Spectroscopic study of dark excitons in $In_xGa_{1-x}As$ self-assembled quantum dots by a magnetic-field-induced symmetry breaking

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The dark states of the heavy-hole ground-state exciton have been studied by photoluminescence spectroscopy of single self-assembled $In_{0.60}Ga_{0.40}As$ quantum dots in magnetic fields of varying strengths and orientations. When tilting B out of the [001] crystal direction the exciton states with angular momenta $|m|=2$ are mixed with the $|m|=1$ states and become observable. In combination with a detailed analysis of the corresponding Hamiltonian a comprehensive picture of the exciton fine structure is obtained. This fine structure is characteristic for direct gap semiconductors with a twofold degenerate valence band.

Excitons play a key role for the optical properties of semiconductor nanostructures, which are of vital interest for present and future optoelectronic devices. Therefore, it is important to understand the properties of these electron-hole complexes, one of which is their fine structure. It consists of a multiplet of states with different angular momenta that are split due to exchange interaction. Among them, only a few with momentum 1 can couple to the light field permitting their spectroscopic study. The other states with angular momenta different from 1 are dark, but still they have strong influence, for example, on the kinetics of the bright excitons. Due to their long lifetime, dark excitons serve as a reservoir out of which they can be converted into optically active states by spin-flip processes.[1]

Spectroscopic access to dark excitons can be obtained by a controlled symmetry breaking, due to which angular momentum is no longer a good quantum number and the dark states are mixed with the bright ones. Such a symmetry breaking can be obtained, for example, by applying a magnetic field. Only a few dark exciton studies[2] have been reported because in typical III-V semiconductors such as GaAs the smallness of the involved energy splittings prevents the resolution of the fine structure, which additionally is also complicated by inhomogeneous broadening. Here spectroscopy on single quantum dots[3–15] opens new possibilities: first, the inhomogeneous broadening is suppressed and second, the Coulomb interaction energies are strongly enhanced by quantum confinement.

Here we study the part of the exciton fine stucture related to the dark excitons in $In_{0.60}Ga_{0.40}As$ self-assembled quantum dots.[16] This fine structure is characteristic for a wide variety of direct gap semiconductors with a twofold degenerate valence-band edge such as quantum wells, wires, or dots. At zero magnetic field the fine structure is given by the electron hole exchange interaction[17–19]

$$\mathcal{H}_{Exchange}=\sum_{i=x,y,z}(a_iJ_{h,i}S_{e,i}+b_iJ_{h,i}^3S_{e,i}) \qquad (1)$$

for an exciton formed by a hole with spin J_h and an electron with spin S_e.[20] The z direction is chosen to point along the heterostructure growth direction. Due to the strain between dot and substrate the heavy and light holes are strongly split in energy so that the light holes can be neglected in this analysis. Therefore, the terms that are linear in the hole momentum along the x and y directions, which cause a mixing of heavy and light holes, can be omitted. With $J_h=\frac{3}{2},J_{h,z}=\pm\frac{3}{2}$ and $S_e=\frac{1}{2},S_{e,z}=\pm\frac{1}{2}$, four exciton states can be formed, which are characterized by their angular momenta projections $M=S_{e,z}+J_{h,z}$. The interaction of the electron and heavy-hole spins with an external magnetic field of arbitrary strength and orientation is given by[17–19]

$$\mathcal{H}_{Zeeman}(B)=\mu_B\sum_{i=x,y,z}\left(g_{e,i}S_{e,i}-\frac{g_{h,i}}{3}\hat{J}_{h,i}\right)B_i$$

with the Bohr magneton μ_B and the direction-dependent electron and hole g factors $g_{e,i}$ and $g_{h,i}$.[20]

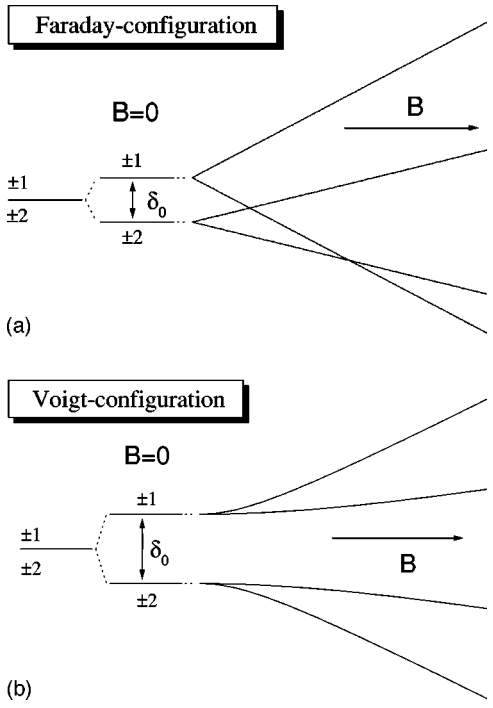

FIG. 1. Scheme of the exciton fine structure in Faraday (a) and Voigt configuration (b).

Using the exciton states $(|+1\rangle,|-1\rangle,|+2\rangle,|-2\rangle)$ as a basis, the following matrix representation is obtained for the $B=0$ Hamiltonian:

$$\mathcal{H}_{Exchange}=\frac{1}{2}\begin{pmatrix}\delta_0 & \delta_1 & 0 & 0\\ \delta_1 & \delta_0 & 0 & 0\\ 0 & 0 & -\delta_0 & \delta_2\\ 0 & 0 & \delta_2 & -\delta_0\end{pmatrix},$$

where the abbreviations $\delta_0=-\frac{3}{4}(a_z+\frac{9}{4}b_z)$, $\delta_1=\frac{3}{8}(b_x-b_y)$, and $\delta_2=\frac{3}{8}(b_x+b_y)$ have been introduced. $\mathcal{H}$ has block diagonal form; bright and dark excitons do not mix. Furthermore, for symmetric structures, which are of interest here, the x direction is equivalent to the y direction $(b_x=b_y)$.[21] Then the off-diagonal matrix elements δ_1 in the subspace of bright excitons vanish and $|+1\rangle$ and $|-1\rangle$ are eigenstates of $\mathcal{H}_{Exchange}$.

The matrix describing the Zeeman-interaction of the spins with a magnetic field $\mathbf{B}=B(\sin\vartheta,0,\cos\vartheta)$ where ϑ is the polar angle relative to the z axis is given by

$$\mathcal{H}_{Zeeman}(B)=\frac{\mu_B B}{2}\begin{pmatrix}(g_{e,z}+g_{h,z})\cos\vartheta & 0 & g_{e,x}\sin\vartheta & g_{h,x}\sin\vartheta\\ 0 & -(g_e+g_{h,z})\cos\vartheta & g_{h,x}\sin\vartheta & g_{e,x}\sin\vartheta\\ g_{e,x}\sin\vartheta & g_{h,x}\sin\vartheta & -(g_e-g_{h,z})\cos\vartheta & 0\\ g_{h,x}\sin\vartheta & g_{e,x}\sin\vartheta & 0 & (g_e-g_{h,z})\cos\vartheta\end{pmatrix}.$$

Here we assume the in-plane magnetic field points along the x direction. Due to the rotational symmetry of the dots, the energies are independent of the choice of this orientation. Different spin-splitting patterns in the magnetic field arise from these Hamiltonians.

Figure 1(a) shows a scheme of the splitting of the exciton fine structure in Faraday configuration ($\vartheta=0$). Bright and dark excitons do not mix, because the magnetic field does not destroy the planar symmetry and angular momentum remains a good quantum number. At $B=0$ the states with $m=\pm1$ and $m=\pm2$ are split by the exchange energy δ_0. Due to the off-diagonal matrix elements for the ±2 excitons these states generally hybridize. However, since these matrix elements are proportional to the b_i, which are small compared to a_z, this mixing will be neglected, in agreement with the experimental data below. With increasing magnetic field,

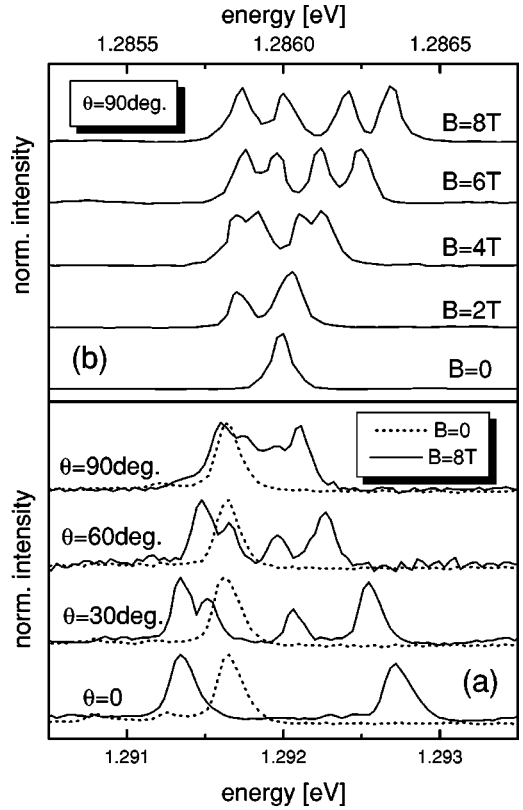

FIG. 2. (a) Photoluminescence spectra of a single quantum dot at $B=0$ (dotted traces) and 8 T (solid traces) for varying orientation of the magnetic field. (b) Same as (a) but for varying magnetic field strengths in a Voigt configuration.

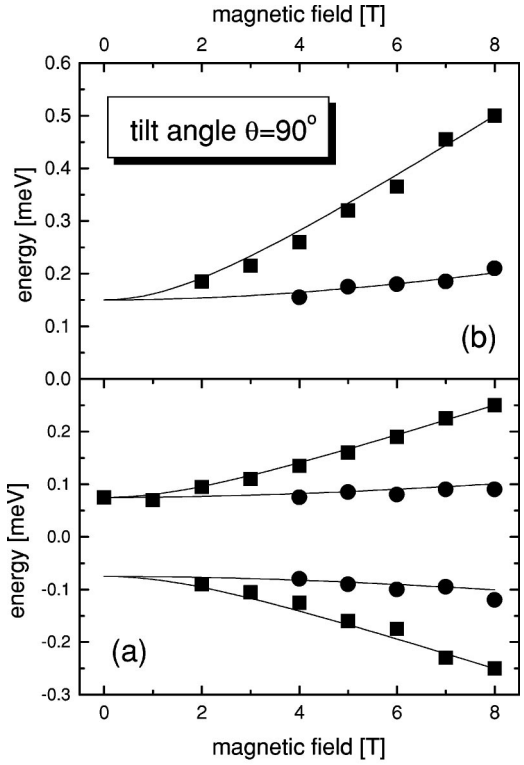

FIG. 3. (a) Energies of the exciton transitions versus magnetic field in a Voigt configuration. (b) Energy splitting between the outer and the inner emission features as function of B. Symbols give experimental data; lines give the results of the calculations.

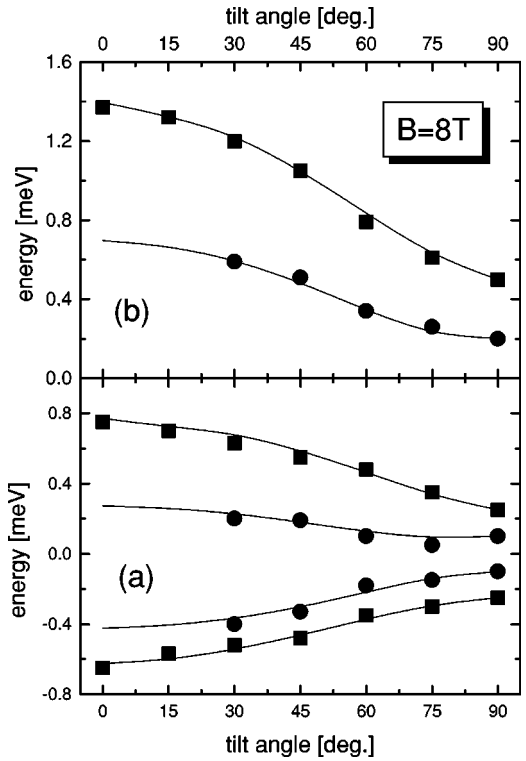

FIG. 4. (a) Exciton transition energies at $B=8$ T plotted against the tilting angle of the magnetic field. (b) Spin splitting between the outer and the inner emission features as function of the tilt angle at $B=8$ T. Symbols give experimental data, lines give the results of the calculations.

states with positive and negative angular momenta split. The splitting depends linearly on B. For the ± 1 excitons it is proportional to the sum, for the ± 2 excitons it is proportional to the difference of the electron and hole g factors.

The magnetic field dependence changes drastically when B is rotated to obtain Voigt configuration ($\vartheta=90°$), as shown in Fig. 1(b). The planar symmetry of the system is now destroyed. Classically, the carrier spins precede around the in-plane field, due to which the $B=0$ angular momentum eigenstates are mixed. From the matrix representation we see that the $m=\pm 1$ states do not mix with each other, but the $m=+1$ mixes with the $m=+2$ exciton through the precession of the electron spin, while it is mixed with the $m=-2$ exciton through the precession of the hole spin (vice versa for the $m=-1$ exciton). Due to the mixing, the dark excitons become optically active. The spin splitting shows a kind of anticrossing behavior. The $|m|=1$ excitons at $B=0$ both shift to higher energies with increasing magnetic field, while the energies of the $|m|=2$ excitons are lowered.

The aim of our study was to resolve this fine-structure pattern in full detail. For that reason we have performed magnetoluminescence spectroscopy of single self-assembled $In_{0.60}Ga_{0.40}As$ quantum dots and have varied the strength and orientation of the magnetic field. The quantum dot structures have been described earlier.[12] Scanning electron microscopy of uncapped samples suggests a planar cylindrical symmetry of the dots as assumed in the theoretical analysis.[21] Single dot spectroscopy was done by fabricating small mesa structures with lateral sizes of about 100 nm using lithography. For optical studies the dot structures were held in the liquid-helium insert ($T=1.2$ K) of an optical split-coil magneto-cryostat ($B\leqslant 8$ T). The orientation of B relative to the dots could be changed by rotating the sample. The power of excitation by an Ar^+ laser was limited to about 20 μW in order to avoid multiparticle effects. The emission of the dots was dispersed by a double monochromator (0.6-m focal length) and detected by a charge-coupled devices camera.

Figure 2(a) shows photoluminescence spectra of a single quantum dot at $B=8$ T for varying field orientations (solid traces). For comparison, also the zero field spectra are shown by the dotted traces. For $\vartheta=0$ the exciton splits into a doublet corresponding to the $m=\pm 1$ excitons, while the excitons with $m=\pm 2$ remain dark. However, when the magnetic field is already tilted, for an angle of $\vartheta=30°$, two additional features appear between the strong emission lines, which arise from mixing of dark and bright excitons.[22] When increasing the tilt angle the energy separation between the exciton states decreases strongly. Figure 2(b) shows the photoluminescence spectra of another $In_{0.60}Ga_{0.40}As$ single quantum dot that were recorded in Voigt configuration for varying magnetic field strength.[23] With increasing B the center of the emission features shifts to higher energies due to the diamagnetic shift of the exciton $\propto B^2$. At $B=0$ a single emission line arising from the recombination of the $|m|=1$ excitons is observed. On its low-energy side an additional spectral line appears at $B=2$ T, which originates from the $|m|=2$ excitons at zero field. For higher fields each of the two emission lines splits into a doublet. The energy splitting between them increases with magnetic field.

Figure 3(a) shows the observed exciton transition energies and the energy splitting between them versus magnetic field in Voigt configuration. To study only the fine-structure effects we have subtracted the energy of the center of the emission features for each field strength individually. The splitting of the energy levels follows the scheme sketched in Fig. 1(b). The splitting between the two inner emission lines is mainly given by the exchange interaction δ_0 and the Zeeman interaction has little influence on their energies. In contrast, it significantly changes the energies of the outer emission features, as seen from Fig. 3(b). The eigenfrequencies and eigenstates for $\vartheta=90°$ have been calculated from the

TABLE I. Excitonic states in a magnetic field in Voigt configuration ($\vec{B}\|\vec{e}_x$).

Energy	Eigenstate
$+\frac{1}{4}[+(\delta_1+\delta_2)+\sqrt{(2\delta_0+\delta_1-\delta_2)^2+4(g_{e,x}-g_{h,x})^2\mu_B^2B^2}]$	$N_1[c_{11}(\|+1\rangle-\|-1\rangle)+c_{12}(\|+2\rangle-\|-2\rangle)]$
$+\frac{1}{4}[-(\delta_1+\delta_2)+\sqrt{(2\delta_0-\delta_1+\delta_2)^2+4(g_{e,x}+g_{h,x})^2\mu_B^2B^2}]$	$N_2[c_{21}(\|+1\rangle+\|-1\rangle)+c_{22}(\|+2\rangle+\|-2\rangle)]$
$-\frac{1}{4}[-(\delta_1+\delta_2)+\sqrt{(2\delta_0+\delta_1-\delta_2)^2+4(g_{e,x}-g_{h,x})^2\mu_B^2B^2}]$	$N_3[c_{31}(\|+2\rangle-\|-2\rangle)+c_{32}(\|+1\rangle-\|-1\rangle)]$
$-\frac{1}{4}[+(\delta_1+\delta_2)+\sqrt{(2\delta_0-\delta_1+\delta_2)^2+4(g_{e,x}+g_{h,x})^2\mu_B^2B^2}]$	$N_4[c_{41}(\|+2\rangle+\|-2\rangle)+c_{42}(\|+1\rangle+\|-1\rangle)]$

fine-structure Hamiltonian $\mathcal{H}=\mathcal{H}_{Exchange}+\mathcal{H}_{Zeeman}$ and are given in Table I. The results are also shown in Fig. 3 by the solid lines.

From the calculations the electron and hole g factors as well as the exchange energies are determined. With $\delta_1=0$ and $\delta_2=0$ we obtain 150 μeV for the electron-hole exchange interaction δ_0. For the g factors we obtain the following values: $g_{e,z}=-0.80$ and $g_{e,x}=-0.65$, $g_{h,x}=-2.20$ and $g_{e,x}=-0.35$. This directionality dependence of the g factors is considerably different from that of quantum wells. There an isotropy of the electron g factor is found, while the in-plane hole g factor is zero. Due to the nonzero g factor $g_{h,x}$ only, a quadruplet splitting occurs, otherwise the energies of the two lower, and higher states, respectively, would coincide.

With these parameters the dependence of the exciton transition energies on the tilt angle of the magnetic field can be calculated. The dependencies at $B=8$ T are shown by solid lines in Fig. 4(a) in comparison with the experimental data, from which good agreement is seen. With increasing tilt angle the energies tend to converge resulting in a reduction of the spin splitting between the emission lines. These splittings are plotted against the tilt angle in Fig. 4(b). The splitting between the two outer lines, i.e., between the bright excitons at zero field, decreases from 1.4 meV at $\vartheta=0$ to 0.5 meV at $\vartheta=90°$. For the splitting between the inner features (dark excitons at $\vartheta=0$) the reduction is only about 0.5 meV.

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[19] E. L. Ivchenko and G. E. Pikus, *Superlattices and Other Heterostructures*, Springer Series in Solid-State Sciences Vol. 110 (Springer, Berlin, 1997).
[20] Geometric confinement causes an enhancement of the coefficients a_i and b_i in the Hamiltonian in comparison to the respective values in bulk or in quantum well. It also leads to a modification of the electron and hole g factors.
[21] The symmetry of the dot structures has been confirmed in the Voigt-configuration experiments by rotating the dot sample by 45°, 90°, and 135° in the x-y plane. Within experimental accuracy, the same B dependence of the exciton transition energies has been observed as described here.
[22] Although the off-diagonal matrix elements are rather small for this field orientation, the emission intensities from the predominantly dark states are rather strong. This arises from the suppression of the spin relaxation due to the discrete energy level structure in strongly quantized dots, in qualitative agreement with the results reported in Ref. 9. To date, however, no conclusive picture of spin-flip processes in quantum dots has been developed, either experimentally or theoretically.
[23] The halfwidth of the emission varies with the studied dot from 50 to 150 μeV. These variations might be related to different influences of charges on the surfaces of the mesa structures.