Longitudinal and transverse fluctuations of magnetization of the excitonic magnetic polaron in a semimagnetic single quantum dot

P. S. Dorozhkin,* A. V. Chernenko,† V. D. Kulakovskii, A. S. Brichkin, and A. A. Maksimov
Institute of Solid State Physics, RAS, 142432 Chernogolovka, Russia

H. Schoemig, G. Bacher, and A. Forchel
Technische Physik, Universitat Wuerzburg, Am Hubland, D-97074 Wuerzburg, Germany

S. Lee
Department of Electronic Materials Engineering, Kwangwoon University, Seoul 139-701, Korea

M. Dobrowolska and J. K. Furdyna
Department of Physics, University of Notre Dame, Notre Dame, Indiana 46556, USA
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Statistical fluctuations of the magnetization on a nanometer scale have been investigated with the use of magneto-optical spectroscopy of quasi-zero-dimensional excitonic magnetic polarons (EMP) in individual pancake-shaped CdSe/ZnMnSe quantum dots (QD's). The EMP emission line demonstrates a qualitatively different behavior in magnetic field B normal and parallel to the QD plane. In the first case, the alignment of the Mn spins in high magnetic fields results in a giant Zeeman shift and linewidth narrowing of almost one order of magnitude to values characteristic of nonmagnetic QD's. No similar effects are observed in the second case where the emission line demonstrates a weak blue shift and small linewidth variation. It is shown that the difference originates from the anisotropy of the hole exchange field in the QD's and can be described using the fundamental fluctuation-dissipation theorem and taking into account the anisotropy of the hole g factor. Both the longitudinal and transverse fluctuations of the magnetization are determined from the analysis of EMP emission lines recorded in the two directions of magnetic field B. We demonstrate that in high magnetic fields the longitudinal fluctuations decrease exponentially whereas the transverse ones decrease as $1/B$.

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I. INTRODUCTION

A variety of new electronic and magnetic properties of semiconductors can be introduced by including magnetic ions in a semiconductor crystal matrix due to the strong *sp-d* exchange interaction between the charge carriers and the magnetic ions. This interaction results in a giant Zeeman splitting of the valence- and conduction-band states, a large Faraday rotation, and the formation of magnetic polarons (MP's) observed in semimagnetic semiconductors [also known as diluted magnetic semiconductors (DMS's)].[1–3] The MP is a small region of crystal with strongly correlated spins of carriers and magnetic ions due to the strong *sp-d* exchange interaction. Optical properties of such MP's have been widely investigated in bulk DMS's (three dimensional, 3D) and DMS quantum wells (2D).[1–9] In QD's, the MP effect is much more pronounced due to the strong 3D confinement of the charge carriers. However to get quantitative access to 0D MP's, one has to avoid inhomogeneous broadening effects caused by the size and/or composition fluctuations, which is possible by experiments on a single QD (SQD).[10–13] Recent magnetooptical studies of individual DMS QD's have revealed a strong influence of thermodynamic fluctuations of the magnetization on the properties of ultrasmall magnetic systems such as QD's.[14,15] This fact imposes a natural limit on functionality and accuracy of possible devices based on controlling the interaction between magnetic ions and carriers spins.

The thermodynamic fluctuations control the width of a photoluminescence (PL) line of an excitonic magnetic polaron from a single DMS QD.[14,15] It was also found that the magnetic field normal to the growth plane (''Faraday geometry'') results in an exponential line narrowing at high magnetic field indicating a strong suppression of magnetic fluctuations.[15] We will show further that in a general case the relation between the linewidth and magnetic fluctuations is not unique. In particular, in EMP's with isotropic g factor of electrons and holes, the transverse (normal to the local magnetization) fluctuations do not contribute in the PL linewidth and thereby the linewidth gives an information only about the longitudinal (parallel to the local magnetization) fluctuations. In structures with an anisotropic carrier g factor, the magnetization does not necessarily follow the magnetic field direction that leads to a well pronounced dependence of the polaron shift on the direction of the magnetic field B and a complicated relation between the linewidth and the magnetic fluctuations.

The effect of the g-factor anisotropy on the magnetic-field dependence of the polaron shift has been investigated in 2D systems in DMS quantum wells (QW's) with a highly anisotropic hole g factor. It was shown that the suppression of the polaron shift in the magnetic field normal to the QW is significantly stronger than in that parallel to the QW plane.[16,17] No analysis of magnetic fluctuations is possible in the QW's because of a relatively strong broadening of QW level caused by fluctuations of its width and composition.

It is obvious that similar anisotropy of the hole g factor will also appear in pancake-shaped QD's whose symmetry is similar to that of quantum wells. Thereby the QD's of such shape are expected to show anisotropy of the magnetic field induced suppression of the polaron shift analogous to that in quantum wells. In addition, that should result in an anisotropy of EMP emission line broadening caused by the fluctuations of the magnetization in the QD.[15] Thus, in order to carry out complete analysis of thermodynamic fluctuations of the magnetization in the QD's one has to use a tilted magnetic field where both longitudinal and transverse fluctuations contribute to the width of EMP emission line.

In this paper we present PL studies on individual pancake-shaped self-assembled QD's in the CdSe/ZnMnSe system in magnetic field **B** parallel and perpendicular to the QD plane. Those measurements allow us to separate contributions from longitudinal and transverse fluctuations of magnetization. PL signal from a DMS QD results from recombination of a localized EMP, i.e., a ferromagnetically aligned spin complex consisting of one exciton and several hundreds Mn^{2+} ions with strongly correlated spins. The investigation of 3D confined EMP in a SQD allows one to completely avoid the effect of inhomogeneous broadening of the PL line due to size fluctuations and spatial inhomogeneity of the Mn content that are characteristic of QW's and multiple QD's. Thereby only two contributions determine the EMP linewidth in the PL spectrum from a single QD in time-integrated PL measurements: fluctuations of the EMP magnetic moment **M** and the transient shift of the PL line due to the EMP energy relaxation.[14,15] Here we study CdSe/ZnMnSe QD's with an EMP lifetime much longer than the EMP formation time that allows us to neglect the contribution from relaxation effects. Thus, these SQD EMP's are ideal objects for exploring magnetic fluctuations.

The paper is organized as follows. In Sec. II we describe our sample and experimental setup. Section III presents the results of measurements of emission spectra on individual pancake DMS QD's in magnetic fields normal and parallel to the QD plane. Finally, in Sec. IV we discuss the anisotropy of the magnetic-field dependence of an EMP ground-state energy and of the fluctuations of magnetization in the QD and their relation to the anisotropy of the hole g factor.

II. EXPERIMENT

The sample under investigation schematically shown in Fig. 1 was grown by molecular beam epitaxy on a (100) GaAs substrate. The QD layer with a nominal CdSe thickness of 2.5 monolayers was grown on a $Zn_{1-x}Mn_xSe$ layer with the Mn content $x=0.25$ and covered with a cap layer of the same content. Typical lateral QD sizes found in similar CdSe/ZnSe and CdMnSe/ZnSe systems by transmission electron microscopy[18,19] and atomic force microscopy,[20] respectively, are about 10 nm. The quantum dots are only a few monolayers in height, thus having a pancakelike form. A strong localization of excitons in our samples is confirmed by a very long, about 580 ps, recombination lifetime determined from time-resolved PL spectroscopy.[21] This time is roughly one order of magnitude longer than that usually found in CdZnSe/ZnSe quantum wells with weak potential fluctuations[22,23] and gives a clear proof of the 3D exciton confinement within a size comparable to the 3D Bohr radius (about 3–4 nm in these materials). In order to obtain PL signal from an individual QD, an opaque aluminum mask was deposited onto the top of the sample. The mask contained small periodically arranged aperture holes down to 100 nm in diameter.[24]

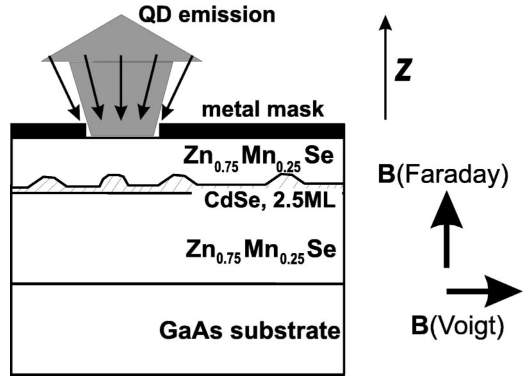

FIG. 1. A sketch of the sample with a metal aperture. Directions of the magnetic field in "Faraday" and "Voigt" geometries are shown by arrows.

PL was excited with ultraviolet lines of Ar^+-ion laser ($\lambda=351$–364 nm). The sample was immersed in superfluid liquid He ($T_{bath}\simeq 1.8$ K) in an optical cryostat with superconducting magnet providing a magnetic field up to 12 T. The PL signal was detected by liquid-nitrogen-cooled charge-coupled device camera. The density of the excitation power was less than 0.1 mW focused on a spot of about 50 μm in order to avoid overheating of the Mn spin system. The EMP formation time in our QD's is $\sim$130 ps, which is more than four times shorter than the EMP lifetime.[21] That allows us to neglect the transient effects in the first approximation.

III. MAGNETIC-FIELD DEPENDENCE OF THE DMS QD EMISSION SPECTRA

Figure 2 displays typical QD PL spectra taken from two different apertures with size as small as 250×250 nm^2 recorded in "Faraday" $\mathbf{B}\|\mathbf{z}$ and "Voigt" $\mathbf{B}\perp\mathbf{z}$ (**z** is the direction perpendicular to the QD plane) geometries in the range of $B=0$–11 T. Low excitation density of 4 W/cm^2 was used in order to ensure that (i) only a single electron-hole pair was generated per QD and (ii) any marked heating of Mn spin system was avoided. At $B=0$ T, the spectra consist of several wide lines on a broad background. Applying a magnetic field normal to the QD plane changes the spectrum drastically. With increasing B the initially wide weakly resolved and unresolved lines exhibit a strong red line shift, become narrower, and finally split into a set of well resolvable lines with full width at half maximum of $\sim$0.5–1 meV. The PL signal is nearly 100% circular polarized already at $B>1.5$ T.

The set of lines and energy separations between different lines changes strongly from one aperture to another. To get a

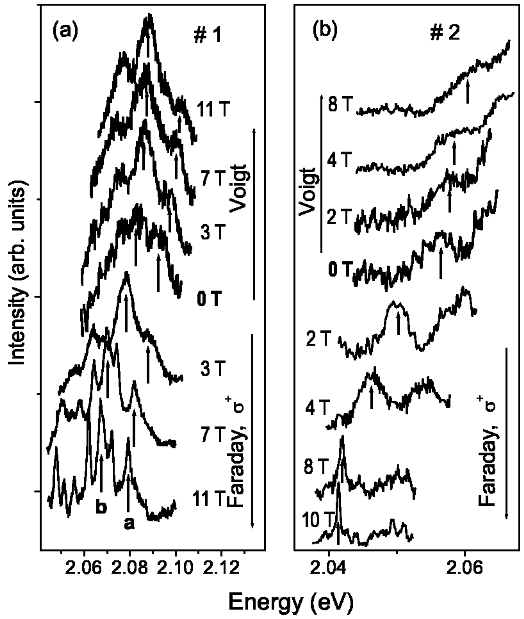

FIG. 2. QD photoluminescence spectra taken from two 250-nm apertures (No. 1 and No. 2) at various magnetic fields in Faraday and Voigt geometries. The arrows point out the PL lines of selected individual quantum dots. [Note, the energy scale is different in (a) and (b).]

spectrum with a relatively small number of separate lines one needs to use aperture with the size smaller than 300 nm. No lines could be resolved with the size larger than 500 nm. Thus, we can ascribe the individual lines observed at high magnetic fields in the Faraday geometry to the emission of excitons localized in different individual QD's. Note that even in high magnetic field $B=11$ T the linewidths are in the range of 0.5–1 meV and are still markedly larger than those in nonmagnetic QD's (less than 0.1 meV).[15,25] The large linewidth of a DMS SQD emission is related to statistical magnetic fluctuations: as the SQD is being probed repeatedly in our experiment, statistical variations of the magnetization within the exciton wave function result in a broadening of the single QD emission peak in time-integrated experiments.[15,26]

The change in the emission linewidth with magnetic field indicates a strong dependence of statistical fluctuations of magnetization in the QD on the magnetic field. The linewidth narrowing in the magnetic field is connected to the suppression of statistical fluctuations. A natural reason for that is the alignment of Mn spins in strong magnetic fields. One could expect a similar strong narrowing of EMP emission line in high magnetic fields in Voigt geometry that also leads to a strong Mn ion spin alignment. However, Fig. 2 shows that the behavior of the EMP emission in Voigt geometry is qualitatively different. In this geometry, the spectra do not reveal any strong changes even in the highest investigated field of 11 T. The lines remain broad demonstrating a small blue shift rather than redshift. They are only weakly linearly polarized even at $B=11$ T with polarization degree less than 0.1. As the Mn spin alignment in high magnetic fields takes place both in the Faraday and Voigt geometries, we conclude that the spin alignment itself is not enough to completely suppress statistical fluctuations of the magnetization.

IV. DISCUSSION

A. Magnetic-field dependence of the EMP ground state

A strong difference in the polaron energy shift in Faraday and Voigt geometries was earlier observed in 2D systems in magneto-optical studies of DMS quantum wells.[16] The difference has been shown to originate from the lowered symmetry of quantum wells and explained in the framework of a model taking into account the anisotropy of the hole g factor.[17] The effect of the latter on the magnetic polaron formation and magnetic fluctuations in the 2D EMP was considered theoretically by Merkulov and Kavokin.[27] The pancake QD's are characterized by the symmetry similar to that of quantum wells and thereby have a similar anisotropy in the hole g factor. Thereby to describe the difference in the polaron shift in a single QD we can follow the theory suggested in this work. The anisotropy of the hole g factor will be also employed in the explanation of the difference in the EMP linewidth behavior in Faraday and Voigt geometries.

The giant Zeeman shift in the Faraday geometry in low-dimensional DMS structures as well as in bulk DMS materials results from the increase of *sp-d* exchange interaction of electrons and holes with Mn ions caused by the Mn spin alignment in magnetic field.[1] The EMP binding energy in a QD is determined by the exchange interaction of an electron and a hole localized in the QD with Mn ions. In bulk diamondlike DMS's the exchange interaction of electrons (e) and holes (h) with magnetic ions is described by the Hamiltonian

$$H_{ex}^{i}=a_i\sum (\mathbf{J}_i\cdot\mathbf{I}_n)\,\delta(\mathbf{r}_i-\mathbf{R}_n), \qquad (1)$$

where $\mathbf{I}_n$ is the spin operator of a magnetic ion with the position vector $\mathbf{R}_n$; a_i, $\mathbf{J}_i$, and $\mathbf{r}_i$ are the electron (hole) exchange integral, spin operator, and position vector, respectively. Electron and hole zone center states are degenerate twofold and fourfold, respectively, and the value of J equals $J_e\equiv s=1/2$ and $J_h=3/2$. In a pancake QD the hole state splits into two twofold degenerate ones due to the lowered symmetry. The hole spin in the lowest energy state is $J_h=3/2$. As this hole state is only twofold degenerate, the holes are usually (for unification with electrons) described as quasiparticles with pseudospin $j_h=1/2$: $\mathbf{J}_h=\hat{g}\mathbf{j}_h$, where $\hat{g}$ is a tensor of the hole g factor. As a consequence, the exchange Hamiltonian of an electron keeps the form given by Eq. (1), whereas that for a hole becomes more complex,

$$H_{ex}^{h}=H_{VV}(\mathbf{I}_1,\ldots,\mathbf{I}_n)-\mu_B g_{\mathrm{Mn}}\mathbf{B}_{ex}^{\mathrm{Mn}}(\mathbf{I}_1,\ldots,\mathbf{I}_n)\cdot\mathbf{j}_h. \qquad (2)$$

Here H_{VV} is the part of the Hamiltonian that depends on the magnetic-ion spin state only (Van-Vleck term). This term leads to an energy shift of both degenerate hole levels. It is usually small and can be omitted in the first approximation.[27] μ_B is the Bohr magneton, $g_{\mathrm{Mn}}=2$ is g factor of Mn^{2+}. The exchange field $\mathbf{B}_{ex}^{\mathrm{Mn}}$ is responsible for the splitting of the hole states and for the rearrangement of the two-component hole wave function. This field plays the main role in EMP formation and its anisotropy leads to a qualitative difference of EMP properties in bulk cubic DMS's and QD's of lower symmetry.

To simplify the problem, for a semiquantitative consideration one can consider an electron (hole) interacting with the total spin $\mathbf{I}=\Sigma_n \mathbf{I}_n$ of all ions inside the electron (hole) orbit. This corresponds to the formal assumption that the electron (hole) wavefunction $\Psi=1/\sqrt{V}$ inside the localization volume V and equal to zero outside it.[2] As long as $I \gg 1$ one can neglect the quantum uncertainty of its components.[27] Finally, let us use the so-called "adiabatic approximation" valid when the binding energy of EMP is substantially higher than both the temperature and all characteristic frequencies in the system of Mn ions.[28] In this approximation the exciton and Mn ion system can be considered separately and it is convenient to introduce an effective exchange field induced by the electron, $\mathbf{B}_{ex}^{e}$ and hole $\mathbf{B}_{ex}^{h}$ acting on Mn spins,

$$\mathbf{B}_{ex}^{e}=\frac{\alpha}{\mu_B g_{\mathrm{Mn}}}\frac{1}{V}\mathbf{s}, \quad \mathbf{B}_{ex}^{h}=\frac{\beta}{3\mu_B g_{\mathrm{Mn}}}\frac{1}{V}\mathbf{J}_h, \tag{3}$$

where $\alpha=a_e$, $\beta=a_h$—their numerical values in ZnMnSe can be found in literature.[29]

In the framework of these approximations spin Hamiltonian of the EMP in the external magnetic field $\mathbf{B}$ can be written as follows:

$$H=g_{\mathrm{Mn}}\mu_B[(\mathbf{B}+\mathbf{B}_{ex}^{e}+\mathbf{B}_{ex}^{h})\cdot\mathbf{I}]=g_{\mathrm{Mn}}\mu_B(\mathbf{B}_\Sigma\cdot\mathbf{I}). \tag{4}$$

Here we have taken into account only the linear in $\mathbf{I}$ terms and omitted the electron-hole magnetic energy $g_e\mu_B(\mathbf{B}\cdot\mathbf{s})+g_h\mu_B(\mathbf{B}\cdot\mathbf{J}_h)$, as it is small $(s,J_h \ll I)$. Equation (4) indicates that the *sp-d* exchange interaction between the 3D confined carriers and the spins of the Mn^{2+} ions leads to a direct dependence of the transition energy of the PL signal from a SQD on the Mn-ion magnetization $\mathbf{M}_{\mathrm{Mn}}$ within the exciton wave function. The EMP transition energy in the QD can be written as

$$\begin{aligned} E_{EMP}(\mathbf{B},T)&=E_0(T)-\mathbf{B}_{\mathrm{ex}}\cdot\mathbf{M}_{\mathrm{Mn}}(\mathbf{B}_\Sigma,T)V_{eff}\\ &=E_0(T)-\mathbf{B}_{\mathrm{ex}}\cdot\mathbf{M}(\mathbf{B}_\Sigma,T). \end{aligned} \tag{5}$$

Here $E_0(T)$ is a field independent part of the QD transition energy, $\mathbf{B}_{ex}=\mathbf{B}_{ex}^{e}+\mathbf{B}_{ex}^{h}$ is the sum exchange field. The effective volume V_{eff}, in which the exciton spin interacts with the spins of the Mn^{2+} ions, is given by $V_{eff}=\gamma V$, where V is the volume occupied by the exciton wave function, and γ (which is less than unity) takes into account the fact that only a part of the exciton wave function overlaps with the Mn^{2+} spins. $\mathbf{M}=\mathbf{M}_{\mathrm{Mn}}V_{eff}=g_{\mathrm{Mn}}\mu_B\mathbf{I}$ is the total magnetic moment of Mn ions within V_{eff}. The Mn-ion magnetization in external and exchange fields depends on the total field $\mathbf{B}_\Sigma$ and is described by the modified Brillouin function:

$$\mathbf{M}_{\mathrm{Mn}}(\mathbf{B}_\Sigma,T)=-xN_0\mu_B g_{\mathrm{Mn}}S_{eff}Br_{5/2}\left(\frac{5\mu_B g_{\mathrm{Mn}}B_\Sigma}{2k_BT_{eff}}\right)\frac{\mathbf{B}_\Sigma}{B_\Sigma}, \tag{6}$$

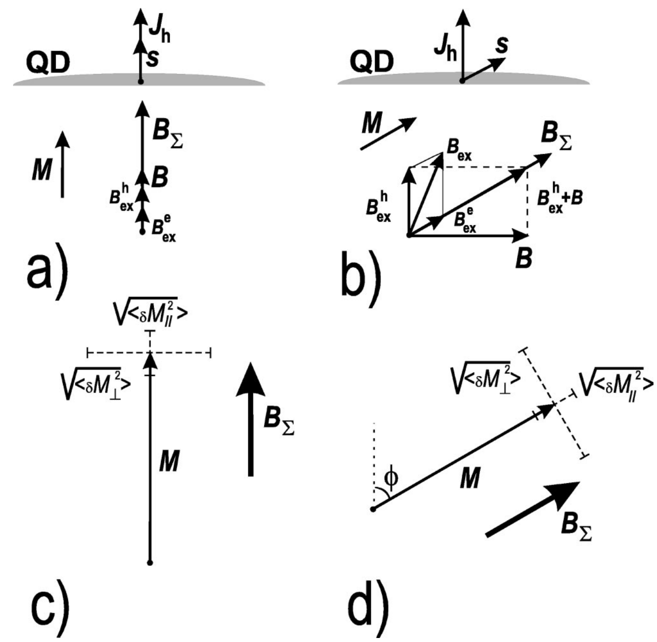

FIG. 3. Electron and hole spins $\mathbf{s}$, $\mathbf{J}_h$, exchange fields $\mathbf{B}_{ex}^{e}$, $\mathbf{B}_{ex}^{h}$, and total field $\mathbf{B}_\Sigma$ in the Faraday (a) and Voigt (b) geometries. Diagrams (c) and (d) explain the meaning of longitudinal and transverse fluctuations of $\mathbf{M}$ in the Faraday and Voigt geometries, respectively.

where N_0 is the number of cations per unit volume. The effective Mn spin $S_{eff}<5/2$, and the effective temperature $T_{eff}=T+T_0$ take into account the antiferromagnetic interaction between neighboring Mn^{2+} spins. k_B is the Boltzmann constant.

Equations (5) and (6) show that the electron and hole exchange fields influence the EMP transition energy not only directly, but also via the magnetization $\mathbf{M}_{\mathrm{Mn}}$ as the Mn ions spin alignment in the EMP is provided by a cooperative action of both the external field and the electron and hole exchange fields. To calculate $\mathbf{M}$ and E_{EMP} one has to determine the direction of $\mathbf{B}_{ex}^{h}$ and $\mathbf{B}_{ex}^{e}$ that is parallel to the hole and electron spins, respectively. The properties of the hole pseudospin and the electron spin in a pancake QD are quite different. The electron spin has no preferable direction in the absence of the magnetic field. In contrast, the properties of the 3/2 hole spin in QD's with the lateral size markedly exceeding that in the normal direction are very similar to those in the quantum wells. In particular, at $B=0$ the hole spin is directed normal to the QD plane. As the hole g factor is highly anisotropic, $g_{zz} \gg g_{xx}$, $g_{yy}=g_\perp$, the hole spin direction coincides with that of an external magnetic field only when the latter is normal to the QD plane, $\mathbf{B}\|\mathbf{z}$. In all other cases the direction of the hole exchange field differs from that of $\mathbf{B}$. In contrast, the electron g factor is isotropic. That means that in the dots with a comparable size of electron and hole wave functions the lowest EMP energy is reached when both the electron spin and Mn spins are directed along the sum of $\mathbf{B}+\mathbf{B}_{ex}^{h}$. In other words, $\mathbf{B}_{ex}^{e}$ always follows the direction of the $\mathbf{B}+\mathbf{B}_{ex}^{h}$. That is illustrated in Figs. 3(a) and 3(b).

The magnitude of the deflection of the hole spin direction from the magnetic field depends on the direction and magnitude of the magnetic field and the ratio g_{xx}/g_{zz}. The magnetic field parallel to the QD plane mixes hole states with $J_h=3/2$ and 1/2 which leads to a superlinear dependence of the exchange field $\mathbf{B}_{ex}^{\mathrm{Mn}}$ on $\mathbf{I}$. In particular, in this case $\mathbf{B}_{ex}^{\mathrm{Mn}}(\mathbf{I})$ can be written as

$$\mathbf{B}_{ex}^{\mathrm{Mn}}(\mathbf{I})=\frac{\beta}{3\mu_B g_{\mathrm{Mn}}}\frac{1}{V}(g_{zz}I_z\mathbf{e}_z+g_\perp I_\perp\mathbf{e}_\perp+G_\perp I_\perp^3\mathbf{e}_\perp), \quad (7)$$

where $g_\perp$ is the in-plane component of the hole g factor, $G_\perp$ is determined by the ratio of characteristic exchange energy and the splitting of the light and heavy hole subbands. If the two last terms are equal to zero, the hole spin is directed normal to the QD plane in any magnetic field. It is natural that no EMP emission line polarization will appear in this case in the magnetic field in the QD plane.[27] That is because there is no preferential hole spin projection J_h^z on z axis.

It is just the last two terms that lead to the splitting of the heavy hole level and the EMP line polarization. As described in the preceding section the linear polarization degree of the EMP emission line in our QD's does not exceed 10% in the highest used magnetic field $\mathbf{B}\perp\mathbf{z}$. That means that we are still far enough from the critical magnetic field where the last two terms in Eq. (7) become comparable to the first one. The estimation of those terms carried out at the end of this section results in the ratio $(g_\perp I_\perp+G_\perp I_\perp^3)/g_{zz}I_z\leqslant 0.1$ at 11 T.

Thereby to simplify the further consideration we have left only the first term in Eq. (7). In this approximation, $\mathbf{B}_{ex}^h$ is always directed along the z axis independently of the magnitude and the direction of the external field $\mathbf{B}$. The electron spin $\mathbf{s}$ and Mn magnetic moment $\mathbf{M}$ tune up along the direction of the sum $\mathbf{B}+\mathbf{B}_{ex}^h$ as shown in Figs. 3(a) and 3(b). In particular, in the Faraday geometry ($\mathbf{B}\|\mathbf{z}$) both exchange fields $\mathbf{B}_{ex}^h$ and $\mathbf{B}_{ex}^e$ are parallel to $\mathbf{z}$. In contrast, in the Voigt geometry ($\mathbf{B}\perp\mathbf{z}$) the hole exchange field $\mathbf{B}_{ex}^h$ is parallel to $\mathbf{z}$ and that of the electron $\mathbf{B}_{ex}^e$ is directed along $(\mathbf{B}+\mathbf{B}_{ex}^h)$. The angle ϕ between $\mathbf{M}$ and $\mathbf{z}$ is determined by the expression

$$\cos\phi=\frac{B_{ex}^h}{\sqrt{B^2+(B_{ex}^h)^2}}.$$

The energy E_{PL} of the detected EMP PL signal is equal to E_{EMP} [Eq. (5)] and can be written as

$$E_{PL}=E_0+E_{mp}\frac{(\beta\cos\phi-\alpha)M(B_\Sigma,T_{eff})}{(\beta-\alpha)M(B_{mp},T_{eff})}, \quad (8)$$

where E_{mp} means the EMP binding energy in zero magnetic field at $T=0$ K; $B_{mp}=|B_{ex}^e|+|B_{ex}^h|$. In the Faraday geometry σ^+ polarized PL signal corresponds to the recombination of the exciton $|J_h^z=+3/2,s^z=-1/2\rangle$. In the Voigt geometry the electron with the spin parallel to $\mathbf{B}_\Sigma$ recombines with the hole whose spin with an equal probability is directed along $\mathbf{z}$ and $-\mathbf{z}$. As a consequence, the PL signal is unpolarized.

Figure 4 shows the results of *simultaneous* fitting of the line shift in two geometries with the use of four adjustable parameters E_{mp}, T_{eff}, B_{mp}, and E_0. The values of the adjustable parameters for two lines are presented in the caption of Fig. 4. The parameters for two lines from the same aperture are slightly different. The same is true for other lines we analyzed. The exchange field B_{mp} was found to be within the range 2.4–3.3 T and $T_{eff}=9.2$–9.6 K. We also found that E_{mp} varies in the range of 14–18 meV, which is in a good agreement with an average value $E_{mp}\approx 17$ meV obtained earlier in time-resolved measurements on the ensemble of CdSe/ZnMnSe QD's.[21]

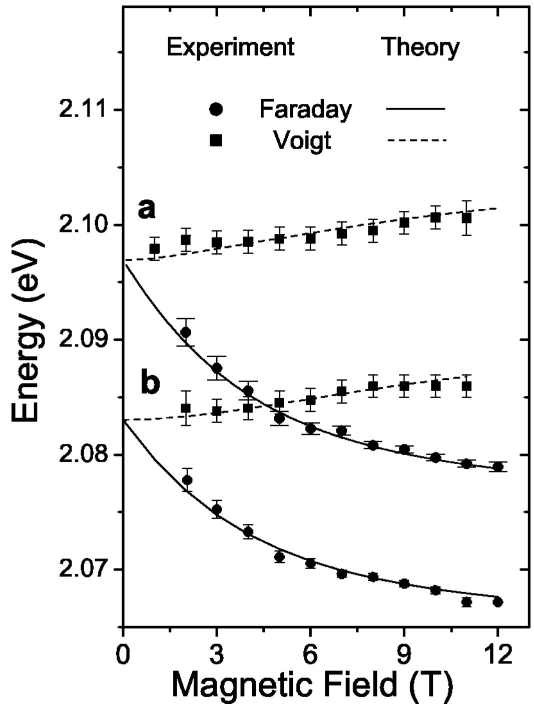

FIG. 4. Magnetic-field dependences of the transition energies of lines a and b from aperture No. 1 in Faraday and Voigt geometries. The solid line shows the results of the fitting with the use of Eq. (8). The best fit for the lines a and b is obtained at $E_{mp}=15.8$ meV, $T_{eff}=9.6$ K, $B_{mp}=2.9$ T and at $E_{mp}=15.0$ meV, $T_{eff}=9.4$ K, $B_{mp}=3.2$ T, respectively

B. Fluctuations of the magnetization in the quantum dot

The PL linewidth of a SQD is not influenced by variations of the Mn content or QD size that are characteristic of a QD ensemble. It is determined only by the magnetic polaron magnetization relaxation and quasiequilibrium fluctuations of its total magnetic moment $\mathbf{M}$. As was mentioned above, time-resolved measurements have shown that the EMP formation time in CdSe/ZnMnSe QD's $\tau_f\approx 130$ ps is sufficiently shorter than the EMP lifetime of $\tau_0\approx 580$ ps.[21] Therefore the influence of the dynamical broadening on the linewidth is negligible.[14] Under these conditions the EMP linewidth is governed only by fluctuations of the EMP magnetic moment $\mathbf{M}$.

The statistical fluctuations of $\mathbf{M}$ lead to a Gaussian shape of the EMP emission line with a full width at half maximum (FWHM)

$$\Delta E = \sqrt{8 \ln 2 \langle \delta E^2 \rangle} = \sqrt{8 \ln 2 \langle \delta (\mathbf{B_{ex}} \cdot \mathbf{M})^2 \rangle},$$

where $\langle \delta E^2 \rangle$ and $\langle \delta(\mathbf{B}_{ex} \cdot \mathbf{M})^2 \rangle$ are the dispersion of E and $\mathbf{B}_{ex} \cdot \mathbf{M}$, respectively. $\langle (\mathbf{B_{ex}} \cdot \mathbf{M})^2 \rangle$ can be presented as a sum of two components, longitudinal and transverse, corresponding to fluctuations of **M** along and normal to **M** respectively: $\langle \delta(\mathbf{B}_{ex} \cdot \mathbf{M})^2 \rangle = B^2_{ex,\parallel} \langle \delta M^2_\parallel \rangle + B^2_{ex,\perp} \langle \delta M^2_\perp \rangle$.

Thus, the equation for FWHM takes the form

$$\Delta E = \{ 8 \ln 2 [\langle \delta M^2_{||} \rangle (B^e_{ex} + B^h_{ex} \cos \phi)^2 + \langle \delta M^2_\perp \rangle \times (B^h_{ex} \sin \phi)^2] \}^{1/2}. \quad (9)$$

According to the fluctuation-dissipation theorem (FDT),[30]

$$\langle \delta M^2_\parallel \rangle = \left. \frac{d M_\parallel(\mathbf{B})}{d B_\parallel} \right|_{\mathbf{B}=\mathbf{B}_\Sigma} k_B T = M'(B_\Sigma) k_B T,$$

$$\langle \delta M^2_\perp \rangle = \left. \frac{d M_\perp(\mathbf{B})}{d B_\perp} \right|_{\mathbf{B}=\mathbf{B}_\Sigma} k_B T = \frac{M(B_\Sigma)}{B_\Sigma} k_B T. \quad (10)$$

$M_{||}(\mathbf{B})$, $B_\parallel$ and $M_\perp(\mathbf{B})$, $B_\perp$ are projections of $\mathbf{M}(\mathbf{B})$ and **B** on the direction of $\mathbf{B}_\Sigma$ and perpendicular to it, respectively.

The meanings of the longitudinal and transverse fluctuations are illustrated in Figs. 3(c) and 3(d). The first ones correspond to the fluctuations of the amplitude of **M**, whereas the latter are due to the fluctuations of its direction. Using the high-temperature limit of the FDT one supposes that the characteristic energy of elementary excitations responsible for the EMP magnetic moment relaxation is smaller than the temperature of the Mn ion spin. Together with the restrictions imposed by the adiabatic approximation it leads to the following limitations of the validity of our model: $E_{mp} > kT \gg \hbar \omega_{\mathrm{Mn}}$, where $\hbar \omega_{\mathrm{Mn}}$ is the energy of any excitation in the system of Mn ions.

According to Eq. (10) longitudinal fluctuations decrease exponentially in high magnetic fields whereas transverse ones decrease as $1/B$. Figure 5 illustrates this difference.

Let us first consider the Faraday geometry. In this geometry $\phi = 0$ and the transverse fluctuations are normal to the exchange magnetic field [cf. Fig. 3(c)]. It follows from Eq. (9) that in this case the transverse fluctuations do not contribute to the linewidth and hence it is completely determined only by the longitudinal fluctuations:

$$\Delta E_{Far}(B) = \sqrt{8 \ln 2 k_B T B_{ex} M'(B_\Sigma)}. \quad (11)$$

ΔE_{Far} can be immediately related to the experimentally determined value of dE_{PL}/dB as it is follows from Eq. (5):[15]

$$\Delta E_{Far}(B) = \sqrt{8 \ln 2 k_B T B_{ex}} \sqrt{- \frac{dE_{PL}}{dB}}. \quad (12)$$

In the Voigt geometry the angle ϕ increases monotonously with external magnetic field. Figure 3(d) shows that in this case both the longitudinal and the transverse fluctuations have a projection onto $\mathbf{B}_{ex}$ and, hence, contribute to the EMP linewidth. Moreover, the relative contribution of the transverse fluctuations increases with magnetic field and becomes dominant at high fields both because of increase of ϕ and decrease of transverse fluctuations. Thus, the investigations of the EMP emission linewidth in the single pancake QD with an anisotropic hole g factor open an unique possibility to measure not only the longitudinal but also the transverse magnetic fluctuations. The observed weak magnetic-field dependence of the EMP linewidth in the Voigt geometry qualitatively confirms the predicted fact that the magnetic field suppresses the longitudinal fluctuations much stronger than transverse ones [Eq. (10)].

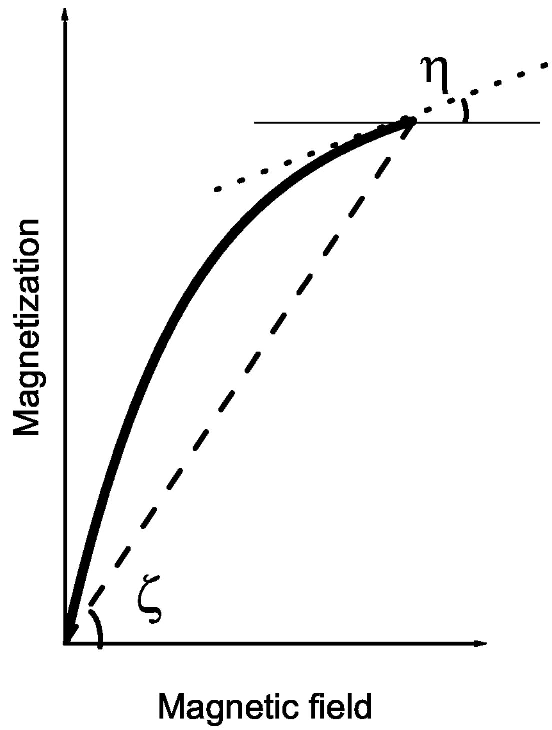

FIG. 5. The plot illustrates the difference in the behavior of longitudinal and transverse fluctuations of **M** in magnetic field. The thick solid line is the magnetization $M(B)$ [Eq. (6)]. η is the angle between the tangent (dotted line) to the magnetization curve and the x axis. $\tan \eta = M'(B) \sim \langle \delta M^2_{||} \rangle$ decreases rapidly in high magnetic fields, whereas $\tan \zeta = M/B \sim \langle \delta M^2_\perp \rangle$ (dashed line) decreases noticeably slower.

The dependences of the EMP linewidth extracted from experimental spectra in the whole range of magnetic fields for two geometries are shown in Fig. 6. It is obvious that single well-resolved lines are required for an accurate determination of their FWHM. Figure 2 shows that such well resolved lines are observed only in the spectra recorded at $B > 4$ T in Faraday geometry (positions of maximums of these lines are observable at lower fields). To extract the width of the lines from the other spectra we have to use the deconvolution of lines from different QD's. In the deconvolution procedure we have fixed the number of lines in the spectrum. This number is well determined from the spectra recorded at high B in the Faraday geometry. In addition, we have chosen for discussion only the lines that are more or less separated from the others (e.g., the lines marked by arrows in Fig. 2). The deconvolution of the line shape of these "separated" lines in the spectrum is rather reliable if their

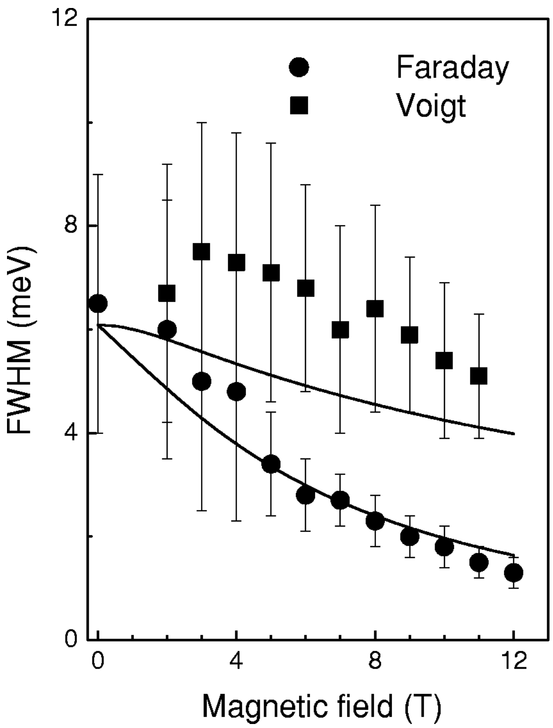

FIG. 6. Magnetic-field dependence of the EMP PL linewidth ΔE (the line a in the aperture No. 1 in Fig. 2) in Faraday and Voigt geometries. The fitting curves (solid lines) are calculated with the use of parameters $E_{mp}=15.8$ meV, $B_{mp}=2.9$, $T_{eff}=9.6$ K, found from the fit of the transition energies. The best fit is found at Mn spin temperature $T=6.2$ K.

FWHM does not exceed the energy separation between the neighboring lines. This is supported by the fact that the extracted FWHM values for such lines weakly depend on the initial parameters. Estimated errors in the FWHM values are shown in Fig. 6. It is natural that they are small only at the high B in the Faraday geometry. In Fig. 6 we have plotted only the points with an estimated error smaller than 30%.

The fitting of the magnetic-field dependences of the EMP line FWHM determined in the Faraday and Voigt geometries is shown in Fig. 6 with solid lines. The theoretical dependences were calculated using Eqs. (9)–(12). Note that we have used in the fitting procedure only one adjustable parameter—the Mn spin temperature T. The values of T_{eff}, B_{mp}, and E_{mp} were taken from the fitting of the EMP transition energy described above. The best fit is obtained for $T\approx 6.2\pm 0.5$ K. Comparison of this value with $T_{eff}=9.6$ K extracted from the fit of transition energies in Fig. 4 leads to the value of $T_0\simeq 3.2$ K which is close to that for bulk ZnMnSe (3.6 K).[31] The simultaneous fit of the magnetic-field dependences of the EMP line FWHM in two geometries provide a rather good description of experimental data in Faraday geometry. In the Voigt geometry, the calculated values are within an experimental error but systematically smaller than experimental ones. However, basing on the presented experimental data we can conclude that in agreement with the theoretical predictions [Eq. (10)] we have found that the magnetic field suppresses strongly only longitudinal fluctuations of **M** in DMS QD's (cf. Ref. 15 as well). Only these fluctuations contribute in the EMP line FWHM at $\mathbf{B}\|\mathbf{z}$. In contrast, the transverse fluctuations decrease much slower. The experimental dependence of the EMP linewidth in the Voigt geometry is in a semiquantitative agreement with the calculated dependence. For a more accurate comparison of the experiment and theory one needs measurements on samples with a smaller amount of QD's in order to separate individual QD lines in the Voigt geometry.

The obtained value of Mn spin temperature $T\simeq 6.2$ K substantially exceeds $T_{bath}\simeq 1.8$ K. The difference is not surprising as the optical excitation of free carriers in DMS's overheats the Mn spin system.[32] Working with a single QD one has to increase the excitation compared to the experiments with quantum wells and bulks. The more the excitation density was used the larger was the Mn spin temperature.

C. Estimation of the in-plane component of the hole g factor

In the previous subsections we used the approximation of an absolutely anisotropic hole g factor for the description of the experimental data. In this subsection we estimate the in-plane component of the hole g factor and show that the above approximation is justified. A magnetic field parallel to the QD plane mixes the heavy hole and light hole states with $J_h=3/2$ and 1/2, respectively, and thus increases an effective in-plane component of a hole g factor. The correction being cubic in B increases with magnetic field [Eq. (7)]. As was mentioned above, this correction leads to the splitting of the $J_h=3/2$ doublet and a deviation of the hole momentum from the z axis. That inevitably leads to an appearance of a linear polarization in the EMP emission.[11] Indeed, when the electron (hole) spin deviates from the z axis by an angle $\varphi_i=\varphi_e$ (φ_h) its wave function can be presented as $\psi_i=|1/2\rangle\cos(\varphi_i/2)+|-1/2\rangle\sin(\varphi_i/2)$. Here $|-1/2\rangle$ and $|1/2\rangle$ are the electron (hole) spin functions with the spin (pseudospin) projection on z equal to $+1/2$ and $-1/2$, respectively. That leads to a linear polarization degree of the emitted light $P=(I_x-I_y)/(I_x+I_y)=-(\sin\varphi_e\sin\varphi_h)/(1+\cos\varphi_e\cos\varphi_h)$ where I_x and I_y are the intensities of the PL signal linearly polarized in x and y directions, respectively.[27] It is seen that P deviates from zero only if $\varphi_h\neq 0$. In a magnetic field $B\gg B_{ex}^h$ the direction of the electron spin is close to the x axis, $\varphi_e\sim\pi/2$, and, hence, $P\sim\sin(\varphi)_h$. By using the value of $P\sim 0.1$ at $B=11$ T found in our measurements we can estimate that $\varphi_h\sim 0.1$ and hence $(g_{xx}=g_\perp+G_\perp I^2)\approx 0.1g_{zz}$. This estimation shows that the approximation of an absolutely anisotropic hole g factor used above is well justified. So small contribution from the mixing of heavy and light hole states in our QD's is due to a large energy splitting of these states because of the very small QD thickness.

V. CONCLUSIONS

In summary, we have studied the Zeeman shift and the linewidth of EMP PL line from individual CdSe/ZnMnSe pancake-shaped QD's in the magnetic field up to 11 T applied perpendicular and parallel to the QD plane. These measurements allowed us to obtain complete information on statistical fluctuations of the EMP magnetization. That turns out to be possible due to a high anisotropy of the hole g factor, which allows one to get information not only on the fluctuations of the magnitude but also of the direction of the magnetization vector. The experimental data are satisfactorily described in the framework of the FDT theorem. The experiment with a magnetic field normal to the QD plane have confirmed that the longitudinal magnetic fluctuations decrease exponentially with magnetic field, whereas the experiments with a magnetic field in the QD plane have shown that the magnetic field weakly influences the transverse fluctuations.

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*Electronic address: pdorozh@issp.ac.ru
†Electronic address: chernen@issp.ac.ru

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