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Highly anisotropic Zeeman splittings of wurtzite Cd$_{1-x}$Mn$_x$Se quantum dots

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The electronic structure and Zeeman splittings of wurtzite Cd$_{1-x}$Mn$_x$Se quantum spheres are studied using the $k \cdot p$ method and mean-field model. It is interesting to find that the Zeeman splittings of some hole states in quantum spheres are highly anisotropic due to the spin-orbit coupling and wurtzite crystal structure. The anisotropy of the Zeeman splittings of hole ground states in large dots is large, while that in small dot is small because the hole ground states vary with radius. An external electrical field can change the Zeeman splitting significantly, and tune the g factor from nearly 0 to about 100. © *2007 American Institute of Physics.* [DOI: 10.1063/1.2784192]

Nowadays, much of the research in semiconductor physics has been shifting toward diluted magnetic semiconductors (DMSs),[1–3] which have extensive applications in spintronics. Manganese-doped II-VI (Ref. 4) and III-V (Ref. 1) compound semiconductors have been widely studied. Meanwhile, the investigations of quantum confinement of carriers in spatially modulated semiconductor structures have been a field of intense activity over the past decades. High-quality CdSe quantum dots were synthesized recently.[5,6] The electronic structure of these quantum spheres under magnetic field were theoretically studied.[7,8] The method to dope Mn ions into CdSe quantum dots was achieved.[9] DMS quantum dots were used as spin-polarized light-emitting diodes.[10] The electron and hole states in zinc-blende DMS quantum dots were studied using $k \cdot p$ method.[11]

The anisotropic Zeeman splitting was earlier found in the GaAs/AlAs quantum wells.[12] The fine structure of excitons in GaAs/AlAs quantum wells showed that the perpendicular (to the well plane) g factors of the heavy hole g_{hz} are in between 2 and 3, while the in-plane values g_{hx} and g_{hy} are smaller than 0.01. This strong anisotropy of the effective hole g value is a consequence of description of the heavy-hole states with $J_{hz} = \pm \frac{3}{2}$ by an effective spin $S_h = \frac{1}{2}$. From the spin Hamiltonian it can be seen that the in-plane splittings can only be due to the cubic hole Zeeman interaction terms. The small values of g_{hx} and g_{hy} correspond to a small q value in a bulk semiconductor. On the other hand, the bulk linear hole Zeeman splitting constant κ, which is about 1.2 for GaAs, results in a large g_{hz} value for GaAs layer samples.

Recently, the anisotropic Zeeman splitting has been found in ballistic one-dimensional hole systems.[13] The splitting of the subband edges is shown in the transconductance gray scale plot when an in-plane magnetic field is applied parallel to the one-dimensional (1D) GaAs hole systems. In contrast, the transconductance gray scale shows that the degenerate 1D subbands are not affected by the perpendicular

magnetic field of up to 8.8 T, i.e., no Zeeman splitting is seen when the magnetic field is aligned perpendicular to the channel. This anisotropy of the effective g factor is a direct consequence of the one-dimensional confinement on a system with strong spin-orbit coupling.

It is expected that the anisotropic Zeeman splitting will also happen in quantum dots, which is very interesting and useful as the carriers' spins in quantum dots can be used as qubits for quantum information processing.[14] In this letter, we use the six-band $k \cdot p$ method of semiconductor quantum dots, taking into account the p-d exchange interaction, to study the electronic structure, Zeeman splittings, and g factors of paramagnetic wurtzite Cd$_{1-x}$Mn$_x$Se quantum spheres under external magnetic and electric fields.

We represent the six-band hole Hamiltonian under external magnetic and electric fields of DMS wurtzite quantum dots in the basis functions $|11\rangle \uparrow$, $|10\rangle \uparrow$, $|1-1\rangle \uparrow$, $|11\rangle \downarrow$, $|10\rangle \downarrow$, $|1-1\rangle \downarrow$ as

$$H_{\text{tot}} = -\begin{pmatrix} H_{h0} & 0 \\ 0 & H_{h0} \end{pmatrix} - H_{\text{so}} + V_{E\text{field}} - H_{\text{mm\_h}} + H_{\text{asym}}$$
$$+ H_{\text{Zeeman\_h}} + H_{\text{pd}}. \qquad (1)$$

H_{h0} is written as

$$H_{h0} = \frac{1}{2m_0} \begin{pmatrix} P_1 & S & T \\ S^\star & P_3 & S \\ T^\star & S^\star & P_1 \end{pmatrix}, \qquad (2)$$

where P_1, P_3, S, T, H_{so}, $H_{\text{mm\_h}}$, H_{asym}, $H_{\text{Zeeman\_h}}$, and the effective-mass parameters of CdSe were given in detail before.[8] $V_{E\text{field}}$ is the electric field potential term. When the electric field F is applied along the x direction, $V_{E\text{field}} = eFx$.

H_{pd} is the p-d exchange interaction Hamiltonian between the hole and magnetic ion, which is written as[2]

$$H_{pd} = \beta \mathbf{s}_h \cdot \mathbf{M} / (g_{\text{Mn}} \mu_B), \qquad (3)$$

where $g_{\text{Mn}} = 2$, $\mathbf{s}_h$ is the spin operator of the hole, β is the p-d exchange constant, $\beta N_0 = -1.11$ eV,[3] N_0 is the number of

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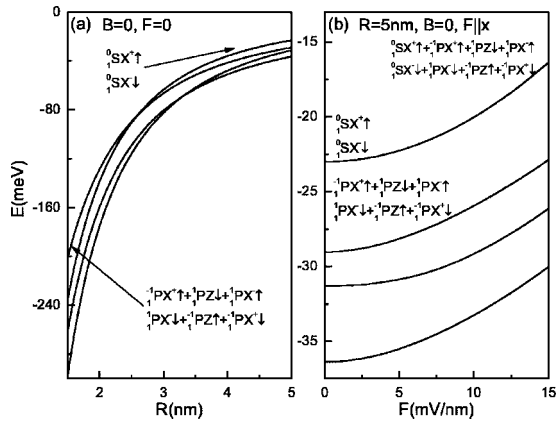
FIG. 1. (a) Hole levels of $Cd_{1-x}Mn_xSe$ quantum spheres at $B=0$ and $F=0$ as functions of R. (b) Hole levels of $Cd_{1-x}Mn_xSe$ quantum spheres with $R=5$ nm at $B=0$ as functions of F ($F\|x$).

FIG. 2. (a) Hole levels of $Cd_{1-x}Mn_xSe$ quantum spheres with $R=5$ nm and $x_{eff}=0.05$ at $F=0$ and $T=10$ K as functions of B ($B\|z$). (b) Similar to (a) but $B\|x$. (c) Hole g factor of $Cd_{1-x}Mn_xSe$ quantum spheres with $R=5$ nm and $x_{eff}=0.05$ at $F=0$ and $T=10$ K as a function of B ($B\|z$). (d) Similar to (c) but $B\|x$.

cations per unit volume, and $\mathbf{M}$ is the magnetization of the localized spins of magnetic ions. In paramagnetic case, $\mathbf{M}$ is parallel with external magnetic field, whose magnitude is given by[2]

$$M = Sg_{Mn}\mu_B N_0 x_{eff} B_S\left[\frac{Sg_{Mn}\mu_B B}{k_B(T+T_{AF})}\right], \quad (4)$$

where $S=\frac{5}{2}$ is the spin of the magnetic ion, x_{eff} is the effective composition of magnetic ions, T_{AF} accounts for the reduced single-ion contribution due to the antiferromagnetic Mn–Mn coupling,[2] for $Cd_{1-x}Mn_xSe$, $T_{AF}=1$ K,[2] and $B_S(x)$ is the Brillouin function.[2]

We assume that the carriers are confined in an infinitely high potential barrier. The envelope function has six components, expanded with the spherical Bessel function and spherical harmonic function $j_l(\alpha_n^l r/R)Y_{l,m}$,[8] where α_n^l is the nth zero point of $j_l(x)$, and R is the radius of quantum dot.

Figure 1(a) shows the hole levels of $Cd_{1-x}Mn_xSe$ quantum spheres as functions of the radius R in the absence of magnetic field. The hole ground states are double degenerated, whose state components are labeled, for example, $^0_1SX^+\uparrow$ means that the state has the envelope function with $n=1$, $m=0$, and $l=0$, the Bloch state $|11\rangle$, and the spin-up state (which is defined in the z direction). We see that the highest two energy levels cross at about $R=2.5$ nm, and when R is small, the hole ground states are P states. Thus, in small wurtzite quantum spheres, the ground hole states are optically dark.[7,8] When R is large, the hole ground states are S states ($^0_1SX^+\uparrow$ and $^0_1SX^-\downarrow$), whose space-wave functions $^0_1SX^+$ and $^0_1SX^-$ are totally different. This difference will affect the Zeeman splitting under magnetic field dramatically, which will be shown later. While the space-wave functions of the ground P states in small R case are almost the same, for example, $^{-1}_1PX^+\uparrow$ and $^{-1}_1PX^+\downarrow$ components have the same space-wave function $^{-1}_1PX^+$. Thus, as the hole ground states vary with R, their spin states vary a lot. Figure 1(b) shows the hole levels of the spheres with $R=5$ nm at $B=0$ as functions of F ($F\|x$). We see that the electric field couples the highest two hole states. Many P state components of the second level are mixed into the first level, and when $F=15$ mV/nm, about half of the state components of the ground states are P states. $^{-1}_1PX^+\uparrow$ and $^{-1}_1PX^+\downarrow$ (whose space-wave functions are same) are mixed into $^0_1SX^+\uparrow$ and $S^0_1X^-\downarrow$ (whose space-wave functions are different), respectively. Thus, the spin states of the hole ground states vary a lot as the electric field increases, which will affect the Zeeman splitting significantly and will be shown later.
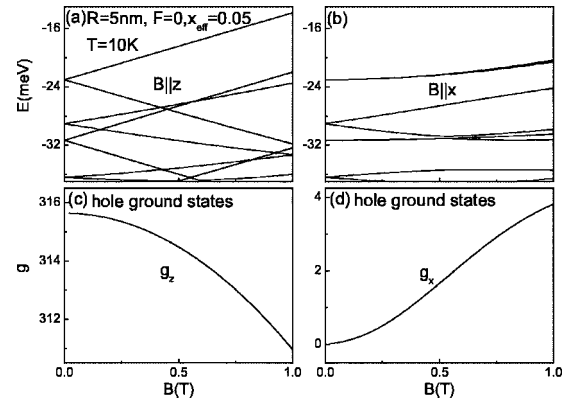
The hole levels of $Cd_{1-x}Mn_xSe$ quantum spheres with $R=5$ nm as functions of B at $F=0$ are shown in Figs. 2(a) and 2(b) for $B\|z$ and $B\|x$, respectively. The double degenerated states split under magnetic field, which is refered to Zeeman splitting. It is interesting to note that the Zeeman splittings of some states are highly anisotropic, for example, the hole ground states, whose space-wave functions are totally different (see Fig. 1). The Zeeman splitting of these states can be as large as 18 meV at $B=1$ T when $B\|z$, and is tiny when $B\|x$. Physically, the highly anisotropic Zeeman splitting is induced by the spin-orbit coupling effect, which makes the spin states couple with the space-wave functions. The space-wave functions are anisotropic due to the crystal field splitting energy [$\Delta_c=25$ meV] (Ref. 8). This makes the spin states anisotropic. In the whole Hamiltonian [Eq. (1)], there are three terms which induce Zeeman splitting, the H_{asym}, H_{Zeeman_h}, and H_{pd}; the last one dominates in our case. When $B\|z$ the three terms can be written as $A\sigma_z$, BI_z,[8] and $C\sigma_z$, respectively, A, B, and C are the coefficients. They have large matrix elements in the states of $^0_1SX^+\uparrow$ and $^0_1SX^-\downarrow$, leading to large Zeeman splitting. When $B\|x$ the three terms can be written as $A\sigma_x$, BI_x, and $C\sigma_x$, respectively. $A\sigma_x$ and $C\sigma_x$ have zero matrix elements between the states of $^0_1SX^+\uparrow$ and $^0_1SX^-\downarrow$ as their space-wave functions are orthogonal. BI_x has zero matrix element between the states of $^0_1SX^+\uparrow$ and $^0_1SX^-\downarrow$ because the spin states are orthogonal. That is why the Zeeman splitting of the ground states in Fig. 2(b) is small. There are some hole states which have nearly same space-wave functions, for example, the first excited states. The anisotropy of their Zeeman splittings is very small. We can define an effective g factor $g=\Delta E/(\mu_B B)$ to indicate the Zeeman splitting energy (ΔE) at a given magnetic field. The calculated effective g factors of the hole ground states are shown in Figs. 2(c) and 2(d) for $B\|z$ and $B\|x$, respectively. The g_z (when $B\|z$) is about 300 and decreases as the magnetic field increases because the magnetization of the localized spins shows a saturation trend as B increases. The g_x is nearly zero at $B=0$ and increases as B increases due to the state coupling. As shown in Fig. 2(b), one of the low excited state ascends close to the ground states and couples with them, leading to their splitting.

Figure 3 shows the hole levels and g factors of $Cd_{1-x}Mn_xSe$ quantum spheres with $R=1.5$ nm and $x_{eff}=0.05$ at $F=0$ and $T=10$ K as functions of the magnetic field B for $B\|z$ (a) and $B\|x$ (b). The difference from Fig. 2 is that

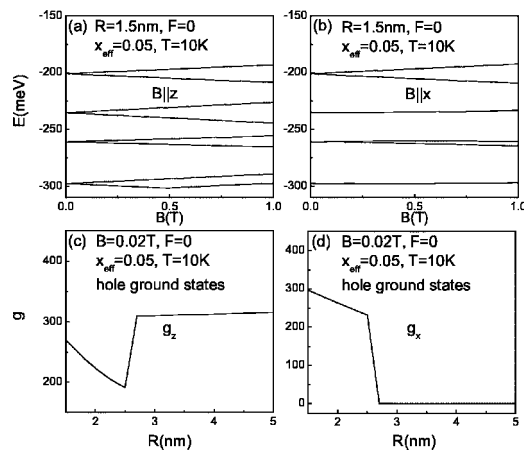FIG. 3. (a) Hole levels of $Cd_{1-x}Mn_xSe$ quantum spheres with $R=1.5$ nm and $x_{eff}=0.05$ at $F=0$ and $T=10$ K as functions of B ($B\|z$). (b) Similar to (a) but $B\|x$. (c) Hole g factor of $Cd_{1-x}Mn_xSe$ quantum spheres with $x_{eff}=0.05$ at $B=0.02$ T ($B\|z$), $F=0$, and $T=10$ K as a function of R. (d) Similar to (c) but $B\|x$.

FIG. 4. (a) Hole levels of $Cd_{1-x}Mn_xSe$ quantum spheres with $R=5$ nm and $x_{eff}=0.05$ at $F=15$ mV/nm ($F\|x$) and $T=10$ K as functions of B ($B\|z$). (b) Similar to (a) but $B\|x$. (c) Hole g factor of $Cd_{1-x}Mn_xSe$ quantum spheres with $R=5$ nm and $x_{eff}=0.05$ at $B=0.02$ T ($B\|z$) and $T=10$ K as a function of F ($F\|x$). (d) Similar to (c) but $B\|x$.

the first excited states instead of the ground states have highly anisotropic Zeeman splitting. It is due to the change of hole ground states, as shown in Fig. 1(a). Figures 3(c) and 3(d) show the g_z and g_x of the hole ground states as functions of R. The sharp changes indicate the level crossing. We see that g_z is always big, while g_x is small when R is large. This significant change of g_x gives a way to test the interesting level crossing in wurtzite quantum dots which causes the dark exciton[7,8] whose exciton recombination time is very long. This long living exciton is useful for investigating the properties of excitons, such as the Bose-Einstein condensation of excitons. As the spin states of the hole ground states of small ($R<2.5$ nm) and large ($R>2.5$ nm) quantum dots are different, we can use them as different types of qubits for quantum information processing.[14]

Therefore, the states which have very different space-wave functions have highly anisotropic Zeeman splitting. The former works on highly anisotropic Zeeman splittings[12,13] focused on the heavy-hole states whose space-wave functions are different (such as $_1^0SX^+\uparrow$ and $_1^0SX^-\downarrow$ in Fig. 1). Our explanation is applicable to their results and suggests investigation of the highly anisotropic Zeeman splitting of other states that have very different space-wave functions.

Figure 4 shows the hole levels and g factors of $Cd_{1-x}Mn_xSe$ quantum spheres with $R=5$ nm in the presence of magnetic field and electric field. The electric field induces state coupling [see Fig. 1(b)], and changes the spin states of the hole ground states. Thus, the Zeeman splittings are changed significantly by the electric field. Compared to the $F=0$ case (see Fig. 2), the Zeeman splitting of the hole ground states when $B\|x$ is increased dramatically. The g factors can be tuned by the electric field, as shown in Figs. 4(c) and 4(d). g_z decreases and g_x increases with increasing electric field due to the state coupling. It is interesting to note that g_x varies from nearly 0 to larger than 100 as the electric field increases. Thus at a fixed magnetic field, we can use the electric field to tune the hole spin to be unpolarized ($g_x=0$) or polarized (g_x is large).

In summary, we studied the electronic structure, Zeeman splittings and g factors of wurtzite $Cd_{1-x}Mn_xSe$ quantum spheres using the six-band $k\cdot p$ method taking into account the p-d exchange interaction between the carrier and the magnetic ion. It is interesting to find that the Zeeman
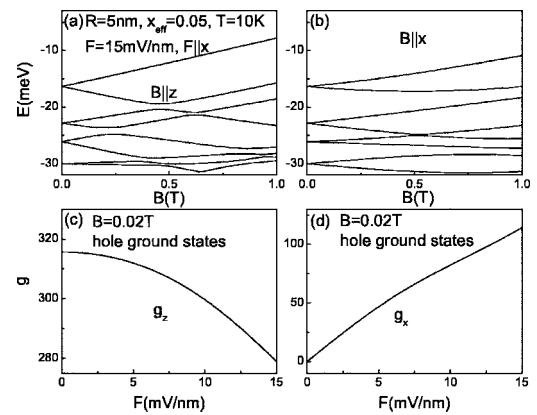
splittings of the hole states in quantum spheres can be highly anisotropic due to the spin-orbit coupling and the wurtzite crystal structure. The states which have very different space-wave functions have highly anisotropic Zeeman splitting. The anisotropy of the Zeeman splittings of hole ground states in large dots is large, while that in small dot is small, due to the change of hole ground states with radius, and this gives us a way to test the interesting level crossing in wurtzite quantum dots which causes the long living dark exciton.[7,8] An external electrical field can change the Zeeman splitting significantly, and tune the g factor from nearly 0 to about 100 due to the coupling of hole states induced by the electric field. Under a fixed magnetic field, we can use the electric field to tune the hole spin to be unpolarized ($g_x=0$) or polarized (g_x is large).

[1] H. Ohno, Science **281**, 951 (1998).
[2] T. Dietl, H. Ohno, and F. Matsukura, Phys. Rev. B **63**, 195205 (2001).
[3] J. K. Furdyna, J. Appl. Phys. **64**, R29 (1988).
[4] A. Wall, A. Franciosi, D. W. Niles, R. Reifenberger, C. Quaresima, M. Capozi, and P. Perfetti, Phys. Rev. B **41**, 5969 (1990).
[5] E. Johnston-Halperin, D. D. Awschalom, S. A. Crooker, Al. L. Efros, M. Rosen, X. Peng, and A. P. Alivisatos, Phys. Rev. B **63**, 205309 (2001).
[6] A. Kasuya, R. Sivamohan, Y. A. Barnakov, I. M. Dmitruk, T. Nirasawa, V. R. Romanyuk, V. Kumar, S. V. Mamykin, K. Tohji, B. Jeyadevan, K. Shinoda, T. Kudo, O. Terasaki, Z. Liu, R. V. Belosludov, V. Sundararajan, and Y. Kawazoe, Nat. Mater. **3**, 99 (2004).
[7] J.-B. Xia and J. Li, Phys. Rev. B **60**, 11540 (1999).
[8] X. W. Zhang and J. B. Xia, Phys. Rev. B **72**, 205314 (2005).
[9] F. V. Mikulec, M. Kuno, M. Bennati, D. A. Hall, R. G. Griffin, and M. G. Bawendi, J. Am. Chem. Soc. **122**, 2532 (2000).
[10] S. Chakrabarti, M. A. Holub, P. Bhattacharya, T. D. Mishima, M. B. Santos, M. B. Johnson, and D. A. Blom, Nano Lett. **5**, 209 (2005).
[11] K. Chang, S. S. Li, J. B. Xia, and F. M. Peeters, Phys. Rev. B **69**, 235203 (2004).
[12] H. W. van Kesteren, E. C. Cosman, W. A. J. van der Poel, and C. T. B. Foxon, Phys. Rev. B **41**, 5283 (1989).
[13] R. Danneau, O. Klochan, W. R. Clarke, L. H. Ho, A. P. Micolich, M. Y. Simmons, A. R. Hamilton, M. Pepper, D. A. Ritchie, and U. Zülicke, Phys. Rev. Lett. **97**, 026403 (2006).
[14] *Semiconductor Spintronics and Quantum Computation*, Nanoscience and Technoloty, edited by D. D. Awschalom, D. Loss, and N. Samarth (Springer, Berlin, 2002), pp. 229–272.