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Tuning of exciton states in a magnetic quantum ring

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HIGHLIGHTS

- The effect of spin interactions on exciton states in a quantum ring is considered.
- Quantum ring containing a single magnetic impurity is studied.
- Multiband approximation has been used for calculation of the exciton states.
- The bright exciton state can be changed to the dark state and vice versa.
- An experimental method is proposed to estimate spin interaction constants.

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ABSTRACT

The exciton states in a CdTe quantum ring subjected to an external magnetic field containing a single magnetic impurity are investigated. We have used the multiband approximation which includes the heavy hole–light hole coupling effects. The electron–hole spin interactions and the s, p–d interactions between the electron, the hole and the magnetic impurity are also included. The exciton energy levels and optical transitions are evaluated using the exact diagonalization scheme. We show that due to the spin interactions it is possible to change the bright exciton state into the dark state and vice versa with the help of a magnetic field. We propose a new route to experimentally estimate the s, p–d spin interaction constants.

1. Introduction

Electronic properties of planar nanoscale semiconductor structures, such as quantum rings (QRs) [1] and quantum dots (QDs) [2] have enjoyed widespread attention in the past few decades due to their novel fundamental effects and for potential technological applications. Experimental advances in creating these structures from a two-dimensional electron gas by using suitable confinements have resulted in confirmation of several theoretical predictions in these systems (see, for example [3,4]). It has been realized lately that QD doped with a single magnetic impurity [5–7] has great potential to contribute significantly in the burgeoning field of single spin manipulation [8], which will eventually lead to important contributions in quantum information processing. Quite naturally, quantum dots, in particular the CdTe QDs containing a single Mn atom has been widely studied in the literature and several mechanisms for manipulation of a single Mn spin have been proposed both theoretically and experimentally [9–12]. The higher spin stability and hence longer lasting of the relaxation and decoherence processes in a QR than in a QD makes QRs more promising candidates for spin manipulations, readout and for realization of spin qubits [13]. Recently, CdTe QRs have been realized experimentally [14]. The problem of two interacting electrons in QR containing one magnetic impurity has been studied previously and it was shown that the scattering on magnetic impurity is responsible for transitions between singlet and triplet states of two electrons [15]. Against the backdrop of these important developments, no studies of an interaction between an exciton and the magnetic impurity in a QR (which can be used for manipulation of a Mn spin orientation in a QR and where unique optical properties of the QR associated with the excitonic Aharonov–Bohm effect can play special role [16]) have been reported yet in the literature.

Here we report on our studies of the exciton states in a CdTe QR in a magnetic field, containing a single magnetic impurity. We have found that due to the resulting spin interactions the bright exciton state can be changed to the dark state and vice versa, with the help of an applied magnetic field. Additionally, we propose

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here an experimental means to estimate the s, p–d spin interaction constants.

2. Theoretical framework

We study the exciton states in a CdTe quantum ring containing a single manganese magnetic impurity (Mn) and subjected to a perpendicular magnetic field. Usually, the thickness of the ring is smaller than the radial dimensions. Therefore, our system can be considered as quasi two-dimensional with the internal radius R_1 and the external radius R_2. The electron and the hole are always in the ground state for the z direction. We chose the confinement potential of the quantum ring in the radial direction with infinitely high borders: $V_{\rm conf}(\rho)=0$ if $R_1 \le \rho \le R_2$ and infinity outside of the QR. The Hamiltonian of the system can then be written as

$$\mathcal{H}=\mathcal{H}_{\rm e}+\mathcal{H}_h+V_{\rm eh}+\mathcal{H}_{\rm eh}+\mathcal{H}_{\rm s-d}+\mathcal{H}_{p-d}+\mathcal{H}_{\rm Mn}, \tag{1}$$

where $\mathcal{H}_{\rm s-d}=-J_{\rm e}\delta(\mathbf{r}_{\rm e}-\mathbf{r}_{\rm Mn})\boldsymbol{\sigma}\mathbf{S}$ and $\mathcal{H}_{\rm p-d}=-J_{\rm h}\delta(\mathbf{r}_{\rm h}-\mathbf{r}_{\rm Mn})\mathbf{j}\mathbf{S}$ describe the electron–Mn and hole–Mn spin–spin exchange interaction with strengths $J_{\rm e}$ and $J_{\rm h}$ respectively, $\mathbf{r}_{\rm Mn}$ is the radius vector of the Mn atom. $\mathcal{H}_{\rm eh}=-J_{\rm eh}\delta(\mathbf{r}_{\rm e}-\mathbf{r}_{\rm h})\boldsymbol{\sigma}\mathbf{j}$ is the electron–hole spin interaction Hamiltonian [17]. The electron and the hole Coulomb interaction term is $V_{\rm eh}=-e^2/\varepsilon|\mathbf{r}_{\rm e}-\mathbf{r}_{\rm h}|$, where ε is the dielectric constant of the system. The last term in Eq. (1) is Zeeman splitting for the impurity spin.

The electron Hamiltonian in our system is

$$\mathcal{H}_{\rm e}=\frac{1}{2m_{\rm e}}\left(\mathbf{p}+\frac{e}{c}\mathbf{A}\right)^2+V_{\rm conf}(\rho,z)+\frac{1}{2}g_{\rm e}\mu_B B\sigma_z, \tag{2}$$

where $\mathbf{A}=\frac{1}{2}B(-y,x,0)$ is the symmetric gauge vector potential, the electron charge is $-e$ and the last term is the electron Zeeman energy. Without the magnetic field the eigenfunctions of $\mathcal{H}_{\rm e}$ is of the form

$$\psi^{\rm e}_{nl\sigma}(\rho,\varphi)=C_{nl}\,{\rm e}^{{\rm i}l\varphi}f^{e}_{nl}(\rho)\chi_\sigma, \tag{3}$$

where C_{nl} is the normalization constant, $n=1,2,\dots,$ and $l=0,\pm 1,\pm 2,\dots$ are the radial and angular quantum numbers, respectively, σ is the electron spin and χ_σ is the electron spin wave function. The functions $f^e_{nl}(\rho)$ are obtained from a suitable linear combination of the Bessel functions: $f^e_{nl}(\rho)=J_l(k_{nl}\rho)-(J_l(k_{nl}R_1)/Y_l(k_{nl}R_1))Y_l(k_{nl}\rho)$, where $k_{nl}=\sqrt{2m_{\rm e}E_{nl}/?^2}$. The corresponding eigenvalues E_{nl} are obtained from the standard boundary conditions of the eigenfunctions.

Taking into account only the Γ_8 states which correspond to the states with the hole spin $j=3/2$ and include the heavy hole–light hole coupling effects, we construct the single-hole Hamiltonian for the ring as

$$\mathcal{H}_{\rm h}=\mathcal{H}_{\rm L}+V_{\rm conf}(\rho)-2\kappa\mu_B Bj_z. \tag{4}$$

Here $\mathcal{H}_{\rm L}$ is the Luttinger hamiltonian in axial representation obtained with the four-band $\mathbf{k}\cdot\mathbf{p}$ theory [18,19]

$$\mathcal{H}_{\rm L}=\frac{1}{2m_0}\begin{pmatrix}\mathcal{H}_{\rm hh} & R & S & 0\\ R^* & \mathcal{H}_{\rm lh} & 0 & S\\ S^* & 0 & \mathcal{H}_{\rm lh} & -R\\ 0 & S^* & -R^* & \mathcal{H}_{\rm hh}\end{pmatrix}, \tag{5}$$

where
$\mathcal{H}_{\rm hh}=(\gamma_1+\gamma_2)(\Pi_x^2+\Pi_y^2)+(\gamma_1-2\gamma_2)\Pi_z^2$, $\mathcal{H}_{\rm lh}=(\gamma_1-\gamma_2)(\Pi_x^2+\Pi_y^2)+(\gamma_1+2\gamma_2)\Pi_z^2$, $R=2\sqrt{3}\gamma_3{\rm i}\Pi_-\Pi_z$, $S=\sqrt{3}\gamma\Pi_-^2$, $\gamma=\frac{1}{2}(\gamma_2+\gamma_3)$, and $\mathbf{\Pi}=\mathbf{p}-\frac{e}{c}\mathbf{A}$, $\Pi_\pm=\Pi_x\pm{\rm i}\Pi_y$. γ_1, γ_2, γ_3 and κ are the Luttinger parameters and m_0 is the free electron mass.

The Hamiltonian (4) is rotationally invariant and we introduce the total momentum $\mathbf{F}=\mathbf{j}+\mathbf{l}_{\rm h}$, where $\mathbf{j}$ is the angular momentum of the band edge Bloch function, and $\mathbf{l}_{\rm h}$ is the envelop angular momentum. Since the projection of the total momentum F_z is a constant of motion, we can find simultaneous eigenstates of (4) and F_z [20].

For a given value of F_z it is logical to seek the eigenfunctions of the Hamiltonian (4) as an expansion [19,21]

$$\Psi_{F_z}(\rho,\varphi)=\sum_{n,j_z}C_{F_z}(n,j_z)f^h_{n,\,F_z-j_z}(\rho){\rm e}^{{\rm i}(F_z-j_z)\varphi}\chi_{j_z}, \tag{6}$$

where χ_{j_z} are the hole spin functions and $f^h_{nl}(\rho)$ are the radial wave functions similar to $f^e_{nl}(\rho)$ with $k^h_{nl}=\sqrt{2m_0E_{nl}/?^2(\gamma_1+\gamma_2)}$. All single hole energy levels and the expansion coefficients are evaluated numerically using the exact diagonalization scheme [21].

To evaluate the energy spectrum of the exciton system we diagonalize the Hamiltonian (1) without spin interactions in a basis constructed as products of the single-electron and single-hole wave functions. The good quantum number is the projection M_z of the exciton total momentum $\mathbf{M}=\mathbf{F}+\mathbf{l}_{\rm e}$. For a given value of M_z and the electron spin σ, the exciton wave function is presented as

$$\Psi_{M_z\sigma}=\sum_{n_el_e}\sum_{F_z}C(n_{\rm e},l_{\rm e},F_z)\psi^e_{n_el_e\sigma}(\rho_{\rm e},\varphi_{\rm e})\Psi_{F_z}(\rho_{\rm h},\varphi_{\rm h}) \tag{7}$$

The numerical calculations were carried out for a CdTe QR with $R_1=100$ Å, $R_2=300$ Å, $L_z=30$ Å (L_z is the height of the ring in the z direction) and with the following parameters: $E_{\rm g}=1.568$ eV ($E_{\rm g}$ is the bandgap energy between valence and conduction bands), $m_{\rm e}=0.096m_0$, $g_{\rm e}=-1.5$, $\gamma_1=5.3$, $\gamma_2=1.7$, $\gamma_3=2$, κ=0.7 [22].

To include the spin–spin interactions, we construct the wave function of the exciton and the magnetic impurity as an expansion of the direct products of the lowest state exciton wave function (7) and eigenfunctions for the magnetic impurity

$$\Psi=\sum_\sigma\sum_{M_z}\sum_{S_z}C(\sigma,M_z,S_z)\Psi_{M_z,\sigma}\times|S_z\rangle. \tag{8}$$

Here $\sigma=\pm 1/2$, $S_z=\pm 1/2,\pm 3/2,\pm 5/2$ and $M_z=\pm 1/2,\pm 3/2,\pm 5/2\dots$. Using the components of this expansion as the new basis functions we calculate the corresponding matrix elements for the electron–hole, the electron–impurity and the hole–impurity interactions. Employing the steps used in [9] for the electron–hole spin interaction matrix element, we get

$$M_{\rm eh}=-J_{\rm eh}\delta_{S_z,S_z'}\sum_{j_z,j_z'}A_{\rm eh}(j_z,j_z')\langle\sigma,j_z|\boldsymbol{\sigma}\mathbf{j}|\sigma',j_z'\rangle, \tag{9}$$

where $A_{\rm eh}$ is obtained by integrating the electron and hole coordinate wave functions, $\boldsymbol{\sigma}$ is the Pauli spin operator and $\mathbf{j}$ is the hole spin operator [9].

For the electron–impurity interaction we have

$$M_{\rm s-d}=-J_{\rm e}\sum_{l_e,l_e'}\delta_{M_z-l_e,M_z'-l_e'}A_{\rm s-d}(\mathbf{r}_{\rm e}=\mathbf{r}_{Mn},l_{\rm e},l_{\rm e}')\times \langle\sigma_z,S_z|\boldsymbol{\sigma}\mathbf{S}|\sigma_z',S_z'\rangle, \tag{10}$$

where $A_{\rm s-d}$ is obtained after the integration of the hole coordinate wave functions and putting $\mathbf{r}_{\rm e}=\mathbf{r}_{\rm Mn}$ in the electron wave function. Similarly, for the case of hole–impurity interaction we have

$$M_{\rm p-d}=-J_{\rm h}\delta_{\sigma,\sigma'}\sum_{j_z,j_z'}A_{\rm p-d}(\mathbf{r}_{\rm h}=\mathbf{r}_{\rm Mn},j_z,j_z')\times\langle j_z,S_z|\mathbf{j}\mathbf{S}|j_z',S_z'\rangle. \tag{11}$$

As the spin interactions are short ranged, the most interesting case is when the magnetic impurity is located in the region of average ring radius. Then we can take $\rho_{\rm Mn}=(R_1+R_2)/2$ and $\varphi_{\rm Mn}=0$. The problem was solved numerically using the exact diagonalization scheme and with interaction parameters $J_e = 15$ meV nm^3, $J_h = -60$ meV nm^3 [6,10].

In order to evaluate the optical transition probabilities, we note that the initial state of the system is that of the magnetic impurity spin with the valence band states fully occupied and the conduction band states being empty. We also assume that the impurity states are $|i\rangle = |S_z\rangle$. Recently there were several experimental reports where the spin orientation of single magnetic impurity in quantum dots was controlled using optical means even in the absence of a magnetic field [10,23]. The final states are the eigenstates of the Hamiltonian (1) presented in (8) $|f\rangle = |\Psi\rangle$. In the electric dipole approximation the relative oscillator strengths for all possible optical transitions are proportional to $P(m) \sim |\langle \Psi | m, S_z \rangle|^2$. Here the values of $m = 1, 0, -1$ characterize the polarization of the light as σ^+, π and σ^- respectively [5]. The impurity spin state remains unchanged during the optical transitions.

3. Results and discussion

In the absence of the magnetic impurity in the QR and without the electron–hole spin interaction, the ground state of the exciton will be four-fold degenerate with values of the total momentum ± 1 and ± 2. The magnetic field lifts that degeneracy due to the Zeeman splitting and as a result two bright ($J_z = \pm 1$) and two dark ($J_z = \pm 2$) exciton states appear. The electron–hole spin exchange interaction in turn gives rise to a further splitting between the bright and dark exciton states and removes the degeneracy between them in zero magnetic field. In Fig. 1(a) the dependence of the 12 low-lying exciton energy levels on the magnetic field is presented with the electron–hole spin interaction included, for a QR without a magnetic impurity. Without electron–hole spin interaction these states would correspond to the states with $\sigma = \pm 1/2$ and $M_z = \pm 1/2, 3/2, 5/2$. The corresponding optical transition probabilities for σ^- and σ^+ polarizations are shown in Fig. 1(b). The sizes of the symbols in Fig. 1(b) indicate the probability of the optical transition to that state. Fig. 1(b) reveals two levels with highest transition amplitude and these levels are similar to the transitions observed for a QD with total momentum $J_z = \pm 1$. Due to the mixing of angular and spin momenta there are also other transitions observable with considerable low intensity. For lower values of the magnetic field, two lowest energy levels in Fig. 1 (a) correspond to the dark exciton states and hence the transition probabilities to that states are very weak. The energies of two bright exciton states with the most important components of the basis functions $|\sigma, j_z\rangle = |-1/2, 3/2\rangle$ and $|1/2, -3/2\rangle$ are shifted upwards by the electron–hole spin interaction, but still are clearly visible optically in Fig. 1(b). In the case of σ^+ polarization we have a strong transition to the state $|-1/2, 3/2\rangle$ (black squares), and for the case of σ^- polarization, the strong transition is for the state $|1/2, -3/2\rangle$ (white squares). We should also mention that with an increase of the magnetic field the transition probabilities remain almost unchanged.

The Mn atom has a spin $S = 5/2$ and there are six possible values of the impurity spin projection S_z. Therefore, due to the s, p–d spin interaction each exciton energy level in Fig. 1(a) will split into six and we have considered 72 energy levels of the system. Many level crossings and anticrossings appear due to the Zeeman and the s, p–d splitting of the energy levels. The presence of the impurity in the ring removes the symmetry of the structure and we do not have any good quantum numbers to describe the states. All states are mixed superpositions with different values of the total momentum of electron, hole and the magnetic impurity.

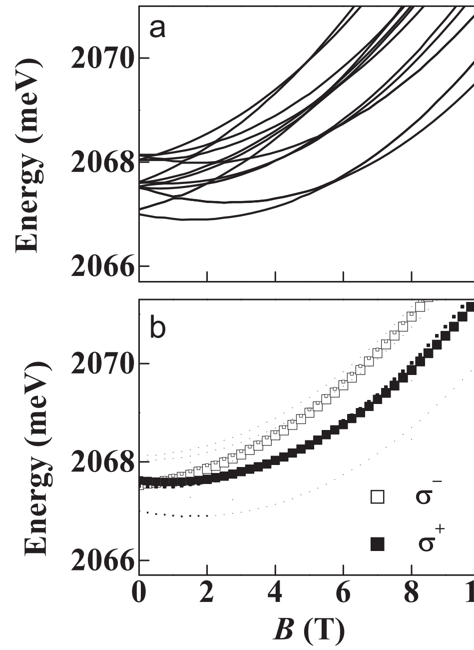

Fig. 1. (a) Magnetic field dependence of the exciton energy levels with electron hole spin-interaction included for the ring without magnetic impurity. (b) Optical transition amplitudes for the σ^+ and σ^- polarizations.

To clarify this complicated situation, we consider here the optical transition spectrum to these 72 states. The high probability transitions will be possible only to the bright exciton states which have the most important components with the same value of the impurity spin S_z as in the initial state. The results for the σ^- polarization of the incident light are presented in Fig. 2. Here the shapes and the colors of the points indicate the initial spin of the impurity and the sizes of the points indicates the probability of the transition to that state. For the σ^- polarization of the incident light the bright exciton states must have the important component with $|\sigma_z, M_z\rangle = |1/2, -3/2\rangle$. For the σ^+ polarization (Fig. 3) the most important component of the bright states must be $|-1/2, 3/2\rangle$ [5]. For example in the case of the σ^- polarization and for the initial state $S_z = -5/2$ in low magnetic fields we have only one strong transition (Fig. 2 black circles). But near $B = 5$–6 T that line weakens and disappears and a new optical mode appears. Similar behavior was seen for other impurity spin states. This is the direct signature of the s, p–d spin interaction. Due to these spin interactions the bright exciton state $|1/2, -3/2\rangle$ is now coupled with the dark state $|1/2, -1/2\rangle$ and we have two coupled energy levels.

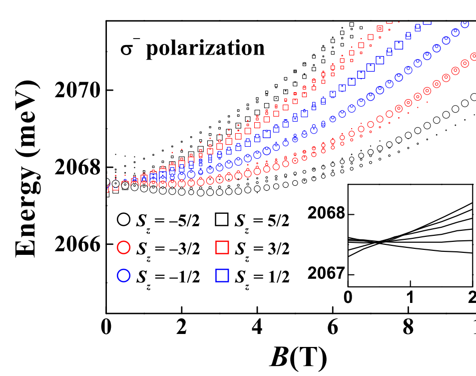

Fig. 2. Magnetic field dependence of the optical transition amplitudes for the case of σ^- polarization and for various values of the initial state impurity spin projection S_z. The crossing point of six bright exciton states is shown as inset. (For interpretation of the references to color in the main text, the reader is referred to the web version of this paper.)

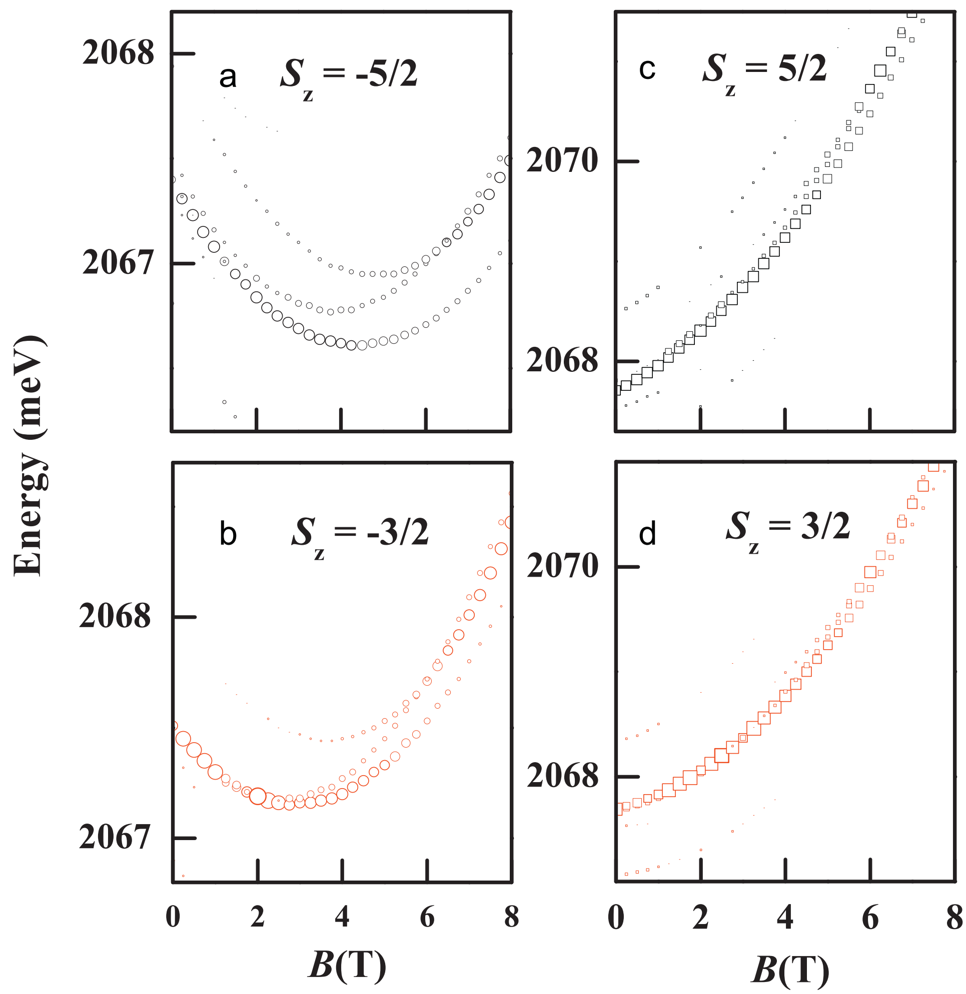

Fig. 3. Magnetic field dependence of the optical transition amplitudes for the case of σ^+ polarization and for various values of the initial state impurity spin projection S_z.

For the first level at $B=0$ the weight of the $|1/2, -3/2, -5/2\rangle$ state is 0.96 and the weight of the $|1/2, -1/2, -5/2\rangle$ state is 0.19. With the increase of the magnetic field, the weight of $|1/2, -3/2, -5/2\rangle$ decreases and the weight of $|1/2, -1/2, -5/2\rangle$ state increases. As a result the bright state changes to dark. For the second level we have an opposite picture. In the case of σ^+ polarization [Fig. 3(a)–(d)] we see similar effects for the case of $S_z = \pm 5/2$ and $\pm 3/2$. Now the bright exciton state $|-1/2, 3/2, S_z\rangle$ is coupled with the dark state $|-1/2, 5/2, S_z\rangle$. For $S_z = \pm 1/2$ the effect is not as pronounced because the energies of the mixed bright and dark states are too close to each other. Although the observed bright-dark state transition is quite similar to the ones observed for QD [6,9], they are different in nature. In a QD the change of bright state to dark or vice versa results in the change of impurity spin projection by unity. For QR besides this mechanism the bright-dark state transition is also related to the change of the ground state angular momentum from zero to nonzero values due to the increase of the magnetic field for all values of the ring radii [16]. Our QR has large radii and width as in [14], which corresponds to strong Coulomb interaction of the exciton [16]. This angular momentum change is a manifestation of the Aharonov–Bohm effect and we believe that for the case of weak Coulomb interaction (QR with small radii and width), the manifestation of the Aharonov–Bohm effect will be more pronounced and we will observe several dark–bright state transition for the same line in the range of magnetic field strength considered in this paper.

An interesting effect was observed for the σ^- polarization. In Fig. 2 there is a crossing point for all energies of the bright exciton states at $B=0.5$ T (see inset in Fig. 2). This is explained as follows: for the σ^- polarization the most important component of the bright exciton states is $|1/2, -3/2, S_z\rangle$, where S_z takes six possible values. For all these states the energy term connected with the s, p–d spin interactions has opposite sign with the Zeeman splitting energy of the magnetic impurity $g_{Mn}\mu_B BS_z$, where $g_{Mn} = 2$. For a certain value of the magnetic field B_0 these two terms will cancel each other and we will see a crossing point. In general, the value of B_0 at the crossing depends on the ring parameters and on the s, p–d interaction constants J_e and J_h. We believe that this effect is experimentally observable. After the detection of the experimental value of the crossing point B_0 one should be able to estimate the real values of the s, p–d interaction constants in a QR. For the σ^+ polarization the most important component of the bright states is $|-1/2, 3/2, S_z\rangle$. Now the s, p–d interaction term and the Zeeman splitting term for the magnetic impurity always have the same sign and there is no crossing point.

In Fig. 4 the angular dependencies of the exciton probability density are presented for the first bright exciton state in the case of σ^- polarization for both, in the absence (Fig. 4(a)) and presence (Fig. 4(b)) of the impurity in the ring under the applied magnetic field $B=2$ T. For the ring without impurity the maximum of the probability density is always on $\varphi_e = \varphi_h$ and it is uniform across the ring. At the presence of the impurity the effective potential of the interaction between the exciton and impurity is repulsive and as a

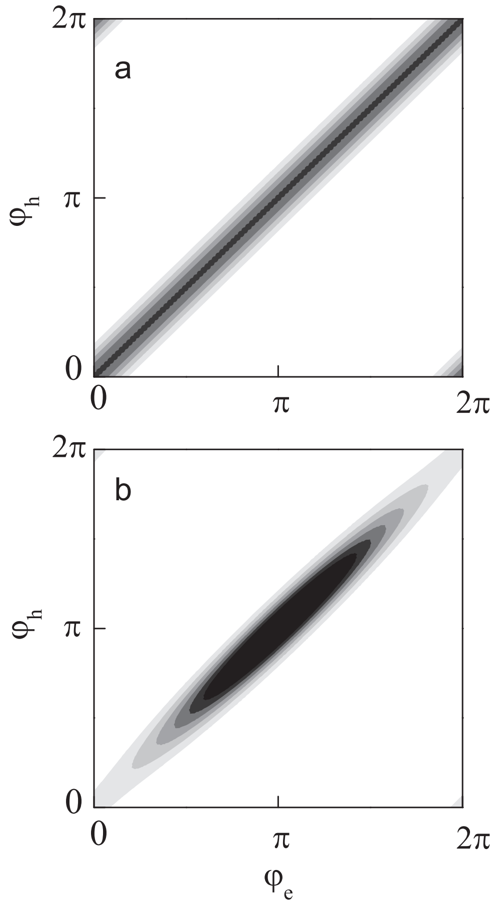

Fig. 4. Angular dependencies of the exciton probability density for the first bright exciton state in the case of σ^- polarization under the applied magnetic field $B=2$ T. a. Ring without magnetic impurity. b. Ring with magnetic impurity.

consequence the maximum of the probability density is shifted to the opposite side of the ring ($\varphi_e = \varphi_h = \pi$). Similar behavior is observed also for dark exciton states.

4. Conclusion

In conclusion, we have studied the effect of spin interactions on the exciton states in a quantum ring containing a single magnetic impurity subjected to a perpendicular magnetic field. The optical properties of such a QR have been investigated. We have shown that due to the s, p–d spin exchange interactions between the electron, the hole and the magnetic impurity it is possible to change the bright exciton state into a dark state and vice versa with the help of the applied magnetic field. Additionally, a new method is proposed for experimental estimation of s, p–d spin interaction constants.

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