Electric-field switching of exciton spin splitting in coupled quantum dots

Xiaojing Li and Kai Chang

SKLSM, Institute of Semiconductors, Chinese Academy of Sciences,
P. O. Box 912, Beijing 100083, China

Abstract

We investigate theoretically the spin splitting of the exciton states in semiconductor coupled quantum dots (CQDs) containing a single magnetic ion. We find that the spin splitting can be switched on/off in the CQDs via the sp-d exchange interaction using the electric field. An interesting bright-to-dark exciton transition can be found and significantly affects the photoluminescence spectrum. This phenomenon is induced by the transition of the ground exciton state, arising from the hole mixing effect, between the bonding and antibonding states.

PACS numbers: 68.65.Hb, 61.72.sd, 81.40.Rs
All-electrical control of electron spin is a central goal in the field of spintronics and quantum information processing. There have been many proposals concerning the experimental realization of all-electrical spintronic devices and qubits in solid state systems in recent years. The electron spin in quantum dots (QD) is a promising candidate for the qubit due to very long spin decoherent time and the feasibility of large-scale integration. There are several different schemes to control carrier spin in semiconductor QDs, e.g., the circular-polarized optical excitation, the spin-orbit interaction, and the $sp-d$ exchange interaction. The $sp-d$ exchange interaction between the carriers and the magnetic ions in a QD leads to giant Zeeman splitting and could be an important testing ground for the realization of a solid-state qubit. The Photoluminescence (PL) spectrum of CdTe QDs doped with a single Mn$^{2+}$ ion have demonstrated the effect of the $sp-d$ exchange interaction on the interband transition. This provides a unique flexibility to tailor the spin splitting of carriers and optical property utilizing external electric fields. However, a strong electric field is required to tune the spin splitting of the exciton in a QD containing a single magnetic ion since the strong confinement of carriers in a single QD prohibit the spatial separation of the electron and hole. Is it possible to control the spin states of exciton easily utilizing weak electric field in the QD system?

Recently, it was demonstrated that the orbital states of carriers in coupled QDs (CQDs) can be tuned by an external electric field in vertically- and laterally-coupled QDs, respectively. An interesting field-induced dissociation of exciton was observed in Photoluminescence experiments. In this work, we consider the CQDs doped with a single Mn$^{2+}$ ion and find that the spin states of the exciton in such CQDs can be easily controlled using electric fields. The spin splitting of the exciton in the CQDs exhibits significant asymmetry with respect to the directions of the electric fields and a switching behavior for the weak coupling case. An interesting bright-to-dark exciton transition arising from the bonding-antibonding hole state transition can be seen by adjusting the parameters, e.g., the spatial separation $d$ between the two QDs.

The CQDs structure is schematically shown in Fig. The electron and the hole are
confined by a square potential well in the z axis,

\[ V_{\perp}^{e,h}(z) = \begin{cases} 0, & [-(d/2 + d_1), -d/2] \\ 0, & [d/2, (d/2 + d_2)] \\ \Delta V_{e,h}^{e,h}, & \text{otherwise} \end{cases} \]  

and laterally by a parabolic potential \( V_{\parallel}^{e,h}(\rho) = m_{e,h} \omega_{e,h}^2 \rho^2 / 2 \), where \( m_e \) and \( m_h \) are the effective mass of the electron and heavy hole, respectively. The electric field is applied perpendicular to the CQDs plane. The Hamiltonian of the system is

\[ H = H_e + H_h + H_{s-d} + H_{p-d} + H_{e-h} + V_{\text{Coul}}, \]

where \( H_e \) (\( H_h \)) is the electron (hole) Hamiltonian, \( H_{s-d} \) (\( H_{p-d} \)) is the \( s-d \) (\( p-d \)) exchange interaction between the electron spin \( s \) (hole spin \( j \)) located at \( \mathbf{r}_e \) (\( \mathbf{r}_h \)) and the Mn\(^{2+} \) spin \( S \) located at \( \mathbf{r}_{Mn} \), and \( H_{e-h} \) is the short-range exchange interaction between the electron and hole. \( V_{\text{Coul}} = -e^2 / 4\pi\varepsilon_0|\mathbf{r}_e - \mathbf{r}_h| \) is the Coulomb interaction between the electron and hole, where \( \varepsilon(\varepsilon_0) \) is the dielectric constant of the material (vacuum), and \( e \) is the charge of the electron. The hole Hamiltonian is \( H_h = H_{LK} - eE_z \), where \( H_{LK} \) is the four-band Luttinger-Kohn Hamiltonian including the heavy hole and light hole bands. The relevant material parameters can be found in Ref. [6]. The eigenstate of the exciton \( \Psi^{e,h}(\mathbf{r}_e, \mathbf{r}_h) \) is expanded in the basis set constructed by the direct product of the eigenstates of the electron, the hole, and the magnetic ion.

Fig. 2 (a) displays the exciton energy spectrum as a function of electric field in a CdTe CQDs with strong interdot coupling (\( d = 2nm \)). We magnify the energy spectrum at weak electric fields (see the inset in Fig. 2 (a)) and find a strong asymmetric spin splitting with respect to the directions of the applied electric fields. The electron and hole distribute equally in the upper and lower QDs in the absence of the electric field. An external electric field pushes the electron and hole in opposite directions and consequently leads to the localization of electron and hole in different QDs. Compared to a single QD tuned by the external field, the wavefunction of carriers in such CQDs is much more easily manipulated electrically due to the weakening of the quantum confinement along the z axis. Since the strength of the \( p-d \) exchange interaction is about four times larger than that of the \( s-d \) interaction, when the hole is pushed into the upper QD that contains a single magnetic ion (see Fig. 1), the spin splitting becomes larger. The spin splitting becomes very small when the electron localizes in the upper QD. The twelve highest exciton energy states in the spectrum consist of antibonding hole-states (the first-excited states of the spin-down hole \( |3/2, -3/2 > \)) and

\[ \Psi^{e,h}(\mathbf{r}_e, \mathbf{r}_h) \]
the electron ground state $|1/2, \pm 1/2 >$. Meanwhile, the hole states of the twelve lowest exciton energy levels are composed of the bonding spin-down hole states $|3/2, -3/2 >$ and the electron ground state $|1/2, \pm 1/2 >$. The energy difference between the bonding and antibonding exciton states can be seen in Fig. 2(b). The electron and hole states in the twelve lowest exciton states are both bonding states, but we can only see six bright lines for the $\sigma^+$ excitation (the other six exciton levels ($J = \pm 2$) are dark states). However, the hole states of the twelve highest exciton energy levels are the antibonding states, therefore they are dark states at the small electric field in the electro-PL spectrum. As the electric field increases, we find the dark antibonding exciton states become bright, arising from the mixing of the bonding-antibonding hole states. There is an energy gap between the bonding and antibonding exciton states because of the interdot tunneling coupling between the two QDs. The bright $\sigma^\pm (\pm \frac{1}{2}, \pm \frac{3}{2}, S_z)$ and dark $\pm 2 (\pm \frac{1}{2}, \pm \frac{3}{2}, S_z)$ exciton states also split because of $s-d$ interaction. Spin splitting of the bonding exciton states increases/decreases oppositely to that of the antibonding exciton states as the electric field varies. In such CQDs, we can realize a switching behavior for the spin splitting utilizing weak electric fields.

For comparison with the CQDs with the strong interdot coupling, we calculate the CQDs for weak interdot coupling case ($d = 4nm$). The exciton energy spectrum in Fig. 3(a) shows that the energy gap between the bonding and antibonding exciton states decreases because of the weakening of the interdot tunneling coupling. The energy spectrum resembles that of the strong coupling case, while the electro-PL spectrum is very different (see Fig. 3(b)). This is because the twelve lowest exciton states are no longer the bright states, since the hole component of the exciton states shows an antibonding feature. In contrast, the highest twelve exciton states become bright, i.e., the bonding hole states in the exciton states. We plot the hole energy as a function of the spatial separation $d$ between the QDs in Fig. 4(b). A crossover between the bonding and antibonding hole states takes place around $d = 2.2nm$ (see the arrow in Fig. 4(b)). The crossover can also be seen in Fig. 4(c), which plots the overlap factor between the electron and hole states as a function of the distance $d$ (see the black lines in 4(c)). There is no crossover between the ground and first-excited hole states, i.e., the bonding and antibonding hole states, without coupling of heavy hole (hh) and light hole (lh) mixing (see the dotted lines in Fig. 4(b)). This behavior can be understood from a four-level model that is schematically shown in Fig. 4(a). In the Luttinger-Kohn Hamiltonian, the off-diagonal element $R \propto k||k_z$ induces the coupling between the HH($L = 0$)
and LH \((L = 1)\) states (see Fig. 4(a)). The lowest two levels \(\lambda_i = E_{01}^i/2 \pm \sqrt{(E_{01}^i)^2 + 4\Delta_i^2}/2\) \((i = a, b)\), where \(E_{01}^a = E_0^a + E_1^b\) \((E_{01}^b = E_0^b + E_1^a)\) is the sum of the energies of the two original coupled levels and \(\Delta_i\) is the coupling term. The competition between \(E_{01}^i\) and \(\Delta_i\) could lead to the bonding and antibonding transition of the ground state. Increasing the distance \(d\) results in the change of the coupling between the HH (bonding) and LH (antibonding) states \(\Delta_i\). As a result, the energy of antibonding exciton states could be lower than bonding exciton states. Therefore, the the twelve lowest exciton states become dark while the twelve highest states are bright in the electro-PL spectrum. This feature can also be understood from the overlap factor of the bonding and antibonding exciton states as a function of electric field (see the red lines in Fig. 4 (c)). We find that the overlap factor of the antibonding (bonding) exciton states increase as the electric field increases (decreases), resulting in the dark-to-bright transition of the ground exciton states. Because the exciton energies of bonding and antibonding states are very close to each other, it is hard to distinguish them from the PL spectrum. The electro-PL spectrum could help us to distinguish them from the intensity of the PL peaks due to the different energy dependences of the bonding and antibonding exciton states on electric fields. (see Figs. 2 and 3)

Finally, we plot the spin splitting as function of the electric field and the distance \(d\) between the CQDs in Fig. 5. The spin splitting is symmetric with respect to the directions of electric fields when the CQDs are strongly coupled. The weak electric fields only result in a negligible small spin splitting since the strong confinement of carriers prohibits the spatial separation of the electron and hole. When the distance \(d\) increases, there is different behavior of the spin splitting at weak electric fields. The spin splitting becomes strongly asymmetric with respect to the directions of the electric fields and shows a switching feature for the opposite directions of the electric fields. We should point out that the position of the magnetic ion in the QD affects heavily the exciton spin splitting, and determines the electric field corresponding to the largest spin splitting, but it will not change the switching behavior in CQDs containing a single magnetic ion.

In summary, we investigated theoretically the energy spectrum and electro-PL spectrum of the CQDs containing a single magnetic ion. For the CQDs with strong interdot coupling, the spin splitting is asymmetric with respect to the directions of the electric fields and can be switched on/off using weak electric fields. For the weak coupling case, we find that the hole mixing effect leads to the crossover between the bonding and antibonding hole states,
consequently resulting in the bright-to-dark transition of the ground exciton states. Our theoretical results could be useful for the designing fresh types of all-electrical spintronic devices.

Acknowledgments

This work was supported by the NSFC Grant No. 60525405.
1. D. P. DiVincenzo, D. Bacon, J. Kempe, G. Burkard, K. B. Whaley, Nature (London) 408, 339 (2000).

2. C. S. Kim, M. Kim, S. Lee, J. Kossut, J. K. Furdyna, and M. Dobrowolska, J. Cryst. Growth 214, 395 (2000).

3. J. Kossut, I. Yamakawa, A. Nakamura and S. Takeyama, Appl. Phys. Lett. 79, 1789 (2001).

4. Kai Chang, J. B. Xia, F. M. Peeters, Appl. Phys. Lett. 82, 2661 (2003); Kai Chang, S. S. Li, J. B. Xia, and F. M. Peeters, Phys. Rev. B 69, 235203 (2004).

5. Y. Léger, L. Besombes, J. Fernández-Rossier, L. Maingault, and H. Mariette, Phys. Rev. Lett. 97, 107401 (2006); L. Besombes, Y. Leger, L. Maingault, and H. Mariette, J. Appl. Phys. 101, 081713 (2007).

6. A. A. Maksimov, G. Bacher, A. MacDonald, V. D. Kulakovskii, A. Forchel, C. R. Becker, G. Landwehr, and L. Molenkamp, Phys. Rev. B 62, R7767 (2000).

7. S. H. Xin, P. D. Wang, A. Yin, C. Kim, M. Dobrowolska, J. L. Merz, and J. K. Furdyna, Appl. Phys. Lett. 69, 2884 (1996).

8. X. J. Li, and Kai Chang, Appl. Phys. Lett. 92, 071116(2008).

9. G. Ortner, M. Bayer, Y. Lyanda-Geller, T. L. Reinecke, A. Kress, J. P. Reithmaier, and A. Forchel, Phys. Rev. Lett. 94, 157401 (2005).

10. B. Szafran and F. M. Peeters, Phys. Rev. B 76, 195442 (2007).

11. H. J. Krenner, M. Sabathil, E. C. Clark, A. Kress, D. Schuh, M. Bichler, G. Abstreiter, and J. J. Finley, Phys. Rev. Lett. 94, 057402 (2005).

12. M. Scheibner, M. F. Doty, I. V. Ponomarev, A. S. Bracker, E. A. Stinaff, V. L. Korenev, T. L. Reinecke, and D. Gammon, Phys. Rev. B 75, 245318 (2007).
FIG. 1: Schematic diagram of a CQD containing a single magnetic ion.

FIG. 2: The exciton energy spectrum as a function of electric field (the upper panel) and the electro-PL spectrum (the lower panel) in CQD. $d_1 = d_2 = 2.4nm$, the radius $l_e = l_h = 5nm$, the distance between the two QDs $d = 2nm$.

FIG. 3: The same as Fig. 2, but for the CQD with larger spatial separation between the two QDs, $d = 4nm$. 

FIG. 4: (Color online) (a) the schematic diagram of the four-level model. (b) The hole energy as a function of distance \(d\) between the two QDs. The solid lines denote the hole energies in CQDs with the HH-LH coupling; the dotted lines denote that without the HH-LH coupling. (c) the overlap factor between the electron ground state and the hole ground states (the solid lines) and first-excited hole states (the dotted lines) as function of the distance \(d\) between the two QDs (the black lines) and the electric fields (the red lines) for \(d = 4\, \text{nm}\).

FIG. 5: The contour plot of the spin splitting of exciton in a CQD as function of the distances \(d\) and external electric fields.
