Optically induced spin polarization and g-factor anisotropy of holes in CdSe/ZnSe quantum dots

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A high degree of the circular polarization of trion emission has been observed in self-organized CdSe/ZnSe quantum dots under quasiresonant optical excitation. The depolarization of emission in a tilted magnetic field was investigated, and it has been established that the observed polarization results from optical spin orientation of heavy holes. The g-factor anisotropy of holes has been measured. It has been shown that due to a specific formation of nanoislands, the quantization axis of a quantum dot declines markedly from the structure growth axis.

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The discrete character of the energy spectrum of electrons determines the peculiarities of great many physical processes in semiconductor quantum dots or, as they are sometimes called, artificial atoms. In particular, recent theoretical and experimental studies of spin dynamics have shown that the basic mechanisms of spin relaxation that take place in three- and two-dimensional systems[1,2] are blocked in quantum dots due to a three-dimensional limitation of the motion of carriers.[3] The spin-relaxation times observed in quantum dots (QDs) at low temperatures[4–6] have proven to be significantly longer than in bulk semiconductors[1] or quantum wells.[7–9] In that way, owing to long spin memory times and the possibility to create and control spin polarization, the QDs are promising objects for future spintronic and quantum information devices.

The present work investigates the behavior of the spin polarized carriers and excitons in self-organized CdSe/ZnSe quantum dots placed in an external magnetic field. A high degree of photoluminescence polarization proves that the long-wavelength part of the emission band is dominated by emission of trions (i.e., of the bound state of an exciton with an electron or hole). The behavior of polarization in a tilted magnetic field bears witness to the fact that there occurs the spin precession of a particle with an anisotropic g factor, i.e., of heavy hole. The study of the spin properties of holes is especially important because the dominant mechanism of electron spin relaxation in quantum dots through the hyperfine interaction with nuclei[10] is blocked for holes owing to a p-type wave function.

It will be recalled that the self-organized quantum dots (distorted usually) have an asymmetric form substantially smaller in height than the size of its base (disklike QDs). The upper valence band in such QDs splits into the states of heavy holes with angular momentum projection $J_z=\pm 3/2$ (ground state) and light holes with $J_z=\pm 1/2$ ($z\parallel[001]$ is the QD growth axis). The heavy-hole states are characterized by the anisotropic g-factor (g-tensor), and in the case of sufficiently high QD symmetry, its components in the structure plane are nearly zero:[11] $g_x=g_y=g_\perp\approx 0$. The electron states are of s type with $s_z=\pm 1/2$ and an almost isotropic g-factor. An electron and a heavy hole form an exciton quartet with projections of angular momentum $J_z=\pm 1$, ± 2, split into two sublevels by isotropic exchange interaction. With decrease in symmetry (usually down to C_{2v}) the anisotropic exchange interaction splits an optically active doublet $J_z=\pm 1$ into two linearly polarized dipoles $|X\rangle$ and $|Y\rangle$ (in the simplest case, the optical transitions are allowed in orthogonal linear polarizations of light $e\parallel X$ and $e\parallel Y$), whose orientation is determined by QD symmetry.[12] Quantum beats between those eigenstates lead to the loss of spin memory since the period of beats $T_b=2\pi h/\delta$ (δ is the anisotropic exchange splitting), as a rule, is small in comparison with the lifetime of exciton. The exciton fine structure manifests itself in magneto-optical polarization experiments with a QD ensemble,[13,14] and is also easily resolved in the photoluminescence spectra of single quantum dots.[15–17]

Experimental procedure. The structures under investigation were grown by molecular-beam epitaxy as pseudomorphic relative to the (001) GaAs buffer layer at a substrate temperature $T_S=280\ ^\circ$C and consisted of an upper and a lower ZnSe barrier layer 60 and 20 nm thick, respectively. They surrounded a CdSe insertion whose nominal thickness was 2.1 monolayers (ML). The precipitation of CdSe was carried out in the regime of multicyclic (0.3 ML per cycle) modified epitaxy with a heightened migration of atoms.[18] The structures were not doped; however, the carriers from residual impurities in the barrier might easily be captured by QDs.

Measurements were performed at a temperature $T=2$ K. Photoluminescence (PL) was excited by an argon ion laser with a pumping density of about 1 W/cm^2. The degree of circular or linear polarization was determined from the following relationships: $\mathcal{P}_c=(I_+-I_-)/(I_++I_-)$ and $\mathcal{P}_L=(I_x-I_y)/(I_x+I_y)$, where I_+, I_- are the intensities of circularly polarized components of radiation and I_x, I_y are the radiation intensities polarized along the axes x and y.

Results. Figure 1(a) presents the low-temperature ($T=2$ K) photoluminescence spectra of CdSe/ZnSe QDs under excitation by the line $\lambda=4965$ Å of an argon laser. Besides a broad luminescence band of excitons, the peculiarities due to the excitation of one or two optical phonons are observed. The large linewidth is usually related to the dispersion of QDs by size. A minor circular polarization of PL ($\sim 2\%$) determined by z projection of the mean spin of the exciton ($\mathcal{P}_c\sim S_z$) has been observed under excitation by circularly polarized light. The small magnitude of the degree of circular polarization is associated with anisotropic exchange splitting in the case of free excitons,[13,14] and with the char-

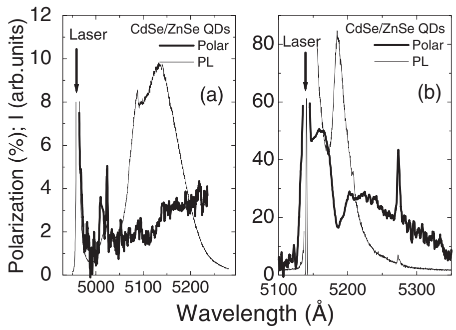

FIG. 1. Luminescence (thin lines) and circular polarization (bold lines) spectra of an ensemble of CdSe/ZnSe quantum dots under excitation by circularly polarized light with wavelength: (a) λ =4965 Å (nonresonant excitation) and (b) λ=5145 Å (quasiresonant excitation).

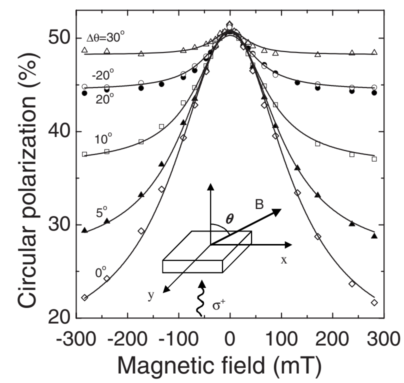

FIG. 2. Dependence of the degree of circular polarization on the magnetic field in different directions of the field. $\Delta\theta=\theta-90°=0°$ corresponds with the Voigt geometry (the magnetic field lies in the plane of the sample). The insertion shows the geometry of the experiment. Solid lines are drawn according to formula (2) with the addition of a constant addendum corresponding to the incomplete polarization at $\Delta\theta=0°$ (see the text).

acter of the energy relaxation of excitations in the case of trions.[19] Figure 1(b) demonstrates a change in the emission spectrum (thin line) under quasiresonant excitation at λ =5145 Å. As seen, the emission line narrows markedly: Not all the dots get excited under quasiresonance conditions but only their "long-wavelength part." The quasiresonant excitation leads to a greater degree of the circular polarization of emission [Fig. 1(b), bold line]. As the anisotropic exchange splitting leads to the almost full loss of the spin polarization, this observation attests to the trion character of emission in the long-wavelength part of the spectrum. The optical excitation of an electron-hole pair in a QD containing a charge carrier generates a trion X^- or X^+. In the ground state of the trion, two similar carriers make up a spin singlet with a total spin $S=0$; as a result, the exchange interaction vanishes,[20] and producing a high degree of spin polarization becomes possible. Under these conditions, the excitation by linearly polarized light does not bring about the optical alignment of excitons (there is no linear polarization of emission), which is a strong argument in favor of trions.[21]

The application of a magnetic field directed under some angle θ to S_z involves the spin precession and the depolarization of emission (Hanle effect). As is known, the precession of the spin S about the magnetic field retains spin projection onto the direction of the field, which results in an incomplete depolarization of emission (if $\theta\neq 90°$).[1,22] Figure 2 shows the curves of the magnetic depolarization of emission for different directions of the magnetic field. As seen from the figure, the greater the deflection of the angle from the value $\theta=90°$ (i.e., the greater $\Delta\theta=\theta-90°$), the narrower the curves and the smaller "the amplitude" of depolarization. Also, it should be noted that the effect does not depend on the sign of $\Delta\theta$. Figure 3 shows the dependence of the polarization of emission in a strong magnetic field [values $\mathcal{P}_c(\infty)$] and of the half-width of the depolarization curve on the angle $\Delta\theta$. Worth noting is the fact that the depolarization of emission is incomplete even in a strictly transverse magnetic field ($\Delta\theta=0°$) (see Figs. 2 and 3).

Hanle effect in a tilted magnetic field. Let the magnetic field B form an angle θ with the z axis, then the z component of the mean spin is described[1,23] by

$$S_z(\Omega)=S_z(0)\left(\frac{\sin^2\theta}{1+\Omega^2T_S^2}+\cos^2\theta\right), \quad (1)$$

where $S_z(0)=S_0T_S/\tau$ is the spin value in the absence of the magnetic field, S_0 is the maximum value of the spin, given by selection rules for interband transitions, $T_S=\tau_S\tau/(\tau_S+\tau)$ is the spin lifetime, determined by lifetime τ and spin-relaxation time τ_S of the trion, $\Omega^2=\Omega_x^2+\Omega_z^2$, $\hbar\Omega=\mu gB$, and $\cos^2\theta=\Omega_z^2/(\Omega_z^2+\Omega_y^2)$. The degree of the circular polarization of emission $\mathcal{P}_c$, measured experimentally, is proportional to the z component of the mean spin: $\mathcal{P}_c\sim S_z$. From formula (1) it follows that the relationship $S_z(B)$ is described by the Lorentz contour: with growing magnetic field, $S_z(B)$ tends to a certain finite value $S_z(\infty)=S_z(0)\cos^2\theta$. The physical sense of this result is simple enough: precession of the spin S_0, oriented along the z axis at the moment of excitation, leads to annihilation of the spin component transverse to the mag-

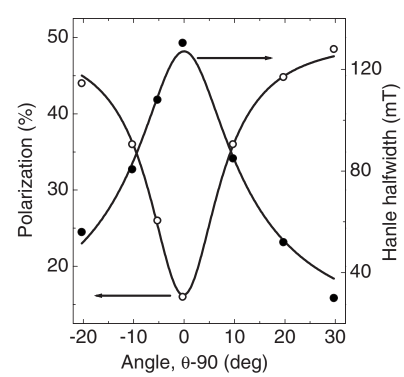

FIG. 3. Dependence of the half-width of the Hanle curve (closed circles) and the degree of PL circular polarization in a strong field [i.e., of $\rho(\infty)$] (open circles) on $\Delta\theta$. The values of $\rho(\infty)$ are obtained via the approximation of the experimental dependencies by the Lorentz contour. Solid lines are calculated dependencies for $B_{1/2}$ and $\rho(\infty)$.

netic field, whereas the component $S_{\parallel}=S_z(0)\cos\theta$, parallel to the magnetic field, is retained. Since the observation is carried on along the z axis, the degree of circular polarization will be proportional to the z component of $S_{\parallel}$: $S_{\parallel}\cos\theta = S_z(0)\cos^2\theta$.

Note that the above consideration is valid at $g_z=g_x=g$ (g_z and g_x are the z and x components of the heavy-hole g tensor). In that isotropic case, the direction of the precession axis (Ω vector) coincides with the direction of the external magnetic field, while the precessional frequency is independent of the θ angle. Experimentally, it manifests itself in that the half-width of depolarization curve does not depend on θ, and the magnitude of $\mathcal{P}_c(\infty)$ is proportional to $\cos^2\theta$. Figure 2 (see also Fig. 3) shows quite another behavior: With the magnetic field deflecting from $\theta=90°$, the magnitude of $\mathcal{P}_c(\infty)$ changes faster than $\cos^2\theta$, and the half-width diminishes markedly (in the investigated range of angles, there occurs a fourfold narrowing of the depolarization curve). It means that the consideration based on the spin precession of a particle with an isotropic g factor is unsatisfactory.

In the case of a particle with an anisotropic g factor, the precession of spin will take place not about the magnetic field $B=(B_x,0,B_z)$, but rather about the vector $\hbar\Omega = (\mu g_x B_x, 0, \mu g_z B_z)$. The polarization of emission in a tilted magnetic field will be described by a formula somewhat different from Eq. (1),

$$S_z(\Omega) = S_z(0)\left(\frac{1}{1+\Omega^2 T_S^2}\frac{g_x^2\sin^2\theta}{g_z^2\cos^2\theta+g_x^2\sin^2\theta} + \frac{g_z^2\cos^2\theta}{g_z^2\cos^2\theta+g_x^2\sin^2\theta}\right). \quad (2)$$

Here, $\hbar^2\Omega^2=(\mu g_x B\sin\theta)^2+(\mu g_z B\cos\theta)^2$. It can be easily seen that now the half-width of the depolarization curve $B_{1/2}$ depends on the angle θ: $B_{1/2}=\hbar/\mu T_S\sqrt{g_x^2\sin^2\theta+g_z^2\cos^2\theta}$. Besides, the dependence of $S_z(\infty)$ on the same angle is more complicated: $S_z(\infty)\sim g_z^2\cos^2\theta/(g_z^2\cos^2\theta+g_x^2\sin^2\theta)$. It is just such behavior that $\mathcal{P}_c(\infty)\sim S_z(\infty)$ and $B_{1/2}$ exhibit in experiments (see Fig. 3). Thus, the experiment reveals unambiguously that here we are dealing with a particle having an anisotropic g factor, $g_z\neq g_x$, and, what is more, the tendency of the angular dependencies involved enables us to infer that $g_z>g_x$. As mentioned above, this corresponds to the case of a heavy hole.

The solid lines in Fig. 2 are drawn in accordance with formula (2), with the addition of a constant term corresponding to an incomplete depolarization at $\theta=90°$. A good agreement with experiment (see Figs. 2 and 3) has been obtained at the following parameters: $g_z=2.5$, $g_x=0.38\pm0.02$, $S_z(0) = S_0T_S/\tau=0.5$, and $T_S=\tau_S\tau/(\tau_S+\tau)=240$ ps. The value $g_z = 2.5$ was obtained for self-organized CdSe/ZnSe QDs in Ref. 17 and was fixed during fitting. The obtained value of the spin lifetime $T_S=\tau_S\tau/(\tau_S+\tau)$ is determined by the least of times τ_S and τ. Direct measurements of the lifetime of trions in CdSe/ZnSe structures carried out in Ref. 6 have shown that $\tau_S\gg\tau$; hence, the spin lifetime is determined by the trion lifetime, $T_S\approx\tau\approx240$ ps. It is noteworthy that the hole g factor g_x along the structure plane has a significant value. Recent magneto-optical experiments on single CdSe/ZnSe QDs have yielded for the transverse g-factor values of 0.2 (Ref. 6) and 0.3 (Ref. 24). The difference between the above transverse g values will be easily understood, taking into account that the appearance itself of a transverse component of the hole g-factor is due to lateral distortions of QDs, which can greatly differ for different samples and even for different QDs in the same sample.

The ground state of a hole in a symmetric QD corresponds to the state of an almost pure heavy hole with $J_z = \pm3/2$. In the first and second orders of perturbation theory, the in-plane magnetic field does not couple the +3/2 and −3/2 states. The corrections to the Luttinger Hamiltonian proportional to J^3 are also small in zinc-blende semiconductors.[25,26] The cause of the appearance of the hole g-factor in the structure plane is, apparently, the low symmetry of the structure. The low symmetry of self-organized QDs (usually C_{2v}) has been corroborated by a great number of experiments.[13,15–17,27] Anisotropy in the structure plane leads to the fact that the states of the angular momentum of a heavy hole are not described any more as pure states $J_z = \pm3/2$. The states of light holes $J_z=\pm1/2$ are admixed to the states of heavy holes so that new wave functions take the form $|\psi_{hh}^{\pm}\rangle=|\pm3/2\rangle+(\gamma^{\pm}/\Delta_{lh})|\mp1/2\rangle$, where $\gamma^{\pm}$ is the parameter of mixing.[28] The magnetic field, in its turn, couples the $|\psi_{hh}^{+}\rangle$ and $|\psi_{hh}^{-}\rangle$ states to the extent of an admixture of light holes. Thus, the admixture of light holes to heavy ones brings about the appearance of the finite value of the heavy-hole g-factor in the structure plane. As is seen, its magnitude is determined by the mixing parameter γ.

Finally, let us discuss the fact that even in a strictly transverse field ($\theta=90°$) depolarization is incomplete, as can be seen from Figs. 2 and 3. The most probable cause of incomplete emission depolarization is, to our opinion, the deflection of the quantization axis z' of QD from the structure growth axis z (for some technological reason). If we assume that there is a scatter of these directions, then the incomplete depolarization will take place for any direction of the magnetic field. Let the angle between z and z' for a given QD be α and the magnetic field be applied along the structure plane. Then, the axis of precession $\Omega\sim(g_{x'}B_{x'},0,g_{z'}B_{z'})$ will not coincide with the external magnetic field. Precession retains the spin projection onto the Ω direction, and this projection gives a residual polarization of emission.

Let us estimate quantitatively what deflection of the QD quantization axis would bring about the above-mentioned effect. When the quantization axis z' deflects from z through an angle α, the precession axis turns out to be inclined to the growth axis by an angle β, which depends on α and on the components of the hole g tensor. The retained projection of the spin S_0 onto the axis of precession equals $S_0\cos\beta$, and the degree of residual polarization $\mathcal{P}_c(\infty)\sim S_z(\infty)=S_0\cos^2\beta$, where $\cos^2\beta$ is determined by

$$\cos^2\beta=\frac{(g_z-g_x)^2\sin^2\alpha\cos^2\alpha}{g_x^2\cos^2\alpha+g_z^2\sin^2\alpha}. \quad (3)$$

In Eq. (3), the primes to indices x and z are omitted for simplicity's sake. Using the values $\mathcal{P}_c(0)=0.5$ and $\mathcal{P}_c(\infty)$

=0.15, we obtain that $\cos^2 \beta = S_z(\infty)/S_z(0) = \mathcal{P}_c(\infty)/\mathcal{P}_c(0) = 0.3$, and the mean value of α must be about 15°.

In conclusion, a high degree of the circular polarization of emission due to spin polarization of holes has been observed in self-organized QDs. It has been found that a decrease in the angle between the magnetic field and the growth axis causes a significant narrowing of the Hanle curve. This bears witness to the anisotropy of the g-factor of a particle (hole) participating in the process. This observation has allowed the emission in the corresponding spectral region to be ascribed to negatively charged trions and the anisotropy of the heavy-hole g-factor to be measured—$g_z/g_x = 6.58 \pm 0.04$. We observed an incomplete magnetic depolarization of trions and interpreted it in terms of deflection of quantization axis from the structure growth axis. Such QD distortion is very important because it limits the attainable value of spin polarization to $\mathcal{P}_c(0) = 0.5$ (via softening of optical selection rules).

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