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Theory of fine structure of correlated exciton states in self-assembled semiconductor quantum dots in a magnetic field

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A theory of the fine structure of correlated exciton states in self-assembled parabolic semiconductor quantum dots in a magnetic field perpendicular to the quantum dot plane is presented. The correlated exciton wave function is expanded in configurations consisting of products of electron and heavy-hole 2D harmonic oscillator states (HO) in a magnetic field and the electron spin $S_z = \pm 1/2$ and a heavy-hole spin $\tau_z = \pm 3/2$ states. Analytical expressions for the short- and long-range electron-hole exchange Coulomb interaction matrix elements are derived in the HO and spin basis for arbitrary magnetic field. This allows the incorporation of short- and long-range electron-hole exchange, direct electron-hole interaction, and quantum dot anisotropy in the exact diagonalization of the exciton Hamiltonian. The fine structure of ground and excited correlated exciton states as a function of a number of confined shells, quantum dot anisotropy, and magnetic field is obtained using exact diagonalization of the many-body Hamiltonian. The effects of correlations are shown to significantly affect the energy splitting of the two bright exciton states.

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I. INTRODUCTION

Since the proposal by Benson *et al.*¹ to use the bi-exciton-exciton cascade for the generation of entangled photon pairs in self-assembled quantum dots, there has been a significant interest in the understanding of exciton states in quantum dots. Each exciton state is a linear combination of electron-hole configurations. Each electron-hole configuration is a product of orbital and spin wave functions. With electron spin $S_z = \pm 1/2$ and only heavy-hole spin $\tau = \pm 3/2$ each configuration is fourfold degenerate due to four possible spin configurations of carriers. With only the direct Coulomb interaction between carriers considered, each exciton level is also fourfold degenerate. As shown experimentally by, e.g., Gammon and coworkers²⁻⁴ and Bayer and coworkers,^{5,6} the four exciton states are split into a low-energy dark multiplet and a bright multiplet at a higher energy. The bright multiplet, which describes the two bright exciton states corresponding to left and right circularly polarized photons, is further split by the long-ranged electron-hole exchange interaction. This long-range exchange interaction (LRE) removes degeneracy of bright exciton states, leads to linear polarization of emitted photons, and prevents emission of entangled photon pairs in the bi-exciton-exciton cascade. Hence, it is important to understand the fine structure of exciton levels in self-assembled semiconductor quantum dots.

The fine structure of the exciton in bulk semiconductors was investigated almost 40 years ago.⁷⁻⁹ In semiconductor self-assembled quantum dots (QDs) the fine structure and exchange interaction are significantly enhanced due to the exciton confinement. This motivated studies of the electron-hole exchange interaction in low-dimensionality systems within the framework of the envelope function approximation by, e.g., Ivchenko and coworkers,^{10,11} Takagahara,^{12,13} as well as others.^{14,15} Zunger and coworkers investigated the exciton fine structure using the empirical atomistic pseudopotential approach¹⁶⁻¹⁸ and Goupalov and Ivchenko¹⁹ and Korkusinski *et al.*²⁰ using an atomistic tight-binding approximation.¹⁹ It

was shown that the strength of the bright exciton splitting depends on the QD in-plane anisotropy.^{5,11,12} Several groups demonstrated tuning of the exciton fine structure by the application of the lateral and vertical electric field^{21,22} or vertical and in plane magnetic field.^{5,23-25} In both cases the splitting of bright exciton states was suppressed by the external field.

In an external magnetic field perpendicular to the plane of the quantum dot the fine -structure splitting of the multiplet is caused by two contributions: the exchange interaction, which couples the spins of the electron and the hole, and their Zeeman interaction with the magnetic field. With an increasing Zeeman interaction, the two exciton states evolve from linear to circular polarization.^{5,26} The exchange interaction depends on the exciton wave function which depends on modification of the direct Coulomb scattering and the single-particle energy levels by the magnetic field.

Here we extend our effective mass theory of electron-hole exchange²⁷ to include electron-hole correlations, quantum dot anisotropy, and a perpendicular magnetic field. The correlated exciton wave function is expanded in configurations consisting of products of electron and heavy-hole 2D harmonic oscillator states (HO) in a magnetic field and the electron spin $S_z = \pm 1/2$ and a heavy-hole spin $\tau_z = \pm 3/2$ states. The key result presented here is the derivation of short- and long-range electron-hole exchange interaction matrix elements expressed in terms of known electron-electron exchange matrix elements. This allows for a simultaneous evaluation of the direct Coulomb, short- and long-range electron-hole exchange interaction matrix elements and matrix elements of the anisotropic confining potential in the HO and spin basis for a perpendicular magnetic field of arbitrary magnitude. Such a formulation of the interacting exciton Hamiltonian allows for a detailed numerical and analytical study of the effect of the electron-hole exchange, direct Coulomb interaction, anisotropy of the confining potential, quantum dot shell structure, and magnetic field on the fine structure

of correlated exciton states. The fine structure of ground and excited correlated exciton states is obtained using exact diagonalization techniques and shows the significant effect of electron-hole correlations.

While effective mass calculations do not include details of the atomistic structure,^{16–18,20,28} they are significantly less computationally expensive and allow for the development of understanding of the role of direct and exchange Coulomb interactions and the quantum dot shell structure on the fine structure of the exciton. This computational tool can help in guiding design of quantum dots with a desired exciton fine structure.

The paper is organized as follows. Section II describes the model. Section III describes an exciton confined in the anisotropic quantum dot. In Sec. IV we derive an expression for the long- and short-range electron-hole exchange matrix elements in the isotropic HO basis as well as conservation rules of the total angular momentum for electron-hole exchange matrix elements. In Sec. V we describe the computational procedure, while results and analysis of the importance of anisotropy for electron-hole exchange are presented in Sec. VI. A summary is contained in Sec. VII.

II. THE MODEL AND SINGLE-PARTICLE STATES

Motivated by experimental demonstrations of the two-dimensional parabolic potential as a good approximation for the self-assembled QDs,^{29–31} we study a quasi-two-dimensional parabolic quantum dot in the in effective mass approximation (EMA) in a magnetic field perpendicular to the x, y plane of the quantum dot. The effect of small anisotropy is included by allowing for the parabolic potentials in the two different in-plane directions to differ.

In EMA, the electron (hole) wave function ϕ is a product of the envelope function $F(H)$ and the “microscopic” wave function $u_c(u_v)$ describing the periodic part of the Bloch functions at the Γ point for the conduction (valence) band states:

$$\phi_{c,l,\sigma}(\mathbf{r}) = F_{l\sigma}(\mathbf{r})u_{c,\sigma}(\mathbf{r}), \quad \sigma \rightarrow j_z = -1/2, 1/2 \quad (\downarrow\uparrow),$$

$$\phi_{v,j,\tau}(\mathbf{r}) = H_{j\tau}(\mathbf{r})u_{v,\tau}(\mathbf{r}),$$

$$\tau \rightarrow j_z = -3/2, -1/2, 1/2, 3/2 \quad (\downarrow\downarrow\uparrow\uparrow).$$

Wave functions are labeled by their band index $c(v)$, spin $\sigma(\tau)$, and envelope quantum numbers $l(j)$. Due to the strain in QDs, light-hole ($\tau_{lh} = \pm 1/2$) and heavy-hole ($\tau_{hh} = \pm 3/2$) levels at the top of the valence band are split.³² This allows us to neglect light holes and consider only heavy-hole states ($\tau_{hh} = \pm 3/2$). We measure the energy in effective Rydbergs ($\text{Ry}^* = m_e^* e^4 / 2\varepsilon^2 \hbar^2$) and distances in Bohr radii ($a_B = \varepsilon \hbar^2 / m_e^* e^2$), where m_e^* is the effective mass of the electron, e is the electron charge, and ε is the dielectric constant of the material. $\hbar$ is the reduced Planck constant.

We define our basis in terms of eigenstates of the isotropic parabolic quantum dot. The envelope functions $F_{l\sigma}(\mathbf{r})(H_{j\tau}(\mathbf{r}))$ are single-particle states of the electron (hole) in an isotropic parabolic confining potential with characteristic frequency $\Omega_0^{e(h)}$ in the presence of the perpendicular magnetic field. The

Hamiltonian of the electron confined in such a QD reads

$$\hat{H}_e = -\left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}\right) + \frac{1}{4}\Omega_h^{e2}(x^2 + y^2) - \frac{i}{2}\Omega_c^e\left(x\frac{\partial}{\partial y} - y\frac{\partial}{\partial x}\right) + g_e\mu_B B\sigma,$$

while for the hole it reads

$$\hat{H}_h = -\frac{m_e}{m_h}\left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}\right) + \frac{1}{4}\frac{m_h}{m_e}\Omega_h^{h2}(x^2 + y^2) + \frac{i}{2}\Omega_c^h\left(x\frac{\partial}{\partial y} - y\frac{\partial}{\partial x}\right) + g_h\mu_B B\tau,$$

where the hybrid frequency $\Omega_h^{e(h)} = \sqrt{\Omega_0^{e(h)2} + \frac{1}{4}\Omega_c^{e(h)2}}$ and the cyclotron energy $\Omega_c^{e(h)} = eB/m_{e(h)}^*c$ are expressed in Rydbergs. The fourth term is the Zeeman energy with $g_{e(h)}$ being the Lande factor of the electron (hole) and μ_B is the Bohr magneton.

After introducing new variables, $z = x - iy$, $z^* = x + iy$, $\partial_z = \partial_x + i\partial_y$, and $\partial_{z^*} = \partial_x - i\partial_y$, we express the harmonic oscillator raising and lowering operators as

$$a = \frac{1}{2}\left(\frac{1}{\sqrt{2}}\frac{z}{l_h^e} + \sqrt{2}l_h^e\partial_{z^*}\right), \quad b = \frac{1}{2}\left(\frac{1}{\sqrt{2}}\frac{z^*}{l_h^e} + \sqrt{2}l_h^e\partial_z\right), \\ a^+ = \frac{1}{2}\left(\frac{1}{\sqrt{2}}\frac{z^*}{l_h^e} - \sqrt{2}l_h^e\partial_z\right), \quad b^+ = \frac{1}{2}\left(\frac{1}{\sqrt{2}}\frac{z}{l_h^e} - \sqrt{2}l_h^e\partial_{z^*}\right), \quad (1)$$

with similar operators for the hole, and $l_h^{e(h)} = \frac{1}{\sqrt{\Omega_h^{e(h)}}}$. The Hamiltonian for the electron (hole) in a magnetic field can be now rewritten as a sum of two harmonic oscillators as follows:

$$\hat{H}_e = \Omega_+^e\left(a^+a + \frac{1}{2}\right) + \Omega_-^e\left(b^+b + \frac{1}{2}\right) + g_e\mu_B B\sigma,$$

where $\Omega_{\pm}^{e(h)} = \Omega_h^{e(h)} \pm \frac{1}{2}\Omega_c^{e(h)}$.

The electron envelope wave function $F_{l\sigma}(\mathbf{r})$ with $l = \{n_+, n_-\}$, written in the language of HO operators, has the form $|n_+, n_-, \sigma\rangle = (a^+)^{n_+}(b^+)^{n_-}|00\rangle/\sqrt{n_+!n_-!}\chi(\sigma)$ (a similar expression holds for the hole) and the eigenenergy $\varepsilon_{n_+, n_-, \sigma}^{e(h)} = \Omega_+^{e(h)}(n_+ + \frac{1}{2}) + \Omega_-^{e(h)}(n_- + \frac{1}{2}) + g_{e(h)}\mu_B B\sigma$. The action of ladder operators on HO states is as follows: $a^+|n_+, n_-\rangle = \sqrt{n_+ + 1}|n_+ + 1, n_-\rangle$, $a|n_+, n_-\rangle = \sqrt{n_+}|n_+ - 1, n_-\rangle$, $b^+|n_+, n_-\rangle = \sqrt{n_- + 1}|n_+, n_- + 1\rangle$, $b|n_+, n_-\rangle = \sqrt{n_-}|n_+, n_- - 1\rangle$.

Since the angular momentum for the electron is defined as $L_e = n_+ - n_-$ while for the hole $L_h = n_- - n_+$, operators $a^+(b^+)$ create an excitation of electron with a positive (negative) angular momentum, while $c^+(d^+)$ create negative (positive) angular-momentum excitations of a hole state. Since the wave function $F_{l\sigma}(\mathbf{r})$ is the same as $F_{l-\sigma}(\mathbf{r})$, we drop the symbol denoting spin σ in the envelope function; however the spin dependence in the Bloch part of the wave function is kept. If $m_e\omega_0^e = m_h\omega_0^h$, the Hamiltonian for the hole is $H_h = \frac{m_e}{m_h}(H_e)^*$. In this case there are the following relations between electron and hole energies and wave functions: $E_h = E_e \frac{m_e}{m_h}$ and $H_h(r) = F_e^*(r)$.

We now introduce a small anisotropy in the confining potential. We assume here that the single-particle lateral confinement of the dot is elliptical and can be described by

two characteristic frequencies, Ω_x and Ω_y . The characteristic frequency of the cylindrically symmetric component of the parabolic confinement can now be expressed in terms of two confinement frequencies as $\Omega_{0,e(h)}^2 = \frac{1}{2}(\Omega_{x,e(h)}^2 + \Omega_{y,e(h)}^2)$.

The anisotropic component is proportional to the anisotropy parameter $\gamma_{e(h)} = (\Omega_{x,e(h)}^2 - \Omega_{y,e(h)}^2)/(\Omega_{x,e(h)}^2 + \Omega_{y,e(h)}^2)$. The anisotropic Hamiltonian $\delta\hat{H}_e$ for an electron (hole) can now be written in terms of matrix elements

$$\begin{aligned} \langle i(n_+, n_-) | \delta\hat{H}_{e(h)} | j(k_+, k_-) \rangle = & \frac{\Omega_{0,e(h)}^2}{4\Omega_{e(h)}^2} \gamma_{e(h)} \{ \sqrt{k_+(k_+ - 1)} \delta_{n_-, k_+ - 2} \delta_{n_+, k_-} + \sqrt{(k_+ + 1)(k_+ + 2)} \delta_{n_-, k_+ + 2} \delta_{n_+, k_-} \\ & + \sqrt{k_-(k_- - 1)} \delta_{n_-, k_+} \delta_{n_+, k_- - 2} + \sqrt{(k_- + 1)(k_- + 2)} \delta_{n_-, k_+} \delta_{n_+, k_- + 2} \\ & + 2\sqrt{(k_+ + 1)k_-} \delta_{n_-, k_+ + 1} \delta_{n_+, k_- - 1} + 2\sqrt{k_+(k_- + 1)} \delta_{n_-, k_+ - 1} \delta_{n_+, k_- + 1} \}. \end{aligned} \quad (2)$$

The anisotropic corrections to the confining potential break the rotational symmetry of the quantum dot and mix single-particle states with angular momenta which differ by 2.

III. EXCITON

The exciton is a correlated state of an electron in the conduction band and a hole in the valence band. Detailed theory of the exciton as a collective state of electron and a hole in a symmetrical parabolic quantum dot has been developed in Ref. 33. The effect of the quantum dot anisotropy on the exciton p shell has been discussed in Ref. 34.

With annihilation (creation) operators $c_{i\sigma}(c_{i\sigma}^\dagger)$ for the electron and $h_{i\tau}(h_{i\tau}^\dagger)$ for the hole acting on the isotropic oscillator state $i = (n_+, n_-)$ with the spin $\sigma(\tau)$, the exciton Hamiltonian $\hat{H}_X$ consists of three terms:

$$\hat{H}_X = \hat{H}_{EH} + \hat{H}_{anis} + \hat{H}_{EHX}. \quad (3)$$

The first term,

$$\begin{aligned} \hat{H}_{EH} = & \sum_{i,\tau} \varepsilon_{i,\tau}^h h_{i,\tau}^\dagger h_{i,\tau} + \sum_{i,\sigma} \varepsilon_{i,\sigma}^e c_{i,\sigma}^\dagger c_{i,\sigma} \\ & - \sum_{ijkl,\sigma\tau} \langle i, j | V | k, l \rangle c_{i\sigma}^\dagger h_{j\tau}^\dagger h_{k\tau} c_{l\sigma}, \end{aligned} \quad (4)$$

is the exciton Hamiltonian of the isotropic parabolic quantum dot with single-particle HO energies $\varepsilon_{i,\tau}^{e(h)}$ of the electron (hole) HO and direct Coulomb interaction matrix elements $\langle i, j | V | k, l \rangle$.

The second term in the Hamiltonian, Eq. (3),

$$\hat{H}_{anis} = \delta\hat{H}_e + \delta\hat{H}_h = \sum_{ij\sigma} t_{ij}^e c_{i\sigma}^\dagger c_{j\sigma} + \sum_{ij\tau} t_{ij}^h h_{i\tau}^\dagger h_{j\tau},$$

introduces the anisotropic confining potential described by Eq. (2).

The third term in the Hamiltonian Eq. (3) is the electron-hole exchange term,

$$\hat{H}_{EHX} = \sum_{ijkl\sigma\sigma'\tau\tau'} \langle i\sigma, j\tau | V_{eh}^X | k\tau', l\sigma' \rangle c_{i\sigma}^\dagger h_{j\tau}^\dagger h_{k\tau'} c_{l\sigma'}.$$

Note that unlike the direct Coulomb matrix element, which has been calculated previously,^{35,36} electron-hole exchange matrix

elements, which depend on spin indices, have not been reported in the HO basis. They are discussed in detail in the next section.

IV. EXCHANGE COULOMB MATRIX ELEMENTS

We now turn to the main goal of this work, the derivation of electron-hole exchange matrix elements. The exchange matrix elements $\langle i\sigma, j\tau | V_{eh}^X | k\tau', l\sigma' \rangle$ can be divided into two groups according to their effect on the excitonic levels: diagonal in spin subspace,²⁷ which cause the splitting between bright ($|\downarrow\uparrow\rangle, |\uparrow\downarrow\rangle$) and dark ($|\downarrow\downarrow\rangle, |\uparrow\uparrow\rangle$) doublets, and off-diagonal in spin subspace, responsible for the bright doublet splitting by coupling states with the spin angular momentum ± 1 .

A. Electron-hole exchange mixing bright exciton states

The electron-hole long-range exchange matrix elements responsible for bright exciton doublet splitting have been derived in Ref. 27 and expressed in terms of electron and hole envelope wave functions:

$$\begin{aligned} \langle i\downarrow, j\uparrow | V_{eh}^X | k\downarrow, l\uparrow \rangle \\ = \mu^2 \int \int d\mathbf{r}_1 d\mathbf{r}_2 \left[\left(\frac{\partial}{\partial x_1} - i \frac{\partial}{\partial y_1} \right)^2 F_i^+(\mathbf{r}_1) H_j^+(\mathbf{r}_1) \right] \\ \times \frac{2}{|\mathbf{r}_1 - \mathbf{r}_2|} (H_k(\mathbf{r}_2) F_l(\mathbf{r}_2)), \end{aligned} \quad (5)$$

where μ^2 is a numerical constant parameterizing the long-range exchange interaction.²⁷ Matrix element $\langle i\uparrow, j\downarrow | V_{eh}^X | k\uparrow, l\downarrow \rangle$ is a Hermitian conjugate of $\langle i\downarrow, j\uparrow | V_{eh}^X | k\downarrow, l\uparrow \rangle$. Note that these matrix elements are not simply matrix elements of the Coulomb interaction $\frac{2}{|\mathbf{r}_1 - \mathbf{r}_2|}$. The long-range exchange interaction involves interaction of dipoles created by two different electron-hole pairs, (i, j) and (k, l) .^{12,13,19,27} The interaction of dipoles has been translated into Coulomb interaction of the charge (k, l) with derivatives of the charges (i, j) .

We can express the differentiation operators in Eq. (5), acting on a product of the electron and hole wave function, in terms of differentiation operations on the electron and on the

hole wave function:

$$\left(\frac{\partial^2}{\partial x_1^2} - \frac{\partial^2}{\partial y_1^2} - 2i\frac{\partial^2}{\partial x_1 \partial y_1}\right) F_i^+(\mathbf{r}_1) H_j^+(\mathbf{r}_1) = \left[\left(\frac{\partial}{\partial x_1} - i\frac{\partial}{\partial y_1}\right)^2 F_i^+(\mathbf{r}_1)\right] H_j^+(\mathbf{r}_1) + F_i^+(\mathbf{r}_1) \left[\left(\frac{\partial}{\partial x_1} - i\frac{\partial}{\partial y_1}\right)^2 H_j^+(\mathbf{r}_1)\right] + 2\left[\left(\frac{\partial}{\partial x_1} - i\frac{\partial}{\partial y_1}\right) F_i^+(\mathbf{r}_1)\right] \left[\left(\frac{\partial}{\partial x_1} - i\frac{\partial}{\partial y_1}\right) H_j^+(\mathbf{r}_1)\right].$$

Expressing differentiation operators in terms of boson creation and annihilation operators as described in Eq. (1) allows us to write

$$\left(\frac{\partial^2}{\partial x_1^2} - \frac{\partial^2}{\partial y_1^2} - 2i\frac{\partial^2}{\partial x_1 \partial y_1}\right) F_i^+(\mathbf{r}_1) H_j^+(\mathbf{r}_1) = \left\{ \left[\frac{1}{\sqrt{2}l_h^e} (a - b^+) \right]^2 F_i^+(\mathbf{r}_1) \right\} H_j^+(\mathbf{r}_1) + F_i^+(\mathbf{r}_1) \left\{ \left[\frac{1}{\sqrt{2}l_h^e} (d - c^+) \right]^2 H_j^+(\mathbf{r}_1) \right\} + 2 \left\{ \left[\frac{1}{\sqrt{2}l_h^e} (a - b^+) \right] F_i^+(\mathbf{r}_1) \right\} \left\{ \left[\frac{1}{\sqrt{2}l_h^e} (d - c^+) \right] H_j^+(\mathbf{r}_1) \right\}.$$

By employing these operators and after some algebra we express the electron-hole long-range exchange matrix elements in terms of the electron-hole Coulomb matrix elements V_{eh}^{EX} :

$$\begin{aligned} & \langle i(n_+^e, n_-^e) \downarrow; j(n_+^h, n_-^h) \uparrow | V_{eh}^X | k(n_+^{h'}, n_-^{h'}) \downarrow; l(n_+^{e'}, n_-^{e'}) \uparrow; \rangle \\ &= -\mu^2 \left\{ \frac{1}{2l_h^e} [\sqrt{n_-^e(n_-^e - 1)} V_{eh}^{EX}(n_+^e, n_-^e - 2; n_+^h, n_-^h | n_+^{h'}, n_-^{h'}; n_+^{e'}, n_-^{e'}) \right. \\ &+ \sqrt{(n_+^e + 1)(n_+^e + 2)} V_{eh}^{EX}(n_+^e + 2, n_-^e; n_+^h, n_-^h | n_+^{h'}, n_-^{h'}; n_+^{e'}, n_-^{e'}) \\ &- 2\sqrt{n_-^e(n_+^e + 1)} V_{eh}^{EX}(n_+^e + 1, n_-^e - 1; n_+^h, n_-^h | n_+^{h'}, n_-^{h'}; n_+^{e'}, n_-^{e'})] \\ &+ \frac{1}{2l_h^h} [\sqrt{n_+^h(n_+^h - 1)} V_{eh}^{EX}(n_+^e, n_-^e; n_+^h - 2, n_-^h | n_+^{h'}, n_-^{h'}; n_+^{e'}, n_-^{e'}) \\ &+ \sqrt{(n_-^h + 1)(n_-^h + 2)} V_{eh}^{EX}(n_+^e, n_-^e; n_+^h, n_-^h + 2 | n_+^{h'}, n_-^{h'}; n_+^{e'}, n_-^{e'}) \\ &- 2\sqrt{n_+^h(n_-^h + 1)} V_{eh}^{EX}(n_+^e, n_-^e; n_+^h - 1, n_-^h + 1 | n_+^{h'}, n_-^{h'}; n_+^{e'}, n_-^{e'})] \\ &+ \frac{1}{l_h^e l_h^h} [\sqrt{n_-^e n_+^h} V_{eh}^{EX}(n_+^e, n_-^e - 1; n_+^h - 1, n_-^h | n_+^{h'}, n_-^{h'}; n_+^{e'}, n_-^{e'}) \\ &- \sqrt{n_-^e(n_-^h + 1)} V_{eh}^{EX}(n_+^e, n_-^e - 1; n_+^h, n_-^h + 1 | n_+^{h'}, n_-^{h'}; n_+^{e'}, n_-^{e'}) \\ &- \sqrt{n_+^h(n_+^e + 1)} V_{eh}^{EX}(n_+^e + 1, n_-^e; n_+^h - 1, n_-^h | n_+^{h'}, n_-^{h'}; n_+^{e'}, n_-^{e'}) \\ &\left. + \sqrt{(n_+^e + 1)(n_-^h + 1)} V_{eh}^{EX}(n_+^e + 1, n_-^e; n_+^h, n_-^h + 1 | n_+^{h'}, n_-^{h'}; n_+^{e'}, n_-^{e'}) \right\}, \end{aligned} \quad (6)$$

where

$$V_{eh}^{EX}(n_+^e, n_-^e; n_+^h, n_-^h | n_+^{h'}, n_-^{h'}; n_+^{e'}, n_-^{e'}) = \iint d\mathbf{r}_1 d\mathbf{r}_2 F_{i(n_+^e, n_-^e)}^+(\mathbf{r}_1) H_{j(n_+^h, n_-^h)}^+(\mathbf{r}_1) \frac{2}{|\mathbf{r}_1 - \mathbf{r}_2|} H_{k(n_+^{h'}, n_-^{h'})}(\mathbf{r}_2) F_{l(n_+^{e'}, n_-^{e'})}(\mathbf{r}_2). \quad (7)$$

This is the central result of this work. In the general case the above expression has to be evaluated numerically; however, in the case of $l_h^e = l_h^h$, the hole wave function $H_{j(n_+^h, n_-^h)}^+(\mathbf{r}) = F_{j(n_+^h, n_-^h)}^+(\mathbf{r})$. In such case the integrals in Eqs. (6) and (7) can be identified as the electron-electron exchange elements,

$$V_{eh}^{EX}(n_+^e, n_-^e; n_+^h, n_-^h | n_+^{h'}, n_-^{h'}; n_+^{e'}, n_-^{e'}) = V_{ee}^{EX}(n_+^e, n_-^e; n_+^{h'}, n_-^{h'} | n_+^{e'}, n_-^{e'}; n_+^h, n_-^h),$$

which can be computed analytically.^{36,37} The quantum numbers corresponding to the hole changed place due to the Hermitian conjugation of the hole wave function, as shown above, and the conventionally chosen order of the wave functions in the element V_{ee}^{EX} ,

$$V_{ee}^{EX}(n_+^e, n_-^e; n_+^{h'}, n_-^{h'} | n_+^{e'}, n_-^{e'}; n_+^h, n_-^h) = \iint d\mathbf{r}_1 d\mathbf{r}_2 F_{i(n_+^e, n_-^e)}^+(\mathbf{r}_1) F_{k(n_+^{h'}, n_-^{h'})}^+(\mathbf{r}_2) \frac{2}{|\mathbf{r}_1 - \mathbf{r}_2|} F_{l(n_+^{e'}, n_-^{e'})}(\mathbf{r}_2) F_{j(n_+^h, n_-^h)}(\mathbf{r}_1).$$

The orbital angular-momentum conservation rules for electron-electron Coulomb integrals stipulate that the z projection of the total orbital angular momentum of a pair on the right and that on the left have to be equal, otherwise the integral is equal to zero. Knowing that, as well as the expression for the orbital angular momentum of electron-electron pair $L^{ee} = (n_+^{(1)} - n_-^{(1)}) + (n_+^{(2)} - n_-^{(2)})$ and for electron-hole pair $L^{eh} = (n_+^{(1)} - n_-^{(1)}) - (n_+^{(2)} - n_-^{(2)})$, we find conservation rules for the matrix elements $\langle i(n_+^e, n_-^e) \downarrow; j(n_+^h, n_-^h) \uparrow | V_{eh}^X | k(n_+^{h'}, n_-^{h'}) \downarrow; l(n_+^{e'}, n_-^{e'}) \uparrow \rangle$. If we denote $\mathcal{L}_L = (n_+^e - n_-^e) - (n_+^h - n_-^h)$ as the angular momentum of the electron-hole pair on the left and $\mathcal{L}_R = (n_+^{e'} - n_-^{e'}) - (n_+^{h'} - n_-^{h'})$ as that of the pair on the right of the matrix element, then the orbital angular-momentum conservation rule for this matrix element is $\mathcal{L}_L = \mathcal{L}_R - 2$. At the same time, the above matrix element is nonzero only for one specific set of spin quantum numbers, $(\sigma, \tau) = (+3/2, -1/2)$

and $(\sigma', \tau') = (-3/2, +1/2)$, for which the electron-hole pair spin conservation rule is $\mathcal{S}_L = \mathcal{S}_R + 2$. Hence, the total angular momentum of a pair, a sum of the orbital and spin angular momenta, is conserved, $\mathcal{J}_L = \mathcal{L}_L + \mathcal{S}_L = \mathcal{L}_R + \mathcal{S}_R = \mathcal{J}_R$. We conclude that the long-range electron-hole exchange can flip the spin of the carriers with simultaneous transfer of at least one particle to a different single-particle orbital so the total angular-momentum conservation rule is fulfilled.

B. Electron-hole exchange responsible for the bright-dark exciton splitting

In the previous section we provided an analysis of the bright exciton splitting. Here we analyze the splitting of bright and dark exciton states, typically attributed to just the short-range exchange.

Electron-hole exchange matrix elements $\langle i\uparrow, j\downarrow | V_{eh}^X | k\downarrow, l\uparrow \rangle = \langle i\uparrow, j\downarrow | \delta_0^{SL} | k\downarrow, l\uparrow \rangle + \langle i\uparrow, j\downarrow | \delta^L | k\downarrow, l\uparrow \rangle$, which are diagonal in the spin subspace, are a sum of the local short- and long-range exchange δ_0^{SLkl} , proportional to the overlap between the two electron-hole pairs and a nonlocal term δ^{Lkl} which involves differentiation operators applied to electron-hole pair envelopes.²⁷ We have $\langle i\downarrow, j\uparrow | V_{eh}^X | k\uparrow, l\downarrow \rangle = \langle i\uparrow, j\downarrow | V_{eh}^X | k\downarrow, l\uparrow \rangle$.

Let us look first at the nonlocal long-range contribution to the diagonal terms

$$\langle i\uparrow, j\downarrow | \delta^L | k\downarrow, l\uparrow \rangle = -\frac{\mu^2}{2} \int \int d\mathbf{r}_1 d\mathbf{r}_2 \left[\left(\frac{\partial^2}{\partial x_1^2} + \frac{\partial^2}{\partial y_1^2} \right) F_i^+(\mathbf{r}_1) H_j^+(\mathbf{r}_1) \right] \frac{2}{|\mathbf{r}_1 - \mathbf{r}_2|} H_k(\mathbf{r}_2) F_l(\mathbf{r}_2), \quad (8)$$

where, as previously, the differentiation operator can be expressed in terms of electron or hole ladder operators depending on the type of envelope it is acting on,

$$\begin{aligned} \left(\frac{\partial^2}{\partial x_1^2} + \frac{\partial^2}{\partial y_1^2} \right) F_i^+(\mathbf{r}_1) H_j^+(\mathbf{r}_1) &= \frac{1}{2l_h^2} [(b - a^+)(a - b^+) F_i^+(\mathbf{r}_1)] H_j^+(\mathbf{r}_1) + \frac{1}{2l_h^2 l_h^h} [(a - b^+) F_i^+(\mathbf{r}_1)] [(c - d^+) H_j^+(\mathbf{r}_1)] \\ &+ \frac{1}{2l_h^2 l_h^h} [(b - a^+) F_i^+(\mathbf{r}_1)] [(d - c^+) H_j^+(\mathbf{r}_1)] + \frac{1}{2l_h^2} F_i^+(\mathbf{r}_1) [(c - d^+)(d - c^+) H_j^+(\mathbf{r}_1)]. \end{aligned}$$

After applying the ladder operators to the envelope functions and substituting this expression to Eq. (8), the matrix element δ^{Lkl} can be written as a sum of electron-hole Coulomb matrix elements V_{eh}^{EX} ,

$$\begin{aligned} &\langle i(n_+^e, n_-^e) \uparrow; j(n_+^h, n_-^h) \downarrow | \delta^L | k(n_+^{h'}, n_-^{h'}) \downarrow; l(n_+^{e'}, n_-^{e'}) \uparrow \rangle \\ &= -\frac{\mu^2}{2} \left\{ \frac{1}{l_h^2} [\sqrt{n_+^e n_-^e} V_{eh}^{\text{EX}}(n_+^e - 1, n_-^e - 1; n_+^h, n_-^h | n_+^{h'}, n_-^{h'}; n_+^{e'}, n_-^{e'}) \right. \\ &\quad + \sqrt{(n_+^e + 1)(n_-^e + 1)} V_{eh}^{\text{EX}}(n_+^e + 1, n_-^e + 1; n_+^h, n_-^h | n_+^{h'}, n_-^{h'}; n_+^{e'}, n_-^{e'}) \\ &\quad - (n_+^e + n_-^e + 1) V_{eh}^{\text{EX}}(n_+^e, n_-^e; n_+^h, n_-^h | n_+^{h'}, n_-^{h'}; n_+^{e'}, n_-^{e'})] \\ &\quad + \frac{1}{l_h^2} [\sqrt{n_+^h n_-^h} V_{eh}^{\text{EX}}(n_+^e, n_-^e; n_+^h - 1, n_-^h - 1 | n_+^{h'}, n_-^{h'}; n_+^{e'}, n_-^{e'}) \\ &\quad + \sqrt{(n_+^h + 1)(n_-^h + 1)} V_{eh}^{\text{EX}}(n_+^e, n_-^e; n_+^h + 1, n_-^h + 1 | n_+^{h'}, n_-^{h'}; n_+^{e'}, n_-^{e'}) \\ &\quad - (n_+^h + n_-^h + 1) V_{eh}^{\text{EX}}(n_+^e, n_-^e; n_+^h, n_-^h | n_+^{h'}, n_-^{h'}; n_+^{e'}, n_-^{e'})] \\ &\quad + \frac{1}{l_h^2 l_h^h} [\sqrt{n_+^e n_+^h} V_{eh}^{\text{EX}}(n_+^e - 1, n_-^e; n_+^h - 1, n_-^h | n_+^{h'}, n_-^{h'}; n_+^{e'}, n_-^{e'}) \\ &\quad - \sqrt{n_+^e (n_-^h + 1)} V_{eh}^{\text{EX}}(n_+^e - 1, n_-^e; n_+^h, n_-^h + 1 | n_+^{h'}, n_-^{h'}; n_+^{e'}, n_-^{e'}) \\ &\quad - \sqrt{n_+^h (n_-^e + 1)} V_{eh}^{\text{EX}}(n_+^e, n_-^e + 1; n_+^h - 1, n_-^h | n_+^{h'}, n_-^{h'}; n_+^{e'}, n_-^{e'}) \\ &\quad + \sqrt{(n_-^e + 1)(n_-^h + 1)} V_{eh}^{\text{EX}}(n_+^e, n_-^e + 1; n_+^h, n_-^h + 1 | n_+^{h'}, n_-^{h'}; n_+^{e'}, n_-^{e'}) \\ &\quad + \sqrt{n_-^e n_-^h} V_{eh}^{\text{EX}}(n_+^e, n_-^e - 1, n_+^{h'}, n_-^{h'} | n_+^{e'}, n_-^{e'}, n_+^h, n_-^h - 1) \\ &\quad - \sqrt{n_+^e (n_+^h + 1)} V_{eh}^{\text{EX}}(n_+^e, n_-^e - 1; n_+^h + 1, n_-^h | n_+^{h'}, n_-^{h'}; n_+^{e'}, n_-^{e'}) \\ &\quad - \sqrt{n_-^h (n_+^e + 1)} V_{eh}^{\text{EX}}(n_+^e + 1, n_-^e; n_+^h, n_-^h - 1 | n_+^{h'}, n_-^{h'}; n_+^{e'}, n_-^{e'}) \\ &\quad \left. + \sqrt{(n_+^e + 1)(n_+^h + 1)} V_{eh}^{\text{EX}}(n_+^e + 1, n_-^e; n_+^h + 1, n_-^h | n_+^{h'}, n_-^{h'}; n_+^{e'}, n_-^{e'}) \right\}, \quad (9) \end{aligned}$$

with the following orbital and spin angular-momentum conservation rules: $\mathcal{L}_L = \mathcal{L}_R$ and $\mathcal{S}_L = \mathcal{S}_R$.

The remaining part of the electron-hole exchange, which is a sum of the short-range exchange and the local part of the long-range exchange, is proportional to the overlap between envelopes of both electron-hole pairs:

$$\langle i\uparrow, j\downarrow | \delta_0^{SL} | k\downarrow, l\uparrow \rangle = \left(2E_{SR} - \frac{16\pi\mu^2}{3} \right) \int d\mathbf{r}_1 F_i^+(\mathbf{r}_1) H_j^+(\mathbf{r}_1) H_k(\mathbf{r}_1) F_l(\mathbf{r}_1), \quad (10)$$

where E_{SR} is a numerical constant parameterizing the short-range exchange interaction.²⁷ This integral can be calculated using $\delta(\mathbf{r}_1 - \mathbf{r}_2) = \frac{1}{4\pi^2} \int d\mathbf{q} \exp[i\mathbf{q}(\mathbf{r}_1 - \mathbf{r}_2)]$ as

$$\int d\mathbf{r}_1 F_i^+(\mathbf{r}_1) H_j^+(\mathbf{r}_1) H_k(\mathbf{r}_1) F_l(\mathbf{r}_1) = \frac{1}{4\pi^2} \int d\mathbf{q} \int d\mathbf{r}_1 F_i^+(\mathbf{r}_1) \exp(i\mathbf{q}\mathbf{r}_1) F_l(\mathbf{r}_1) \int d\mathbf{r}_2 H_j^+(\mathbf{r}_2) H_k(\mathbf{r}_2) \exp(-i\mathbf{q}\mathbf{r}_2). \quad (11)$$

Evaluating the integrals over the wave vector q gives the final result for the short-range exchange contribution,

$$\begin{aligned} \int d\mathbf{r}_1 F_i^+(\mathbf{r}_1) H_j^+(\mathbf{r}_1) H_k(\mathbf{r}_1) F_l(\mathbf{r}_1) &= \frac{1}{4\pi^2} \frac{1}{\sqrt{n_i! m_i! n_l! m_l! n_k! m_k! n_j! m_j!}} \sum_{p_1=0}^{\min(n_l, n_i)} \sum_{p_2=0}^{\min(m_l, m_i)} \sum_{p_3=0}^{\min(n_k, n_j)} \sum_{p_4=0}^{\min(m_k, m_j)} \binom{n_i}{p_1} \binom{n_l}{p_1} \\ &\times \binom{m_i}{p_2} \binom{m_l}{p_2} \binom{n_k}{p_3} \binom{n_j}{p_3} \binom{m_k}{p_4} \binom{m_j}{p_4} \times I \end{aligned} \quad (12)$$

where

$$I = \left(\frac{l_h}{l_e} \right)^{n_k + m_k + n_j + m_j - 2p_3 - 2p_4} \frac{2\pi}{l_e^2} \delta_{L_L, L_R} (-1)^{n_k + m_k + n_j + m_j} \left[1 + \left(\frac{l_h}{l_e} \right)^2 \right]^{-(p+1)} \Gamma(p+1). \quad (13)$$

V. COMPUTATIONAL PROCEDURE

The Hamiltonian of an exciton in a QD with an isotropic confining potential without the electron-hole exchange interaction $\hat{H}_{EH}$ [Eq. (4)] conserves both the electron and hole spin and the angular momentum of the e-h pair. The exciton states in each L, σ, τ subspaces can be constructed, and the Hamiltonian matrix can be built in the space of these configurations and diagonalized separately. However, the presence of anisotropy in the QD as well as the electron-hole exchange interaction mixes different angular momentum as well as spin subspaces.

The complete Hamiltonian matrix has to be built using all electron-hole configurations in the form $|i\tau j\sigma\rangle = h_{i\tau}^+ c_{j\sigma}^+ |\text{vac}\rangle$, where i, j denotes the single-particle levels $i = (n_+, n_-)$. From diagonalization of the exciton Hamiltonian matrix one obtains the set of eigenstates in the form of linear combinations of basis configurations, $|X_k\rangle = \sum_{ij\sigma\tau}^N A_{ij\sigma\tau}^{(k)} |i\tau j\sigma\rangle$, with energy E_k , where $k = 1, \dots, N$, N is the size of the basis, and $A_{ij\sigma\tau}^{(k)}$ is the amplitude of the configuration $|i\tau j\sigma\rangle$ in the state $|X_k\rangle$. Coefficients $A_{ij\sigma\tau}^{(k)}$ as well as the exciton energies depend on the magnetic field B and are evaluated for each magnetic field separately.

Having obtained the eigenenergies and eigenfunctions of the electron-hole pair, one can calculate the emission/absorption spectra from the Fermi's golden rule, which, in the dipole approximation, can be written as

$$I(\omega) = \sum_f \sum_i P_i |\langle f | \hat{P}^\pm | i \rangle|^2 \delta(E_i - E_f - \omega), \quad (14)$$

where $|i\rangle$ ($|f\rangle$) is the initial (final) state with corresponding energy E_i (E_f), P_i is the probability of the initial-state occupation given by the distribution $P_i = \exp\{-\frac{E_i - E_0}{kT}\} / P_{\text{SUM}}$, with $P_{\text{SUM}} = \sum_i \exp\{-\frac{E_i - E_0}{kT}\}$, k being the Boltzman constant and T the temperature. The interband polarization operator $\hat{P}^\pm$ annihilates ($-$) or creates ($+$) one electron-hole pair from the

initial state. Depending on the spin σ of removed particles, emitted photons have circular [σ_+ ($\hat{P}_+ = \sum_i c_{i\downarrow} h_{i\uparrow}$), σ_- ($\hat{P}_- = \sum_i c_{i\uparrow} h_{i\downarrow}$)] or linear [X ($\hat{P}_X = \hat{P}_+ + \hat{P}_-$), Y ($\hat{P}_Y = \hat{P}_+ - \hat{P}_-$)] polarization.

VI. RESULTS AND DISCUSSION

The key result of this work is the detailed derivation of long- and short-range exchange Coulomb matrix elements in the HO basis, Eq. (6), for an arbitrary magnetic field. These matrix elements can now be included in the exciton Hamiltonian and combined with the direct Coulomb interaction, anisotropy, and magnetic field. This methodology allows us to introduce and understand the influence of electron-hole correlations and their modification by the magnetic field on the exciton fine structure. In this section, we illustrate our procedure with numerical and analytical calculations for a model quantum dot. The model isotropic quantum dot discussed below has the electron shell spacing $\Omega_e = 24$ meV, the hole shell spacing $\Omega_h = 6$ meV, the dielectric constant $\varepsilon = 10.6$, the effective mass of an electron $m_e^* = 0.1m_0$ and hole $m_h^* = 0.4m_0$, and Lande factors $g_e = -0.7$ and $g_h = 0.38$ [Refs. 38–40 and 41]. With these parameters the effective Rydberg is $Ry = 12.11$ meV, the effective Bohr radius $a_B = 5.61$ nm, $\Omega_h + \Omega_e = 2.48$ Ry, and $\Omega_e = 4\Omega_h$. The anisotropy of the model quantum dot is given by the ratio Ω_y/Ω_x and the anisotropy parameter γ . Results for the exciton fine structure will be illustrated for the anisotropy of $\Omega_y/\Omega_x = 0.72$ ($\gamma = 0.32$). The exciton Zeeman energy in high magnetic field dominates over the exchange splitting of the two exciton bright states and, hence, the Zeeman energy is not included in the results of calculations presented in what follows.

In order to understand the effect of electron-hole correlations we will study the exciton fine structure as a function of the number of confined electron and hole electronic shells for a fixed shell energy spacing. The more electronic shells

FIG. 1. (Color online) Magnetic field evolution of the selected electron-hole exchange matrix elements, namely $\langle 00\uparrow; 00\downarrow | V_{eh}^X | 00\uparrow; 20\downarrow \rangle$ (a), the local part of the $\langle 00\uparrow; 00\downarrow | V_{eh}^X | 00\downarrow; 00\uparrow \rangle$ matrix element (b), the nonlocal part of the $\langle 00\uparrow; 00\downarrow | V_{eh}^X | 00\downarrow; 00\uparrow \rangle$ matrix element (c), and the full $\langle 00\uparrow; 00\downarrow | V_{eh}^X | 00\downarrow; 00\uparrow \rangle$ matrix element (d).

present, the stronger the electronic correlations. For a very deep quantum dot with large number of confined shells such an analysis is equivalent to the analysis of convergence. In reality, dots with different numbers of confined shells can be grown. Understanding the role of different shells, $s, p, d, \dots$, on the exciton fine structure might allow us to design the exciton fine structure by designing quantum dot shells.

Let us, first, analyze the dependence of several important electron-hole exchange matrix elements involving the s, p , and d shells, $\langle 00\uparrow; 00\downarrow | V_{eh}^X | 20\uparrow; 00\downarrow \rangle$, $\langle 00\uparrow; 00\downarrow | \delta^{SL} | 00\downarrow; 00\uparrow \rangle$, $\langle 00\uparrow; 00\downarrow | \delta^L | 00\downarrow; 00\uparrow \rangle$, and $\langle 00\uparrow; 00\downarrow | \delta^{SL} + \delta^L | 00\downarrow; 00\uparrow \rangle$ on the magnetic field in the symmetric electron-hole case where $I_h^h = I_h^e$. Their dependence on the magnetic field is shown in Fig. 1. We note that, from Eq. (6) and Ref. 29, $V_{eh}^{EX} \sim \sqrt{\pi\omega_h} = \frac{\sqrt{\pi}}{l_h}$, the matrix element $\langle 00\uparrow; 00\downarrow | V_{eh}^X | 20\uparrow; 00\downarrow \rangle$, which mixes bright excitonic configurations, evolves like l_h^{-3} . The magnetic field dependence of the diagonal matrix element $\langle 00\uparrow; 00\downarrow | \delta^{SL} + \delta^L | 00\downarrow; 00\uparrow \rangle$ [Fig. 1(d)] is more complicated due to the interplay between the l_h^{-2} dependence of δ^{SL} (b) and the l_h^{-3} dependence of δ^L (c). Let us now use the electron-hole exchange matrix elements in the calculations of the correlated exciton states (procedure described in Sec. V). We will analyze here how the bright exciton level splitting depends on the number of electron and hole single-particle energy shells, the anisotropy of the confining potential, and the magnetic field.

Figure 2 (right inset) shows the exciton emission spectrum calculated for s, p, d confined single-particle shells in an anisotropic QD. There are two emission peaks corresponding to the two bright exciton states. Figure 2 (left inset) shows the energy of the lowest bright exciton state for different number of confined single-particle shells in an anisotropic QD. As expected, the more HO shells are confined in the QD, the lower the exciton energy due to the direct Coulomb scattering.

Due to the fact that we are working in the basis of isotropic HO, if we confine only the s shell for electrons and holes, we do not observe any electron-hole exchange splitting (Fig. 2). As explained in Sec. IV, the electron-hole exchange matrix elements are nonzero only between pairs of states for which the orbital angular momentum differs by 2. For this reason we observe a strong increase in the electron-hole exchange splitting for a quantum dot with shells containing states with angular momenta 2, 4, $\dots$, i.e., d and f shells, than when an odd shell is added (p, e , and g shells).

FIG. 2. (Color online) The bright exciton state splitting for QDs with different number of confined shells and with anisotropy parameter $\gamma = 0.32$. (Left inset) Energies of bright exciton states for QDs with different number of confined shells. (Right inset) Exciton emission spectrum from anisotropic QD confining s, p , and d single-particle states.

FIG. 3. The percentage of the long-range exchange matrix element covered by the three first terms of Eq. (16).

To understand why the anisotropy in the confining potential of the QD is necessary to observe electron-hole exchange splitting of the bright states, let us consider, first, an anisotropic QD in the absence of the long-range electron-hole exchange. The wave function of the bright state with the spin angular momentum $\tau + \sigma = 1$ in the presence of anisotropy has the form

$$\begin{aligned}
 |\text{GS}\uparrow\downarrow\rangle = & A_{00,00}|00\uparrow,00\downarrow\rangle - A_{00,11}|00\uparrow,11\downarrow\rangle \\
 & + A_{01,01}(|01\uparrow,01\downarrow\rangle + |10\uparrow,10\downarrow\rangle) \\
 & - A_{11,00}|11\uparrow,00\downarrow\rangle + A_{11,11}|11\uparrow,11\downarrow\rangle \\
 & - A_{20,00}(|02\uparrow,00\downarrow\rangle + |20\uparrow,00\downarrow\rangle) \\
 & - A_{00,02}(|00\uparrow,20\downarrow\rangle + |00\uparrow,02\downarrow\rangle) \\
 & - A_{10,01}(|01\uparrow,10\downarrow\rangle + |10\uparrow,01\downarrow\rangle). \quad (15)
 \end{aligned}$$

A similar expression can be written for the second bright state with spin angular momentum equal to $\tau + \sigma = -1$.

We now turn to evaluating the coupling between these states due to the long-range exchange interaction

$$\begin{aligned}
 \langle \text{GS}\uparrow\downarrow | V_{eh}^X | \text{GS}\uparrow\downarrow \rangle &= -2(A_{00,00}A_{20,00}\langle 00\uparrow,00\downarrow | V_{eh}^X | 20\uparrow,00\downarrow \rangle \\
 &+ A_{00,00}A_{10,01}\langle 00\uparrow,00\downarrow | V_{eh}^X | 10\uparrow,01\downarrow \rangle \\
 &+ A_{00,00}A_{00,02}\langle 00\uparrow,00\downarrow | V_{eh}^X | 00\uparrow,02\downarrow \rangle \\
 &+ A_{11,00}A_{20,00}\langle 00\uparrow,11\downarrow | V_{eh}^X | 20\uparrow,00\downarrow \rangle \\
 &+ A_{11,00}A_{10,01}\langle 00\uparrow,11\downarrow | V_{eh}^X | 10\uparrow,01\downarrow \rangle \\
 &+ A_{11,00}A_{00,02}\langle 00\uparrow,11\downarrow | V_{eh}^X | 00\uparrow,02\downarrow \rangle + \dots). \quad (16)
 \end{aligned}$$

Because we find all $\langle i\uparrow, j\downarrow | V_{eh}^X | k\uparrow, l\downarrow \rangle$ matrix elements of the same order, their contribution is largely determined by the magnitude of the coefficients $A_{ij}A_{kl}$ in the exciton wave function. Since the lowest-energy single-particle state is $|00,00\rangle$, the coefficient $A_{00,00}$ is the biggest. We find that the first three terms of the sum from Eq. (16) contribute $\approx 70\%$ to the bright exciton exchange splitting for a broad range of values of the anisotropy parameter, as shown in Fig. 3.

The first three most important terms representing the three most important coupling mechanisms between the two bright excitonic states $|\text{GS}\downarrow\uparrow\rangle$ and $|\text{GS}\uparrow\downarrow\rangle$ involving electron-hole exchange matrix elements: (a) $\langle 00\uparrow,00\downarrow | V_{eh}^X | 20\uparrow,00\downarrow \rangle$, (b) $\langle 00\uparrow,00\downarrow | V_{eh}^X | 10\uparrow,01\downarrow \rangle$, and (c) $\langle 00\uparrow,00\downarrow | V_{eh}^X | 00\uparrow,02\downarrow \rangle$ are shown schematically in Fig. 4.

We now turn to the effect of the magnetic field on the exciton fine structure. The magnetic field changes the single-particle energy levels of both electrons and holes and

FIG. 4. (Color online) The three most important paths of coupling between two bright excitonic states $|\text{GS}\downarrow\uparrow\rangle$ and $|\text{GS}\uparrow\downarrow\rangle$ involving e-h matrix elements $\langle 00\uparrow,00\downarrow | V_{eh}^X | 20\uparrow,00\downarrow \rangle$ (a), $\langle 00\uparrow,00\downarrow | V_{eh}^X | 10\uparrow,01\downarrow \rangle$ (b), and $\langle 00\uparrow,00\downarrow | V_{eh}^X | 00\uparrow,02\downarrow \rangle$ (c).

FIG. 5. (Color online) Magnetic field evolution of the splitting of bright exciton states calculated for the anisotropic QDs with anisotropy parameter $\gamma = 0.32$.

increases the strength of direct Coulomb interactions.^{31,42} In Fig. 5 we show the energy splitting of the two bright exciton levels for anisotropic QD ($\gamma = 0.32$) as a function of increasing magnetic field obtained by exact diagonalization of the exciton Hamiltonian, Eq. (3), for different numbers of confined electronic shells. Alternatively, we can think of different dots, each confining a different number of electronic shells. We see in Fig. 5 that the splitting increases with the increasing magnetic field and its dependence on the magnetic field changes with the increase of the number of confined single-particle shells. While the splitting changes little with the magnetic field for shallow quantum dots with s, p shells, with an increasing number of shells the magnetic field dependence becomes a nonlinear function of the magnetic field. Since the exciton Zeeman energy is a linear function of the magnetic field, the measurement of the exciton fine-structure splitting in a magnetic field should allow the extraction of the nonlinear part and hence of the magnetic field dependence of the electron-hole exchange. According to the results presented here, this dependence should correlate with the number of confined electronic shells as measured in high-excitation emission spectroscopy.

The nonlinear dependence of the bright exciton splitting can be also traced to the exciton fine structure of isotropic QDs. For isotropic QDs in the absence of the magnetic field,

FIG. 6. (Color online) Magnetic field evolution of the splitting of bright exciton states calculated for the isotropic QDs.

FIG. 7. (Color online) Important electron-hole configurations in the absence of the magnetic field (upper row) and in the finite magnetic field (lower row).

there is no electron-hole exchange-induced splitting as shown previously^{5,11–13,27}; however, as the field increases, the nonzero splitting of the bright states occurs (Fig. 6) due to symmetry breaking by the magnetic field.

This can be understood on a simple example in which we consider only four electron-hole configurations, as shown in Fig. 7. Two of these configurations, 1 and 3, are the lowest-energy configurations with both carriers on the s shell, and they differ only by the spin projection of the carriers. The other two configurations (2 and 4) are examples of configurations with angular momentum $L = \pm 2$, which are coupled by the electron-hole exchange interaction with the lowest-energy configurations (1–2) and (3–4). In the absence of the magnetic field the configurations 2 and 4 from the upper row in Fig. 7 have the same kinetic energy. Since all diagonal and off-diagonal matrix elements are the same, the resulting eigenstates will have the same energy, leading to a lack of electron-hole exchange splitting. However, as the field increases, it shifts the single-particle levels and causes a splitting of the shells similar to the splitting induced by the anisotropy (Fig. 7). Since the energies of single-particle levels change in the magnetic field, the kinetic energies of the electron-hole configurations change as well so the second and third configurations in the lower row no longer have the same kinetic energy. For these reasons the nonzero electron-hole exchange splits the bright exciton levels of isotropic quantum dots in finite magnetic fields.

VII. SUMMARY

In summary, theory of fine structure of a correlated exciton state in self-assembled parabolic semiconductor quantum dots in a perpendicular magnetic field is presented. The main result is the derivation of the short- and long-range electron-hole exchange interaction matrix elements in the harmonic oscillator basis for an arbitrary magnetic field. The long-range matrix elements are expressed in terms of Coulomb exchange matrix elements. This allows the inclusion of the direct, short- and long-range electron-hole exchange interaction matrix elements and quantum dot anisotropy, all expressed in the harmonic oscillator basis, into a microscopic exciton Hamiltonian. The fine structure of the ground and excited correlated exciton states as a function of the number of confined shells, quantum dot anisotropy, and magnetic field is

obtained using exact diagonalization techniques. The effects of correlations are shown to significantly affect the energy splitting of the two bright exciton states. The theory developed here is expected to help in designing quantum dots with a desired fine structure.

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