Diluted magnetic semiconductor quantum dots: an extreme sensitivity of the hole Zeeman splitting on the aspect ratio of the confining potential

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The valence band states confined in infinitely deep quantum dots made of diluted magnetic semiconductors (DMSs) are considered theoretically. A complex anisotropic structure of the valence bands in DMSs with cubic symmetry described by the full Luttinger Hamiltonian is taken into account. It is found that the Zeeman splitting is very sensitive to the shape of the confining potential and, in particular, to its orientation relative to the direction of an external magnetic field. This sensitivity has its origin in a mixing of different spin components of a hole wave function which takes place for finite hole wave vectors \( \mathbf{k} \). Several consequences of the effect are discussed, including a possibility to control the inter-dot tunneling by an external magnetic field. It is shown also that the polarizations of optical transitions in a single DMS quantum dot depend on details of geometry of its confining potential as well as on the strength of the magnetic field.

I. INTRODUCTION

From the moment of initial stages of their investigation in mid-seventies (see, e.g., [1, 2, 3]) until present diluted magnetic semiconductors (DMSs) and quantum structures involving them, continue to attract a considerable attention of the scientific community (see, e.g., [4, 5]). The interest has recently gained impetus with new focus on spintronic materials (see, e.g., recent review [4]). The part of this interest stems from the fact that the presence of an exchange interaction between the band carriers and electrons from the partially-filled conduction electrons, and the band carriers and electrons from the partially-filled conduction band states and \( N \), that the presence of an exchange interaction between the band carriers and electrons from the partially-filled conduction band states and \( N \) is considerable greater than \( N_0 \). For example, in Cd\(_{1-x}\)Mn\(_x\)Te \( N_0 \alpha = 0.22 \) eV and \( N_0 \beta = -0.88 \) eV [8]. Thus the giant spin splitting (GSS) of excitonic states in DMSs is mainly determined by the spin splitting of the hole states.

The excitonic GSS in DMS quantum structures is determined by the hole splitting to an even greater extent than in bulk crystals. The reason for this is that the electron and the hole states in low-dimensional semiconductor structures are energy dependent mixtures (stemming from the \( k \)-dependence of the mixing in the bulk) of the Bloch amplitudes with \( \Gamma_6 \) and \( \Gamma_8 \) symmetries of the center of the Brillouin zone point group representations. Since the negative exchange constant \( N_0 \beta \) is several times greater in magnitude than the positive \( N_0 \alpha \), even a small admixture of \( \Gamma_8 \) states to \( \Gamma_6 \) state can reduce significantly (up to 30%) the value of total effective exchange constant of the conduction electrons [8]. For quantum wires (QWRs) and, especially, for quantum dots (QDs) this reduction of the conduction band exchange constant is expected to be even more significant. A degree of a reciprocal process of a reduction of the value of the effective exchange constant of the holes due to an admixture of \( \Gamma_6 \) symmetry to \( \Gamma_8 \) states is less pronounced because of the mentioned difference of the magnitudes of the exchange constants \( N_0 \alpha \) and \( N_0 \beta \). Therefore, the exciton spin splitting in DMS quantum structures, which combines the splitting of the electron and the hole comprising it, is dominated by what happens in the valence band.

An example of such processes, that affects the Zeeman splitting of the holes, is an additional heavy hole-light hole mixing induced by the QD confining potential. This has been noted and exploited in Ref. [10], where author used an exactly solvable model, namely of the Luttinger Hamiltonian in a spherical approximation with spatial confinement given by a spherically symmetric quantum dot potential. The reduction of the Zeeman splitting of an optically active ground state of an exciton due to

\[ \hat{H}_{s-a} = N_0 \alpha x \langle \hat{\mathbf{S}} \rangle \cdot \hat{\mathbf{s}}, \]

in the case of the conduction electrons, and the \( p-d \) interaction in the same approximation

\[ \hat{H}_{p-d} = \frac{1}{3} N_0 \beta x \langle \hat{\mathbf{S}} \rangle \cdot \hat{\mathbf{J}}, \]

in the case of the valence band holes. Here \( \hat{\mathbf{s}} \) and \( \hat{\mathbf{J}} \) are operators of the electron spin and the total angular momentum of the hole, respectively, \( x \) is the molar fraction of magnetic ions introduced substitutionally into the semiconductor matrix and \( \langle \hat{\mathbf{S}} \rangle \) represents their average localized spin.

The corresponding exchange constants \( N_0 \alpha \) for the conduction band states and \( N_0 \beta \) for the valence band states in majority of II-VI DMSs are usually of opposite signs [8]. Usually also, the absolute value of \( N_0 \beta \) is considerably greater than \( N_0 \alpha \). For example, in Cd\(_{1-x}\)Mn\(_x\)Te \( N_0 \alpha = 0.22 \) eV and \( N_0 \beta = -0.88 \) eV [8]. Thus the giant spin splitting (GSS) of excitonic states in DMSs is mainly determined by the spin splitting of the hole states.

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heavy-light hole mixing was found to be independent of the QD size. The degree of the reduction depends only on the relation between the heavy and the light hole effective masses in the crystal and for CdTe it amounts to about $\rho \approx 0.8$.

In the present paper we pursue the study of the consequences of the heavy hole-light hole mixing induced by the localization of the holes by a QD potential. However, we concentrate mainly on the dependence of the mixing on the shape or, more specifically, on the aspect ratio of the confining potential. We demonstrate that in different QDs, giving rise to similar confinement energies, the symmetry and, thus, the total angular momentum of the hole ground state might be completely different. To illustrate this point, let us imagine that a characteristic length of a quantum dot, say, $z$-direction $L_z$, is much smaller than that in $x$- and $y$-directions $L_x \ll L_y, L_z$. The situation becomes then similar to the case of a quantum well (QW), where the ground state of the hole is purely of a heavy hole in character with $J_z = \pm 3/2$. In the opposite case, when $L_z \gg L_x, L_y$, the structure resembles a QWR with the ground state of holes being mainly the light hole in character due to the greater masses in $x$-$y$ plane \cite{[1][2]}. Yet, from the point of view of the confinement energy only both these two extreme cases may be the same.

One of the consequences of different symmetries of the hole ground states in different structures having the same confinement energy is a strong dependence of the expectation value $\langle \hat{J}_z \rangle$ on the shape of the confining potential. On the other hand, the Zeeman splitting of the hole states produced by the interaction Hamiltonian \cite{4} in the first order of perturbation theory is proportional to the value of $\langle \hat{J}_z \rangle$ (we shall always assume that external magnetic field $\mathbf{B}$ that induces the magnetization appearing in Eqs. \cite{1} and \cite{3} is along the $z$-axis). One can expect, thus, in the case of DMS a strong dependence of the effective exciton splitting on the aspect ratio of quantum dots. We will show also that due to the strong anisotropy of the hole states the orientiation of the QD with respect to an external magnetic field affects dramatically the Zeeman splitting.

It was shown in Ref. \cite{12} that due to the $k$-dependence of $s(p)$-$d$ exchange constants their values in quantum structures are reduced to a certain extent. Nevertheless, for the sake of simplicity, to describe $s$-$d$ and $p$-$d$ exchange interactions we use in this paper the values of these constants equal to their values in the bulk. This simplification becomes invalid only in very small QDs (with radius smaller than about 30Å), while here we focus our attention on much greater structures and we show that even in those the dependence of the Zeeman splitting on their geometry is significant. Admittedly, for small QDs one should replace bulk $N_0\beta$ by the values of $p$-$d$ exchange constants, calculated in \cite{13}. This does not change, of course, the qualitative results obtained in the present paper.

To sum up, we are considering here the shape-induced changes of $\langle \hat{J}_z \rangle$, and their various consequences, in DMS QDs while neglecting the variation of exchange constant $N_0\beta$ itself since we expect it to be of smaller importance.

**II. THEORETICAL MODEL**

We consider the valence band states in a diluted magnetic semiconductor QD in a shape of a rectangular parallelepiped with dimensions $L_x, L_y$, and $L_z$. This particular choice of the form of confining potential is motivated by two reasons. First, by changing the relations between $L_x, L_y$ and $L_z$ one can control the asymmetry of the confining potential. This is convenient both for calculations and for interpretation of the results obtained. The second reason is that the symmetry of QD potential in this particular case is rather low. In a general case, this is $D_{2h}$ symmetry. Such a low symmetry prevents appearance of additional constants of motion, such as the angular momentum which "creeps in" in the case of spherically symmetric QDs.

For simplicity we consider the case of a dot with infinitely high potential barriers surrounding it. The parameters $L_x, L_y$ and $L_z$ can be treated then as characteristic length scales of the hole localization in a real QD (i.e., with finite potential barriers) in $x, y$ and $z$-directions.

Our basic starting Hamiltonian is the Luttinger Hamiltonian \cite{14} describing the hole states in a cubic semiconductors which can be presented in the form

$$\hat{H}_L = \frac{\hbar^2}{2m_0} \left[ \left( \gamma_1 + \frac{5}{2} \gamma_2 \right) (\hat{K}_x^2 + \hat{K}_y^2 + \hat{K}_z^2) - 2\gamma_2 (\hat{K}_x^2 \hat{J}_x^2 + \hat{K}_y^2 \hat{J}_y^2 + \hat{K}_z^2 \hat{J}_z^2) - 2\gamma_3 \left( \hat{K}_x \hat{K}_y (J_x J_y + J_y J_x) + \hat{K}_z (J_x J_z + J_z J_x) + \hat{K}_z \hat{K}_y (J_z J_y + J_y J_z) \right) \right],$$

where $\hat{K}_x = -i\partial/\partial x$, $\hat{K}_y = -i\partial/\partial y$, $\hat{K}_z = -i\partial/\partial z$, with the $z$-axis is parallel to [001] crystallographic direction, $\gamma_i$ are the Luttinger parameters, while $J_x, J_y$ and $J_z$ being $4 \times 4$ operator matrices of the projections of the angular momentum $J = 3/2$ in the basis of $\Gamma_8$ Bloch amplitudes

$$\psi_{3/2} = \frac{1}{\sqrt{2}} (X + iY) \uparrow,$$

$$\psi_{1/2} = \frac{1}{\sqrt{6}} \left[ (X + iY) \downarrow - 2Z \uparrow \right],$$

$$\psi_{-1/2} = \frac{1}{\sqrt{6}} \left[ -(X - iY) \uparrow - 2Z \downarrow \right],$$

$$\psi_{-3/2} = -\frac{1}{\sqrt{2}} (X - iY) \downarrow.$$

We expand the orbital part of each of the spin components \cite{11}-\cite{14} of the hole wave function in the basis of
the solutions of the Schrödinger equation for an infinitely deep QD. The hole wave function is then represented by

$$\Psi = \sum_{J_z=-3/2}^{3/2} \sum_{N_x} \sum_{N_y} \sum_{N_z} A_{J_z}^{n,m,l} \sqrt{\frac{8}{L_xL_yL_z}} \times \sin(nk_xx) \sin(nk_yy) \sin((k_z + \phi)z) \cdot \Psi_J,$$

where

$$A_{J_z}^{n,m,l} = \sin(nk_x x) \sin(nk_y y) \sin((k_z + \phi)z)$$

This approximation is equivalent to neglecting of term proportional to $\gamma_3$ in the Hamiltonian (3) or, more strictly, to a reduction of the spherical term $\propto \gamma_3 (\hat{K} \cdot \hat{J})^2$ to a one with cubic symmetry $\propto \gamma_3 (K_x^2 J_z^2 + K_y^2 J_z^2 + K_z^2 J_z^2)$. In fact, the terms proportional to $K_z^2$ in Eq. (3) do not mix states with different orbital functions in the expansion (8) and the hole ground state has the form [3].

With the above approximation the last term in Eq. (3), being linear in the operators $\hat{K}_z$, has only vanishing matrix elements. At the same time, the second term in Eq. (3) is proportional to the squares of the matrices $J_z$. Thus, the non-vanishing matrix elements of this term involve states with $\Delta J_z = \pm 2$ only. As a result, the $4 \times 4$ matrix of the Hamiltonian (3) splits into two $2 \times 2$ identical submatrices in the basis $\{+3/2, -1/2\}$ and $\{-3/2, +1/2\}$. These matrices have the form

$$H = \frac{\hbar^2}{2m_0} \begin{pmatrix} P + Q & R \\ R & P - Q \end{pmatrix},$$

where

$$P = \gamma_1(k_x^2 + k_y^2 + k_z^2),$$

$$Q = \gamma_2(k_x^2 + k_y^2 - 2k_z^2),$$

$$R = -\sqrt{3}\gamma_2(k_x^2 - k_y^2).$$

In the case of a strictly cubic QD ($k_x = k_y = k_z$), the ground state of the hole is four-fold degenerate. The hole states can be characterized by the projection of the angular momentum $J_z = \pm 3/2, \pm 1/2$. We can remove this degeneracy by reducing the symmetry of the Hamiltonian from $O_h$ to $D_{4h}$ ($k_x \neq k_y = k_z$). This leads to the heavy hole-light hole splitting, $J_z$ remains a good quantum number. The hole ground state doublet is then the heavy hole doublet with $J_z = \pm 3/2$ if $k_x^2 + k_y^2 < 2k_z^2$, while it corresponds to the light hole doublet with $J_z = \mp 1/2$, if $k_x^2 + k_y^2 > 2k_z^2$.

In the general case, $k_x \neq k_y \neq k_z$. (the symmetry of the Hamiltonian being $D_{2h}$) the hole states are the mixtures of the components of the Hamiltonian being $J_z = \pm 3/2$ and $J_z = \mp 1/2$. In the absence of an external magnetic field the ground state with the energy

$$E = \frac{\hbar^2}{2m_0} (P - \sqrt{Q^2 + R^2})$$

is twofold degenerate. This effective spin doublet is characterized by an expectation value of the projection of the total angular momentum

$$\langle J_z \rangle = \pm \left( \frac{2}{1 + (\delta + \sqrt{\delta^2 + 1})} - \frac{1}{2} \right),$$

where

$$\delta = \frac{Q}{R} = \frac{k_x^2 + k_y^2 - 2k_x^2}{\sqrt{3}(k_z^2 - k_x^2)}.$$

For degenerate states the choice of the wave functions is not unique. To obtain Eq. (12) we use the correct zero-order functions with respect to the perturbation $GJ_z$. For each value of $k_x$ and $k_y$ it is possible to find a value of $k_z$ that leads to vanishing of $\langle J_z \rangle$. The corresponding condition is $\delta = 1/\sqrt{3}$. In the case of $k_x > k_y$ to satisfy this equality one has to choose $k_z = k_y$. This result easy to see: the equality $k_z = k_y$ under the condition $k_x > k_y$ means that the hole ground state is a superposition of the states with $J_z = \pm 3/2$ only. The average value $\langle J_z \rangle$ for such states is equal to zero, similarly to the case when the states are the mixtures of states with $J_z = \pm 3/2$ and then $\langle J_x \rangle$ and $\langle J_y \rangle$ vanish.

It is seen, thus, that the expectation value of $J_z$ for the hole ground state in a QD varies in a wide range from $\langle J_z \rangle = \pm 3/2$ to $\langle J_z \rangle = 0$ depending on the exact shape of the confining potential, i.e., relation between its extension in three crystallographic directions.

The situation changes somewhat if we take into account more than one term in the expansion (8). The projection $J_z$ ceases to be a good quantum number for the holes even in the case when $k_x = k_y$. This is due to the presence of the term proportional to $\gamma_3$ in Eq. (3). Moreover, the hole states now are the mixtures of all four spin components. Nevertheless, as before, one can vary effectively the value of $\langle J_z \rangle$ in the ground hole state from nearly $\pm 3/2$ to $0$ by changing the shape of the confining potential. To illustrate this feature we plot in Fig. [3] the expectation value of the projection of the total angular momentum $J_z$ for the ground spin doublet in the QD having $L_x = 50A$ and $L_y = 70A$ (solid line) and $L_x = 40A$ and $L_y = 80A$ (dashed line) as a function of $L_z$. Hereafter we use the number of basis functions in the expansion $N_x = N_y = N_z = 6$, which were checked to be sufficient to ensure numerical accuracy (about $1 \text{meV}$ in calculation of hole energy). In our calculations we use the CdTe Luttinger parameters $\gamma_1 = 5.3$, $\gamma_2 = 1.62$ and
As mentioned above, the value of the Zeeman splitting of the valence band states in the magnetic field \( \mathbf{B} \parallel z \) is proportional, in the first order of perturbation, to \( \langle J_\parallel \rangle \). However, the perturbation theory is not quite justified if the value of the Zeeman splitting becomes comparable to the energy separation between the size-quantized states in a QD. To calculate the Zeeman splitting in such case we perform a numerical diagonalization of the full Hamiltonian matrix for the holes, which is represented by the sum of the Luttinger Hamiltonian (3) and the \( p-d \) exchange interaction (2). Note, that we neglect here the direct influence of the magnetic field on the hole states. The magnetic field in our model just induces the magnetization on the sample, which is proportional, in turn, to the average spin (\( \hat{S} \)) of the magnetic ions appearing in (3). In the calculations that are presented below we use the parameterization of the value (\( \hat{S} \)) in real DMS structures, given in (4).

### III. DISCUSSION

The evolution of the positions of the two lowest energy levels in the valence band in \( \text{Cd}_{0.97}\text{Mn}_{0.03}\text{Te} \) DMS quantum dots having \( L_x = 50\,\text{Å} \), \( L_y = 70\,\text{Å} \) and \( L_z = 65\,\text{Å} \) (solid lines) and \( L_x = 65\,\text{Å} \), \( L_y = 70\,\text{Å} \) and \( L_z = 50\,\text{Å} \) (dashed lines) in the magnetic field \( \mathbf{B} \parallel z \) is presented in Fig. 2. Such QDs provide exciton confinement energy of about 400 meV, that is, of the correct magnitude as compared to existing experimental observations in real self-assembled QDs made of this material (7). In a small magnetic field the value of the Zeeman splitting of the ground state is proportional to the expectation value of the projection of the total angular momentum of the hole on the direction of the magnetic field. This quantity is \( \langle J_z \rangle = 0.003 \) for the first structure and \( \langle J_z \rangle = 1.2 \) for the second structure, which is just the first one merely rotated by \( \pi/2 \) around the \( y \)-axis. Due to the fact that \( J_z \) is not a good quantum number, the magnetic field \( \mathbf{B} \parallel z \) induces the mixing of the hole states corresponding to different size-quantized levels. With an increase of the magnetic field the admixture of the higher energy states to the ground levels becomes significant and it produces a deviation of the value of the Zeeman splitting from that predicted by the first order of perturbation theory. Nevertheless, Fig. 2 shows that due to a strong dependence of \( \langle J_z \rangle \) on the actual geometry it is possible to obtain DMS quantum dots with the same, homogeneous distribution of the magnetic impurities, that have the same space-quantized energies but, nevertheless, differ essentially in their effective \( g \)-factors.

This sensitivity of the effective \( g \)-factor to the exact shape and orientation of the confining potential can be used to control hole tunneling between DMS quantum dots. To clarify this point, let us consider two QDs localizing spatially the holes in DMS QW in two in-plane directions. In the third direction, say \( x \)-direction, the holes are localized due to the presence of a QW confining potential. Such a pair of the QDs is, thus, made of the same material and have the same dimensions in the \( x \)-direction. The remaining two spatial dimensions of these QDs are chosen in such a way as to result in different expectation values of some component of the angular momentum of a hole, say \( \langle J_z \rangle \), in the plane of the QW. Moreover, the ground state energy of the hole in the QD having the greater value \( \langle J_z \rangle \) should be somewhat higher than that in the other QD, see Fig. 3. The
energy difference between these two ground states and the distance between the QDs may prevent the tunneling of a hole between these two QDs. Therefore, we refer to these QDs as uncoupled in this case. Now, the magnetic field \( B \parallel z \) shifts the ground states of holes in QDs. This shift is different for each dot and is proportional to \( \langle J_z \rangle \). This is due to the fact that the \( \langle J_z \rangle \) are different for our two dots. At some magnetic field energies of ground states of holes in two QDs may become the same, see Fig. 3. One can achieve, thus, the condition favorable for the resonant tunneling. Let us assume now that the hole originally is localized in the first QD (e.g., where the ground state in the absence of the field is lower) and that the time length of the magnetic field pulse is made equal to one half (three halves, five halves etc.) of the period of the hole oscillations between the two QDs. Then, after such a pulse the hole will be localized in the second QD.

The system is scalable to some extent. It is possible, for example, to add a third QD with another value and/or direction of the resonant magnetic field that couples the second and the third QD by the tunneling. As a result, by applying the magnetic field that effectively couples the second and the third QD one can also transfer the hole between these dots without affecting the hole present in the first QD. Such an arrangement can be envisaged as useful in quantum logic gates.

Of course, the quantitative analysis of a possibility of such hole tunnel transport as well as the quantitative description of the Zeeman splitting of excitons in DMS QDs requires the consideration of the finite potential barriers and the valence band-conduction band mixing.

Along with the Zeeman splitting, the polarization characteristics of optical transitions in QDs have to be strongly affected by the shape of the confining potential.

One can expect this even by inspecting Fig. 4. A strong dependence of the character of the hole ground state (i.e., the relative contribution of different spin components to the wave function) on the shape of a QD should result not only in a variation of \( \langle J_z \rangle \) but also of probabilities of optical transitions. The values describing the probabilities of various optical transitions in different linear polarizations of light associated with the fourfold degenerate exciton ground state as a function of \( L_z \) for QD having \( L_x = 70 \text{ Å} \) and \( L_y = 50 \text{ Å} \) is plotted in Fig. 4. With an increase of \( L_z \), the situation changes from QW-like \( (L_z \ll L_x, L_y) \) to QWR-like \( (L_z \gg L_x, L_y) \). At the same time, the intensity of the optical transition in the \( E \parallel z \) polarization increases, while the intensities of the remaining two polarizations, considered here, decrease.

Finally, let us mention yet one more effect that takes place in DMS quantum dots. It was shown in Ref. [12] that the localization of the holes in two spatial directions in the case of DMS quantum wires results in a possibility to control the character of the ground state in the valence band by means of a magnetic field. This leads, in turn, to a strong magnetic field dependence of the polarization of optical transitions in such structures. A similar situation occurs, of course, in the case of DMS quantum dots. In Fig. 5 we plot the magnetic field dependence of the probabilities of optical transitions in circular \( \sigma^+ \), \( \sigma^- \), and in two linear polarizations for ground (non-degenerate) exciton state in \( \text{Cd}_{0.97}\text{Mn}_{0.03}\text{Te} \) QD with \( L_x = 70 \text{ Å} \), \( L_y = 60 \text{ Å} \) and \( L_z = 50 \text{ Å} \). It is seen, that by application of a magnetic field one can effectively control the polarization characteristics of the light emitted from such DMS QD.

Let us emphasize, that we use in our calculations the full, anisotropic Luttinger Hamiltonian and a rectangular profile of a QD, so the selection rules for the optical transitions differ substantially from those obtained assuming spherically symmetric quantum dots. For example, in
FIG. 5: The probability of optical transitions in two circular, and two linear polarizations for the ground state of exciton in Cd\textsubscript{0.97}Mn\textsubscript{0.03}Te DMS QD with \( L_x = 70\,\text{Å} \), \( L_y = 60\,\text{Å} \) and \( L_z = 50\,\text{Å} \) in the magnetic field \( B \parallel z \) at \( T=2\,\text{K} \).

our case the ground state of the exciton in a QD is not a dark state. Moreover, all valence band states in our case are linear combinations of all four different spin (or more appropriately, of all four projections of the total angular momentum) components, resulting in a possibility of observation of optical transitions in all polarizations. Nevertheless, in Fig. 5 we do not plot the probabilities of optical transitions in \( \pi \) polarization (\( E \parallel z \)). Such transitions are forbidden, which is connected to the fact, that we have dealt here with QDs having infinitely high barriers. This is analogous to the situation of QWs, when the optical transitions, such as \( e_1-h_3 \), are forbidden only in the case of infinite barriers.

The effects, considered in the present paper, do exist also in the case of non-magnetic quantum dots. However, in the case of diluted magnetic semiconductor QD the effects are much more pronounced, as a direct consequence of the large values of the effective g-factor of the holes.

Acknowledgments

This work was partially supported by FENIKS RTD (EC: G5RD-CT-2001-00535).

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