Spin depolarization of holes and lineshape of the Hanle effect in semiconductor nanostructures

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We report the statistical effect of random in-plane deformations on the magnetic field depolarization of photoluminescence (the Hanle effect) in quasi-two-dimensional semiconductor nanostructures such as quantum wells or disklike quantum dots. Provided that the optical orientation signal from the sample is due to the nonequilibrium spin polarization of holes, the lineshape of the Hanle effect becomes non-Lorentzian. This results from a scatter of hole g-factor values, which is immanent to the hole ensembles in such systems. Analysis of the lineshape of the Hanle effect holds promise as a tool for distinguishing between X^+ and X^- trion states in nanostructures.

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In light of the enhanced interest in studying spin phenomena in low-dimensional semiconductor nanostructures such as quantum wells (QWs) and quantum dots (QDs), including details of spin structure, it is convenient to employ optical spin orientation measurements. For bulk semiconductor crystals, the optical orientation signal usually arises due to the nonequilibrium spin polarization of the conduction-band or donor-bound electrons. In the case of holes there exists a strong correlation between spin and momentum, so that the nonequilibrium spin polarization of the holes dissipates after a few momentum changing collisions.[1] Such depolarization typically occurs on a subpicosecond time scale, while typical hole lifetimes are of the order of hundreds of picoseconds. As a rule, therefore, on average the nonequilibrium spin polarization is nearly absent in an ensemble of holes, so that the holes do not contribute to the optical orientation signal in continuous-wave (cw) experiments.

The situation is, however, quite different in two-dimensional (2D) quantum nanostructures. In that case the motion of the holes is restricted by the confinement potential, and the energy spectrum of the valence band is thus fundamentally changed as compared to that of the bulk crystal. Typically, the lowest-energy valence-band state in such 2D situations is the heavy-hole subband, while the light-hole subband is split off due to the difference in effective masses and to biaxial strain in the 2D layer. The Kramers sublevels of the heavy hole $\pm 3/2$ differ by the value of 3 in the angular-momentum projection. This fact restricts the number of random interactions which can mix these sublevels and initiate the spin flip of the hole. Indeed, while for "warm" holes in a QW spin relaxation remains quite efficient, it slows down noticeably for "cold" holes at the bottom of the band.[2]

The possibilities for spin relaxation are further restricted in QDs, where the motion of holes is limited in all three dimensions and the energy spectrum is discrete. One should bear in mind that in QDs (and in fact also in QWs) the excitonic effects, particularly the electron-hole exchange interaction, become strong, and can mask the pure spin properties of holes of interest in this Brief Report. A convenient way to eliminate the influence of the exchange interaction on spin relaxation of holes (and thus on optical properties) is by investigating the photoluminescence of negatively charged excitons, i.e., X^- trions. The ground state of the X^- trion contains two electrons with opposite spins, so that their mean exchange field on the hole is zero. Examination of the dynamics of the optical orientation signal of the single X^- trion states shows unambiguously that the inhomogeneous spin of the hole indeed does not relax on the time scale of typical X^- radiative lifetimes (hundreds of ps).[3,4] The same conclusion can be drawn from the analysis of cw photoluminescence spectra of single X^- states excited by unpolarized light and subject to a magnetic field. The intensities of Zeeman-split transitions involving hole states with opposite spin orientation are close to each other, although the Zeeman energy exceeds the thermal energy.[5,6] In other words, in the presence of a magnetic field the *equilibrium* spin polarization of the holes cannot be reached within the trion lifetime, thus providing further evidence of long hole spin-relaxation times.

Thus the hole spin relaxation in QDs is a relatively slow process, making it possible to observe the effects of hole spin polarization in cw optical orientation experiments. In a recent paper by Kusrayev *et al.*,[7] an optical polarization signal measured with quasiresonant excitation of CdSe/ZnSe QDs was ascribed to optical orientation of holes in X^- trions. The proof for this interpretation was the observed dependence of the contour width of magnetic-field-induced depolarization of luminescence (the Hanle effect) on the tilt angle of the field between the Faraday and Voigt configurations. In the present Brief Report we continue the investigation of the properties of optical spin orientation of holes. Specifically, we show that the shape of the Hanle effect contour in the Voigt configuration differs from the standard Lorentz shape.

In an optical orientation experiment the sample is excited by circularly polarized light, which creates a nonequilibrium spin polarization of charge carriers (usually electrons). Subsequently, as a result of the competition between the processes of recombination (governed by the lifetime τ) and spin relaxation (governed by spin-relaxation time τ_s), the time-averaged z projection of the mean spin is given by $S_z = S_{z0} T_s/\tau$, where S_{z0} denotes the initial spin, and $T_s^{-1} = \tau^{-1} + \tau_s^{-1}$. The main tool of the cw optical orientation method is

the investigation of the spin depolarization in transverse magnetic field B (the Hanle effect). Precession of the mean spin in the external field results in a decrease in S_z, given by[1]

$$S_z = \frac{T_s}{\tau} S_{z0} \frac{1}{1 + (B/B_{1/2})^2} \equiv \frac{T_s}{\tau} S_{z0} L(b), \tag{1}$$

where $b = B/B_{1/2}$ and $B_{1/2}$ is the half-width of the Lorentz contour of depolarization, $B_{1/2} = \hbar/\mu_B g T_s$, which depends on the g factor of the particles and, again, on the spin lifetime T_s. This allows separate determination of the characteristic times τ and τ_s if the value of g factor is known.[1]

To clearly establish the main idea of the present report, it is useful to recall the g-factor anisotropy of holes in epitaxial semiconductor nanostructures—especially QWs and disklike QDs. In such objects, the lowest-energy subbands (or levels in the case of QDs) correspond to heavy-hole states. The magnetic field applied along the growth axis then leads to the Zeeman splitting of the heavy-hole doublet, while in the transverse magnetic field (i.e., when the field is in the plane of the structure), the energy splitting is much smaller, as follows from theoretical models, and as was observed experimentally.[8] Indeed, for nanostructures with high in-plane symmetry (D_{2d}), e.g., for ideal QWs grown along the [001] crystallographic direction, the transverse g factor of heavy holes is determined by the Luttinger spin parameter q. This parameter accounts for the cubic structure of the semiconductor and is usually small in value, $q \ll 1$.[9]

In practice, however, the transverse g factor of heavy holes in QDs and in QWs is often determined by structural distortions in the plane of the layer.[10–12] Possible sources of such distortions are the anisotropy of the localizing potential (e.g., shape anisotropy of the QDs), fluctuations in local distribution of strain of the atomic bonds (uniaxial in-plane deformations), or a combination of both these factors. Recent studies have shown that the effect of deformations is probably more pronounced in QDs.[13] Whatever the microscopic origin, these distortions result in admixing of light-hole states to heavy-hole states, and this admixing induces a transverse component in the g factor of the heavy hole, its value depending linearly on the value of the local distortion.[14,15]

Thus, while in case of optical orientation of electrons, the Hanle effect reveals the spin depolarization of particles with a well-defined g factor whose value depends on the band-structure parameters of the material, in the case of heavy holes we deal with particles having random g-factor values induced by random local distortions. The distribution of the g-factor values turns out to have the same order of magnitude as the mean value, which is to be taken into account while analyzing the Hanle effect on holes.

Consider the effect of random deformations on hole states (which initially are isotropic in the layer plane). It is natural to choose Gaussian functions for the probability densities of the Cartesian components of the two-dimensional random vector of the deformation. This choice is equivalent to assuming a uniform distribution of the deformation vector over possible directions, an assumption that is reasonable for the CdSe/ZnSe QD ensemble.[16] The required g-factor distribution coincides with that of the *absolute value* of the 2D random deformation vector having the form

$$p(g) = \frac{\pi}{2\bar{g}^2} g \exp\left(-\frac{\pi}{4\bar{g}^2} g^2\right), \tag{2}$$

where $\bar{g}$ is the mean value of the g factor. Using the distribution in Eq. (2) to average $S_z(B)$ given in Eq. (1), we obtain the heavy-hole Hanle effect in the form

$$S_z = \frac{T_s}{\tau} S_{z0} \tilde{b}^{-2} \exp(\tilde{b}^{-2}) \mathrm{Ei}(\tilde{b}^{-2}) \equiv \frac{T_s}{\tau} S_{z0} A(\tilde{b}), \tag{3}$$

where $\tilde{b} = B/B_{\bar{g}}$, $B_{\bar{g}} \equiv \sqrt{\pi}\hbar/2\mu_B \bar{g} T_s$, and $\mathrm{Ei}(x)$ is a standard exponential integral.[17] Similar to Eq. (1), Eq. (3) presents a simple one-parameter family of contours [however, now the parameter is not the g factor itself, as in Eq. (1), but its ensemble mean value]. The difference between the function $A(B)$ in Eq. (3) and the Lorentz function $L(B)$ in Eq. (1) lies in their asymptotic behavior: at $B \to \infty$, the Lorentz curve decays as $L(b) \approx 1/b^2$, while the new function has long "tails," $A(\tilde{b}) \approx \ln \tilde{b}/\tilde{b}^2$.

Aiming at experimental verification of the result obtained above, we have studied the Hanle effect in CdSe/ZnSe QD samples. It had been shown that in sample QD1 (grown on (001)GaAs substrate at 280 °C, nominal thickness of CdSe 2.1 ML, nominally undoped[7]), the resonant optical excitation by the 514.5 nm line of an argon-ion laser results in the optical orientation of negatively charged trions X^- [the corresponding emission and polarization spectra are shown in Fig. 1(b) of Ref. 7]. In this situation the spin-unpaired hole of the trion is responsible for the nonequilibrium spin polarization. Application of an external magnetic field then leads to the Hanle effect due to the depolarization of the holes (Fig. 1). The depolarization curve contains two parts: a bell-shaped contour and a contribution which is field independent in the presented range of fields. A reliable identification of the bell-like signal as belonging to the heavy holes is provided by the dependence of the width of the contour on the tilt angle of the magnetic field to the growth axis (defined as z) of the sample. The linewidth near the Faraday configuration (i.e., for field close to the z direction) is about four times as narrow as that observed in the pure Voigt configuration (field normal to z).[7] This is due to the g-factor anisotropy of the heavy hole, the longitudinal component g_{zz} being noticeably larger than the in-plane components.[8]

The interpretation of the field-independent contribution is more ambiguous. For tilted fields its origin is obvious and well known:[1] in the precession of the spin in an applied magnetic field, the spin component parallel to the field is conserved. In the exact Voigt configuration, however, the field-independent contribution becomes smaller in value, but does not vanish. A possible explanation of the effect is based on the idea that the quantization axis of each QD has several degrees of random inclination from the growth axis of the sample.[7,18] Then at every field orientation, even including the exact Voigt configuration, the situation of "tilted field" is realized for a fraction of the QDs, giving rise to a nonzero optical polarization at strong magnetic fields.

However, one should allow for a possible alternative in-

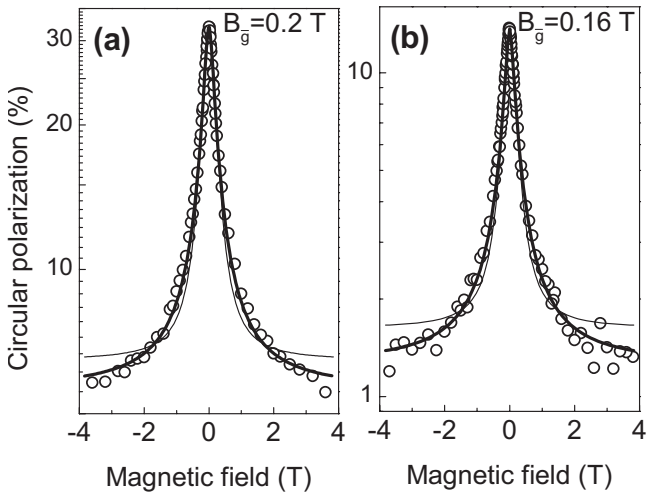

FIG. 1. Suppression of optical orientation by a transverse magnetic field (the Hanle effect) detected at 2.0 K using negative trion (X^-) photoluminescence in CdSe/ZnSe QDs in the Voigt configuration. The experimental data (points) were taken (a) on the "wing" of resonance fluorescence near the laser line and (b) at a distance from the laser line. Solid lines show the results of fitting: thin lines—fitting using Eq. (1); bold lines—using Eq. (3) (see text for details).

terpretation of the field-independent part of the polarization, namely, an optical orientation signal from the excitons. Indeed, the neutral excitons make a finite contribution to the luminescence at the detection energies used in the measurement. This can be concluded from the nonzero "optical alignment" signal, which is observed when the excitation beam is linearly polarized.[16,19] Moreover, due to the isotropic exchange interaction between the electron and the hole of the exciton, one could expect a significant width of the excitonic Hanle effect, which may appear to be field independent in the range of fields used in the experiment. However, this excitonic interpretation seems less probable, for two reasons. First, the excitons in CdSe/ZnSe QDs typically show a strong anisotropic exchange interaction, which is unfavorable for their optical orientation. As was shown in Refs. 16 and 19, the optical orientation of excitons in CdSe/ZnSe QDs is zero (with a 1 to 2% precision), unless it is stabilized by the strong magnetic field along the z axis. Thus the observed values of the field-independent part of the polarization [up to 6%, depending on the detection energy; see Fig. 1(a)] appear to be too large to ascribe this signal to excitons. Second, the value of the field-independent part scales with the amplitude of the bell-shaped part [cf. Figs. 1(a) and 1(b)], which argues in favor of their common (trion-related) origin.

Whatever is the nature of the field-independent part of the polarization, this is of secondary importance in the fitting procedure discussed below. We have fitted the experimental dependences in Fig. 1 by the sum of contour Eq. (3) and an additive constant, using the value of $B_{\bar{g}}$ and of the additive constant as fitting parameters. The agreement with experiment is evidently better than if one used the Lorentz contour Eq. (1) instead of Eq. (3). Assuming that the spin lifetime is nearly equal to the exciton lifetime (a consequence of long hole spin-relaxation time) and taking 240 ps for T_s, as deduced in Ref. 7, we obtained 0.21 for the mean value of the

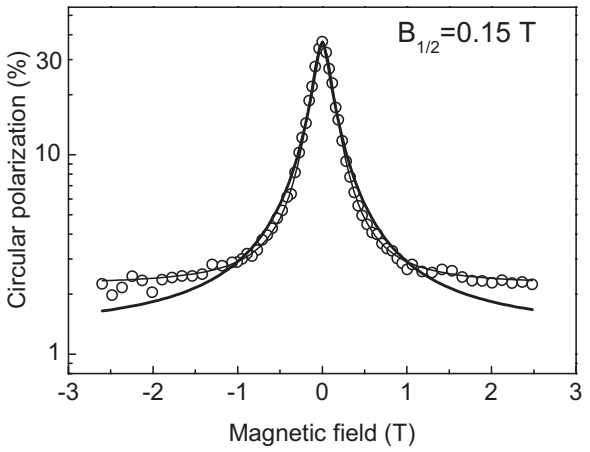

FIG. 2. The Hanle effect measured using positive trion (X^+) luminescence in a CdTe/(Cd,Mg)Te QW. Voigt configuration, temperature 2.0 K. Lines show the fitting results as in Fig. 1.

hole g factor (hole Zeeman splitting $\sim$0.012 meV/T). These estimates are fairly close to the corresponding results reported in Refs. 6 and 11 based on the direct observation of hole Zeeman splitting in single CdSe/ZnSe QDs. Thus the observed result confirms the expected lineshape of the Hanle effect due to the holes. Similar results were obtained on sample QD2 [fully developed undoped CdSe/ZnSe QDs on (001)GaAs substrate, nominal thickness of CdSe 3 ML, fabricated by a different group than QD1, see Ref. 20 for details] using resonant excitation by the 532 nm line of a YAG:Nd laser. This suggests that in sample QD2 the trion Hanle effect is also due to holes, i.e., that the X^- trions dominate. Thus in principle the shape of the depolarization contour in the Voigt configuration allows one to determine the type of the trion state—even without using experiments with tilted magnetic fields. Here one should note that in optical studies of semiconductors the problem of determining the type of the dominant trion state (X^- or X^+) is important, and is not trivial; and that so far only a few experimental solutions have been suggested to do this successfully.[7,21]

As a complementary test we have measured the trion Hanle effect in the structure containing a 40 Å CdTe/(Cd,Mg)Te quantum well [sample QW—nominally undoped, grown on a (001)GaAs substrate]. The reason for this choice was that in a similar specimen, the trion state was identified as X^+ (two holes and one electron),[22] which would provide valuable comparison with the effects of X^- present in samples QD1 and QD2. Unlike the case of CdSe/ZnSe QDs, in sample QW the exciton and the trion states are represented by separate spectrally resolved lines. In spite of significant differences between the objects, the emission spectrum of the QW sample (with resonant excitation of the trion state using a Ti-sapphire laser) is qualitatively similar to the spectra observed on samples QD1 and QD2, exhibiting a highly polarized "wing" of quasiresonant fluorescence near the laser line along with a separate broad peak, the degree of polarization decreasing as the difference between the detection energy and the laser line increases. The results of measurements and calculations of the Hanle effect (Fig. 2) show that for the sample QW fitting with the Lorentz contour Eq. (1) gives

better results than that obtained with using Eq. (3). This is as expected for positive trions, where the optical orientation signal is due to electron spins. We note that for the sample QW, fitting with a nonzero value of the additive constant again provides better agreement with the experiment. However, here the value of the additive constant used in the fitting process amounts about 2% and can be ascribed to experimental imperfections.

In summary, we have shown that the scatter in the value of the heavy-hole in-plane g factor in a statistical ensemble of hole states results in a non-Lorentzian shape of the photoluminescence depolarization curve (the Hanle effect curve). The shape of the contour is described by Eq. (3). This result should apply generally to 2D semiconductor nanostructures, such as quantum wells and disklike quantum dots. In the present Brief Report, the effect was revealed using the luminescence of CdSe/ZnSe quantum dots arising from the negatively charged trion states X^-. The analysis of the lineshape of the Hanle effect developed in this paper holds promise as a tool for distinguishing between X^+ and X^- trion states, and thus for establishing which of these states is dominant in a particular low-dimensional semiconductor system.

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