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Strong variation of the exciton *g* factors in self-assembled $In_{0.60}Ga_{0.40}As$ quantum dots

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The magnetic-field dependence of energy and polarization of the low excitation photoluminescence from self-assembled $In_{0.60}Ga_{0.40}As$ quantum dots has been studied by single dot spectroscopy. For the spin splitting of the emission lines at 8 T three different characteristic values, -1.5 meV, -0.3 meV, and $+0.3$ meV are observed. For dots with small spin splittings, no indication for ground shell biexciton emission can be found, in contrast to dots with a large splitting. These findings suggest that the emission in the dots with a weak Zeeman interaction originates from charged excitons whose formation becomes possible from dopants in the nearest surrounding of the dot structures. The presence of these dopants modifies the electronic band structure and thus also the *g* factors of the carriers. [S0163-1829(99)50236-1]

During the last few years considerable interest has been focused on quantum dot (QD) structures.[1] The interest arises from the three-dimensional confinement of their electronic levels, which gives rise to a discrete density of states. QD's have been fabricated by using different techniques, among which self-assembled growth has been proven particularly successful because of the high optical quality of the dot structures.[1,2] Optical spectroscopy addressing large ensembles of QD's has clearly demonstrated the confinement effects on the electron and hole states. For example, photoluminescence (PL) spectra recorded using high optical excitation show several distinct emission features, which can be attributed to the recombination of electrons and holes from several confined levels.[3]

However, despite the huge progress in the fabrication of QD's the largest challenge for studying their basic physical properties still are the variations of, e.g., dot size and shape in QD ensembles. These variations give rise to energetically broad emission spectra, from which it is difficult to obtain information about the electronic fine structure. A particularly interesting topic related to this fine structure is the Zeeman interaction of the electron and hole spins with an external magnetic field, which can be described by their effective *g* factors. *g* factors in confined geometries depend very sensitively on the electronic band structure and thus furnish detailed insight into the electronic levels.

The difficulties in the resolution of the fine structure in studies on QD ensembles were motivation for developing spectroscopic techniques with a high spatial resolution.[4–15] These tools permit addressing individual QD's and therefore inhomogeneous broadening effects are suppressed. Among these tools are sophisticated spectroscopy techniques such as near-field optical spectroscopy with a resolution below one wavelength of light. Other techniques require a further processing of the as-grown QD samples. For example, the QD's can be covered by a mask that contains holes through which only a single or a few dots are optically excited by a laser.

Here we present a magneto-optical study of the spin splitting of the low excitation emission from single self-assembled $In_{0.60}Ga_{0.40}As$ QD's. By comparative spectroscopy of a large number of individual QD's we find three distinct levels for the Zeeman splitting. In combination with the results of high excitation spectroscopy we attribute the observed spectral lines to emission either from neutral or from charged excitons:[16–21] The *g* factor of the trion emission deviates from that of the neutral exciton complexes due to the modification of the band structure, in particular the band mixing, by the dopants.

The $In_{0.60}Ga_{0.40}As$ QD's have been fabricated using the Stransky-Krastanow growth mode.[22] Details of the fabrication have been given earlier,[15] except for a growth interruption of 10 s after the QD material deposition for the particular samples in the present experiments. From scanning electron microscopy of an uncapped sample a QD density of 1×10^{10} cm^{-2} is estimated. The emission of the two-dimensional reference occurs at about 30 meV lower energy than the emission from a sample without growth interruption. This indicates that the QD size is slightly increased by the growth interruption, which enables surface diffusion of QD material towards the structures.

Lithography was used to fabricate small mesa structures with lateral sizes of about 100 nm, for which we estimate a mean occupation by one QD from the above dot density. For optical studies only mesas were selected whose low excitation spectra consist of a single emission line in an energy range equivalent to the full width at half maximum ($\sim$25 meV) around the center of emission of an unpatterned reference sample. From this observation we conclude that only one QD is contained in such a mesa that is to be contrasted with other structures, whose spectra consist of a few lines in this energy range.

For spectroscopic studies the QD's were held in the liquid helium insert ($T=1.5$ K) of a split-coil magnetocryostat ($B \leqslant 8$ T). All experiments were performed in Faraday configuration. For optical excitation a cw Ar^+ laser (514.5 nm, linear polarization) was used. The emission of the QD's was dispersed by a double monochromator with a focal length of 0.6 m and detected by a liquid nitrogen cooled charge coupled devices camera. The polarization of the emission could be analyzed by a quarter wave retarder and linear polarizers.

Figure 1 shows characteristic PL spectra of different single QD's recorded at an excitation power of 100 μW. In magnetic field the emission in each case splits into a doublet. The lowest panel shows emission spectra at $B=0$ (solid trace) and 8 T (dotted trace) of a QD representing the ma-

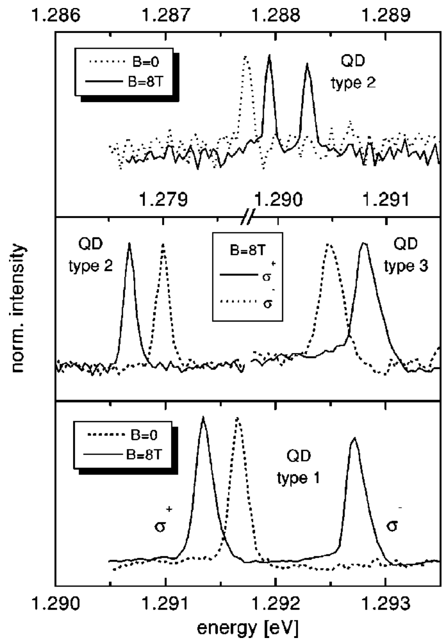

FIG. 1. PL spectra of single self-assembled $In_{0.60}Ga_{0.40}As$ QD's. The lowest panel shows a QD with a large spin splitting, the two other panels QD's with small splittings.

jority of structures (type 1). The spin splitting $\Delta_{\pm}=E(\sigma^{+})-E(\sigma^{-})$ at 8 T in this case is -1.5 meV. Two other types of QD's with a strongly reduced Zeeman splitting can also be found for these samples. The middle panel shows polarized PL spectra of such QD's at 8 T. For type 2 (left trace) the splitting is about -0.3 meV, while for type 3 (right trace) a splitting of $+0.3$ meV is observed.[23] The diamagnetic shift of the center of the spectral lines is the same for all structures and is 0.4 meV from $B=0$ to 8 T, as seen from the comparison of a type 2 QD (top panel) with a type 1 QD.

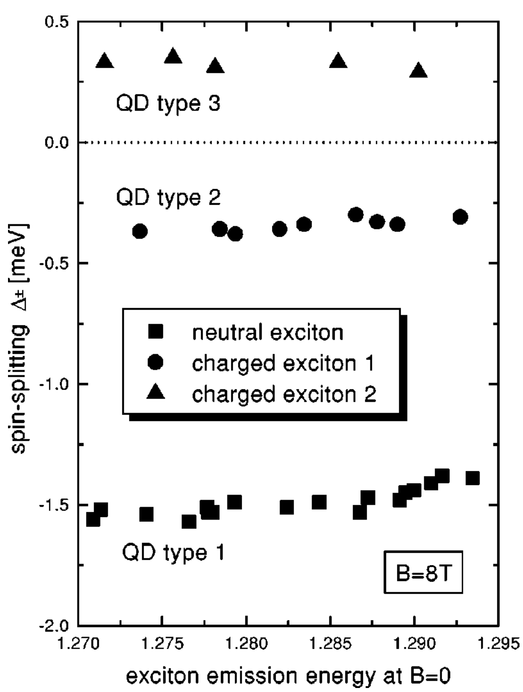

FIG. 2. Spin splitting at $B=8$ T observed for a large number of single $In_{0.60}Ga_{0.40}As$ QD's. The splitting is plotted against the QD emission energy at zero magnetic field.

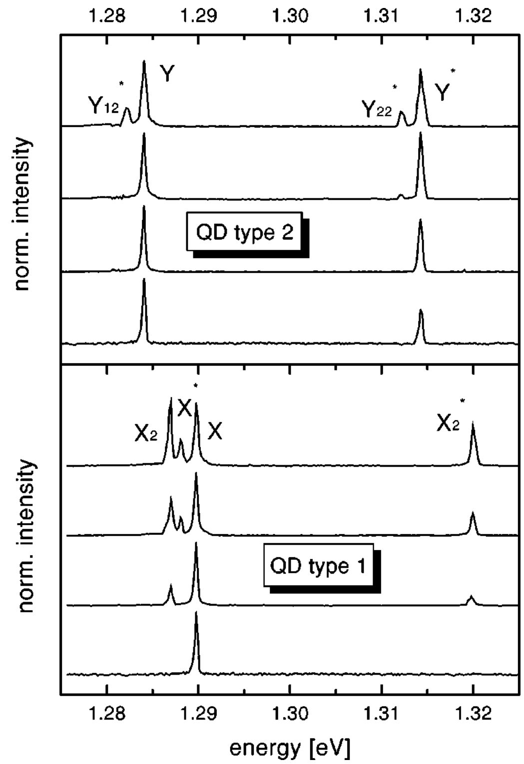

FIG. 3. PL spectra of single $In_{0.60}Ga_{0.40}As$ QD's recorded at $B=0$ for varying excitation powers. The lower panel shows spectra for a QD of type 1, the upper one spectra for a QD of type 2. The excitation powers from bottom to top were 100, 200, 300, and 400 μW.

The different behaviors are summarized in Fig. 2, which shows the spin splitting $\Delta_{\pm}$ for a number of single QD's at $B=8$ T. $\Delta_{\pm}$ is plotted versus the energy of the QD emission at $B=0$. Each symbol at a different energy corresponds to a different single QD. The energy range extends over 25 meV, which is about the half width of the emission from an unpatterned reference. Three different levels are observed for the spin splitting, one at -1.5 meV (QD type 1), one at -0.3 meV (QD type 2) and another one at $+0.3$ meV (QD type 3).

Additional information about the origin of the different spectral features can be obtained from studies using varying excitation powers. Figure 3 shows PL spectra from single QD's of type 1 (lower panel) and of type 2 (upper panel). The behavior of type 3 dots is very similar to that of type 2. When increasing the excitation, for QD type 1 emission from biexcitons X_2 is observed, which are formed by two excitons of opposite spin structure in the ground shell. This emission is shifted to lower energies by the biexciton binding energy of 3 meV.

Simultaneously with the appearance of X_2, luminescence from the first excited exciton shell is detected more than 30 meV above the ground shell. Its appearance is related to the suppression of spin relaxation in QD's: Due to Pauli blocking the relaxation of excitons in excited shells is prevented, if the ground shell is already populated with an exciton of the same spin structure.[12] Therefore an excited biexciton complex is formed by an exciton in the ground and one in the excited shell. This complex can decay by recombination of one of these excitons. Recombination of the excited exciton leads to the high-energy feature X_2^*. Decay of the ground exciton, on the other hand, leads to the emission X^*. In comparison to X, its energy is lowered by the Coulomb interaction energy of the two excitons. This excited biexciton binding energy of about 2 meV is smaller than that of a ground biexciton. In contrast, for the other QD types emission from the first excited shell is detected even at the lowest excitation powers.[24] In addition, no ground-state biexciton emission can be observed at any excitation level, whereas excited biexciton emission appears both for the ground and the excited shell.

As explanation for these observations we suggest the following model: From the data in Fig. 3 the formation of ground biexcitons in QD's of types 2 and 3 is suppressed in a situation in which the lowest shell is occupied by a single optically generated exciton. This means that relaxation of excitons is not possible, although in the excited shells excitons with spin structures opposite to that in the ground shell are present due to the nonresonant, linearly polarized laser excitation. This can occur, if the ground state is populated by either an excess electron or hole.[25]

Let us first consider the case of low excitation. If the ground shell is occupied by a carrier, a first exciton can only relax, if the spin of the corresponding carrier of the exciton is different from that of the excess carrier. In case of relaxation, emission from the ground shell (Y) will appear. In case of blocked relaxation, emission from the excited shell is detected (Y^*). Furthermore, at increased excitation, relaxation of a second exciton is forbidden so that ground biexciton formation is impossible. Instead, excited biexcitons are formed leading to the emission features Y^*_{21} and Y^*_{22}. By contrast, type 1 QD's are apparently free of excess carriers, so that biexcitons can be formed in the ground shell.

Thus we assign the low excitation emission in QD's of type 1 to the radiative decay of neutral excitons,[26] while the emission from QD types 2 and 3 is attributed to the recombination of charged excitons. The electron/hole for the trion formation most probably is released from residual impurities in the QD samples. Due to the interruption of the QD growth the probability of incorporating impurities on the QD surfaces is considerably enhanced in comparison to noninterrupted growth.[27] Despite of the ultraclean environment it is well known that MBE-fabricated crystals show a background doping of about 10^{14}–10^{15} cm^{-3}. The dopants release their carriers, which are transferred into the QD confinement regions. Together with optically excited electron-hole pairs these equilibrium carriers will form excitonic trions.

The consistency of this explanation has to be checked with the magnetic-field data. In particular, the question of whether an excess carrier can modify the exciton spin splitting has to be addressed. The excess carrier with its given spin orientation is present in the initial and the final states of the optical transition. Since the spin splitting is given by the difference of the Zeeman interaction energies in the initial and final states, an excess carrier is expected not to affect the spin splitting of the charged exciton emission. Thus the spin splitting of positive or negative trion emission should be the same as that for a neutral exciton complex, which is given by the sum of the electron and hole g factors. This has recently been observed for charged excitons in modulation doped quantum wells.[28] In this case the residual impurity is located rather far from the QD.

If the dopant is, however, located in the nearest surrounding of the QD, it is expected to change the g factors. The electron and hole g factors are determined by the details of the electronic band structure. The presence of an impurity represents a strong perturbation of this band structure, which will change the mixing of the bands, in particular of the valence band. This might explain the strong deviation of the Zeeman splitting in the QD's of types 2 and 3 in comparison to type 1 dots. The observation of two spin splitting levels different from that of the exciton most probably is explained by the presence of two different impurities.

In previous studies[15] we have obtained values for electron and hole g factors in QD's fabricated without growth interruption and we have shown that the spin splitting is mostly given by the hole g factor. These QD's show almost the same spin splitting for neutral excitons as the present structures. Assuming that the presence of a dopant modifies mainly the valence-band structure, but leaves the electron g factor unchanged, we can use the value of $g_e = -0.8$ determined previously also for the different QD types observed here. In this way, we obtain a hole g factor of about -2.4 for dot type 1, $+0.1$ for dot type 2, and $+1.4$ for dot type 3.

For self-assembled QD's one might also expect strong variations of the g factors due to variations of size, shape, or strain. However, the observation of three distinct levels for the spin splitting shows that these variations do not affect the g factors strongly. For example, for QD's of type 1, $\Delta_\pm$ tends to decrease with increasing emission energy. The variation of the spin splitting with emission energy is only of the order of 0.1 meV within each level, from which geometry variations can be ruled out as origin for the three-level splitting.

Finally let us discuss the equal diamagnetic shifts δ of the emission lines in the different QD types. In analogy to the spin splitting, δ is given by the difference of the shifts of the initial and the final states. In strongly confined QD's the diamagnetic shift of a carrier configuration is given by the sum of the shifts of the individual constituting carriers, which are proportional to $e^2\langle \vec{r}^2\rangle/m$. Here $\langle \vec{r}^2\rangle$ is the square of the lateral extension of the single particle wave function with m being the carrier mass. Initial and final states of both neutral and charged exciton complexes differ from one another by the recombining electron-hole pair. Therefore the diamagnetic shift of the emission from each configuration that is given by the sum of the electron and hole diamagnetic shifts is identical.

In summary, we have reported a detailed spectroscopic study of the exciton spin splitting of single QD's. The experimental data suggest that the variations of the splitting originate from the observation of neutral or charged excitons. The g factor of the trions most probably deviates from that of the neutral complexes due to the modification of the band mixing in the QD's by the presence of dopants. These results also might give an explanation for recent reports of observations of strong fluctuations of the spin splitting for self-assembled QD's.[29]

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[1] For an overview of the extensive work on QD's, see L. Jacak, P. Hawrylak, and A. Wojs, *Quantum Dots* (Springer Verlag, Berlin, 1998); D. Bimberg, M. Grundmann, and N. Ledentsov, *Quantum Dot Heterostructures* (John Wiley & Sons Ltd., New York, 1998).

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[16] See, for example, K. Kheng *et al.*, Phys. Rev. Lett. **71**, 1752 (1993); G. Finkelstein *et al.*, *ibid.* **74**, 976 (1995); H. Buhmann *et al.*, Phys. Rev. B **51**, 7969 (1995).

[17] For self-assembled QD's Landin *et al.* have reported the observation of emission from charged excitons X^- and X^+ (Ref. 11). The Coulomb interaction energies in trion complexes have been studied by transmission spectroscopy on ensembles of QD's (Ref. 18). Theoretically, calculations of the energies and oscillator strengths of trions in QD's have been done. (Refs. 19–21).

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[23] The emission intensities and spectral widths vary with the studied QD. The origin of these variations is not yet understood, but might be related to different influences of the lateral surfaces of the mesa structures on the QD's.

[24] The excited state emission for the QD types 2 and 3 cannot be attributed to the ground-state emission from a second QD in the mesa stucture. Besides its appearance already at low excitation, it shows a spin splitting that is considerably larger than that of a ground-state exciton.

[25] The excess carrier is an equilibrium carrier because the observed splitting for a single QD does not vary with time. If the QD would be randomly charged, then at some time emission from excitons, respectively, biexcitons would be observed at characteristic energies. These emission features would disappear in case of the appearance of a charge in the QD. Instead, a new emission line would appear at different energies due to the Coulomb interaction of the exciton with the momentary excess carrier. Such behavior was observed at very high excitation powers significantly exceeding those used for the present studies.

[26] The magnetic-field dependence of the type 1 QD emission is very similar to the behavior observed for QD's fabricated without growth interruption (Ref. 12). There, a spin splitting of $\Delta_{\pm} = -1.4$ meV was found at 8 T. However, for these QD's only very few splittings with magnitudes as small as those for QD types 2 and 3 have been observed. The frequency of observing charged exciton emission is $<5\%$ for QD's fabricated without, whereas it is $\sim 25\%$ for the structures with growth interruption.

[27] In spatially inhomogeneous crystals as QD samples are, compensation effects between donors and acceptors will not necessarily occur, because the carriers will be captured in the QD's so that diffusion is suppressed.

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