Intertwining of Zeeman and Coulomb interactions on excitons in highly symmetric semiconductor quantum dots

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We present an experimental study and develop a group theoretical analysis of the Zeeman effect on excitons in pyramidal semiconductor quantum dots possessing the symmetries of the C_{3v} point group. The magnetic field dependence of the emission pattern originating from neutral exciton states is investigated in both the Faraday and Voigt configurations. The Zeeman doublet splitting of the "bright" exciton states varies linearly with the magnetic field strength in each configuration while the intensity of the "dark" exciton transitions exhibit a nonlinear dependence. We demonstrate that these observations originate from the intertwining of the Zeeman and Coulomb interactions, which provides clear spectral signatures of this effect for highly symmetric quantum dots. We uncover a large anisotropy of the Zeeman doublet splittings for longitudinal and transverse magnetic fields, revealing the ubiquitous role of a symmetry elevation in our pyramidal quantum dots. These results suggest that the common description of the Zeeman effect based on effective g factors for electrons and holes must be revised when dealing with exciton complexes.

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I. INTRODUCTION

The discovery of the electron spin has long been associated with the "anomalous" Zeeman effect in the atomic spectra of hydrogen[1,2] and with the deflection of an atomic beam of silver atoms in the Stern-Gerlach experiment.[3] In semiconductors the Zeeman effect has been first studied on the optical spectra of acceptor impurities in bulk germanium and, subsequently, on the optical spectra of excitons in bulk and in semiconductor heterostructures. In an applied magnetic field, the optical spectra of the elementary excitations associated with the substitutional impurities or with the excitons have a rich multiplet structure, which reveals the splitting of degenerate levels into sublevels and depends on the symmetry of the crystal field for an impurity or of the confinement potential for an exciton confined in a heterostructure. The quantum states of an exciton in a semiconductor are determined by the Coulomb interaction between the electron and the hole composing this quasiparticle. While in a bulk semiconductor the degeneracy of the exciton states is solely dependent on the symmetry of the band edges of the crystalline structure, in a semiconductor quantum dot (QD), it is particularly sensitive to the nanostructure's shape, size, and composition, and to the presence of strain, electric, or magnetic field.

A general theoretical description of the interplay between the Coulomb and the Zeeman interaction has so far treated the applied magnetic field as a perturbation acting on the individual Bloch states of either the electrons or the holes at the band edges of the semiconductor. This general approach was initially developed by Kohn and Luttinger in order to describe the cyclotron resonances of holes in a germanium or a silicium crystal.[4,5] The magneto-optical properties of excitons in GaAs/$Al_xGa_{1-x}As$ quantum wells have been extensively studied both experimentally[6–9] and theoretically,[10–12] thereby highlighting the importance of valence-band mixing and the paramagnetic and diamagnetic contributions to both the Zeeman splitting energy and the diamagnetic shift of the excitons. Despite these results on magnetoexcitons in quantum wells, the interplay between the Coulomb and the Zeeman interactions has largely eluded the observation in magnetophotoluminescence studies of the fine-structure splitting of excitons in single semiconductor QDs.[13–15] In these studies the Zeeman interaction of a hole (or an electron) with an applied magnetic field is commonly described at the level of the single-particle eigenstates at the valence- (conduction-) band edge of the semiconductor using the valence-band parameters (q,κ) introduced by Luttinger for cubic crystalline structures.[5] Most experimental studies of excitons confined in QDs were performed on self-assembled quantum dots (SAQDs) grown on $\{100\}$ surfaces and were concerned with polarization anisotropy,[16–18] in-plane anisotropy of carrier effective g factors,[19,20] fine-structure splitting of neutral and charged exciton complexes,[21–24] which were induced by the shape asymmetry, and the existence of anisotropic strain and piezoelectric fields in these dots. Only recently, highly symmetric pyramidal QDs grown on $\{111\}$ surfaces have emerged as interesting quantum structures in which zero fine-structure splitting of the neutral exciton was predicted[25,26] and eventually observed,[27–30] and for which the entanglement of polarized photon pairs was demonstrated.[31]

It is our purpose in this paper to develop a general theoretical model of the Zeeman effects on the quantum states of excitons in highly symmetric QDs and to compare its predictions to experimental studies performed on magnetoexcitons confined in pyramidal QDs grown on (111)B GaAs substrates.[32,33] The underlying idea is to account for the interplay between Coulomb and Zeeman interactions by implementing an effective Hamiltonian formalism in the basis of the exciton states that is solely based on symmetry arguments. In Sec. II, we present the group theoretical derivation of the Zeeman Hamiltonian for the two different configurations of the applied magnetic field (Voigt and Faraday geometries) in the case of excitons confined in pyramidal QDs possessing C_{3v} point group symmetry. We derive explicit expressions of the Zeeman doublet splittings for neutral exciton states and identify spectral features resulting from the interplay between Coulomb and Zeeman interactions. In Sec. III, we report on a magneto-optical study of excitons in single pyramidal quantum dots and analyze the Zeeman doublet splittings and the diamagnetic shifts measured in a transverse (Voigt configuration) and in a longitudinal (Faraday configuration) magnetic field. All the experimental results are analyzed and discussed within the framework of the effective Zeeman Hamiltonian developed in Sec. II to which is added the electron-hole exchange terms resulting from the actual symmetry of the pyramidal QD. Sec. IV brings concluding remarks on the Zeeman effects in QDs in relation with their intrinsic symmetry and the diamagnetic and paramagnetic contributions to the Zeeman splittings.

II. THEORY

A. Formulation of Zeeman Hamiltonian

The application of a magnetic field to a quantum dot modifies the optical spectrum of excitonic complexes and allows the study of their quantum states and their degeneracies for a given symmetry of the structure. To each excitonic complex, we associate an effective Zeeman Hamiltonian H_Z, which is defined in the subspace spanned by the quantum states of the excitonic complex under consideration. The point group of the nanostructure is determined by the common symmetry operations mapping the crystalline structure and the quantum dot to themselves. In this paper, the pyramidal quantum dots are assumed to have symmetries of the C_{3v} point group as we consider semiconductor QDs made of III-V compounds with a zinc-blende crystalline structure. The electrons and the holes states are assumed to be associated, respectively, with the Γ_6 and Γ_8 points of the Brillouin zone (symmetry T_d). The description of an exciton confined to a QD will thus necessitate a basis of four vector states in order to account for the orbital and spinorial degrees of freedom of one electron and one hole. We assume that the electron and hole states are strongly confined into the QD and that the electron (hole) ground states correspond to irrep $E_{1/2}$ (${}^iE_{3/2}$).[34] The other case of interest corresponds to hole excited states transforming according to the irrep $E_{1/2}$ (the results are given in the Appendix). For the point group (PG) C_{3v}, the exciton ground states transform according to the two-dimensional irreducible representation (irrep) E when the electron and hole states transform according to irrep $E_{1/2}$ and ${}^iE_{3/2}$, respectively.[28] We recall that the irrep E is obtained from the direct product of the electron and hole irreps, $E = E_{1/2} \otimes {}^iE_{3/2}$. The exciton basis (also called the standard basis) is formally written as a set of basis vectors $\{|b,E,1\rangle,|b,E,2\rangle,|d,E,1\rangle,|d,E,2\rangle\}$, where the first label identifies one of the two energy levels of the exciton (b or d) that are split in energy by the electron-hole exchange interaction, the second label identifies the irrep, and the third one is an integer index of the irrep partners. Within this exciton subspace, the Zeeman Hamiltonian is expressed as a sum of terms invariant under all symmetry operations of the C_{3v} point group. For deriving the invariant terms of the Hamiltonian, we implement a modified version of the procedure that was first used by Luttinger[35] to study the cyclotron motion of electrons in the valence band of a cubic crystal, and then generalized by Cho[36] to deal with any effect of symmetry breaking introduced by an external perturbation of the motion of electrons and holes in cubic crystals (such as a magnetic field, an electric field, or an external stress). An invariant term of the linear Zeeman Hamiltonian is a scalar product of one of the components of the external magnetic field and of the corresponding component of a irreducible tensorial operator of order 1;[37] both components must transform as basis functions of the same irrep or as basis functions of the irrep and its complex conjugate irrep if the two irreps are not equivalent. In the general case of a magnetic field with Cartesian coordinates (B_x,B_y,B_z), the linear part of the Zeeman Hamiltonian is formally written as

$$H_Z^{\text{lin}} = \tfrac{1}{2}\mu_B g_\perp\big[(B_x + iB_y)L_2^{[E]} + (B_x - iB_y)L_1^{[E]}\big] + \tfrac{1}{2}\mu_B g_\parallel B_z L_1^{[A_2]}, \quad (1)$$

where $(L_1^{[E]},L_2^{[E]})$ and $L_1^{[A_2]}$ are the components of irreducible tensorial operators of order 1, which transform under the PG C_{3v} as partners of irreps E and A_2, respectively. In the exciton basis given above, the invariant terms of the Zeeman Hamiltonian are those for which the associated irrep is contained in the direct product of the representations $\Gamma_i^* \otimes \Gamma_i$, where $\Gamma_i^*,\Gamma_j \equiv E$. For instance, the direct product of irrep E with itself gives $E \otimes E = A_1 + A_2 + E$; this implies that the components $(B_x + iB_y, B_x - iB_y)$ transforming as basis functions of E appear in the Zeeman Hamiltonian with the same transverse effective g factor denoted as $g_\perp$, while the component B_z transforming as irrep A_2 appears with another effective g factor denoted as $g_{||}$. It is worthwhile noting that the specific form of the basis functions of irrep E depends on the matrix representations of the PG C_{3v}: When using the point group tables given by Altmann,[34] the basis functions of a Cartesian tensor of order 1 that span the irrep E are $(-x - iy, x - iy)$ for the components of a polar vector and $(l_x + il_y, l_x - il_y)$ for those of an axial vector (e.g., magnetic field, angular momentum).

Assuming that the magnetic field is oriented along the y axis (taken to be perpendicular to one of the reflection symmetry planes of C_{3v}), the matrix form of the Zeeman Hamiltonian in the standard exciton basis given above is written as

$$M_Z = \frac{1}{2}\mu_B B_y \begin{pmatrix} 0 & g_1 & 0 & g_3 \\ g_1 & 0 & g_3 & 0 \\ 0 & g_3^* & 0 & g_2 \\ g_3^* & 0 & g_2 & 0 \end{pmatrix}, \quad (2)$$

where g_1,g_2 are real numbers, and g_3 may be a complex number. If the magnetic field is oriented along the z axis, the matrix form of the Zeeman Hamiltonian is changed to

$$M_Z = \frac{1}{2}\mu_B B_z \begin{pmatrix} g_4 & 0 & g_6 & 0 \\ 0 & -g_4 & 0 & -g_6 \\ g_6^* & 0 & g_5 & 0 \\ 0 & -g_6^* & 0 & -g_5 \end{pmatrix}, \quad (3)$$

where g_4,g_5 are real numbers, and g_6 may be a complex number.

The elements of the Zeeman Hamiltonian matrices for each orientation of the magnetic field are derived by using the Wigner-Eckart theorem that relates a given matrix element $\langle d,E,i|L_k^{[E]}|d,E,j\rangle$ with a reduced matrix element $\langle d,E||L^{[E]}||d,E\rangle$, which is independent of the partner functions of the irrep basis. The formal expressions of the transverse effective g factors g_i are written as

$$g_1 = \langle b,E\|L^{[E]}\|b,E\rangle, \quad g_2 = \langle d,E\|L^{[E]}\|d,E\rangle,$$
$$g_3 = \langle b,E\|L^{[E]}\|d,E\rangle, \tag{4}$$

and those of the longitudinal effective g factors as

$$g_4 = \langle b,E\|L^{[A_2]}\|b,E\rangle, \quad g_5 = \langle d,E\|L^{[A_2]}\|d,E\rangle,$$
$$g_6 = \langle b,E\|L^{[A_2]}\|d,E\rangle, \tag{5}$$

where the operators $L^{[E]}$ and $L^{[A_2]}$ in the reduced matrix elements refer to irreducible tensorial operators of order 1 corresponding to irreps E and A_2, respectively. We stress that the reduced matrix elements take different values for each pair of eigenstates of the ground exciton; likewise, there will be other sets of reduced matrix elements for an excited exciton or another exciton complex (an extension of the model to an excited exciton is given in the Appendix).

In addition to the linear Zeeman terms, it is essential to consider the diamagnetic Zeeman terms for their contributions may be as large or larger depending on the strength of the magnetic field and may introduce distinct coupling terms between excitonic states. For instance, the set of basis functions $(2xy, x^2 - y^2)$ transforms according to the irrep E and, hence, there are other off-diagonal diamagnetic Zeeman terms in addition to the linear Zeeman terms considered above. In the general case of a magnetic field with Cartesian coordinates (B_x, B_y, B_z), the diamagnetic Zeeman Hamiltonian is derived with the method of invariants and is given by the following expression:

$$\begin{aligned} H_Z^{\rm dia} &= \left[\alpha^{\perp}\left(B_x^2 + B_y^2\right) + \alpha^{\parallel} B_z^2\right]\left[L_1^{[A_2]}L_1^{[A_2]} + L_1^{[E]}L_2^{[E]} + L_2^{[E]}L_1^{[E]}\right] + \left[\omega(B_x + iB_y)^2 + \varepsilon(B_x - iB_y)B_z\right]\left[L_2^{[E]}L_2^{[E]}\right] \\ &+ \left[\omega(B_x - iB_y)^2 + \varepsilon(B_x + iB_y)B_z\right]\left[L_1^{[E]}L_1^{[E]}\right] + \left[\gamma(B_x + iB_y)B_z + \eta(B_x - iB_y)^2\right]\left[L_2^{[E]}L_1^{[A_2]} + L_1^{[A_2]}L_2^{[E]}\right] \\ &+ \left[\gamma(B_x - iB_y)B_z + \eta(B_x + iB_y)^2\right]\left[L_1^{[E]}L_1^{[A_2]} + L_1^{[A_2]}L_1^{[E]}\right]. \end{aligned} \tag{6}$$

The matrix form of the diamagnetic Zeeman Hamiltonian for a magnetic field along the y axis is then calculated with the Wigner-Eckart theorem in the standard exciton basis, and it is written as

$$M_Z^{\rm dia} = B_y^2 \begin{pmatrix} \alpha_1 & 0 & \alpha_3 & 0 \\ 0 & \alpha_1 & 0 & \alpha_3 \\ \alpha_3^* & 0 & \alpha_2 & 0 \\ 0 & \alpha_3^* & 0 & \alpha_2 \end{pmatrix} - B_y^2 \begin{pmatrix} 0 & \eta_1 & 0 & \eta_3 \\ \eta_1 & 0 & \eta_3 & 0 \\ 0 & \eta_3^* & 0 & -\eta_1 \\ \eta_3^* & 0 & -\eta_1 & 0 \end{pmatrix}, \tag{7}$$

where $\alpha_1(\eta_1)$ and α_2 are real diamagnetic coefficients coupling the bright and the dark exciton states, respectively, and where α_3 and the η_3 are complex off-diagonal coefficients that couple the *bright* exciton states $\{|b,E,1\rangle, |b,E,2\rangle\}$ to the *dark* exciton states $\{|d,E,1\rangle, |d,E,2\rangle\}$ (the justification for the names *bright* and *dark* is given later in this section). It is worthwhile discussing first the respective role of these coefficients in order to retain only the relevant ones. The off-diagonal coefficient η_1 would introduce a quadratic dependence of the Zeeman energy splitting with the magnetic field. Since the magnetic field dependence of the experimentally measured Zeeman splittings are best fitted with a linear function, this coefficient will thus be discarded in the later analysis. The other off-diagonal coefficients (α_3, η_3) could separately contribute to the coupling between the dark states and the bright states. While the diamagnetic terms with α_3 couple identically a bright state $|b,E,i\rangle$ to a dark state $|d,E,i\rangle$, the diamagnetic terms with η_3 couple them differently. We will only retain the diamagnetic terms with α_3, however, and show that the contribution of all the terms with η_i's is negligible for reasons of a symmetry elevation from C_{3v} to D_{3h}.

Including the dominant diamagnetic Zeeman terms, the matrix form of the complete Zeeman Hamiltonian for a magnetic field oriented along the y axis is written in the standard exciton basis as

$$M_Z^{\rm lin} + M_Z^{\rm dia} = \frac{1}{2}\mu_B B_y \begin{pmatrix} 0 & g_1 & 0 & g_3 \\ g_1 & 0 & g_3 & 0 \\ 0 & g_3^* & 0 & g_2 \\ g_3^* & 0 & g_2 & 0 \end{pmatrix} + B_y^2 \begin{pmatrix} \alpha_1 & 0 & \alpha_3 & 0 \\ 0 & \alpha_1 & 0 & \alpha_3 \\ \alpha_3^* & 0 & \alpha_2 & 0 \\ 0 & \alpha_3^* & 0 & \alpha_2 \end{pmatrix}. \tag{8}$$

When the magnetic field is aligned instead with the x axis, the matrix elements of the diamagnetic terms are unchanged if the second term of expression (7) is neglected. The matrix form of the linear Zeeman terms differs, however, by a phase factor. For the sake of completeness, we also give the linear Zeeman matrix form for a magnetic field oriented along the x axis; it is written as

$$M_Z = \frac{1}{2}\mu_B B_x \begin{pmatrix} 0 & -ig_1 & 0 & -ig_3 \\ ig_1 & 0 & ig_3 & 0 \\ 0 & -ig_3^* & 0 & -ig_2 \\ ig_3^* & 0 & ig_2 & 0 \end{pmatrix}. \tag{9}$$

In the standard exciton basis, the matrix form of the unperturbed exciton Hamiltonian is given by the diagonal matrix

$$M_{\rm exch} = \frac{1}{2}\begin{pmatrix} \delta_0 & 0 & 0 & 0 \\ 0 & \delta_0 & 0 & 0 \\ 0 & 0 & -\delta_0 & 0 \\ 0 & 0 & 0 & -\delta_0 \end{pmatrix}, \tag{10}$$

where δ_0 is the fine structure splitting between the set of degenerate exciton eigenstates $\{|b,E,1\rangle, |b,E,2\rangle\}$ and

TABLE I. Eigenenergies of neutral exciton states in a transverse magnetic field (Voigt configuration, $B = B_x$ or B_y) ordered by decreasing values when $g_1', g_2' \geqslant 0$. In these expressions, the prime value of the effective g factors and diamagnetic coefficients is a shortened notation defined by $\alpha_i' = \alpha_i B^2$ and $g_i' = \frac{1}{2} g_i \mu_B B$ for $i = 1,2,3$.

$E_1 = \frac{1}{2}(\alpha_1' + \alpha_2' + g_1' + g_2') + \frac{1}{2}\sqrt{(g_1' - g_2' + \alpha_1' - \alpha_2' + \delta_0)^2 + 4\lvert g_3' + \alpha_3'\rvert^2}$
$E_2 = \frac{1}{2}(\alpha_1' + \alpha_2' - g_1' - g_2') + \frac{1}{2}\sqrt{(g_2' - g_1' + \alpha_1' - \alpha_2' + \delta_0)^2 + 4\lvert g_3' - \alpha_3'\rvert^2}$
$E_3 = \frac{1}{2}(\alpha_1' + \alpha_2' + g_1' + g_2') - \frac{1}{2}\sqrt{(g_1' - g_2' + \alpha_1' - \alpha_2' + \delta_0)^2 + 4\lvert g_3' + \alpha_3'\rvert^2}$
$E_4 = \frac{1}{2}(\alpha_1' + \alpha_2' - g_1' - g_2') - \frac{1}{2}\sqrt{(g_2' - g_1' + \alpha_1' - \alpha_2' + \delta_0)^2 + 4\lvert g_3' - \alpha_3'\rvert^2}$

$\{|d,E,1\rangle,|d,E,2\rangle\}$, which originates from the electron-hole (e-h) exchange interaction.

B. Analysis of Zeeman splittings

The 4×4 secular equation $\det||H_{ij} - E\delta_{ij}|| = 0$ can be solved exactly and yields analytical expressions for the eigenstates and eigenvalues of the total Hamiltonian $H = H_Z^{\text{lin}} + H_Z^{\text{dia}} + H_{\text{exch}}$ in the Voigt and in the Faraday configurations.

1. Voigt configuration

For a transverse magnetic field, the eigenenergies and eigenstates are given in Tables I and II, respectively. The optical selection rules are derived by applying the Wigner-Eckart theorem to the point group C_{3v}. Using the tables of Altmann's Clebsch-Gordon coefficients for C_{3v},[34] we determined the nonzero dipole matrix elements; these are given by

$$\langle A_1 | x - iy | b,E,1\rangle = \langle A_1 | -(x+iy) | b,E,2\rangle \equiv \mu_b,$$

where $|A_1\rangle$ corresponds to the irrep of the vacuum state, $(x - iy)$ is the component of the dipole moment operator for left circularly polarized light, and $-(x+iy)$ is that for right circularly polarized light. Hence, it follows that an optically active transition from any of the four exciton states is characterized by a linear polarization with a direction given in Table III.

For the Voigt configuration, we then expect that the optical spectrum of the exciton is composed of two doublets of linearly polarized lines corresponding to the two sets of quantum states $\{|\psi_1\rangle,|\psi_2\rangle\}$ and $\{|\psi_3\rangle,|\psi_4\rangle\}$. Since in the limit of low magnetic field, the linear Zeeman energies are typically much smaller than the e-h exchange energy δ_0, we derived an approximate expression of the Zeeman splitting energies $\Delta E_Z(\text{up}), \Delta E_Z(\text{low})$ for the upper and lower Zeeman doublet, respectively; these expressions are valid in the limit $(g_i \mu_B B_y, \alpha_i B_y^2 \ll \delta_0,\ i = 1,2)$ and are given by

$$\Delta E_z(\text{up}) \cong g_1 \mu_B B_y + \frac{1}{\delta_0}\left(\left|\frac{1}{2} g_3 \mu_B B_y + \alpha_3 B_y^2\right|^2 - \left|\frac{1}{2} g_3 \mu_B B_y - \alpha_3 B_y^2\right|^2\right) \tag{11}$$

and

$$\Delta E_z(\text{low}) \cong g_2 \mu_B B_y + \frac{1}{\delta_0}\left(\left|\frac{1}{2} g_3 \mu_B B_y - \alpha_3 B_y^2\right|^2 - \left|\frac{1}{2} g_3 \mu_B B_y + \alpha_3 B_y^2\right|^2\right). \tag{12}$$

Consequently, we predict that the Zeeman energy splitting of each doublet varies linearly with the magnetic field strength for a QD possessing the C_{3v} symmetry. This linear dependence of the Zeeman splittings with the magnetic field arises from the intertwining between the Coulomb and the Zeeman interactions in a transverse magnetic field. We emphasize that this linear dependence provides a unique signature of this intertwining as the standard theoretical description of the Zeeman interaction at the carrier level predicts instead a hyperbolic dependence of the Zeeman doublet splitting with a transverse magnetic field. The expression of this dependence can be easily obtained from the diagonalization of the Zeeman and e-h exchange Hamiltonian given in Ref. 27, taking into account that the anisotropic electron-hole exchange parameters ($\delta = \delta^* = 0$) are equal to zero in the case of C_{3v} symmetry; it is then written as

$$\Delta E_z = \frac{1}{2}\left|\sqrt{\delta_0^2 + (g_\perp^e - g_\perp^h)^2 \mu_B^2 B_y^2} - \sqrt{\delta_0^2 + (g_\perp^e + g_\perp^h)^2 \mu_B^2 B_y^2}\right|, \tag{13}$$

TABLE II. Eigenstates of neutral excitons in a transverse magnetic field ($B = B_y$). Prime values are defined in the caption of Table I.

Eigenstates	Eigenvalues
$\lvert\psi_1\rangle = \frac{1}{\sqrt{2}}(\lvert b,E,1\rangle + \lvert b,E,2\rangle) - \frac{(\delta_0/2 + \alpha_1' + g_1' - E_1)}{\sqrt{2}(g_3' + \alpha_3')}(\lvert d,E,1\rangle + \lvert d,E,2\rangle)$	E_1
$\lvert\psi_2\rangle = \frac{1}{\sqrt{2}}(\lvert b,E,1\rangle - \lvert b,E,2\rangle) + \frac{(\delta_0/2 + \alpha_1' - g_1' - E_2)}{\sqrt{2}(g_3' - \alpha_3')}(\lvert d,E,1\rangle - \lvert d,E,2\rangle)$	E_2
$\lvert\psi_3\rangle = \frac{1}{\sqrt{2}}(\lvert d,E,1\rangle + \lvert d,E,2\rangle) - \frac{(-\delta_0/2 + \alpha_2' + g_2' - E_3)}{\sqrt{2}(g_3' + \alpha_3')^*}(\lvert b,E,1\rangle + \lvert b,E,2\rangle)$	E_3
$\lvert\psi_4\rangle = \frac{1}{\sqrt{2}}(\lvert d,E,1\rangle - \lvert d,E,2\rangle) + \frac{(-\delta_0/2 + \alpha_2' - g_2' - E_4)}{\sqrt{2}(g_3' - \alpha_3')^*}(\lvert b,E,1\rangle - \lvert b,E,2\rangle)$	E_4

TABLE III. Orientation of polarization vector for an optical transition stemming from an exciton eigenstate $|\psi_i\rangle$, where $i = 1,2,3,4$.

Eigenstate	$\vert\psi_1\rangle$	$\vert\psi_2\rangle$	$\vert\psi_3\rangle$	$\vert\psi_4\rangle$
Polarization	y	x	y	x

where $g_\perp^e$ and $g_\perp^h$ are the transverse effective g factors for an electron and a hole, respectively.

In addition to the Zeeman splittings of the cross-linearly polarized doublets, we also calculated the energy splitting of the collinearly polarized Zeeman doublets. Their expressions are given by

$$\Delta E_z(x-\text{pol}) = \left\{\sqrt{\left[(\alpha_1-\alpha_2)B_y^2 - \frac{1}{2}(g_1-g_2)\mu_B B_y + \delta_0\right]^2 + 4\left|\alpha_3 B_y^2 - \frac{1}{2}g_3\mu_B B_y\right|^2}\right\} \quad (14)$$

and

$$\Delta E_z(y-\text{pol}) = \left\{\sqrt{\left[(\alpha_1-\alpha_2)B_y^2 + \frac{1}{2}(g_1-g_2)\mu_B B_y + \delta_0\right]^2 + 4\left|\alpha_3 B_y^2 + \frac{1}{2}g_3\mu_B B_y\right|^2}\right\}, \quad (15)$$

where $\Delta E_z(x-\text{pol})$ and $\Delta E_z(y-\text{pol})$ correspond to the energy splittings of the x-polarized and y-polarized transitions, respectively. In contrast with the cross-polarized Zeeman doublet splittings, the energy splittings of the copolarized doublets extrapolate to the *e-h* exchange energy at zero magnetic field, thereby providing a means to measure it experimentally. A direct determination of the *e-h* exchange energy is not possible because the exciton states $\{|d,E,1\rangle,|d,E,2\rangle\}$ are, in general, not radiatively active.

The contribution of the off-diagonal terms in the Zeeman Hamiltonian give rise, on the one hand, to a quartic and a quadratic correction of the Zeeman doublet splittings (as discussed above) and, on the other hand, to a hybridization of the exciton states $\{|b,E,1\rangle,|b,E,2\rangle\}$ to $\{|d,E,1\rangle,|d,E,2\rangle\}$ (see Table II). The first consequence of this hybridization is a coupling between exciton states, which have their own optical dipole moment (μ_d or μ_b). As will be demonstrated in this section, the optical transitions originating from the exciton states $\{|d,E,1\rangle, i = 1,2\}$ are completely "dark" in quantum dots for which the symmetry is elevated from C_{3v} to D_{3h}. Hence, the hybridization between the so-called "dark" excitonic states and the "bright" exciton states leads to a significant transfer of oscillator strength to the "dark" state transitions. When both the linear and quadratic terms contribute to the coupling, the intensity of the two optical transitions originating from the dark states is state specific. In the limit of $\mu_d^2 \ll \mu_b^2$, it is straightforward to derive from Table II the intensity ratio between the optical transitions from the "dark" and from the "bright" exciton states for a given orientation of the linear polarization vector. The intensity ratios for the linearly polarized transitions (x- and y-pol) are given in the limit of low magnetic field by the following expressions:

$$\frac{I_3}{I_1}(y-\text{pol}) \simeq \frac{\left|1/2g_3\mu_B B_y + \alpha_3 B_y^2\right|^2}{\delta_0^2} \quad (16)$$

and

$$\frac{I_4}{I_2}(x-\text{pol}) \simeq \frac{\left|1/2g_3\mu_B B_y - \alpha_3 B_y^2\right|^2}{\delta_0^2}. \quad (17)$$

From these expressions, one readily predicts an uneven transfer of oscillator strength to the "dark" exciton states, which originates from an interference between the off-diagonal linear and quadratic Zeeman terms g_3 and α_3.

2. *Faraday configuration*

For a longitudinal magnetic field, the matrix form of the diamagnetic Zeeman Hamiltonian is given in the standard exciton basis by

$$M_Z^{\text{dia}} = B_z^2 \begin{pmatrix} \alpha_4 & 0 & \alpha_6 & 0 \\ 0 & \alpha_4 & 0 & \alpha_6 \\ \alpha_6^* & 0 & \alpha_5 & 0 \\ 0 & \alpha_6^* & 0 & \alpha_5 \end{pmatrix}. \quad (18)$$

The eigenenergies and eigenstates of the 4×4 secular equation are given in Tables IV and V, respectively.

For the Faraday configuration, we then find that the optical spectrum of the exciton is composed of two doublets of circularly polarized lines corresponding to the two sets of quantum states $\{|\psi_1\rangle,|\psi_2\rangle\}$ and $\{|\psi_3\rangle,|\psi_4\rangle\}$ as summarized in Table V. In the limit of low magnetic field, the off-diagonal elements of the Zeeman interaction are typically much smaller than the *e-h* exchange energy δ_0; we can thus derive an approximate expression of the Zeeman splitting energies $\Delta E_Z(\text{up}),\Delta E_Z(\text{low})$ for the upper and lower Zeeman doublets, respectively; these expressions are valid in the limit ($g_6\mu_B B_z,\alpha_6 B_z^2 \ll \delta_0$) and are given by

$$\Delta E_z(\text{up}) = g_4\mu_B B_z + \frac{1}{\delta_0}\left(\left|\alpha_6 B_z^2 + \frac{1}{2}g_6\mu_B B_z\right|^2 - \left|\alpha_6 B_z^2 - \frac{1}{2}g_6\mu_B B_z\right|^2\right) \quad (19)$$

and

$$\Delta E_z(\text{low}) = g_5\mu_B B_z + \frac{1}{\delta_0}\left(\left|\alpha_6 B_z^2 - \frac{1}{2}g_6\mu_B B_z\right|^2 - \left|\alpha_6 B_z^2 + \frac{1}{2}g_6\mu_B B_z\right|^2\right). \quad (20)$$

TABLE IV. Eigenenergies of neutral exciton states in a longitudinal magnetic field (Faraday configuration, $B = B_z$) ordered by decreasing values when $g'_4, g'_5 \geqslant 0$. In these expressions, the prime values of the effective g factors and diamagnetic coefficients are defined by $\alpha'_i = \alpha_i B^2$ and $g'_i = \frac{1}{2} g_i \mu_B B$ for $i = 4,5,6$.

$$E_1 = \frac{1}{2}(g'_4 + g'_5 + \alpha'_4 + \alpha'_5) + \frac{1}{2}\sqrt{(g'_4 - g'_5 + \alpha'_4 - \alpha'_5 + \delta_0)^2 + 4|\alpha'_6 + g'_6|^2}$$
$$E_2 = -\frac{1}{2}(g'_4 + g'_5 - \alpha'_4 - \alpha'_5) + \frac{1}{2}\sqrt{(g'_5 - g'_4 + \alpha'_4 - \alpha'_5 + \delta_0)^2 + 4|\alpha'_6 - g'_6|^2}$$
$$E_3 = \frac{1}{2}(g'_4 + g'_5 + \alpha'_4 + \alpha'_5) - \frac{1}{2}\sqrt{(g'_4 - g'_5 + \alpha'_4 - \alpha'_5 + \delta_0)^2 + 4|\alpha'_6 + g'_6|^2}$$
$$E_4 = -\frac{1}{2}(g'_4 + g'_5 - \alpha'_4 - \alpha'_5) - \frac{1}{2}\sqrt{(g'_5 - g'_4 + \alpha'_4 - \alpha'_5 + \delta_0)^2 + 4|\alpha'_6 - g'_6|^2}$$

Hence, we predict that the Zeeman splitting energies of the doublets depend linearly on the strength of the longitudinal magnetic field in the low-field limit. A cubic magnetic field dependence of the Zeeman splittings is expected to arise in the intermediate-field limit, which would indicate a significant hybridization between the bright and the dark exciton states. Nevertheless, the hybridization is determined by the relative strength of the off-diagonal versus the diagonal matrix elements of the Zeeman Hamiltonian and will be revealed by an optical transition from a "dark" exciton state, the intensity of which will grow quadratically with the magnetic field strength in the weak-field limit. In the intermediate-field limit, the intensity of one of the dark exciton transition increases monotonically as a function of the magnetic field while the other one varies nonmonotonically due to the interference between the off-diagonal Zeeman terms in matrices (3) and (18) (see expressions of the eigenstates ψ_3 and ψ_4 in Table V).

C. Symmetry elevation to D_{3h}

In this section, we consider the consequences of symmetry elevation to D_{3h} on the Zeeman interaction terms of the effective Zeeman Hamiltonian derived above for C_{3v} symmetry. Several quadratic and linear terms with the magnetic field were discarded in this derivation. The quadratic terms denoted after their parameters (η_1,η_3) in the diamagnetic Zeeman matrix are expected to be negligible because the symmetry of excitonic quantum states of C_{3v} QDs are well approximated by a symmetry elevation to D_{3h}. Previous studies[38,39] of the optical spectra of excitonic complexes in C_{3v} QDs revealed the presence of this symmetry elevation; it manifested itself by an extinction of the optical activity of the "dark" exciton states, although both "dark" and "bright" exciton states are labeled with the same irrep E of the C_{3v} PG. We recall that the two exciton states $\{|d,E,1\rangle,|d,E,2\rangle\}$ are associated with the irrep E'' of D_{3h} and thus are optically inactive and have been called "dark" states so far, whereas the exciton states $\{|b,E,1\rangle,|b,E,2\rangle\}$ are associated with irrep E' and have been called "bright" states because there are optically active.

When the global symmetry of the QD is D_{3h}, an analysis of the invariant terms of the Zeeman Hamiltonian leads to fewer linear and quadratic terms. In the D_{3h} PG, the set of quadratic basis functions $(2xy, x^2 - y^2)$ transforms according to irrep E' and, likewise, the function $(x^2 + y^2)$ transforms according to A'_1; hence, the invariant terms containing $(B_x^2 - B_y^2)$ only couples excitonic states $|E',i\rangle$ (or $|E'',i\rangle$) within themselves since the decomposition of the direct products $E' \otimes E'$ (or $E'' \otimes E''$) contains E', but that of $E' \otimes E''$ does not contain it. In order to couple together "dark" and "bright" exciton states, the Zeeman interaction term must contain basis functions that transform according to an irreducible representation contained in the direct product $E' \otimes E'' = A''_1 \oplus A''_2 \oplus E''$. Looking for a quadratic Zeeman interaction term, the only set of basis functions that transforms according to E'' is (zx,zy), while for a linear Zeeman term the only set of basis functions is given by the components of the axial vector (R_x,R_y), which also transforms according to the irrep E''.

If the symmetry elevation to D_{3h} is effective in the C_{3v} QDs, the quadratic part of the Zeeman Hamiltonian will thus contain two terms associated with the effective parameters (α_1 and α_2) while its linear part will contain a single term associated with g_3 when the transverse magnetic field is oriented along the x or y axis. We emphasize that the linear Zeeman terms that are associated with the effective g factors g_1 and g_2 are strictly equal to zero in the case of D_{3h}, implying that the Zeeman doublet splittings are equal to zero in a transverse magnetic field. In a longitudinal magnetic field, the Zeeman splittings are, however, not equal to zero because the linear Zeeman terms containing the effective g factors g_4 and g_5 are allowed for symmetry reasons in the case of the PG, D_{3h}, as they were in the case of C_{3v}.

In summary, by considering the symmetry elevation to D_{3h} we are able to identify the dominant contributions to the Zeeman Hamiltonian in C_{3v} QDs for a transverse magnetic field. These contributions include a single effective g factor (g_3) and three quadratic Zeeman terms with α_1, α_2, and α_3, the third parameter being allowed only if the symmetry elevation to D_{3h} is incomplete.

TABLE V. Eigenstates of neutral excitons in a longitudinal magnetic field and corresponding polarization of the emission line. Prime values are defined in the caption of Table IV.

Eigenstates	Eigenvalues	Polarization
$\|\psi_1\rangle = \|b,E,1\rangle + \frac{(\alpha'_6+g'_6)^*}{E_1-g'_5-\alpha'_5+\delta_0/2}\|d,E,1\rangle$	E_1	σ^-
$\|\psi_2\rangle = \|b,E,2\rangle + \frac{(\alpha'_6-g'_6)^*}{E_2+g'_5-\alpha'_5+\delta_0/2}\|d,E,2\rangle$	E_2	σ^+
$\|\psi_3\rangle = \|d,E,1\rangle + \frac{(\alpha'_6+g'_6)}{E_3-g'_4-\alpha'_4-\delta_0/2}\|b,E,1\rangle$	E_3	σ^-
$\|\psi_4\rangle = \|d,E,2\rangle + \frac{(\alpha'_6-g'_6)}{E_4+g'_4-\alpha'_4-\delta_0/2}\|b,E,2\rangle$	E_4	σ^+

III. EXPERIMENTAL RESULTS

A. Sample and experimental conditions

We have studied pyramidal InGaAs/AlGaAs quantum dots by magnetophotoluminescence spectroscopy in both the Voigt and the Faraday configuration. All the quantum dots were measured from the same sample, which permitted the simultaneous observation of positively and negatively charged excitons in a single luminescence spectrum.[27] The sample was grown by low-pressure organometallic chemical vapor deposition on 2° off-(111)B GaAs substrates prepatterned with a 5-μm pitch array of inverted tetrahedral recesses.[40] The growth on a (111)B-oriented substrate is perfectly suited to obtain semiconductor QDs possessing the symmetry properties of the C_{3v} point group because the growth axis is a threefold rotation axis of cubic crystals.[25,39,41] We focused our photoluminescence study on selected pyramidal QDs for which the fine-structure splitting (FSS) of the neutral exciton was equal to zero. These pyramidal QDs were also shown in previous studies[28] to exhibit the characteristic polarized emission patterns of biexcitonic complexes resulting from optical selection rules for the C_{3v} point group. Other pyramidal QDs of the same sample had a small FSS in the range of 10–60 μeV, indicating a notable symmetry breaking from C_{3v} to C_s. The sample was mounted on the cold finger of a cryostat and maintained at a temperature of 10 K unless specified otherwise. The cryostat was inserted in the room-temperature bore of a superconducting magnet, and the magnetic field was varied between 0 and 6.5 T. The photoluminescence (PL) was excited with a cw Ti-sapphire laser tuned to 700 nm or with a Nd-YVO$_4$ laser at 532 nm; it was dispersed in a spectrograph of 55-cm focal length equipped with a nitrogen-cooled Si charged-coupled-device (CCD) array detector. The spectral resolution was 40 μeV and the spectral accuracy was ± 5 μeV by fitting the peaks with a symmetric spectral line shape. For all the investigated QDs, the linewidth corresponding to full width at half maximum was found to vary in a range between 80 and 110 μeV. Individual QDs were optically selected by focusing the laser light on the sample with a high numerical aperture microscope objective (N.A. = 0.55). The polarization analysis was realized by means of a linear polarizer mounted in front of the spectrograph slit; the linear polarization direction of the incident light was chosen by rotating a $\lambda/2$ plate positioned in front of the linear polarizer.

B. Photoluminescence of a single QD

A typical PL spectrum from a C_{3v} QD at zero magnetic field is displayed in Fig. 1(a). The spectrum of the QD is composed of a set of four major lines corresponding to radiative emission of the negatively charged exciton (X^-), of the biexciton ($2X$), of the neutral exciton (X), and of the positively charged exciton (X^+). The linear polarized emission spectra attest to the absence of any energy splitting of the neutral exciton line; this observation is the spectral signature of the doubly degenerate nature of the exciton eigenstates in a pyramidal QD with C_{3v} symmetry.[25,28] We emphasize that a symmetry lower than C_{3v} (e.g., C_{2v},C_s) is characterized by a doublet of linearly polarized emission lines, which is generally observed in the PL spectra of self-assembled InGaAs/AlGaAs QDs grown on (100) substrates, for CdSe/ZnSe QDs,[13,42] and of interfacial QDs in GaAs/AlGaAs quantum wells.[43] The weaker lines in the PL spectrum have been identified in previous works as emission lines from excitonic complexes, for which one hole occupied an excited state confined in the QD.[28,44]

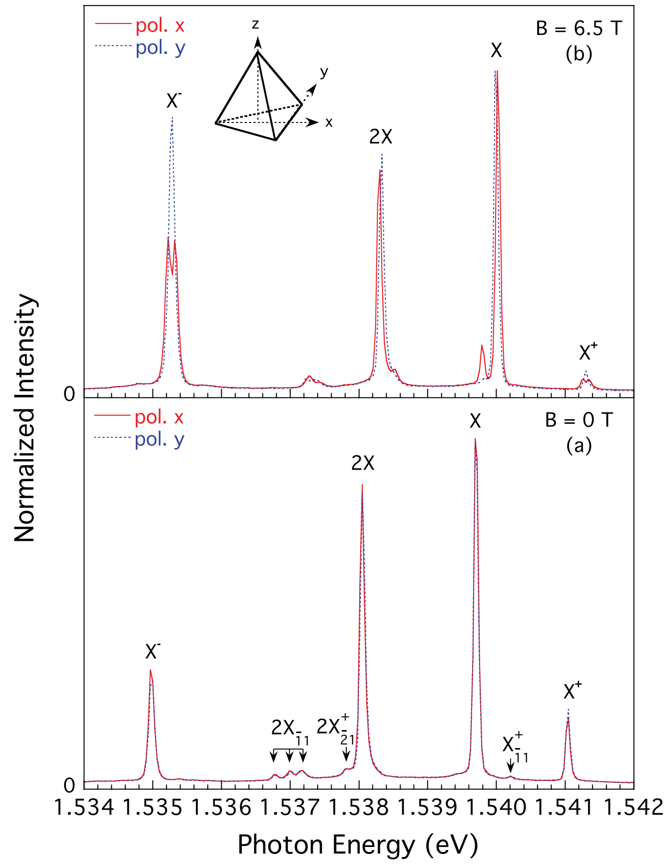

FIG. 1. (Color online) Photoluminescence spectra of a single pyramidal quantum dot in two orthogonal linear polarizations measured at 10 K (a) without and (b) with an external magnetic field of 6.5 T in the Voigt configuration. The inset shows a pyramidal QD and the orientation of the Cartesian coordinate system with respect to its threefold axis of rotation, denoted as the z axis. The external magnetic field is applied along the $[-110]$ crystalline axis of the zinc-blende lattice (y axis) and the photons are collected along the [111] axis (z axis).

C. Magnetophotoluminescence in the Voigt configuration

The application of a magnetic field in the Voigt configuration allows a clear distinction between lines originating from neutral and charged excitons. The emission pattern of the charged excitons is characterized by a set of two doublets of lines that are linearly polarized in a direction either parallel or perpendicular to the applied magnetic field. While the Zeeman energy splittings of the doublets vanish at zero magnetic field for both the charged and the neutral excitons, the energy separation between the doublets converges to zero for a charged exciton, whereas it takes a finite value for the neutral exciton.[14,20,27] In Fig. 1(b), we show the polarized photoluminescence spectra of the same QD that were recorded at a magnetic field of 6.5 T in the Voigt configuration. The pattern of emission lines from the charged excitons is composed of two doublets of linearly polarized emission lines that have nearly identical intensities, while that of the neutral exciton (or the neutral biexciton) consists of a weak doublet on the low-energy side of an intense emission doublet (note that the ordering of lines is opposite for the emission pattern of the biexciton). In preceding magneto-optical studies of QDs,[27] the

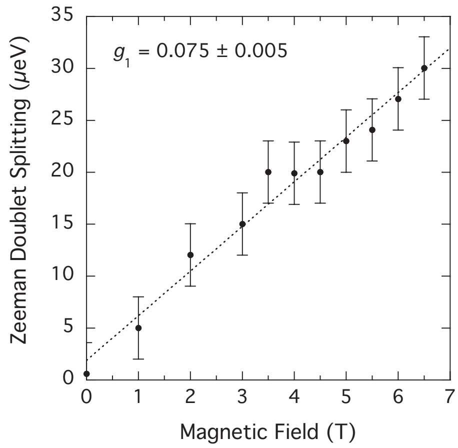

FIG. 2. Energy splitting of the Zeeman doublet associated with the "bright" exciton states of QD1 [the intense Zeeman doublet is labeled with an X in Fig. 1(b)].

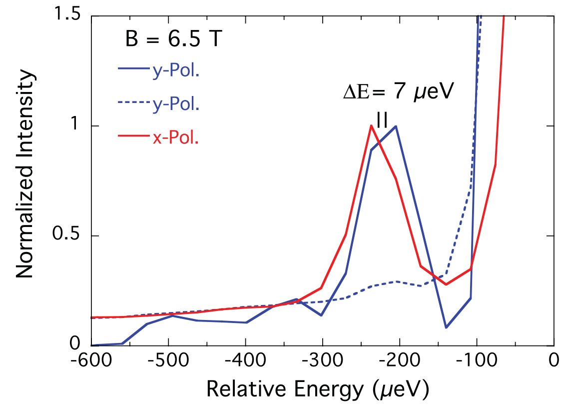

FIG. 3. (Color online) Polarized photoluminescence spectra of QD1 showing on an expanded energy scale the weak Zeeman doublet on the low-energy side of the intense Zeeman doublet. The dotted line corresponds to the original spectrum for y-polarization while the solid line is the same spectrum normalized to the weak doublet component in the x-polarization spectrum. The origin of the energy scale is taken at the position of the x-polarized component of the intense Zeeman doublet.

weak doublet was assigned to the radiatively forbidden exciton states (also known as the "dark" exciton states) of total angular momentum projection $M_z = \pm 2$, and the intense doublet was associated with the radiatively allowed exciton states (also known as the "bright" exciton states with $M_z = \pm 1$). Although seemingly similar to the optical spectra of self-assembled QDs, the analysis of their evolution in an increasing magnetic field reveals a behavior with the transverse magnetic field that is specific to the point group symmetry of C_{3v} QDs.

In Fig. 2, we display the dependence of the Zeeman energy splitting of the intense emission doublet with the magnetic field. There are two remarkable features in these data: The Zeeman splitting remains very tiny at the highest magnetic field of 6.5 T ($\approx$30 μeV), and the doublet splitting varies linearly with the magnetic field strength. This observation is in full agreement with the predicted behavior from our theoretical model of the Zeeman interaction in C_{3v} QDs. The linearity of the Zeeman splitting dependence with the magnetic field proves that the description of the interplay between the Coulomb and Zeeman interactions is crucial to the understanding of the exciton states in a transverse magnetic field. A linear fit to these data yields a value of the effective g factor $|g_1| = 0.075$, which is extremely small in comparison to typical values of effective g factors for the Zeeman splitting in the Faraday configuration (see the results in the next section). The sign of the effective g factor g_1 is determined unequivocally by the ordering of the linear polarization orientation of the intense Zeeman doublet: As the component of the Zeeman doublet on its high-energy side is linearly polarized along the x axis, the sign of g_1 is negative (see Tables I and III). Figure 3 shows an expanded view of the polarized PL spectra on the low-energy side of the intense Zeeman doublet in order to estimate the Zeeman splitting energy of the weak Zeeman doublet. At a magnetic field of 6.5 T, the intensity of the weak Zeeman doublet reaches a maximum, and the estimated value of its splitting energy is about 7 μeV. In order to evaluate the effective g factor g_2, one first needs to determine the values of the three diamagnetic coefficients (α_1,α_2,α_3) and of the effective g factor g_3 because the intensity of the weak Zeeman doublet prevents the determination of the splitting energy at other values of the magnetic field.

The off-diagonal terms of the Zeeman Hamiltonian containing g_3 and α_3 can be determined from the magnetic field dependence of the intensity of the so-called dark state transitions. In Fig. 4, we display the polarized PL spectra obtained for various values of the magnetic field in the Voigt configuration. The emission of the weak Zeeman doublet becomes observable as soon as the magnetic field strength is larger than about 3 T in this sample. Furthermore, the linearly polarized components of the weak Zeeman doublet do not have the same intensity, the intensity of the x-polarized line being always much larger than that of the y-polarized line at a given value of the magnetic field. This striking asymmetry in the intensity of the weak Zeeman doublet originates from an interference between the linear and quadratic Zeeman terms associated with the off-diagonal terms of the Zeeman Hamiltonian containing g_3 and α_3. The magnetic field dependence of each linearly polarized component of the weak Zeeman doublet is shown in Fig. 5. From a fit to expressions (16) and (17) for the intensity ratio of the linearly polarized transitions, we obtain consistent values for g_3 and α_3; they are equal to -0.204 ± 0.015 and 0.63 ± 0.02 μeV/T^2, respectively. The sign of g_3 is opposite to that of α_3 because the larger intensity corresponds to that of the x-polarized component of the weak Zeeman doublet. Note that the sign of the diamagnetic term is taken to be positive, as one would expect it. From expression (12) for the energy splitting of the "dark" exciton Zeeman doublet, it is then possible to estimate the effective g factor g_2 at a fixed magnetic field of 6.5 T: This yields the value of $g_2 = -0.038 \pm 0.02$ [the large error is compounded by the measurement accuracy of the transition energy (± 5 μeV) and the error in the determination of g_3 and α_3].

In Fig. 6, we show the magnetic field dependence of the average energy of each Zeeman doublet. Making use of the expressions for the eigenenergies given in Table I, one obtains from the fit to the data the values of the diamagnetic coefficients

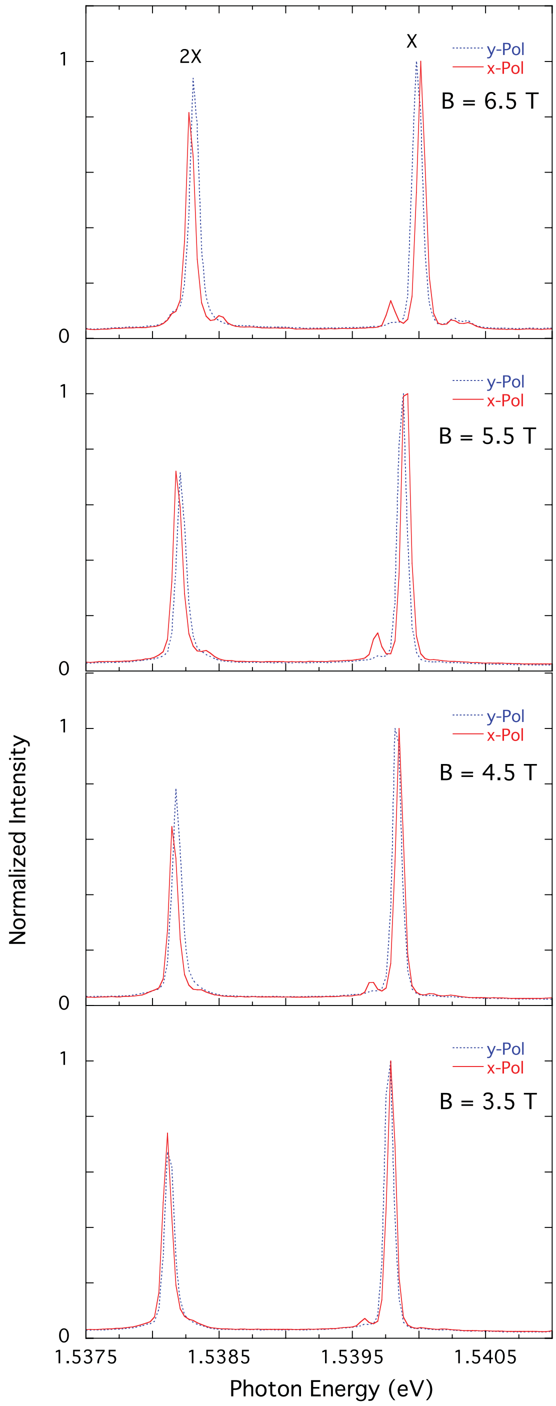

FIG. 4. (Color online) Polarized photoluminescence spectra of the pyramidal QD1 for intermediate strengths of the external magnetic field. The x-polarized component of the weak Zeeman doublet is always more intense than its y-polarized component.

$\alpha_1 = 7.05 \pm 0.06\ \mu\text{eV/T}^2$, $\alpha_2 = 7.47 \pm 0.06\ \mu\text{eV/T}^2$, and the e-h exchange parameter $\delta_0 = 191\ \mu$eV. It is instructive to note that these diamagnetic coefficients have nearly the same value; this is not surprising since the "dark" and the "bright" excitons correspond to the same carrier configuration (electron and hole in the ground state) if Coulomb correlation is neglected.

Figure 7 displays the magnetic field dependence of the Zeeman splitting of the outer lines of the quadruplet, which

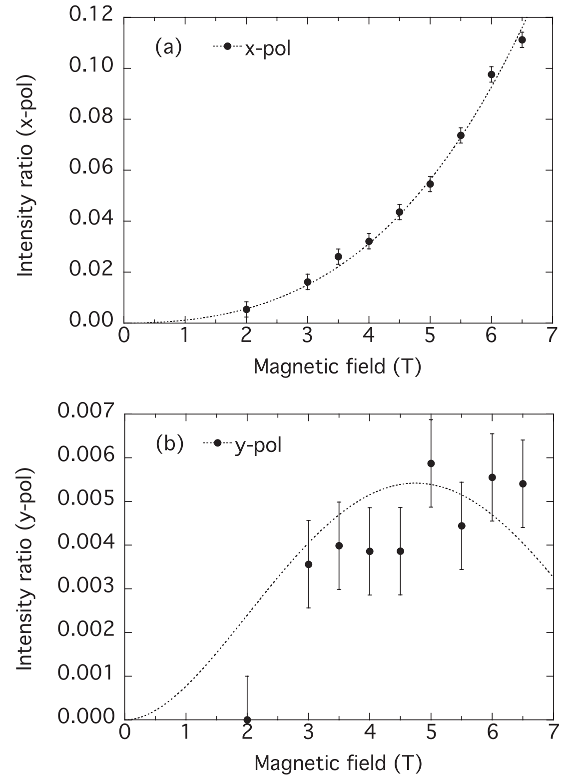

FIG. 5. Intensity ratio between one of the components of the weak Zeeman doublet and the component of the intense Zeeman doublet corresponding to the same linear polarization (a) polarization direction along the x axis and (b) polarization direction along the y axis. The dotted lines are the best fits according to theoretical expressions given in the text.

are linearly polarized along the x axis, corresponding to a direction perpendicular to the applied magnetic field. From a fit according to expression (14), one obtains a value of the e-h exchange parameter δ_0 that is identical to that obtained above, and consistent values for the parameters g_3 and α_3 (with a relative error of 60%, which is relatively large because of the size of the error bars for the photon energy).

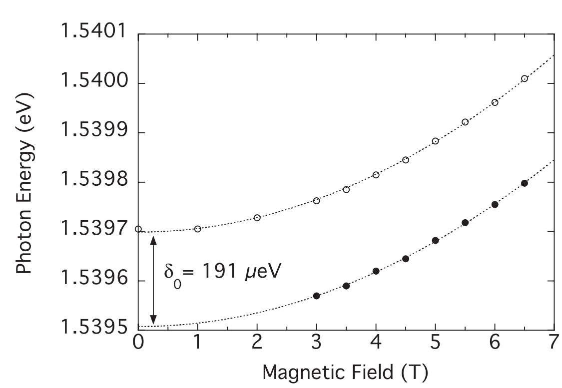

FIG. 6. Diamagnetic shift of the Zeeman doublets of QD1. Open and solid dots correspond respectively to the intense and weak doublets. The energy separation extrapolated at zero magnetic field yields the value of the electron-hole exchange energy δ_0.

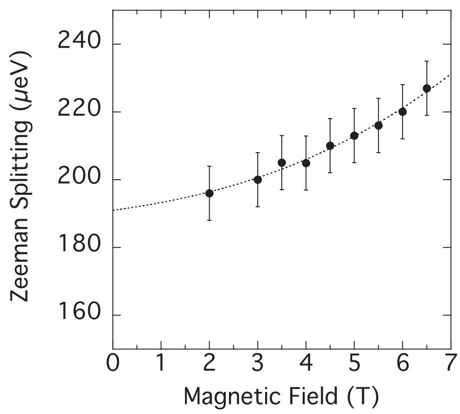

FIG. 7. Magnetic field dependence of the measured Zeeman splitting between the x-polarized lines of the neutral exciton in QD1. The dotted line is the best fit from expression (13) given in the text. The extrapolated value at zero magnetic field is the electron-hole exchange energy δ_0.

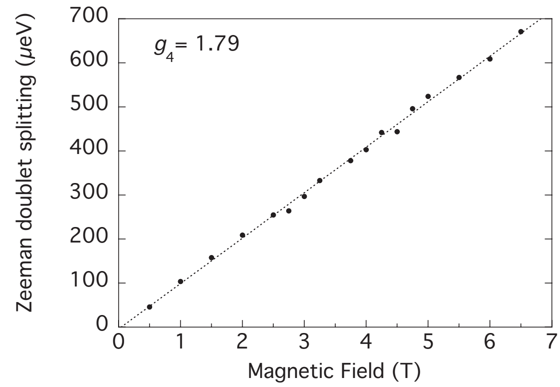

FIG. 9. Zeeman doublet splitting of the neutral exciton as a function of the applied longitudinal magnetic field B_z for another C_{3v} QD, pyramidal QD3.

D. Magnetophotoluminescence in the Faraday configuration

In Fig. 8(b), we present a typical spectrum of a C_{3v} QD in the Faraday configuration at a magnetic field of 6.5 T. The C_{3v} symmetry of this QD is identified by the characteristic emission pattern of the excited biexciton complex $2X_{11}$, which is shown in the unpolarized spectrum of Fig. 8(a) in the absence of a magnetic field. The specificity of this emission pattern is a set of three prominent lines for the radiative cascade starting from the $2X_{11}$ states and ending on the X_{01} exciton states, as we reported in Ref. 28. The other weak emission lines in the spectra were identified by the analysis of their excitation power dependences and their spectral positions (see, e.g., Ref. 44). In a longitudinal magnetic field, we find that the emission pattern of the main excitonic complexes ($X^-, 2X, X, X^+$) is composed of only one Zeeman doublet. We observe that all the emission lines are circularly polarized and that the highest-energy line of each Zeeman doublet is left circularly polarized for a magnetic field pointing upward along the z axis. These results in the Faraday configuration are in sharp contrast with those obtained in the Voigt configuration, for which the emission pattern was a quadruplet of Zeeman lines. The generality of these results was confirmed in the study of more than ten other pyramidal QDs.

In Fig. 9, we show the magnetic field dependence of the Zeeman doublet splitting of the neutral exciton. From the best fit to the data we find that the Zeeman doublet splitting varies linearly with the applied magnetic field, which yields a value of g_4 equal to 1.79 ± 0.02; we can then exclude any significant contribution to the splitting of a cubic dependence with the longitudinal magnetic field. It implies that the parameters g_6 and α_6, which were introduced in the effective Zeeman Hamiltonian in the case of C_{3v} symmetry, are equal to zero for this QD. Moreover, if these parameters had taken very small values, one would have observed a small contribution from the "dark" states $\{|d,E,1\rangle,|d,E,2\rangle\}$ to the emission pattern of the neutral exciton at the highest values of the applied magnetic field. A close inspection of the PL spectra from QD1 and QD2 displayed, respectively, in Figs. 8(b) and 10, does not reveal any weak emission lines at proximity to the Zeeman doublet associated with the neutral exciton states. The absence of the off-diagonal terms associated with the parameters g_6 and α_6 in the Zeeman Hamiltonian likely arises from the symmetry elevation from C_{3v} to D_{3h}. As the longitudinal magnetic field in the direction of the z axis transforms according to the irrep A_2' of the PG D_{3h}, the off-diagonal terms that are linear and quadratic with B_z do not exist because the irreps A_2' and A_1' are not found in the irrep decomposition of the direct product $E' \otimes E''$. Hence, the absence of hybridization in the Faraday configuration between the "bright" and the "dark" exciton states is attributed to the symmetry elevation

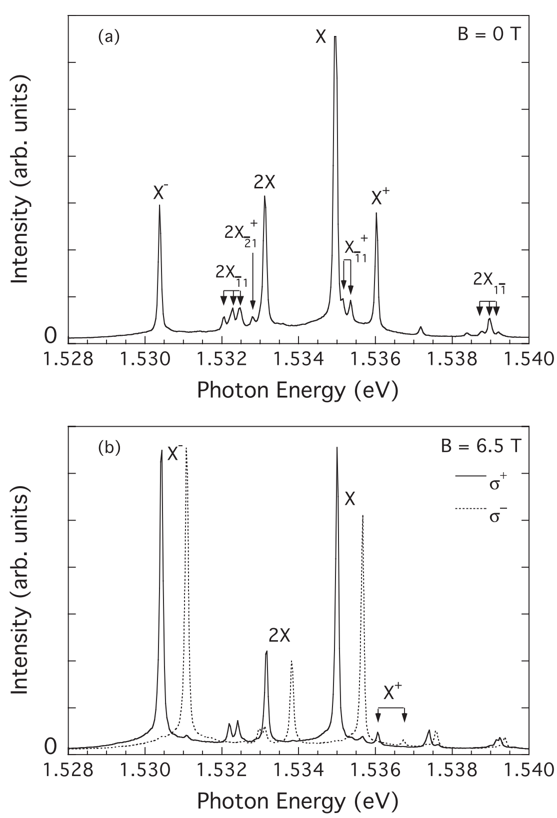

FIG. 8. Photoluminescence spectra of a C_{3v} QD, pyramidal QD2, measured in the Faraday configuration at a temperature of 32 K: (a) for 0 T and (b) for 6.5 T. The Zeeman splittings of the doublets labeled X, X^-, X^+, and $2X$ are identical in the Faraday configuration.

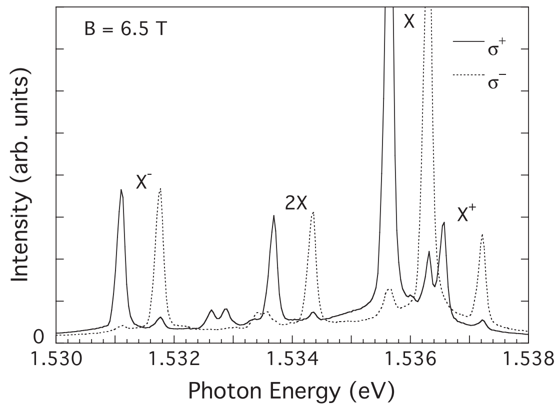

FIG. 10. Polarized photoluminescence spectra of QD3 at a magnetic field of 6.5 T and a temperature of 30.5 K. The scaling of intensity is chosen to enhance the weaker lines from X^-, $2X$, and X^+.

to D_{3h} that is present in the pyramidal QDs. Nevertheless, the hybridization between the "dark" states and the "bright" states could be restored if the magnetic field were tilted with respect to the threefold rotation axis of the QD; this would allow the measurement of the effective g factor g_5. This was not attempted, however, in our experiment because of the fixed orientation of the sample holder.

We also investigated the dispersion of the Zeeman doublet splitting of the neutral exciton in a set of 15 pyramidal QDs at the maximum value of the magnetic field. We estimated a mean value of the effective g factor associated with the neutral exciton $g_4 = 1.75$ with a standard deviation of about 0.1. We did not find any correlation between the value of the effective g factor and the emission energy of the exciton, which suggests that the dispersion among the values of the g factor is not simply related to a size variation of the QDs. The diamagnetic coefficient α_4 was determined in QD3, yielding a value of $6.3 \pm 0.1\ \mu\text{eV/T}^2$, which was typical of all the measured QDs in the Faraday configuration. This value is also comparable with diamagnetic coefficients measured in other QD systems in the intermediate to strong confinement regime.[45]

A comparison of our results with a recent study[46] of the Zeeman effect in symmetric QDs is enlightening as to the origin of the dark-bright mixing of the exciton states that is induced by a longitudinal magnetic field. In this study, the GaAs QDs were grown by droplet epitaxy on a GaAs (111)A substrate. Typical values of the fine-structure splitting (FSS) of the neutral exciton emission were of a few μeV, which led the authors to conclude that these QDs had a high symmetry corresponding to the C_{3v} point group. Nevertheless, a nonzero value of the FSS indicates an anisotropy of the electron-hole exchange interaction that results from a symmetry breaking to C_s. The radiative pattern of the neutral exciton confined to a symmetric GaAs QD was composed of four emission lines when a longitudinal magnetic field was applied, thereby revealing a mixing between the bright and the dark states of the exciton. Sallen and co-workers attributed this mixing to a coupling between the heavy-hole states with spin projections $+3/2$ and $-3/2$ onto the [111] growth axis, which is induced by a Zeeman interaction term varying linearly with the magnetic field.[27,46] Although this Zeeman interaction term is sufficient to explain the circular polarization and the Zeeman splitting of the emission lines, it alone cannot account for their relative emission intensities. Indeed, the emission intensity of the σ^+ polarized dark transition is much weaker than the σ^- polarized one. In our model, we can explain this asymmetric coupling by the interference between the contribution of the diamagnetic interaction term $\alpha_6 B_z^2$ and the linear Zeeman term $g_6 \mu_B B_z$, the existence of which is accounted for by the hole effective g factor g_{h2} in their theoretical approach.

IV. CONCLUDING REMARKS

We have studied experimentally and theoretically the radiative emission pattern of neutral excitons in highly symmetric quantum dots and their evolution in an external magnetic field. The measurements of the Zeeman doublet splittings of the exciton quantum states revealed a large anisotropy of the exciton effective g factors when the magnetic field was oriented in directions parallel and perpendicular to the [111] symmetry axis of the C_{3v} QD. We interpreted the very small Zeeman splitting of the "bright" exciton states in a transverse magnetic field to an effect of symmetry elevation from C_{3v} to D_{3h}, which nullifies the Zeeman splitting. The origin of the observed Zeeman splitting anisotropy should be distinguished, however, from the known effect of the QD shape asymmetry in self-assembled InGaAs/GaAs QDs that gives rise to a strong anisotropy of the hole effective g factor (and to a lesser extent to the electron g factor) when the magnetic field is oriented along the strong confinement direction or perpendicular to it.[18,47] The diamagnetic shifts of the "bright" exciton states were measured and corresponded to comparatively similar values of the diamagnetic coefficients ($\alpha_1 \approx 7\ \mu\text{eV/T}^2$, $\alpha_4 \approx 6\ \mu\text{eV/T}^2$) in a transverse and a longitudinal magnetic field. As the diamagnetic shift is a sensitive probe of the spatial confinement in a direction transverse to the magnetic field,[48] we infer from its insensitivity to the magnetic field orientation that the spatial extent of the exciton wave function is close to isotropic in our pyramidal quantum dots. This qualitative analysis of the diamagnetic shifts corroborates our interpretation of the small values of the Zeeman splitting for a transverse magnetic field as resulting from a peculiar effect of symmetry elevation, which is particularly effective in these pyramidal quantum dots, as was demonstrated in the analysis of their photoluminescence spectra.[28]

For the Voigt configuration, we observed that the Zeeman effect on the neutral exciton radiative pattern had two remarkable characteristics in highly symmetric QDs: a linear dependence of the Zeeman doublet splitting with the magnetic field and a nonlinear hybridization of the "dark" states with the "bright" states of the exciton. These observations in pyramidal QDs should be contrasted with those made on the neutral exciton in self-assembled QDs, for which the Zeeman splittings followed a hyperbolic dependence with a transverse magnetic field.[14] In addition to the role played by the high symmetry of the neutral exciton states in C_{3v} QDs, we highlighted the predominant intertwining between the Coulomb and Zeeman interactions to explain the linearity of the Zeeman splittings. The excellent agreement with our theory demonstrates that this is not merely an interplay between the electron-hole exchange and the Zeeman interactions. In order to fully account for this intertwining, we chose the four exciton states as the vector basis to formulate the effective Zeeman Hamiltonian. Moreover, we included in the invariant expression of the effective Zeeman Hamiltonian all the linear and diamagnetic Zeeman terms that are allowed for symmetry reasons. It should be clear that the given expression of the effective Zeeman Hamiltonian is specific to the symmetry properties of the exciton states, and might then require other invariant terms if one were to formulate an effective Zeeman Hamiltonian for another exciton complex. For instance, the common description of the charged exciton radiative pattern in an external magnetic field makes use of different effective g factors for electrons and holes, and predicts Zeeman doublet splittings that are independent of the sign of the extra charge. This prediction was, however, not validated by recent experimental studies of the Zeeman effect on charged excitons in various QD systems.[23,24,46] The strong sensitivity of the radiative pattern of charged exciton in a transverse magnetic field was attributed to a Coulomb interaction among the three carriers composing a charged exciton, or, in other words, to a charge-dependent extension of the complex wave function.[27]

In summary, our theory provides the framework to analyze the experimental radiative pattern of neutral excitons confined in pyramidal quantum dots of C_{3v} symmetry for different orientations of an applied magnetic field. We highlighted the intertwining of the Zeeman and Coulomb interactions by analyzing the magnetic field dependence of the Zeeman splittings in the neutral exciton emission spectra of single pyramidal QDs. We uncovered an interference between a linear Zeeman term and a diamagnetic term that resulted in an asymmetrical coupling of the optically forbidden exciton states to the optically allowed ones in a transverse magnetic field. Although our theoretical formalism was developed to describe the Zeeman effect on the optical spectra of the neutral exciton confined in highly symmetric QDs of C_{3v} symmetry, it can be extended to other exciton complexes, such as the charged exciton and biexciton states and to quantum dots of other symmetries. To assess the Zeeman effect on other exciton complexes, we expect the contribution of the diamagnetic Zeeman terms to be equally important as the diamagnetic shift is mainly determined by the difference of wave-function radial extension of the complex between the initial and final states of the radiative process.

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APPENDIX: ZEEMAN SPLITTINGS FOR AN EXCITED EXCITON

We consider the case of an exciton for which the electron and the hole states transform according to the same irrep $E_{1/2}$. Since the direct product of the electron and hole irreps is decomposed into $E_{1/2} \otimes E_{1/2} = A_1 + E + A_2$, the set of exciton states can be formally written as $\{|A_1\rangle, |E,1\rangle, |E,2\rangle, |A_2\rangle\}$, to which are associated the eigenenergies $(\delta_1, \delta_3, \delta_3, \delta_2)$. These eigenenergies account for the Coulomb exchange interaction between the electron and the hole.

The calculation of the matrix form of the linear part of the Zeeman Hamiltonian in the exciton state basis given above yields:

$$M_Z^{\text{lin}} = \frac{1}{2\sqrt{2}}\mu_B B_x \begin{pmatrix} 0 & g_1 & g_1 & 0 \\ g_1^* & 0 & \sqrt{2}g_3 & -g_2^* \\ g_1^* & -\sqrt{2}g_3 & 0 & g_2^* \\ 0 & -g_2 & g_2 & 0 \end{pmatrix} + \frac{i}{2\sqrt{2}}\mu_B B_y \begin{pmatrix} 0 & g_1 & -g_1 & 0 \\ -g_1^* & 0 & \sqrt{2}g_3 & g_2^* \\ g_1^* & \sqrt{2}g_3 & 0 & g_2^* \\ 0 & -g_2 & -g_2 & 0 \end{pmatrix} + \frac{1}{2}\mu_B B_z \begin{pmatrix} 0 & 0 & 0 & g_5 \\ 0 & g_4 & 0 & 0 \\ 0 & 0 & -g_4 & 0 \\ g_5^* & 0 & 0 & 0 \end{pmatrix}. \tag{A1}$$

The effective g factors are defined by the following expressions:

$$g_1 = \langle A_1 \| L^{[E]} \| E \rangle, \quad g_2 = \langle A_2 \| L^{[E]} \| E \rangle,$$
$$g_3 = \langle E \| L^{[E]} \| E \rangle, \quad g_4 = \langle E \| L^{[A_2]} \| E \rangle,$$
$$g_5 = \langle A_1 \| L^{[A_2]} \| A_2 \rangle.$$

The Hermiticity of the Zeeman Hamiltonian implies that g_4 is real and that g_3 is purely imaginary. These five parameters constitute a full set of effective g factors that completely define the linear part of the Zeeman effect for the exciton under consideration.

When the magnetic field is oriented along a symmetry axis (x, y, or z axis), the matrix form of the diamagnetic part of the Zeeman Hamiltonian depends on fewer effective parameters than for an arbitrary orientation of the magnetic field. We will

TABLE VI. Eigenenergies of the C_{3v} exciton in a transverse magnetic field (B=B_y).

$E_1 = \frac{1}{2}(\delta_1+\delta_3) - \frac{i}{2}g_3\mu_B B + \frac{1}{2}(\alpha_1+\alpha_3+\omega_2)B^2 - \frac{1}{2}\sqrt{[(\alpha_3-\alpha_1+\omega_2)B^2+(\delta_3-\delta_1)-ig_3\mu_B B]^2+\lvert 2\sqrt{2}(\omega_1-\eta_1)B^2+ig_1\mu_B B\rvert^2}$
$E_2 = \frac{1}{2}(\delta_1+\delta_3) - \frac{i}{2}g_3\mu_B B + \frac{1}{2}(\alpha_1+\alpha_3+\omega_2)B^2 + \frac{1}{2}\sqrt{[(\alpha_3-\alpha_1+\omega_2)B^2+(\delta_3-\delta_1)-ig_3\mu_B B]^2+\lvert 2\sqrt{2}(\omega_1-\eta_1)B^2+ig_1\mu_B B\rvert^2}$
$E_3 = \frac{1}{2}(\delta_3+\delta_2) + \frac{i}{2}g_3\mu_B B + \frac{1}{2}(\alpha_3+\alpha_2-\omega_2)B^2 + \frac{1}{2}\sqrt{[(\alpha_3-\alpha_2-\omega_2)B^2+(\delta_3-\delta_2)+ig_3\mu_B B]^2+\lvert 2\sqrt{2}(\omega_3-\eta_2^*)B^2+ig_2\mu_B B\rvert^2}$
$E_4 = \frac{1}{2}(\delta_3+\delta_2) + \frac{i}{2}g_3\mu_B B + \frac{1}{2}(\alpha_3+\alpha_2-\omega_2)B^2 - \frac{1}{2}\sqrt{[(\alpha_3-\alpha_2-\omega_2)B^2+(\delta_3-\delta_2)+ig_3\mu_B B]^2+\lvert 2\sqrt{2}(\omega_3-\eta_2^*)B^2+ig_2\mu_B B\rvert^2}$

TABLE VII. Eigenenergies of the C_{3v} exciton in a longitudinal magnetic field (B_z).

$$E_1 = \frac{1}{2}(\beta_1+\beta_2)B_z^2 + \frac{1}{2}(\delta_1+\delta_2) + \frac{1}{2}\sqrt{[(\beta_1-\beta_2)B_z^2+(\delta_1-\delta_2)]^2+|g_5\mu_B B_z|^2}$$
$$E_2 = \delta_3 + \beta_3 B_z^2 + \frac{1}{2}g_4\mu_B B_z$$
$$E_3 = \delta_3 + \beta_3 B_z^2 - \frac{1}{2}g_4\mu_B B_z$$
$$E_4 = \frac{1}{2}(\beta_1+\beta_2)B_z^2 + \frac{1}{2}(\delta_1+\delta_2) - \frac{1}{2}\sqrt{[(\beta_1-\beta_2)B_z^2+(\delta_1-\delta_2)]^2+|g_5\mu_B B_z|^2}$$

only give explicit expressions for these three cases:

$$M_Z^{\mathrm{dia}}(B_z) = B_z^2\begin{pmatrix}\beta_1 & 0 & 0 & 0\\ 0 & \beta_3 & 0 & 0\\ 0 & 0 & \beta_3 & 0\\ 0 & 0 & 0 & \beta_2\end{pmatrix}, \quad \text{(A2)}$$

$$M_Z^{\mathrm{dia}}(B_y) = B_y^2\begin{pmatrix}\alpha_1 & 0 & 0 & 0\\ 0 & \alpha_3 & 0 & 0\\ 0 & 0 & \alpha_3 & 0\\ 0 & 0 & 0 & \alpha_2\end{pmatrix} - B_y^2\begin{pmatrix}0 & \eta_1-\omega_1 & -\eta_1+\omega_1 & 0\\ \eta_1^*-\omega_1^* & 0 & \omega_2 & \eta_2-\omega_3^*\\ -\eta_1^*+\omega_1^* & \omega_2 & 0 & \eta_2-\omega_3^*\\ 0 & \eta_2^*-\omega_3 & \eta_2^*-\omega_3 & 0\end{pmatrix}, \quad \text{(A3)}$$

$$M_Z^{\mathrm{dia}}(B_x) = B_x^2\begin{pmatrix}\alpha_1 & 0 & 0 & 0\\ 0 & \alpha_3 & 0 & 0\\ 0 & 0 & \alpha_3 & 0\\ 0 & 0 & 0 & \alpha_2\end{pmatrix} + B_x^2\begin{pmatrix}0 & \eta_1-\omega_1 & -\eta_1+\omega_1 & 0\\ \eta_1^*-\omega_1^* & 0 & \omega_2 & \eta_2-\omega_3^*\\ -\eta_1^*+\omega_1^* & \omega_2 & 0 & \eta_2-\omega_3^*\\ 0 & \eta_2^*-\omega_3 & \eta_2^*-\omega_3 & 0\end{pmatrix}. \quad \text{(A4)}$$

The effective parameters, α_i, β_i, and ω_2 are real, and all the other parameters are complex numbers.

When symmetry elevation to D_{3h} is considered, the symmetry of the exciton states corresponds to irreps E', A_1'', and A_2'' instead of irreps E, A_1', and A_2'; it is then straightforward to show that the effective g factor $g_3 = 0$ and that the diamagnetic coefficients $\omega_1 = 0$ and $\omega_3 = 0$.

An analytical solution of the secular equation can be found, and the eigenenergies are given in Tables VI and VII for a transverse and a longitudinal magnetic field, respectively. We note that a simple analytical solution does not exist for a transverse magnetic field aligned along the x axis.

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