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Zeeman splitting of excitons and biexcitons in single $In_{0.60}Ga_{0.40}As/GaAs$ self-assembled quantum dots

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Self-assembled $In_{0.60}Ga_{0.40}As/GaAs$ quantum dots have been studied by single dot photoluminescence spectroscopy at $T=1.5$ K. Emission from the biexciton state is observed, for which we find a binding energy of 3.1 meV. In magnetic field we observe equal Zeeman splittings for the exciton and biexciton spectral lines. The splitting of the biexciton emission is given by the splitting of the exciton in the final state of the biexciton transition. The circular polarization of the quantum dot luminescence decreases with increasing magnetic field, although the spin splitting is clearly larger than the thermal energy. This indicates that the spin relaxation is strongly suppressed in these quantum dots. [S0163-1829(98)50736-9]

During recent years the spectroscopic investigation of semiconductor quantum dots[1] (QD's) has clearly revealed the three-dimensional confinement of electronic states in these structures.[2–7] In particular, QD's fabricated by self-organized growth have attracted considerable attention. Self-organized growth typically provides QD's with dimensions comparable to the exciton Bohr radius.[3,6,7] Therefore the effects of the geometric confinement and the Coulomb interaction effects have to be investigated in detail to obtain a quantitative understanding of the optical spectra.

When studying arrays of QD's, fine-structure effects arising for example from exciton-exciton interaction can often hardly be resolved in the optical spectra. Fluctuations of the dot size cause a broadening of the spectral lines, which is comparable or larger than typical Coulomb interaction energies and prevents the study of such effects. In case of self-assembled QD's the spectral width of the emission from an ensemble of dots typically is a few tens of meV.[3]

Therefore several experimental techniques have been developed which allow the investigation of a single QD. For example, a fine-structure splitting of the ground and excited exciton states was reported which was attributed to a shape asymmetry of the studied natural QD's.[8] Emission from biexcitons has been observed in such systems[5] as well as in self-assembled QD's, for which also emission from charged excitons[9] and from multiexciton complexes[10] was found. Time-resolved studies on laser-induced single QD's gave rise to speculations about a Fermi-gas like behavior of the excitons in a QD.[11] For strongly confined nanocrystals random intermittency of the QD luminescence was found and explained by Auger emission processes of carriers out of a QD.[12] In a magnetic field the Zeeman splitting of the ground and excited exciton emission was observed.[13] Further, its hyperfine structure was resolved in the excitonic luminescence from natural QD's which arises from the dynamic polarization of the lattice nuclei (Overhauser effect).[14]

In this paper we report on magnetophotoluminescence studies of excitons (X) and biexcitons (X_2) in self-assembled $In_{0.60}Ga_{0.40}As$ QD's. To suppress the effects of inhomogeneous broadening, the experiments were performed on single QD's allowing to observe fine-structure effects in the optical spectra. In magnetic field a large Zeeman splitting of the exciton emission is observed, which corresponds to an exciton g factor of about 3. Using high optical excitation, emission from biexcitons is observed. The spin splitting of the biexciton is equal to the splitting of the exciton emission and originates from the splitting of the exciton in the final state of the optical transition.

The $In_{0.60}Ga_{0.40}As$ QD's were fabricated by molecular beam epitaxy using Stransky-Krastanov quasi-three-dimensional growth. From scanning electron microscopy of a sample in which the QD's were not covered by a cap layer we find that the dots are approximately hemispherically shaped and have a base radius of about 10 nm. For spectroscopic investigation of small numbers of QD's small fields were fabricated by electron beam lithography and wet chemical etching on an unpatterned sample. The lateral sizes of these fields ranged from 600 down to 100 nm. The smallest fields contain only a few quantum dots, and for magnetic field investigation only those were chosen, whose low excitation spectra consist of a single exciton emission line.

The samples were held at a temperature of 1.5 K in the helium insert of an optical cryostat with a superconducting split-coil magnet ($B<8$ T). The magnetic field was aligned normal to the heterostructure. For optical excitation we used a cw Ar^+ laser. The laser spot could be focused to a diameter of about 30 μm. In order to avoid sample heating, the average laser power density incident on the sample was limited to 10 W cm^{-2}. The emission was dispersed by a double monochromator ($f=0.6$ m) and detected with a LN_2-cooled Si charge-coupled-devices camera. The polarization of the luminescence could be analyzed by a quarter-wave-retarder and a linear polarizer.

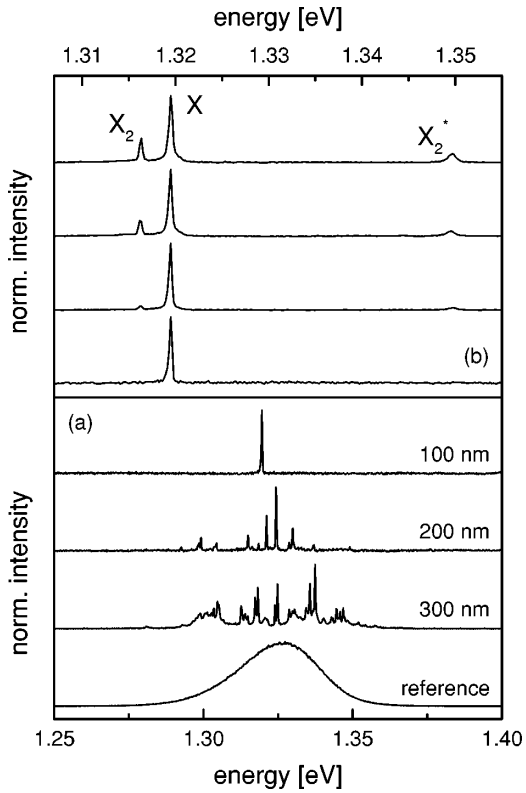

FIG. 1. (a) Photoluminescence spectra of self-assembled $In_{0.60}Ga_{0.40}As/GaAs$ QD's in small fields of different lateral sizes (100, 200, and 300 nm) and from an unpatterned dot sample. (b) Photoluminescence spectra from a 100 nm wide field containing only a single QD for different excitation powers.

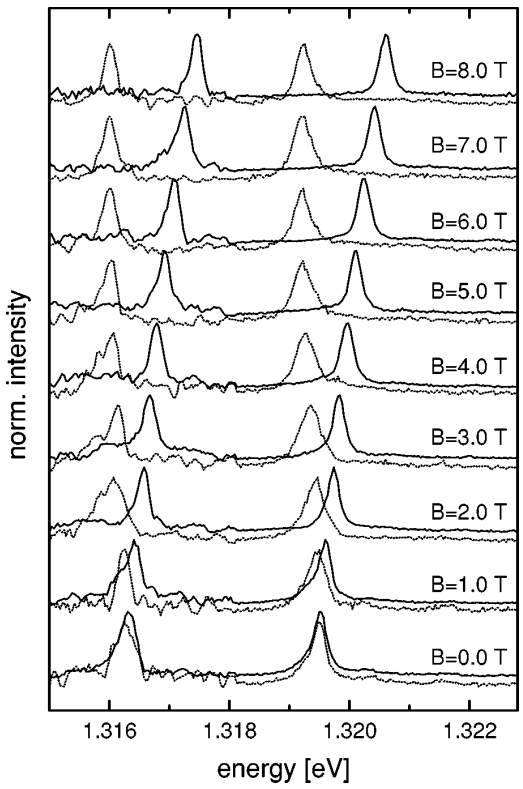

FIG. 2. Polarized photoluminescence spectra of a self-assembled $In_{0.60}Ga_{0.40}As/GaAs$ single QD for various magnetic fields increasing from bottom to top. The solid and the dotted lines give the σ^- and the σ^+ polarized spectra, respectively.

Figure 1(a) shows photoluminescence spectra of an unpatterned dot sample (lowest trace), of fields with lateral sizes of 300 and 200 nm (middle traces) and finally of a field with a size of 100 nm (uppermost trace). The spectra were recorded using low excitation powers of about 20 μW. The emission spectrum of the unpatterned sample consists of a broad emission band with a spectral width of about 30 meV arising from dot size fluctuations. In the large field the broad emission is broken up into a set of sharp lines each having a width of about 200 μeV limited by the spectral resolution of the setup. However, the number of spectral features is still too large for clear assignments when varying for example the excitation power. In contrast, for the smallest field only one sharp emission line X is observed at 1.3196 eV, which originates from the recombination of one electron-hole pair in the dot. From this observation we can conclude that this field contains only a single QD.

Photoluminescence spectra of a single $In_{0.60}Ga_{0.40}As/GaAs$ QD are shown in Fig. 1(b) for various excitation powers. With increasing excitation (from bottom to top) a new emission line labeled by X_2 appears on the low energy side of the X emission at $E=1.3165$ eV. Simultaneously a high energy feature $X_2^\star$ is observed about 30 meV above the exciton line, the origin of which will be discussed below. The intensity of X_2 increases superlinearly with excitation power. In combination with the results of magnetic field studies described below this power dependence indicates that this emission can be attributed to biexcitonic recombination.

The low energy shift of X_2 relative to the X emission arises from the X-X Coulomb interaction in the dot and can be considered as biexciton binding energy, the difference between the energies of two uncorrelated excitons and the energy of the two exciton complex. This energy difference is equal to the splitting between the exciton and biexciton recombination lines. From the spectra we find a biexciton binding energy $\Delta(X_2)$ of 3.1 meV, which is clearly larger than the typical binding energies of less than about 1 meV observed in GaAs-based quantum wells or of 0.13 meV in bulk GaAs. The enhancement of $\Delta(X_2)$ is a consequence of the strong geometric confinement in self-assembled QD's which causes a pronounced increase of the inter-particle Coulomb interactions.[16]

Figure 2 shows photoluminescence spectra of a single $In_{0.60}Ga_{0.40}As/GaAs$ QD at varying magnetic fields. The σ^+ polarized spectra are given by the dotted lines, the σ^- polarized ones by the solid lines, respectively. To observe the X_2 features clearly, they have been normalized to unity. With increasing magnetic field a splitting of the exciton emission, is observed. The splitting arises from the Zeeman splitting of the exciton, $\Delta E_\pm = g_X \mu_B B$, where g_X is the exciton g factor and μ_B is the Bohr magneton. Similarly to the exciton, also the biexciton emission is seen to split with increasing magnetic field.

In Fig. 3 the energies of the spin-polarized exciton and biexciton emission lines are plotted versus magnetic field. The center of the exciton doublet (indicated by the solid line) shifts diamagnetically to higher energies with increasing

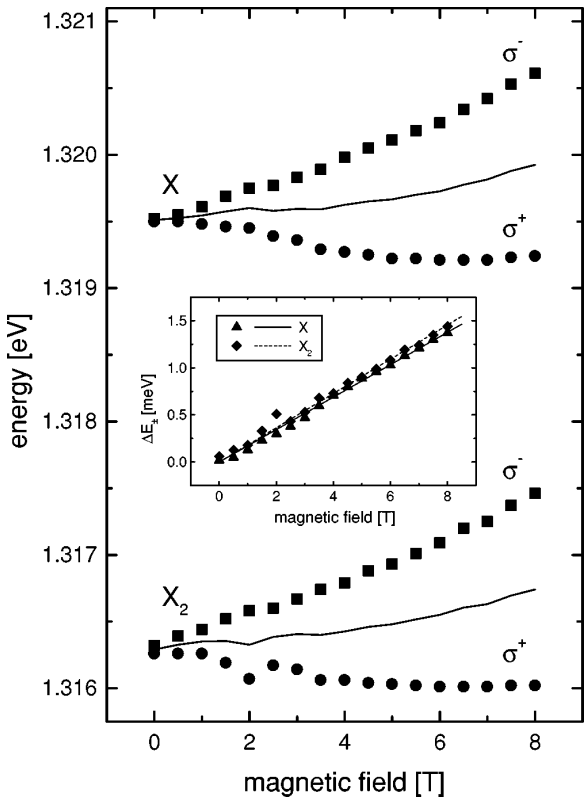

FIG. 3. Exciton and biexciton energies as functions of the magnetic field. The transitions are labeled corresponding to their different circular polarizations. The inset shows the Zeeman splitting of the exciton and the biexciton emission versus magnetic field.

magnetic field. For strong geometric confinement the diamagnetic shift can be approximated[15] by

$$\Delta E^{X}_{diamag} = \frac{e^2}{8}\left(\frac{\langle r_e^2\rangle}{m_e} + \frac{\langle r_h^2\rangle}{m_h}\right)B^2, \tag{1}$$

where $\langle r^2_{e/h}\rangle$ are the mean square lateral extensions of the electron and hole wavefunctions in the dot, respectively. Assuming equal extensions for electron and hole we obtain $\langle r^2_{e,h}\rangle^{1/2} \sim 12$ nm from the experimental shift which is in good agreement with the dot radius of 10 nm from scanning electron microscopy.

For low magnetic fields the σ^+ polarized exciton emission line shifts to lower energies because of the spin splitting and for $B > 4$ T its energy is almost constant, because the Zeeman splitting and the diamagnetic shift are about equal. In contrast, the σ^- polarized line shifts smoothly to higher energies. Within the experimental accuracy the spin splitting between the two emission lines increases linearly with B, as shown by the triangles in the inset of Fig. 3, and at $B = 8$ T the splitting is as large as 1.4 meV. From the slope of the increase we obtain an excitonic g factor of $g_X = 3.02 \pm 0.05$.

The magnetic field dependence of the biexciton transition energies is quite similar to that of the exciton. In particular, the diamagnetic shift of the center of the X_2 spectral lines is equal to the diamagnetic shift of the exciton emission. This shift is given by the difference of the biexciton diamagnetic shift in the initial state of the optical transition and of the exciton shift in the final state. Thus we find that the biexciton diamagnetic shift is two times the diamagnetic shift of the exciton. This is in agreement with calculations, which yield for the biexciton diamagnetic shift in strongly confined QD's:

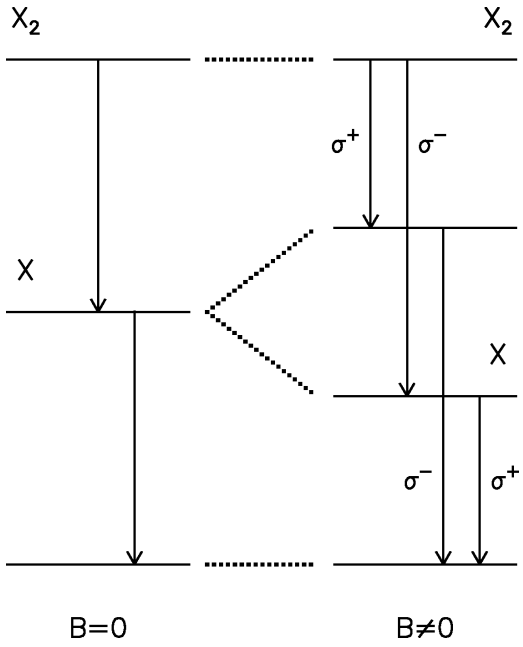

FIG. 4. Scheme of optical transitions from the exciton and biexciton states at zero and nonzero magnetic field.

$$\Delta E^{X_2}_{diamag} = \frac{e^2}{8}\left(\frac{\langle r_{e_1}^2\rangle}{m_e} + \frac{\langle r_{e_2}^2\rangle}{m_e} + \frac{\langle r_{h_1}^2\rangle}{m_h} + \frac{\langle r_{h_2}^2\rangle}{m_h}\right)B^2 = 2\Delta E^{X}_{diamag}. \tag{2}$$

Further, the magnetic field dependence of the biexciton spin splitting is equal to the dependence for the exciton spin (diamonds in the inset of Fig. 3). The biexciton is a spin singlet state and therefore its energy cannot be split by a magnetic field. However, the final state of the biexciton transition is an exciton, as indicated by the transition scheme in Fig. 4. Therefore the spin splitting of the biexciton emission is given by the Zeeman splitting of the exciton, in agreement with the experimental findings. The reduction of the energy separation between the σ^- polarized biexciton and the σ^+ polarized exciton emission can be interpreted as reduction of the biexciton binding energy with increasing magnetic field. This means, that for the present QD's B reduces the biexciton binding energy from about 3.1 to 1.6 meV.[16]

We have also investigated the circular polarization of the spin-polarized exciton emission from these QD's. For that reason spectra were recorded without using quarter wave retarder and linear polarizer. The spin splitting can be observed starting from magnetic fields of about 2 T. In Fig. 5 the circular polarization

$$P = \frac{I(\sigma^+) - I(\sigma^-)}{I(\sigma^+) + I(\sigma^-)} \tag{3}$$

is plotted versus magnetic field. At $B = 2$ T the polarization is about 25%. For higher B it then surprisingly drops to zero up to 8 T, although the spin splitting for high fields is clearly larger than the thermal energy at the low temperatures in the

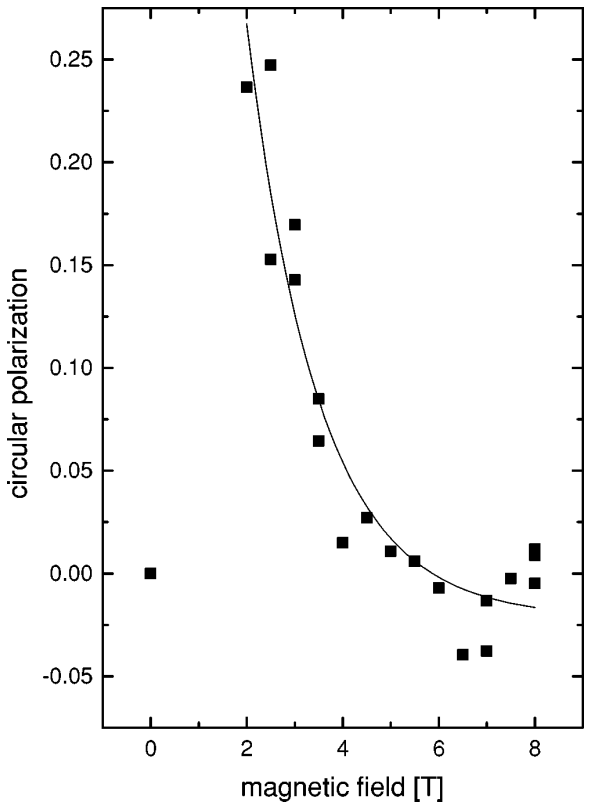

FIG. 5. Circular polarization of the exciton luminescence from a single self-assembled $In_{0.60}Ga_{0.40}As/GaAs$ QD. The solid line is a guide to the eye.

present experiments. We assume that the excitons are unpolarized when they relax into the spin-split exciton ground state because we are exciting nonresonantly with linearly polarized light. Therefore the spins cannot be in thermal equilibrium and the spin relaxation has to be suppressed in these strongly confined QD's at high B. This means that the spin relaxation time increases with B and at high fields it is significantly longer than the radiative decay time, for which we find about 500 ps from time-resolved spectroscopy. This ''bottleneck'' for spin relaxation presumably is a direct consequence of the discrete energy level structure in the QD's, which suppresses strongly energy exchange processes related with a spin flip.

This observation is confirmed by the high excitation spectra in Fig. 1(b), where simultaneously with the appearance of the biexciton feature a high energy spectral line $X_2^{\star}$ is observed. The high energy emission originates from the recombination of an electron and a hole in the first electron and hole excited states. The splitting of 30 meV from the exciton line agrees well with the splitting which we obtain from calculations for the energies of the QD single particle states. When the ground state is occupied by a carrier with a certain spin orientation, an additional carrier in the dot can only relax into the ground state when it has opposite spin and biexciton formation is possible. However, when the spin orientation is the same as that of the ground state carrier, relaxation into the ground state is prevented by the Pauli-principle and an excited biexciton state is formed, which consists of an electron-hole pair in the ground and one in the first excited states.

In conclusion, we have investigated self-assembled $In_{0.60}Ga_{0.40}As/GaAs$ QD's by single dot magneto-photoluminescence spectroscopy. Due to the strong quantum confinement we find a greatly enhanced biexciton binding energy. In magnetic field the fine structure of the biexciton emission is equal to that of the exciton, because the biexciton is a spin-singlet state and the spectrum of the X_2 emission is fully determined by the exciton in the final state of the biexciton transition.

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[16] Strictly, the biexciton binding energy is given by the energy separation of the biexciton emission from the dark exciton state. The dark exciton state has a lower energy than the optically active one due to the electron-hole exchange interaction. However, in III-V semiconductors the typical exchange interaction energy is rather small, for example 20 μeV in bulk GaAs. This energy might be enhanced by quantum confinement by about one order of magnitude, which still is small compared to the energy separation of 3.1 meV observed here.