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Optical and magnetic anisotropies of the hole states in Stranski-Krastanov quantum dots

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Using the trion as a monitor we investigate the anisotropy of the single-hole state in epitaxial CdSe/ZnSe quantum dots. Heavy-light hole mixing caused by a symmetry reduction below D_{2d} results in elliptical polarization of the optical transitions with a specific axis for each dot defined by strain and shape. In a transverse magnetic field, a quartet of strictly linearly polarized lines appears that reveals the off-diagonal coupling of both electron and hole states. Although induced by the field, the linear polarization is not related to the field orientation, but either along or perpendicular to the dot axis seen at zero field. We find an in-plane hole g factor as large as 0.3 with distinct anisotropic behavior.

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Semiconductor quantum dots (QDs) are often called "artificial atoms." However, crystal symmetry and specific band structure of the semiconductor make the energy eigenstates utterly different from those of the simple particle-in-a-box problem.[1,2] Detailed knowledge of the energy levels and the corresponding wave functions is of immense fundamental and practical interest. In the present work, the hole eigenstates of epitaxial Stranski-Krastanov (SK) QDs are examined in this respect. We consider the prototype case of a semiconductor with zinc-blende symmetry. Unlike colloidal nanocrystals, SK QDs exhibit a defined quantization axis (z) given by the growth direction. In the frequent case of [001] growth, the symmetry of the bulk semiconductor implies that x and y direction are equivalent (D_{2d}). That equivalence is generally lifted in a QD due to shape and/or strain. The splitting of the optically allowed exciton into a line doublet caused by the electron-hole exchange interaction is a well-studied consequence of this symmetry reduction.[3,4] The question of whether the in-plane anisotropy is also manifested on a single-particle level has not been addressed so far. As long as the confinement energy is much smaller than the band gap, the electron with its s-like Bloch function can be safely treated as an isotropic effective-mass particle with spin $\pm 1/2$. On the other hand, such an approximation fails even qualitatively for the p-type holes of total angular momentum $\pm 3/2$ connected with the fourfold-degenerate edge of the valence band. In what follows, we demonstrate that the single-hole states of a SK QD are indeed substantially affected and that the anisotropy translates into a specific coupling with a transverse magnetic field. Our experimental concept is based on the trion feature representing the fundamental optical excitation of charged QDs.[5,6] In case of a single negative resident charge, the trion consists of one hole and two electrons. As the total spin of the latter is zero in the singlet ground state, the trion represents a direct monitor of the angular momentum configuration of the hole.[7]

The CdSe/ZnSe QD samples used in this study are grown by molecular-beam epitaxy using a thermal activation procedure.[8,9] The height and diameter of the pure CdSe core are about 2 and 5–10 nm, respectively, as revealed by transmission electron microscopy.[8,9] The samples are naturally n doped so that some of the QDs are occupied with a resident electron. In order to study individual QDs, mesa structures with linear dimensions of 1 μm are prepared by electron-beam lithography and wet chemical etching. The samples are placed on a rotation holder in a split-coil magneto-optical cryostat and are excited with the 488-nm line of a cw Ar^+ laser at a temperature of 2 K. The experiments are carried out in backward geometry with the propagation direction of incident and emitted light parallel to the [001] growth axis. Magnetic fields B up to 9 T are applied in Voigt ($\vec{B} \perp z$) geometry. The photoluminescence (PL) in a certain linear polarization is selected by rotation of a half-wave plate introduced in front of the analyzer. The PL signal is dispersed in a monochromator with a linear dispersion of 0.24 nm/mm and detected with a nitrogen cooled charge-coupled-device matrix providing a spectral resolution of about 50 μeV. After careful inspection of many PL lines, we have selected three representative QDs for this paper.

As a consequence of the zero total electron spin, the electron-hole exchange interaction vanishes in the trion ground state. Unlike the exciton, the trion states are twice degenerate independent on the symmetry—in accordance with the Kramers theorem for half integer spin particles.[10] The optical recombination of the trion leaves a single electron behind. For a heavy-hole (HH) trion with projection $J_z = \pm 3/2$, only the two transition $|\pm 3/2\rangle \rightarrow \sigma^{\pm} + |\pm 1/2\rangle$ are allowed from the four possibilities where $\sigma^{\pm}$ denotes the respective circular photon polarization. For excitation with linearly polarized light, the trion PL is thus expected to be totally unpolarized. Figure 1 depicts spectra taken on QD#1 for different positions of the polarization analyzer $\vec{e} = (\cos\phi, \sin\phi, 0)$ relative to [100] direction. In addition to the main line of 70-μeV spectral width, an acoustic-phonon feature is present on the low-energy side. In marked contrast to the expectation, the emission exhibits noticeable linear polarization. The normalized line strength follows closely a relation $I(\phi) = 1 + \rho_L \cos[2(\phi - \theta)]$ with a polarization degree of $\rho_L = 0.36$. The characteristic angle θ is independent on the excitation polarization and defines, hence, a direction intrinsic to the QD. Both degree of polarization and direction scatter from QD to QD, even when located in the same mesa

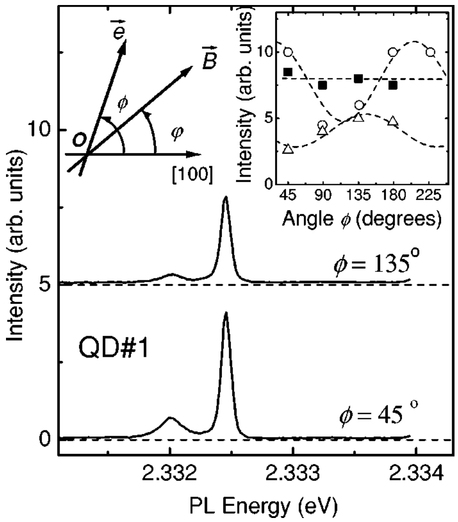

FIG. 1. Zero-field PL feature of a single trion (QD#1) detected in linear polarization along the [110] and $[1\overline{1}0]$ directions, respectively. Upper left: Definition of the angle ϕ (as well as φ in an external magnetic field, see Figs. 3 and 4). Inset: PL signal of the trion as a function of ϕ, circles: QD#1, triangles: QD#2, full rectangles: QD#3. The dashed lines are fits with $A+B\cos[2(\phi-\theta)]$. Variation of the excitation polarization (in the figure along $[1\overline{1}0]$) leaves the data unchanged.

(inset of Fig. 1). Although θ is close to [110] and $[1\overline{1}0]$ for QD#1 and QD#2, respectively, there is no clear correlation with the principal crystal axes. In QD#3, the effect is even below the resolution of our polarization set up ($\rho_L < 0.1$).

The above observation reflects directly the impact of in-plane anisotropy on the hole ground state where the angular momentum is no longer a good quantum number. In leading order, a light-hole (LH) contribution with $J_z = \pm 1/2$ is created and the wave function reads as $|\psi_h^{\pm}\rangle = |\pm 3/2\rangle - (\gamma^{\pm}/\Delta E_{l-h})|\mp 1/2\rangle$, ΔE_{l-h} being the energy separation between the HH and LH ground state. The normalized optical dipole element of the trion transition, accounting for a phase shift of π between the two electron configurations, can be expressed as $\vec{d} \propto (\vec{x}-i\vec{y})(|+3/2\rangle\langle+1/2| - \kappa|+1/2\rangle\langle-1/2|) + (\vec{x}+i\vec{y})(|-3/2\rangle\langle-1/2| - \kappa|-1/2\rangle\langle+1/2|)$.[11] Here, $\vec{x}$ and $\vec{y}$ are unit vectors along the [100] and [010] direction, respectively, $|J_z\rangle\langle S_z|$ is the projection operator between the electron state of spin S_z and the trion state with angular momentum J_z, and κ measures the difference in strength between HH and LH radiative coupling. The LH admixture is thus associated with circular polarization $|\mp 1/2\rangle \rightarrow \sigma^{\mp} + |\pm 1/2\rangle$ just opposite to $|\pm 3/2\rangle$ so that each of the transitions becomes elliptically polarized. Writing $\gamma^{\pm} = \gamma \exp(\pm i 2\theta)$, the preferential polarization direction can be identified with the phase of the HH-LH coupling matrix element, while the linear degree is given by $\rho_L = 2\gamma\kappa\Delta E_{l-h}/(\Delta E_{l-h}^2 + \gamma^2\kappa^2)$. Shape, as well as strain, both including chemical composition, are sources of in-plane anisotropy. For C_{2v} symmetry, shear strain provides a mixing coefficient $\gamma_{\varepsilon}^{\pm} \propto \mp i\langle\psi_{lh}^0|\varepsilon_{xy}|\psi_{hh}^0\rangle$[12] and, thus, a polarization along [110] or $[1\overline{1}0]$. The same holds when the QD shape has no inversion symmetry in the growth direction where the

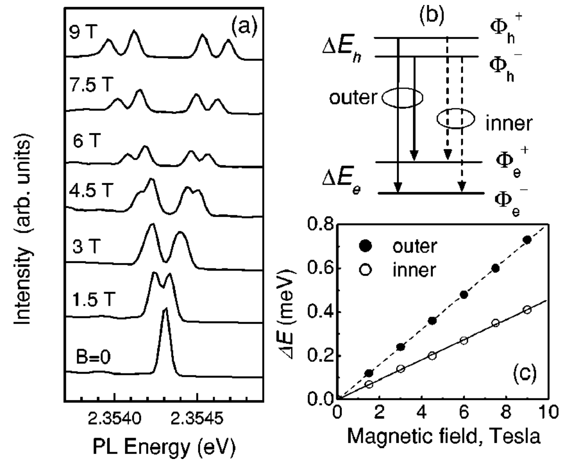

FIG. 2. Splitting of the trion PL line (QD#2) in a transverse magnetic field. (a) PL spectra in unpolarized detection mode. (b) Scheme of optical transitions. (c) Separation between inner and outer lines as a function of the field strength. The data points for $B < 6$ T are taken from polarized detection measurements (see Fig. 3) where the splitting is better seen.

role of ε_{xy} is replaced by the derivative $\partial V/\partial z$ of the confinement potential.[13] The Luttinger Hamiltonian[14] gives in spherical approximation $\gamma_S^{\pm} \propto \langle\psi_{lh}^0|(\partial/\partial x \pm i\partial/\partial y)^2|\psi_{hh}^0\rangle$ where ψ_{hh}^0 and ψ_{lh}^0 are the zero-order orbital wave functions defined by the confinement potential. In C_2 symmetry with principal axes rotated by an angle α relative to the [100]-[010] frame, this yields preferential polarization along $\theta = \alpha$ or $\theta = \alpha + \pi/2$ depending on what direction is that of stronger carrier localization. Additional strain components can further change this angle. From the fact that θ scatters noticeably across the QD ensemble, we conclude that strain and shape anisotropy combine in a given QD and create a specific polarization axis. From the experimental polarization degree we estimate $\gamma \approx 0.3\Delta E_{l-h}$ ($\kappa^2 = 1/3$) for QD#1.

A reduction of the in-plane symmetry shows up even more prominently in the coupling with a transverse magnetic field.[15–17] The Zeeman interaction of the isotropic electron with an external field $\vec{B}$ is described by the Hamiltonian $H_B^e = (1/2)\mu_B g_e \vec{\sigma}\vec{B}$ with σ_i denoting the Pauli matrices. The magnetic coupling of the hole in D_{2d} is given by $H_B^h = \mu_B g_0[k\vec{J}\vec{B} + q(J_x^3 B_x + J_y^3 B_y + J_z^3 B_z)]$ (g_0: free-electron g factor) where $\vec{J}$ is the 3/2 angular momentum operator.[14] The second non-Zeeman term is consistent with the symmetry of the Luttinger Hamiltonian. The coefficients k and q are parameters of the bulk material, and no information is available on q for ZnSe or CdSe. Unlike the electron, no coupling of the heavy hole arises from the Zeeman part $H_{BZ}^h = \mu_B g_0 k\vec{J}\vec{B}$ in first and second order in a transverse field $\vec{B} = B(\cos\varphi, \sin\varphi, 0)$. In contrast, as shown in Fig. 2(a), a line quartet exposing all four possible transitions emerges for the trion PL in this geometry. The energy separation between the inner (ΔE_i) as well as outer (ΔE_o) quartet components is a linear function of B [see Fig. 2(c)]. These findings unambiguously demonstrate the existence of an effective HH in-plane g factor. A line splitting of the trion PL in Voigt geom-

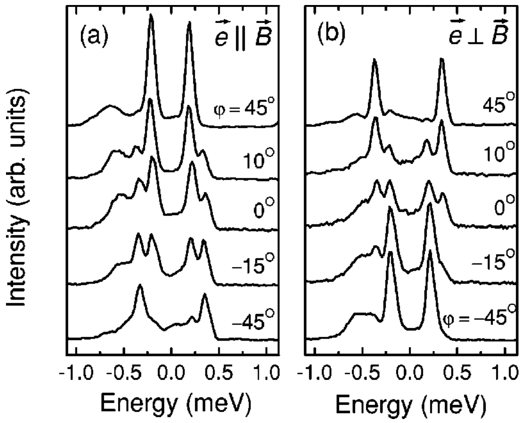

FIG. 3. (a) and (b) Reconstruction of PL spectra in QD#1 under rotation of the sample around the z axis for ($\vec{B} \perp z$). The spectra are taken in linear polarizations along ($\vec{e} \parallel \vec{B}$) and perpendicular ($\vec{e} \perp \vec{B}$) to the magnetic-field direction (B=9 T). The angle φ is defined in Fig. 1.

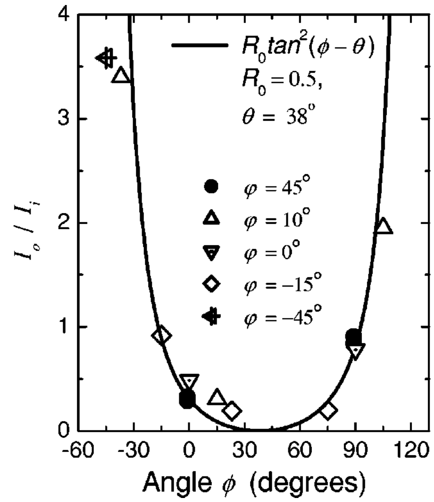

FIG. 4. Signal ratio of the outer and inner components for QD#1 as a function of the analyzer position ϕ for various field orientations φ. The data demonstrate that the lines are fully linearly cross polarized. The polarization axes are independent on the magnetic field but specific for the respective QDs. θ=38° for QD#1 is in accord with the zero-field measurements of Fig. 1.

etry has been also observed on III-V QDs,[6] however, its origin has not been elucidated so far.

More information is revealed when the polarization of the PL lines is analyzed in conjunction with the orientation of the in-plane magnetic field. For the spectra depicted in Fig. 3, the polarization analyzer axis $\vec{e}$ is set parallel and perpendicular to $\vec{B}$, respectively, while the sample is rotated around the quantization axis. Inasmuch as it concerns the spectral position of the components, there is no change of the line pattern. However, the inner and outer pair of the quartet are orthogonally polarized to each other and the linear polarization degree of the lines is now practically 100%. In contrast to particles with isotropic Zeeman coupling, where the polarization would be either along or perpendicular to $\vec{B}$, a distinct dependence of the line strength on the rotation angle is found. In fact, there is no correlation with the field orientation at all. This is demonstrated by measurements where the polarization analyzer is rotated, while a certain angle φ between $\vec{B}$ and the [100] direction is adjusted. The polarization axes are most accurately determined from the intensity ratio $R=I_o/I_i$ of the outer and inner lines, since deviations in the total signal level caused by the different adjustments are canceled out here. As seen in Fig. 4, this ratio is independent on the choice φ and can be well described by a function $R(\phi)=R_0\tan^2(\phi-\theta)$. Within the accuracy of our set up, the angle θ agrees well with that deduced for the built-in polarization without magnetic field for the respective QD. Both for QD#1 and QD#2, the inner lines are stronger ($R_0 \cong 0.5$). In QD#3, where no built-in polarization is found, a line doublet is merely observed signifying that the in-plane HH g factor is very small here.

A transverse field introduces off-diagonal couplings between the Kramers-degenerate states. The eigenstates are hence phase-rotated superpositions of the type $\Phi_p^{\pm}=(1/\sqrt{2})\times(\pm e^{-i\varphi_p/2}|\psi_p^{+}\rangle+e^{i\varphi_p/2}|\psi_p^{-}\rangle)$, $p=e,\ h$, with energies shifted by $\pm(1/2)\mu_B g_p^{\perp} B$ relative to their zero-field position.[17] For the isotropic electron, $|\psi_e^{\pm}\rangle=|\pm 1/2\rangle$ and $\varphi_e=\varphi$. Without HH-LH mixing, i.e., for $|\psi_h^{\pm}\rangle=|\pm 3/2\rangle$, using the explicit expressions of the J_i^3,[14] the D_{2d} non-Zeeman term provides a coupling $\langle +3/2|H_B^h|-3/2\rangle=-3/4\mu_B g_0 qB\exp(i\varphi)$ within the HH subspace. That is, $\varphi_h=\pi-\varphi$ (instead of $\varphi_h=\pi+\varphi$ in the isotropic case) and by calculating $|\langle\Phi_h^{\pm}|\vec{e}\vec{d}|\Phi_e^{\pm}\rangle|^2$, it is readily seen that the trion PL becomes indeed strictly linearly polarized, however, with axes $\phi=-\varphi$ and $\phi=\pi/2-\varphi$ defined by the magnetic field. As this behavior is in clear contradiction to the experimental findings in Figs. 3 and 4, we conclude $q \ll 1$ so that the D_{2d} interaction does not contribute significantly to $g_h^{\perp}$. A similar result (q=0.04) has been found on GaAs/(Ga,Al)As quantum well structures.[18] In the presence of HH-LH mixing, the Zeeman term generates a first-order magnetic coupling $\langle\psi_h^{+}|H_{BZ}^h|\psi_h^{-}\rangle=\sqrt{3}\mu_B g_0 k(\gamma/\Delta E_{l-h})B\times\exp[-i(\varphi+2\theta)]$ in the hole ground state. That is, $\varphi_h=\varphi+2\theta$ for the magnetic phase shift or, defining an effective g tensor by $H_{BZ}^h=\mu_B\tilde{S}_i g_{ij}^h B_j$ where $\tilde{S}_x=-\sigma_x/2$, $\tilde{S}_y=\sigma_y/2$ are the components of the hole pseudospin,

$$\hat{g}_h=g_h^{\perp}\begin{pmatrix}-\cos 2\theta & \sin 2\theta\\ \sin 2\theta & \cos 2\theta\end{pmatrix},\quad g_h^{\perp}=\frac{2\sqrt{3}\gamma}{\Delta E_{l-h}}kg_0. \tag{1}$$

While the zero-field transitions are elliptically polarized along the same axis, pairs of linearly cross-polarized lines are obtained with intensities of $I_o\propto(1-\kappa\gamma/\Delta E_{l-h})^2\sin^2(\phi-\theta)$ and $I_i\propto(1+\kappa\gamma/\Delta E_{l-h})^2\cos^2(\phi-\theta)$ for the outer and inner quartet components, respectively. This result is in perfect agreement with the experiment, also regarding the larger intensity of the inner lines. From $(\Delta E_o\pm\Delta E_i)/\mu_B B$ we find $g_e^{\perp}=1.1\pm0.1$ in all three QDs and $g_h^{\perp}=0.3\pm0.1$ for QD#1 and QD#2.

The in-plane hole g tensor is diagonal in the coordinate system defined by the intrinsic polarization axes. Here, it has pseudoisotropic character $g_{xx}=-g_{yy}$. The magnetic anisotropy can be, in principal, also observed on the exciton in uncharged QDs. However, the exciton transitions are already energetically split into linearly polarized components due to electron-hole exchange. Thus, significantly stronger magnetic fields are required. In a previous study on diluted magnetic quantum wells,[15] where the Zeeman coupling is giantly enhanced, a pseudoisotropic in-plane g-factor has been deduced from polarized PL measurements. However, the polarization degree is only in the 10% range and no splitting into the characteristic quartet pattern is seen.

The above magnetic-field data has also important implications for the hole spin relaxation in QDs. If the QD symmetry is even below C_2, the hole ground state becomes $|\psi_h^{\pm}\rangle = \alpha^{\pm}|\pm 3/2\rangle + \beta^{\pm}|\mp 3/2\rangle + \gamma^{\pm}|\mp 1/2\rangle$, enabling spin flip via the admixture of the J_z state of opposite sign. In a transverse field, this will give rise to a diagonal magnetic coupling in the hole ground state associated with a specific dependence of the PL lines on the field orientation φ. The fact that such behavior is not observed experimentally shows that deviations from C_2 symmetry are indeed small, making low spin-flip rates feasible[7].

In conclusion, distinct anisotropies of the hole states in SK QDs have been uncovered by studying the trion feature. In particular, a significant coupling to a transverse magnetic field with a strongly anisotropic in-plane g-tensor is found. While the magnetic field introduces strict linear optical polarization, the direction of the polarization is independent of the field direction, but defined by shape and strain anisotropies. Those anisotropies are hardly seen by direct morphological techniques. Our findings have various implications for the use of QDs as single-spin elements. The anisotropies enable to tailor magnetic superpositions in a desired way.

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