e = -3.02 \pm 0.05$. Combining both relations we obtain $g_e = -0.81$ and $g_h = -2.21$ [19].

At high B the splittings between the two σ^- polarized emission lines and between the σ^+ polarized ones are not identical. The $m = \pm 2$ spectral features are shifted to lower energies towards the σ^+ polarized bright X. This asymmetry of the quadruplet arises from the singlet triplet splitting E_{ex}, for which we obtain 200 μeV from the spectra. This exchange energy is 1 order of magnitude larger than the singlet-triplet splitting reported for bulk GaAs [20,21]. This comparison shows the strong enhancement of the exchange interaction by the three-dimensional confinement in the QD's.

The short ranged part of the e-h exchange energy in an X is given by the probability of the e and h being at the same position [22]

$$\frac{E_{ex}}{E_{ex}^{3D} \times \pi a_{X,3D}^3} = \int d^3 r \, |\Psi(\vec{r}, \vec{r})|^2. \quad (3)$$

Here E_{ex}^{3D} is the exchange energy in bulk, and $a_{X,3D}$ is the bulk X Bohr radius. We have represented the present QD's as cylinders of $In_{0.60}Ga_{0.40}As$ having a height to width ratio of 1:3 embedded in GaAs [23]. The results do, however, not depend sensitively on the assumed shape of the dots. For calculating the right-hand side of Eq. (3) we have made multiparameter variational calculations of the X wave functions [24,25] in the QD's. As shown in Fig. 4 by the solid line, the calculation gives a strong increase of the exchange energy with decreasing dot size as a consequence of the increased e-h overlap for the smaller dots.

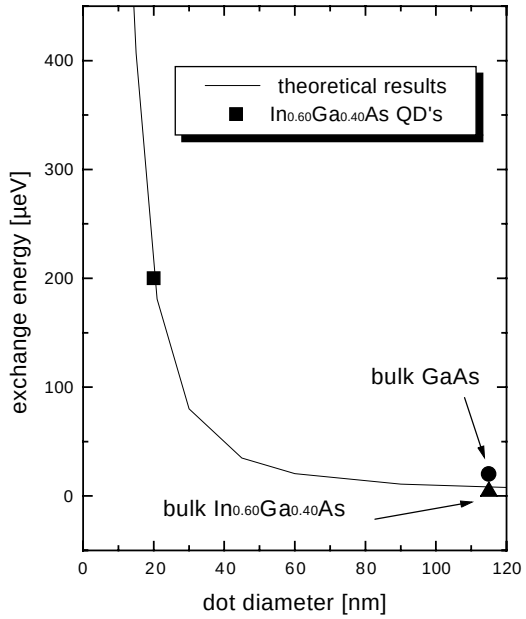

FIG. 4. *e-h* exchange interaction plotted versus QD size.

The experimental result for the present QD's is included in Fig. 4 by a square, and the result for bulk GaAs of 20 μeV from Ref. [21] is shown by a circle. We have also included an estimate for bulk $In_{0.60}Ga_{0.40}As$ obtained by scaling the bulk GaAs value using the corresponding material parameters. The theoretical curve is taken to go through this bulk value. The calculation describes the enhancement of the exchange energy in good accord with the experimental data.

Currently the shape of self-assembled QD's is controversial [26]. The spectral features observed here arise from asymmetries of the QD confinement potential. The formation of differently shaped dots most probably originates from inhomogeneous strain distributions during the growth of the QD's which may result in changes of the dot shape and also of the local material composition. Thus the asymmetry effects might depend on the growth parameters of the QD's but they should occur independently of the QD material. Because of the quantum confinement, these effects become more important for smaller size of the dot. Single dot spectroscopy can provide detailed insight into asymmetries of the QD confinement potential. However, further investigations are required to obtain a still more detailed picture of the geometry of the QD's and its relation to the optical spectra.

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