Optical orientation and alignment of excitons in self-assembled CdSe/ZnSe quantum dots: The role of excited states

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We studied the optical orientation of excitons in an ensemble of self-organized CdSe/ZnSe quantum dots (QDs) via photoluminescence (PL) under quasiresonant excitation. The observed PL spectra clearly show the high efficiency of phonon-assisted emission processes. The results demonstrate unambiguously that the QD symmetry is lower than D_{2d}, which results in the splitting of optically active exciton states into two linearly polarized dipoles. The corresponding QD axes have different directions, defined by shape and strain anisotropies. It is shown that the spin-relaxation time of excitons in the ground state noticeably exceeds the exciton lifetime. We have further observed that the degree of optical orientation is small at zero magnetic field, but dramatically increases as the field is increased. The experimental results are interpreted in terms of a cascade model, in which the exciton is initially created in the excited state (which is believed to be a coupled state with phonons) and emits from the ground state after energy relaxation. This model provides an explanation for the experimentally observed conversion of the linear polarization from one set of axes to another. It was shown that the excited states have a significant influence on the polarization of the ground-state emission, and thus it is possible to measure their short characteristic times. We were able to estimate the lifetime τ_2 of the exciton in the excited state to be $\sim$1 ps and the value of the anisotropic exchange-splitting $\Omega_{x,y}$ to be $\sim$0.5 meV.

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I. INTRODUCTION

Investigation of spin phenomena in semiconductors and their nanostructures continues to attract attention in the current literature, prompted both by the fundamental aspects of the role of electron spin in physical processes and by the possibility of making use of the spin of free carriers in electronic devices. Since, in such devices, the electronic (or the excitonic) spin state carries information, it is essential that the orientation of the spin remain intact as long as possible. Recently, it was shown that the electron spin relaxation in bulk GaAs crystals can be as long as 100 ns.[1,2] However, the *hole* spin-relaxation time in bulk semiconductors as well as in semiconductor quantum wells (QWs) is rather short—as a rule, in the range of picoseconds.[3] Furthermore, for excitons in wide band-gap semiconductors, the strong electron-hole exchange interaction results in a rather fast spin relaxation.[4] In quantum dots (QDs), however, the situation can be more favorable to longer spin-relaxation times, since the discrete excitonic spectrum hinders the relaxation process due to the absence of suitable final states (the "bottleneck" effect). Recent experiments have shown that the spin-relaxation (spin-coherence) times for charge carriers and excitons in InAs/GaAs QDs are much longer than their respective lifetimes.[5]

Detailed knowledge of the spin properties of semiconductors and semiconductor nanostructures is a necessary prerequisite for the study of spin-based effects on these structures. A great deal of such data has already been obtained by means of optical spin orientation and optical alignment techniques.[6] Optical orientation of excitons was described in Ref. 7. Optical alignment of excitons under excitation by the linearly polarized light was theoretically predicted in Ref. 8, and was subsequently discovered in bulk CdSe crystals.[9] Excitation of a crystal (as well as a QW or a QD) by circularly polarized light creates excitons with the preferential orientation of spins, while linearly polarized light produces excitons with the definite direction of the oscillating dipole moment. The quantum state of an exciton with a definite direction of the dipole moment is the coherent superposition of states characterized by the projections $m=\pm 1$ of the angular momentum on the direction of the wave vector of the exciting light.

An in-plane deformation of the nanostructure (as well as any other asymmetry of the localizing potential) will lead to a splitting of the $m=\pm 1$ states into two levels, E_X and E_Y, which are dipole active in the two perpendicular directions x and y. Experiments using polarized excitation thus provide possibilities for studying the parameters of the ground and excited excitonic states, as well as details of the interaction of carriers or excitons with phonons.

In the present paper, we investigate the details of optical orientation and optical alignment of excitons in an ensemble of self-organized CdSe/ZnSe quantum dots under quasiresonant excitation. The experimental results are interpreted in terms of a model, which assumes an anisotropy of the QDs in the (001) layer plane of the structure, in such a way that

some of the QDs are elongated along ⟨110⟩ directions, while other QDs are elongated along ⟨100⟩ directions. We have also studied the behavior of optical alignment of the two QD families in the presence of an external magnetic field. It will be shown that the excited states influence the polarization of the ground-state emission in a fundamental way. The experimental results also show that the spin-relaxation times of the states involved in the emission process noticeably exceed the lifetime of excitons.

II. EXPERIMENTAL PROCEDURE AND RESULTS

The structure under study was grown by molecular-beam epitaxy on a (001)-GaAs semi-insulating substrate. The GaAs surface had been first covered by a 1-μm ZnSe buffer layer. The QD ensemble itself was then formed by depositing CdSe to a nominal thickness of three monolayers (MLs). The growth was completed by depositing a ZnSe cap layer of 50 nm. The deposition of the CdSe layer was preceded and followed by a 2-s growth interruption. The described growth procedure corresponds to fully developed quantum dots.[10] The PL measurements were carried out at sample temperature $T=2.0$ K. The PL was excited either by an argon ion laser (lines 5145 Å and 4880 Å) or by a second harmonic of a Nd:yttrium aluminum garnet (YAG) laser (5320 Å). The pump intensity did not exceed 1 W/cm^2. The direction of incidence of the exciting beam was close to normal to the layer plane (i.e., along the growth axis z), and the PL signal was collected along the backward direction. The linear or circular polarizations of the exciting light were achieved using either a linear polarizer or a linear polarizer combined with a quarter-wave plate, respectively. The magnetic field of up to 6 T was applied along the growth axis using a superconducting solenoid.

The linear polarization of the emitted PL can be completely described by the relations

$$P_L = \frac{I_x - I_y}{I_x + I_y}, \quad P_{L'} = \frac{I_{x'} - I_{y'}}{I_{x'} + I_{y'}},$$

where I_α imply the intensities of the radiation polarized along the axis $\alpha = x, y, x', y'$. Here the x axis coincides with the exciting light polarization, defined by the unit vector e. The (x', y') reference frame is rotated with respect to the (x, y) frame by 45° around the z axis. Definition of the degree of circular polarization is analogous, $P_C = (I_+ - I_-)/(I_+ + I_-)$, where I_+, I_- are the two circularly polarized components of radiation.

Figure 1(a) presents the PL spectrum emitted from the QDs, together with the wavelength dependence of the degree of circular polarization P_C (optical orientation). The data in Fig. 1(a) was obtained using circularly polarized excitation at λ=5320 Å (2.3305 eV). The intensity spectrum shows several "shoulders" on the short-wavelength side of the inhomogeneously broadened band.[11] This structure is also observed for excitation at λ=5145 Å (2.4097 eV). However, for excitation by λ=4880 Å (2.5406 eV), i.e., at an energy noticeably higher than the exciton ground state of the QDs, the shoulders disappear. In the optical orientation (or optical alignment) spectrum, the structure becomes much more distinct [see Figs. 1(a)–1(c)]. The polarization spectra show maxima on the Stokes side at distances of 2, 3, 4, and 5 optical phonons from the laser frequency. This result demonstrates the efficiency of energy relaxation of the excited states by means of LO phonons. Having emitted an integer number of optical phonons, the excitons radiate from the QD ground-state energies.[12] The degree of polarization (both linear and circular) is seen to monotonically decrease as the number of emitted phonons increases. The degree of circular polarization is vanishingly small at zero magnetic field [Fig. 1(a), full circles], but increases noticeably when the magnetic field is applied [Fig. 1(a), open circles]. Note that the circular polarization changes sign at the long-wavelength side of the luminescence band, which may be a consequence of impurity bound or charged excitons (trions) participating in the recombination.[13]

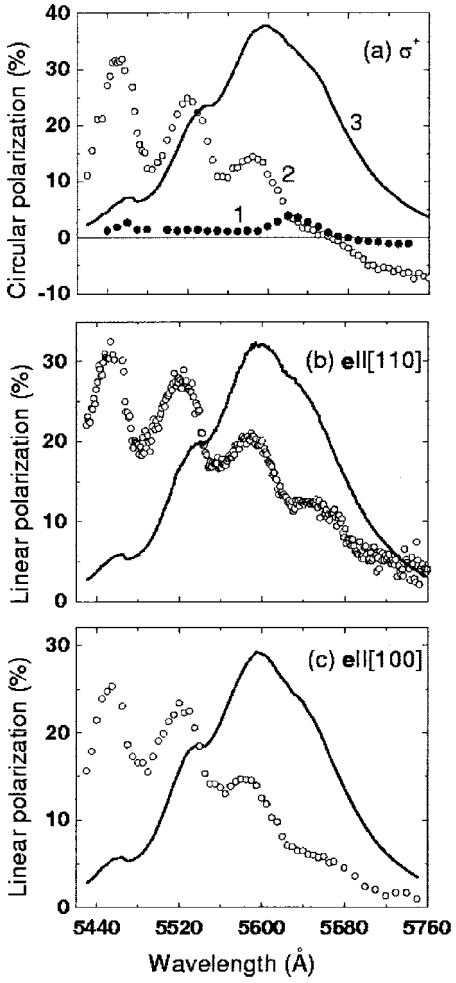

FIG. 1. Total PL intensity (solid lines) and the degree of polarization of the PL emission (circles) observed for the CdSe/ZnSe QDs. The exciting light was at a wavelength λ=5320 Å (2.3305 eV). Circularly polarized PL observed under circularly polarized excitation (optical orientation) at $B=0$ (closed circles) and at $B=4$ T (open circles) is shown in panel (a). Panels (b) and (c) show linearly polarized PL observed under linearly polarized excitation (optical alignment) for $e\|[110]$ and $e\|[100]$, respectively.

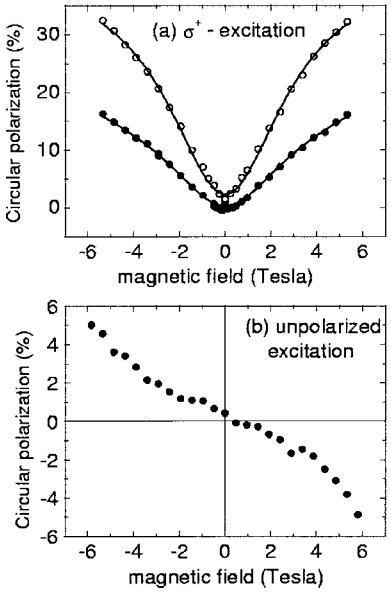

FIG. 2. (a) Dependence of the degree of circular polarization of the PL emission on a magnetic field under circularly polarized excitation. Open circles correspond to data detected at $\lambda=5455$ Å; full circles to detection at $\lambda=5600$ Å. (b) The same as (a) but observed with unpolarized excitation, with detection at $\lambda=5600$ Å.

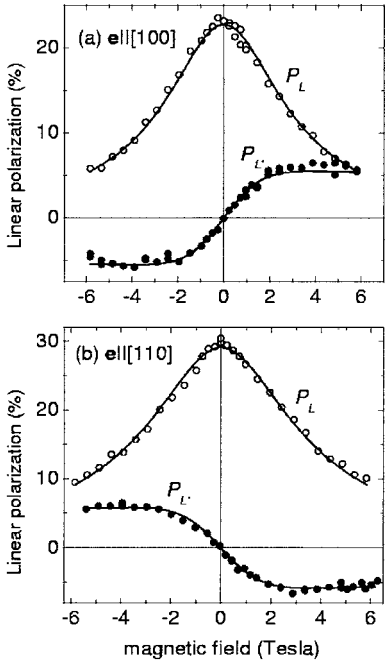

FIG. 3. Dependence of the degree of linear polarization of the PL emission on a magnetic field. Data for the (x,y) frame (where the x axis coincides with the direction defined by the unit vector e) are shown by open circles. Data for the (x',y') reference frame [rotated with respect to the (x,y) frame by 45° around the z axis] are given by full circles. Panel (a) of the figure corresponds to excitation with $e\|[100]$; panel (b) corresponds to excitation with $e\|[110]$.

Figure 2(a) shows the dependence of the degree of circular polarization of the PL on the magnetic field when the sample is excited by a circularly polarized laser beam (clearly demonstrating the "restoration of the optical orientation" by the applied field[14]). The magnetic field in which the degree of polarization reaches one half of its saturation value (see the following formula), which we will define as the characteristic scale of the field, is about 3 T. At higher fields, a tendency of the polarization P_C to saturate is observed, with maximum P_C reaching a value of only 35%.

One should note that in a magnetic field, circularly polarized PL is observed even when the excitation is unpolarized [see Fig. 2(b)]. This effect is caused by thermal redistribution of the excitons over the available Zeeman sublevels. One can see that, while the degree of circular polarization increases with magnetic field for linearly polarized excitation, its value is only a few percent—much smaller than expected for the conditions of thermal equilibrium (see the following discussion).

The phenomenon of optical alignment is observed for any direction of the exciting light polarization in the plane of the structure, the degree of linear polarization being nearly independent of the angle (isotropic optical-alignment effect). Figures 1 and 3 demonstrate only two of the possible situations—$e\|[110]$ and $e\|[100]$. When a magnetic field is applied, the degree of linear polarization of excitons seen in Fig. 3 is being suppressed in the same characteristic field range in which the increase in P_C is observed to be most rapid [see Fig. 2]. The value of the characteristic field varies a little from the case $e\|[110]$ to the case $e\|[100]$. While the PL is depolarized by the magnetic field [$P_L(B)$ decreases], this is accompanied by a conversion of linear polarization from the orientation along the $\langle 110\rangle$ to $\langle 100\rangle$ axes and vice versa, i.e., when magnetic field is increased, there arises a linear polarization $P_{L'}(B)$ in the reference frame rotated by 45° to the direction of e.

III. DISCUSSION

A. Circular PL polarization observed for unpolarized and for polarized excitations

For unpolarized excitation, the circular polarization of the PL emission usually arises due to the redistribution of excitons over the Zeeman sublevels corresponding to the spin projections $m=\pm 1$. At thermal equilibrium, the degree of circular polarization is determined by the expression $P_c^{eq}=\tanh(\mu g_{ex}B/2kT)$, where μ is the Bohr magneton and g_{ex} is the g factor of the exciton. At $B=6$ tesla, $T=2$ K, and for $g_{ex}=1.6$,[15] the expected value of polarization $P_c^{eq}\approx 0.92(92\%)$, whereas the value experimentally measured is $P_c^{exp}=0.04(4\%)$, 23 times smaller than the expected value. This indicates that the thermal equilibrium between the spin sublevels is not reached within the exciton lifetime τ. In this case, the degree of polarization will be lower by the factor $(\tau+\tau_S)/\tau$, so the overall degree of circular polarization becomes $P_C=[\tau/(\tau+\tau_S)]\tanh(\mu g_{ex}B/2kT)$.[6] This gives $\tau_S/\tau\approx 22$, i.e., the spin-relaxation time is 22 times longer than the exciton-recombination lifetime. The preceding result agrees qualitatively with direct measurements of τ and τ_S in InAs QDs by means of time-resolved PL.[5]

The relation between τ and τ_S established here should also result in large values (nearly 100%) of optical orientation signal, since the nonequilibrium spin of the exciton does not have time to relax within the exciton-recombination lifetime ($\tau_S \gg \tau$). The experiment shows, however, that at zero magnetic field the degree of circular polarization of exciton emission under circularly polarized excitation does not exceed a few percent [see Fig. 1(a), full circles]. The reason for this discrepancy is that the shape asymmetry of the QDs leads to a splitting of the optically active degenerate exciton doublet $m=\pm 1$ (corresponding to $X \pm iY$ polarizations) into two linearly polarized dipoles X and Y, whose orientation is specified by the distortion of the respective QDs.[16–18] Circularly polarized light excites the coherent superposition of X and Y eigenstates, namely, the exciton with nonzero spin projection along the z axis. The time evolution of X and Y results in quantum beats between the eigenstates, which is equivalent to the precession of exciton spin about the exchange field. If the exciton lifetime is longer than the period of the quantum beats $\tau_b = 2\pi\hbar/\delta$ (where δ stands for the "anisotropic" exchange splitting between the states X and Y), then the steady-state PL polarization will be small, as is indeed observed.[19] Then, as a magnetic field is applied along the growth direction, the linear dipoles X and Y will be gradually turned toward the states $X \pm iY$ with an angular momentum projection $m=\pm 1$ on the z axis, resulting in the increase of the degree of circular polarization P_C, as seen in Fig. 2(a)[20] (restoration of optical orientation.[14]). The characteristic magnetic field, which produces the restoration of the optical orientation, will depend on the value of the doublet splitting by the relation $B_0 = \delta/\mu g_{ex}$. It should be noted that in case of a charged exciton complex X^+ or X^- (i.e., a trion involving either two holes or two electrons with antiparallel spins, $S_h=0$ or $S_e=0$, respectively), the sublevels are *not split* at $B=0$, and optical orientation can then be observed even at zero magnetic field.[21]

B. Linear PL polarization observed in the Faraday geometry ($\mathbf{k} \| \mathbf{B} \| \mathbf{z}$)

When the magnetic field is applied in the Faraday geometry ($\mathbf{k} \| \mathbf{B} \| \mathbf{z}$, where $\mathbf{k}$ is the wave vector of incident light), the behavior of the degree of linear polarization of the excitonic PL emission depends on the ratio between the inverse exciton lifetime τ^{-1} and the anisotropic exchange splitting δ. If $\delta \ll \hbar/\tau$, the depolarization of the emitted radiation is governed by the precession of the excitonic dipole moment around the magnetic field at a frequency $\Omega_\| = \mu g_{ex} B/\hbar$, and the dependence of P_L on B is described by a Lorentzian line with half width at half maximum (HWHM) $B_{1/2} = \hbar/\mu g_{ex} T_L$, where $T_L^{-1} = \tau_L^{-1} + \tau^{-1}$, and τ_L is the time in which the optical alignment of the exciton is depolarized.[6] The precession of the exciton dipole moment around the magnetic field defines in turn the polarization plane given by the average angle $\varphi = \frac{1}{2}\arctan(\Omega_\| T_L)$, which is consistent with the increase of the linear polarization $P_{L'}(B)$ in the reference frame rotated by 45° with respect to the direction of e. If $\delta \gg \hbar/\tau$ and $\tau_L \gg \tau$, then the dependence $P_L(B)$ is determined by the precession of the exciton dipole moment in the effective magnetic field $B_{eff} = \sqrt{(\delta/\mu g_{ex})^2 + B^2}$ at a frequency $\Omega = \sqrt{(\delta/\hbar)^2 + \Omega_\|^2}$, and the HWHM of the corresponding Lorentz contour $B_{1/2} = \delta/\mu g_{ex}$ yields the value of the anisotropic exchange splitting. As for the linear polarization in the primed axes, it does not appear, $[P_{L'}(B)=0]$.[19] Physically, the decrease of the linear polarization $P_L(B)$ in the magnetic field (for the case $\delta \gg \hbar/\tau$) occurs because of the field-induced reconstruction of the X and Y dipoles into the pure states $m=\pm 1$, similar to the mechanism of restoration of optical orientation (see the preceding subsection).

The absence of the optical orientation of excitons at $B=0$, together with its restoration when a magnetic field is applied [see Fig. 2(a)] indicates that it is the latter inequality, $\delta \gg \hbar/\tau$, which characterizes the ground-state excitonic transitions. Indeed, the absence of optical orientation at $B=0$ implies that either $\tau_S \ll \tau$ or $\delta \gg \hbar/\tau$. We have already shown, however, that $\tau_S \gg \tau$ in the system under study. It therefore necessarily follows that $\delta \gg \hbar/\tau$. This last conclusion agrees with recent time-resolved experiments,[12] which revealed that the degree of linear excitonic polarization in CdSe/ZnSe QDs does not change in the time scale of the exciton lifetime. The fact that $\delta \gg \hbar/\tau$ in the structure under study automatically suggests that no linear polarization should be expected in the primed reference frame. This, however, is in clear contradiction to the experiment, as seen in Fig. 3. This contradiction can be resolved via the cascade model, as discussed in Sec. III C.

Up until now, we have assumed that all the QDs are elongated in a single direction, e.g., along [110]. This means that the eigenstates of the exciton are linear dipoles X and Y, polarized along [110] and $[1\bar{1}0]$, respectively. But such a model cannot account either for the observed isotropic optical alignment effect or for the magnetic-field-induced conversion of linear polarization from one set of axes to another. It is intuitively clear (as we will show later) that these results can be interpreted by assuming the nearly isotropic distribution of QD elongation. However, the isotropic distribution of QD elongation is not the only possibility to explain these results. Alternatively, we can assume the existence of two equal groups of QDs in the ensemble, one group having their main anisotropy axes along [110] and/or $[1\bar{1}0]$, and the other along [100] and/or [010]. As an illustration, we have chosen the $\langle 110 \rangle$ and $\langle 100 \rangle$ orientations for our analysis. Indeed, this choice suggests itself as physically plausible, because these directions are structurally distinct, and it is natural that the growth process may distinguish between them more easily than between others, favoring one or the other as the dots are forming.

C. The cascade model

To allow for the appearance of the linear polarization of excitons in the primed axes $P_{L'}$, one must assume that the process has a cascade nature. It is clear that at nonresonant (or quasiresonant) excitation, the exciton reaches the ground (radiative) state from some higher-lying state. Clearly, the magnetic field will affect the excitonic spin not only in the ground state (from which the recombination takes place), but in the higher-lying state as well. As we already argued, the

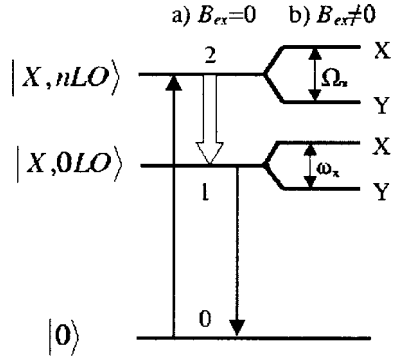

FIG. 4. A schematic picture of cascade excitation of excitons through a coupled state with optical phonons. (a) Corresponds to the case without anisotropic exchange interaction, $B_{ex}=0$; (b) corresponds to $B_{ex}\neq 0$.

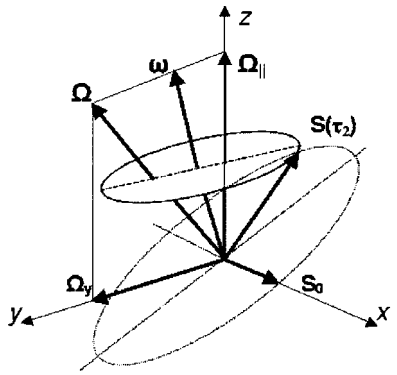

FIG. 5. Schematic representation of exciton pseudospin precession in the cascade process for QDs of the $\langle 100\rangle$ type ($\Omega_y\neq 0$). S_0 is the initial pseudospin created under [110] linearly polarized excitation, and Ω and ω are the Larmor precession frequencies at the first and second stage of the cascade process, respectively. During the first stage of the cascade, S_0 turns by a mean angle $\varphi=\Omega\tau<2\pi$. During the second stage ($\omega\tau_1\gg 1$), the pseudospin lies along the direction of ω, so it has projections S_y and S_z. For QDs of the $\langle 110\rangle$ type ($\Omega_x\neq 0$), the components S_x and S_z survive. Note that if $\Omega\tau_2\gg 1$, the precession of $\mathbf{S_0}$ around $\mathbf{\Omega}$ results in the destruction of the pseudospin, since $\mathbf{S_0}\perp\mathbf{\Omega}$.

excitonic dipole moments will precess about the magnetic field, with the mean value of the dipole moment over the ensemble depending on the ratio between $\hbar/\tau$ and δ. As those excitons with *rotated* dipole moments reach their ground state, their subsequent evolution can result in the appearance of PL emission a finite linear polarization along the primed axes. (Note, however, that in order for this to happen, it is essential that the rotation of the exciton dipole moment in the ground state starts from a direction other than vector e).

Next, we analyze the experimental results using a three-level model, in which the exciton is initially excited to state 2 from the vacuum level—state 0 (the state representing the crystal without an exciton)—and then falls to the emitting ground state 1 (see Fig. 4). A similar cascade model has been used in Ref. 22 to explain the results on the optical orientation of excitons in type-II GaAs/AlAs superlattices. Under the conditions of our present experiments, state 2 may correspond to a coupled state of the exciton with one or several optical phonons $|X,nLO\rangle$. The anharmonic decay of the optical phonons then promotes the exciton to the ground state $|X,0\rangle$ (state 1, as shown in Fig. 4),[23,24] from which it recombines and can be detected.

The lifetime of the exciton in level 2 is much shorter than in level 1, owing to the rapid decay of the LO phonons.[25] Therefore, in contrast to level 1, the inequality $\delta\gg\hbar/\tau$ may remain unfulfilled for level 2. In what follows, we do not impose any limitations over the ratio between δ and $\hbar/\tau$ for level 2. Instead, we proceed based on the assumption that the optical orientation and alignment of excitons on level 1 are both determined primarily by the value of δ, while on level 2 both parameters (δ *and* $\hbar/\tau$) influence the polarization of the exciton emission as a function of the magnetic field.

D. The pseudospin model

The polarization properties of excitons are most conveniently treated by a model using the pseudospin $S=1/2$.[22] In accordance with this model, the excitonic states $|+1\rangle$ and $|-1\rangle$ correspond to the pseudospin projections $S_z=+\frac{1}{2}$ and $S_z=-\frac{1}{2}$ on the z axis. The linearly polarized dipoles $|X\rangle=(|+1\rangle+|-1\rangle)/\sqrt{2}$ and $|Y\rangle=(|+1\rangle-|-1\rangle)/\sqrt{2}i$ are described by the projections $S_x=+\frac{1}{2}$ and $S_x=-\frac{1}{2}$, while the states $|X'\rangle=(|X\rangle+|Y\rangle)/\sqrt{2}$ and $|Y'\rangle=(-|X\rangle+|Y\rangle)/\sqrt{2}$ polarized along the axes x' and y' correspond to the projections $S_y=+\frac{1}{2}$ and $S_y=-\frac{1}{2}$, respectively. It can be readily shown that the relationships of the components of $\mathbf{S}$ with the polarization characteristics of the excitonic emission have the form $P_c=2S_z$, $P_L=-2S_x$, and $P_{L'}=2S_y$. For excitation of excitons by the $e\|[110]$ linearly polarized light, the initial value of the pseudospin $\mathbf{S}$ (at $B=0$) equals $\mathbf{S}_0=(S_{0x},0,0)$. In a magnetic field, the vector $\mathbf{S}$ precesses around the vector $\boldsymbol{\omega}$ [defined by $\boldsymbol{\omega}=(\omega_x,0,\Omega_\parallel)$] at the effective Larmor frequency $\omega=\sqrt{(\delta/\hbar)^2+\Omega_\parallel^2}$, where $\delta=\hbar\omega_x$ is the anisotropic exchange splitting. Assuming that $\omega\tau\gg 1$ (i.e., that the exciton pseudospin completes many cycles around $\boldsymbol{\omega}$ before recombination), one can find the components of $\mathbf{S}$ from its projections on the direction of $\boldsymbol{\omega}$. Thus,

$$S_x=S_{0x}\cos^2\varphi=S_{0x}\frac{\omega_x^2}{\omega_x^2+\Omega_\parallel^2},\quad S_y=0,$$

$$S_z=S_{0x}\cos\varphi\sin\varphi=S_{0x}\frac{\omega_x\Omega}{\omega_x^2+\Omega_\parallel^2},\qquad(1)$$

where φ is the angle between ω and the x direction. If the condition $\omega\tau\gg 1$ is not satisfied (say, $\omega\tau\approx 1$), then within its lifetime the exciton has time to precess only by some angle $\varphi=\omega\tau<2\pi$. Obviously, in this case all components of $\mathbf{S}$ have nonzero values.

The preceding model allows an intuitive visualization of the evolution of the excitonic pseudospin in a magnetic field. Figure 5 illustrates the motion of the pseudospin subject to an external magnetic field and the "exchange field" $B_y=\hbar\omega_y/\mu g_{ex}$. One can see that the absorption of linearly polarized light $e\|[110]$ by $\langle 100\rangle$ QDs results in the creation of

an exciton on the level 2 (see Fig. 4), having its initial pseudospin S_0 directed along the x axis. Because of the Larmor precession in the plane perpendicular to vector $\mathbf{\Omega}=(0,\Omega_y,\Omega_\parallel)$(see Fig. 5; note that Ω_y is the anisotropic exchange splitting of level 2), at $\Omega\tau\leqslant 1$, the spin $\mathbf{S}_0\perp\mathbf{\Omega}$ will deviate from the x axis by some angle $\varphi<2\pi$. At time $t\cong\tau_2$ (τ_2 being the lifetime of level 2), the precessive motion of the excitonic spin on level 2 is interrupted by the transition to level 1, and any further evolution of the pseudospin $\mathbf{S}(\tau_2)$ will then occur in the ground state. If the exchange splitting at level 1 differs from that at level 2, the pseudospin will now precess about a different direction, i.e., about the vector $\boldsymbol{\omega}=(0,\omega_y,\Omega_\parallel)$, where ω_y stands for the value of the anisotropic exchange splitting at level 1. Since at level 1 the condition $\omega\tau_1\gg 1$ is well satisfied (where τ_1 is the exciton lifetime on level 1), the resultant pseudospin is obtained by projecting $\mathbf{S}(\tau_2)$ onto the direction of $\boldsymbol{\omega}$ (see Fig. 5), $\mathbf{S}(t\gg\tau_1)=[\mathbf{S}(\tau_2)\boldsymbol{\omega}]\boldsymbol{\omega}/\omega^2$. The important consequence of this picture is that the stationary value of the pseudospin has nonzero projections on the y and z axes, while $S_x=0$—in spite of the fact that, initially, the pseudospin vector was directed precisely along x. These results indicate that the application of a magnetic field results in finite values of both circular (S_z) and converted linear polarizations (S_y). It is important to note that the latter effect takes place only if the directions $\boldsymbol{\omega}$ and $\mathbf{\Omega}$ do not coincide, i.e., if $\omega_y\neq\Omega_y$. At $\boldsymbol{\omega}\parallel\mathbf{\Omega}$, the pseudospin always precesses in the plane perpendicular to this direction, and in steady state ($\omega\tau_1\gg 1$), the mean value $S\equiv 0$. One can see that the process just described allows for the appearance of linear polarization $P_{L'}=2S_y$ in the rotated reference frame (the magneto-optical conversion of linear polarization from the [110] to the [100] orientation), as is indeed experimentally observed. The process involving $\langle 110\rangle$ QDs, where the anisotropic exchange field ($B_x=\hbar\omega_x/\mu g_{ex}$) is directed along x, yields a different but qualitatively analogous result. In this case, the precession of the dipole moment occurs around the direction lying in the (x,z) plane, and in steady state, the pseudospin therefore has finite projections on both the x and the z axes, S_x and S_z, respectively.

E. Calculations and comparison with experiment

Next we perform a quantitative analysis of the proposed model. First we calculate the projections of pseudospin $\mathbf{S}_2=(S_{2x},S_{2y},S_{2z})$ in state 2 (where the condition $\Omega\tau\gg 1$ is not satisfied) by solving the Bloch equation for the mean pseudospin.[6,26,27] Then the projections of the pseudospin in state 1 (by virtue of the condition $\omega\tau\gg 1$) can be found from the simple projection of $\mathbf{S}_2$ on the direction of $\boldsymbol{\omega}$. In what follows, it is important to bear in mind that the projections of $\mathbf{S}$ correspond to specific polarizations of emitted light (specifically, S_z denotes circular polarization, and S_x and S_y stand for linear polarizations P_L and P_L, as discussed in detail in Ref. 22).

In the case of linearly polarized excitation $e\parallel[110]$, i.e., $\mathbf{S}_0=(S_{0x},0,0)$ in the upper step of the cascade, for the $\langle 110\rangle$-type QDs [$\mathbf{\Omega}=(\Omega_x,0,\Omega_\parallel)$], the components of pseudospin in state 2 then become

$$S_{2x}^{110}(B)=S_{0x}\frac{\tau_{L2}}{\tau_{L2}+\tau_2}\frac{1+\Omega_x^2T_L^2}{1+\Omega_x^2T_L^2+\Omega_\parallel^2T_L^2},$$

$$S_{2y}^{110}(B)=S_{0x}\frac{\tau_{L2}}{\tau_{L2}+\tau_2}\frac{\Omega_\parallel T_L}{1+\Omega_x^2T_L^2+\Omega_\parallel^2T_L^2}. \tag{2}$$

Similarly, for $\langle 100\rangle$-type QDs, we have $\mathbf{\Omega}=(0,\Omega_y,\Omega_\parallel)$, and

$$S_{2x}^{100}(B)=S_{0x}\frac{\tau_{L2}}{\tau_{L2}+\tau_2}\frac{1}{1+\Omega_y^2T_L^2+\Omega_\parallel^2T_L^2},$$

$$S_{2y}^{100}(B)=S_{0x}\frac{\tau_{L2}}{\tau_{L2}+\tau_2}\frac{\Omega_\parallel T_L}{1+\Omega_y^2T_L^2+\Omega_\parallel^2T_L^2}, \tag{3}$$

where τ_2, τ_{L2} are exciton lifetime and alignment decay time, respectively; $T_L^{-1}=\tau_2^{-1}+\tau_{L2}^{-1}$ defines the "alignment lifetime;" and $S_{0x}\tau_{L2}/(\tau_2+\tau_{L2})=S_{0x}T_L/\tau_2$ gives the value of the pseudospin at $B=0$. Here the superscript of S identifies the dot type, and the subscript refers to the type of polarization. It is interesting to note that for excitation by linearly polarized light $e\parallel[110]$, there appears (in addition to the linearly polarized PL emissions corresponding to S_{2x} and S_{2y}) also mean projections S_{2z} that lead to circular polarizations $P_C=2S_{2z}$, as follows:

$$S_{2z}^{110}(B)=S_{0x}\left(\frac{T_L}{\tau_2}\right)\frac{\Omega_\parallel T_L}{1+\Omega_x^2T_L^2+\Omega_\parallel^2T_L^2},$$

$$S_{2z}^{100}(B)=S_{0x}\left(\frac{T_L}{\tau_2}\right)\frac{\Omega_y T_L}{1+\Omega_y^2T_L^2+\Omega_\parallel^2T_L^2}. \tag{4}$$

One can see from these relations that there already appears a circular polarization in the first step of the cascade. For the $\langle 110\rangle$-type QDs, this gives $S_{2z}^{110}(B)\propto\Omega_\parallel\propto B$, while for the $\langle 100\rangle$-type QDs, the circular polarization is nonzero even at zero magnetic field, $S_{2z}^{100}(0)\neq 0$. The appearance of the circular polarization at linearly polarized excitation in the absence of a magnetic field (as is the case for the $\langle 100\rangle$-type QDs for $e\parallel[110]$ excitation) is a consequence of the low symmetry of the QDs; it takes place when the e vector of the exciting light is not parallel to either of the main axes of the QD. The case of excitation $e\parallel[100]$ can be treated analogously.

Excitation by circularly polarized light gives, for $\langle 110\rangle$ QDs,

$$S_{2x}^{110}(B)=S_{0z}\left(\frac{T_C}{\tau_2}\right)\frac{\Omega_x\Omega_\parallel T_C^2}{1+\Omega_x^2T_C^2+\Omega_\parallel^2T_C^2},$$

$$S_{2y}^{110}(B)=-S_{0z}\left(\frac{T_C}{\tau_2}\right)\frac{\Omega_x T_C}{1+\Omega_x^2T_C^2+\Omega_\parallel^2T_C^2},$$

$$S_{2z}^{110}(B)=S_{0z}\left(\frac{T_C}{\tau_2}\right)\frac{1+\Omega_\parallel^2T_C^2}{1+\Omega_x^2T_C^2+\Omega_\parallel^2T_C^2}, \tag{5}$$

where $T_C^{-1}=\tau_2^{-1}+\tau_S^{-1}$, with τ_S being the spin-relaxation time of the level-2 exciton.

In the cascade model under consideration, the polarization characteristics of the excitonic radiation from the ground state (level 1) can be deduced from the projection of $\mathbf{S}_2 = (S_{2x}, S_{2y}, S_{2z})$ on the direction of $\boldsymbol{\omega}$. In the case of linear polarization from the ground state of $\langle 110 \rangle$-type QDs, where $\boldsymbol{\omega} = (\omega_x, 0, \Omega_\parallel)$, we obtain ($e \| [110]$)

$$P_L^{110}(B) = P_L^0 \frac{(\omega_x + \omega_x \Omega_x^2 T_L^2 + \Omega_x \Omega_\parallel^2 T_L^2)\omega_x}{(1 + \Omega_x^2 T_L^2 + \Omega_\parallel^2 T_L^2)(\omega_x^2 + \Omega_\parallel^2)}, \quad P_{L'}^{110} = 0, \tag{6}$$

and for the $\langle 100 \rangle$-type QDs [where $\Omega = (0, \omega_y, \Omega_\parallel)$], we get

$$P_L^{100} = 0, \quad P_{L'}^{100}(B) = P_L^0 \frac{\omega_y \Omega_\parallel T_L (\omega_y - \Omega_y)}{(1 + \Omega_y^2 T_L^2 + \Omega_\parallel^2 T_L^2)(\omega_y^2 + \Omega_\parallel^2)}. \tag{7}$$

One can see from (7) that the polarization of excitons in the rotated axes $P_{L'}$ appears only due to the cascade process, as was shown qualitatively in the preceding section, the value of $P_{L'}$ being proportional to the difference between ω_y and Ω_y. We recall that, by virtue of the inequality $\delta \gg \hbar/\tau$, the alignment in the rotated axes $P_{L'}^{100}(B)$ would have been absent if level 1 were excited resonantly. Assuming equal contributions from the $\langle 110 \rangle$- and $\langle 100 \rangle$-type QDs to the total emission of the QD ensemble, for the resultant polarization we obtain $P_L = (P_L^{110} + P_L^{100})/2 = P_L^{110}/2$, $P_{L'} = (P_{L'}^{110} + P_{L'}^{100})/2 = P_{L'}^{100}/2$.

Equation (7) shows that the converted linear polarization is proportional to the ω_y squared as long as Ω_y remains larger than ω_y. From this we can conclude that the averaging over an isotropic distribution of ω_y conserves conversion of linear polarization from one set of axes to another. Therefore, the model can account for the observation of linear-to-linear polarization conversion.

The circularly polarized excitation by, e.g., σ^+-light [i.e., $\mathbf{S} = (0, 0, S_{0z})$] finally leads to the following relations for the $\langle 110 \rangle$-type QDs:

$$P_C^{110} = 2S_z = 2S_0 \frac{T_C}{\tau_2} \frac{(1 + \omega_x \Omega_x T_C^2 + \Omega_\parallel^2 T_C^2)\Omega_\parallel^2}{(1 + \Omega_x^2 T_C^2 + \Omega_\parallel^2 T_C^2)(\omega_x^2 + \Omega_\parallel^2)}, \tag{8}$$

$$P_L^{110} = -2S_x = -2S_0 \frac{T_C}{\tau_2} \frac{(1 + \omega_x \Omega_x T_C^2 + \Omega_\parallel^2 T_C^2)\omega_x \Omega_\parallel}{(1 + \Omega_x^2 T_C^2 + \Omega_\parallel^2 T_C^2)(\omega_x^2 + \Omega_\parallel^2)}, \quad P_{L'}^{110} = 0. \tag{9}$$

Analogous expressions are valid for the $\langle 100 \rangle$-type QDs, with the only difference being that ω_x and Ω_x should be replaced by ω_y and Ω_y, respectively. Assuming again equal contributions from both types of QDs to the total PL signal, for the resultant circular polarization, we obtain $P_c = (P_c^{110} + P_c^{100})/2$. The calculated magnetic field dependences of the polarizations are shown by the solid curves in Figs. 2 and 3. It is seen that, for a fixed choice of parameters ω_x, ω_y, Ω_x, Ω_y, T_L, T_C, and τ_2, the theory fits the experimental results rather well.

Let us now compare the derived expressions (6)–(9) with experimental results in greater detail. It follows from (8) that in a high magnetic field, P_C saturates at a level $P_C(\infty) = 2S_0 T_C/\tau_2$. On the other hand, we know from Eqs. (6) and (7) that at zero magnetic field, $P_L(0) = S_0 T_L/\tau_2$. Comparison with experiment leads to the result $S_0 T_L/\tau_2 = 0.28$ and $2S_0 T_C/\tau_2 = 0.4$. As the value of S_0 is of the order of unity, the constants T_C and T_L are comparable and have the same order of magnitude as τ_2. (The former statement agrees with the fact that each process of exciton spin flip is accompanied by the dephasing of the states $m = \pm 1$.) Our analysis shows that the expressions (6) and (8) for P_L and P_C are mainly determined by the parameters ω_x and ω_y. The variation of the values of Ω_i, T_L, and T_C taken in reasonable limits (T_L and T_C in the picosecond range, and Ω_i comparable to ω_i) does not influence the value of ω_x and ω_y. From fitting to the experimental results for P_L shown in Fig. 3 (open circles), one then obtains the parameters $\hbar\omega_x = 0.28 \pm 0.02$ meV, $\hbar\omega_y = 0.24 \pm 0.02$ meV. It is important to note that—using Eq. (9) and its analog for the $\langle 100 \rangle$ QDs—we are able to describe the results for $P_C(B)$ [Fig. 2(a)] with the same values of the $\hbar\omega_x$ and $\hbar\omega_y$ parameters.

The results for the linearly polarized emission in the primed axes ($P_{L'}$) are the most sensitive to the values of the fitting parameters of the excited state 2. In fact, as has been shown, the appearance of $P_{L'}$ in a magnetic field is a consequence of the pseudospin evolution in state 2. By comparing Eq. (7) with its $\langle 110 \rangle$ QD analog, we can extract the parameters $\hbar\Omega_x = 0.46 \pm 0.06$ meV, $\hbar\Omega_y = 0.46 \pm .06$ meV, $T_L \sim 1$ ps, and $\tau_2 \sim 2$ ps. The very short exciton lifetime on level 2 is not surprising in light of the rapid anharmonic decay of optical phonons into acoustical phonons. The analysis of Raman experiments in bulk AlN (Ref. 28) has shown that phonon lifetimes at low temperatures are indeed in the picosecond range. An unexpected result is that, for the excited state that is coupled to the phonons, the exchange splitting of $\hbar\Omega_x = 0.46$ meV turns out to be larger than the same parameter corresponding to the ground state, $\hbar\omega_x = 0.28$ meV. But the model developed can only describe the experiment with such relation of the exchange-splitting parameters ($\Omega_i > \omega_i, i = x, y$). We tentatively explain this result by assuming that the phonons may additionally distort the shape of the QDs. The same parameters were used to fit all other experimental dependences reported here.

As we have seen, the value of τ_2 is in the picosecond range. On the other hand, the long spin lifetime statement ($\tau_S \gg \tau_2$) requires that the values of $T_C/\tau_2 \equiv \tau_S/(\tau_S + \tau_2)$ and $T_L/\tau_2 \equiv \tau_L/(\tau_L + \tau_2)$ should be close to unity. It follows immediately from these values that $S_0 \approx 0.28$. Meanwhile, the optical selection rules between p and s QD ground states give the value $S_0 = 0.5$. This discrepancy can be resolved, however, by the possible violation of selection rules for optical transitions involving higher-lying states. Such violation would result in S_0 no longer being equal to 0.5. The authors of Ref. 12 have used the same argument in order to account for the overall reduction of the degree of linear polarization with increasing excess energy.

One can see from the preceding analysis that the magnetic field not only leads to the conversion of the linear polarization from one reference frame to another, but also to conversion from optical orientation to optical alignment, and vice

versa. Indeed, both latter conversions have been observed in qualitative agreement with calculations. However, here the quantitative analysis of the results was more complicated because of the simultaneous presence of two types of QDs in the ensemble. Perhaps the results for these conversions are sensitive to the variation of ω_i from dot to dot and to directional distribution of the principal axes of the QDs. A detailed description of these experiments and the corresponding analysis will be carried out in a future publication.

IV. CONCLUSIONS

In conclusion, we list the main results of the paper. We have studied the characteristics of optical orientation and alignment of excitons in CdSe/ZnSe QDs. We have established that the excitonic spin-relaxation time τ_S is noticeably longer than the exciton lifetime τ. However, in spite of the inequality $\tau_S \gg \tau$, the degree of optical orientation at zero magnetic field is small. The origin of this unexpected feature can be traced to the strong anisotropic exchange interaction that splits the excitonic doublet $m=\pm 1$ into two orthogonally polarized linear dipoles. In the external magnetic field, we observed the effect of restoration of the optical orientation—an effect somewhat analogous to Van Vleck paramagnetism. Fitting model-based calculations to our experimental results allowed us to determine the value of this anisotropic exchange splitting of the exciton state. Experiments on the optical alignment of excitons allowed us to conclude that there were two groups of anisotropic QDs—in one group, the QDs were elongated along the $[110]$ (or, equivalently, the $[1\bar{1}0]$) crystallographic directions, and in the other, the elongation was along $[100]$ (or $[010]$). We note that our model—which assumes the existence of the two types of QDs, elongated either in $\langle 110\rangle$ or in $\langle 100\rangle$ directions—is just a simplification. In fact, there should be a directional (in-plane) distribution of the principal axes of the QDs, a picture that has also been encountered in other study of the CdSe/ZnSe QDs.[29] Thus, the averaging procedures required to account for the details of such a distribution would only obscure the essential physics of the system. The model of the two ensembles of QDs is convenient for two reasons. First, the model is able to reproduce qualitatively (and often even quantitatively) the main magneto-optical phenomena occurring in the system. And second, the model is considerably simpler than the more rigorous treatment, thus allowing a more transparent presentation of the spin dynamics that take place in those phenomena.

Conversion of the linear polarization of excitonic radiation from one set of axes to axes rotated by 45° was observed when a magnetic field was applied. This phenomenon was interpreted in terms of a cascade process, i.e., a process in which the exciton first created by the laser is in an excited state (which is believed to be a coupled exciton-phonon state), but the observed PL is emitted from the exciton ground state, after the exciton relaxes to that state by phonon emission. The observation of peaks separated from the laser excitation energy by an integer number of optical phonons appearing prominently in the polarization spectra distinctly reveals this cascade process by which the emission takes place. We were able to estimate the lifetime of the exciton in the excited state to be $\tau_2 \approx 1$ ps, and the value of the anisotropic exchange splitting to be $\hbar\Omega_{x,y} \approx 0.5$ meV. The analysis of our experimental results shows that the value of the exchange splitting in the ground state is noticeably smaller than that in the excited state. We tentatively attribute this result to an additional distortion of the shape of the QD by phonons.

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