Voltage control of the magnetic properties of charged semiconductor quantum dots containing magnetic ions

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We describe a model device allowing voltage control of the magnetic properties of magnetic ions in III-V self-assembled semiconductor quantum dots. The applied voltage, combined with Coulomb blockade, allows the control of the number of holes in the quantum dot. The spins of the holes interact with the spins of the magnetic ions via *sp-d* exchange interactions. The spectrum of a Mn ion in a *p*-type InAs quantum disk in a magnetic field is calculated as a function of the number of holes described by the Luttinger-Kohn Hamiltonian. For a neutral Mn acceptor, the spin of the hole leads to an effective magnetic field which strongly modifies the magnetization of the ion. The magnetization can be modified further by charging the dot with an additional hole. The interacting holes form a singlet parity ground state, suppress the effective field and modify the magnetic moment of the charged complex.

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There is currently interest in developing voltage control of magnetic properties[1] of diluted magnetic III-V semiconductors[2–6] (DMS). The interaction of electrons and holes with magnetic impurities in nanocrystals and quantum dots has been investigated by several authors in the last years.[5–12] Building on this work we describe here a model device allowing voltage control of the magnetic properties of Mn ions in III-V self-assembled semiconductor quantum dots[13] by controlling the number of holes in the vicinity of the ion. The schematic device structure is shown in Fig. 1(a). A layer of quantum dots containing magnetic ions is sandwiched between a metallic gate on the surface and a *p*-type back contact. The potential applied to the gate, Vg, changes the position of the quantum dot level with respect to the Fermi level of the contact and, combined with Coulomb blockade, allows precise control of the number of holes in each dot.[14] The spin of the holes interacts with the spin of the Mn ion via *sp-d* exchange interaction and modifies the magnetic properties of the complex. The exchange interaction can be modified by changing the dot radius,[6,7] by the number of additional carriers and by the magnetic field. To understand such interactions we assume a very diluted Mn concentration and study a model of a dot containing one Mn impurity. Figure 1(b) shows the interacting spins in the quantum dot as a function of the voltage. In dots based on III-V semiconductors, Mn ions act as acceptors.[15–18] Therefore, in the absence of external bias our model consists of a negatively charged acceptor and a positively charged hole. By applying positive bias the dot can be emptied and by applying negative bias an additional hole enters the quantum dot. The spin of the second hole almost cancels the spin of the first hole and strongly modifies the magnetic properties of the Mn center.

We start with our device at zero bias. The substitutional Mn^{+2} ion in a III-V semiconductor quantum dot of radius R and thickness W corresponds to an electronic configuration $3d$,[5] and with one weakly bound valence hole forms a neutral Mn^0 acceptor.[15–18] The Hamiltonian of the Mn^0 complex in a magnetic field applied along the growth axis is

$$H_{\mathrm{Mn}^0} = g\mu_B B S_z - \mathcal{J}(r)\vec{\sigma}\vec{\mathbf{S}} + H_h, \tag{1}$$

where g is the Landé factor of the Mn ion, μ_B is the Bohr magneton, B is the external magnetic field and $\vec{\mathbf{S}}$ is the spin of the Mn ion ($S=5/2$). The first term describes the Hamiltonian of an isolated Mn spin, the second term $H_{\mathrm{ex}}=-\mathcal{J}(r)\vec{\sigma}\vec{\mathbf{S}}$ corresponds to the *sp-d* exchange interaction between the spin of the hole, $\vec{\sigma}$, and the Mn ion *d*-shell

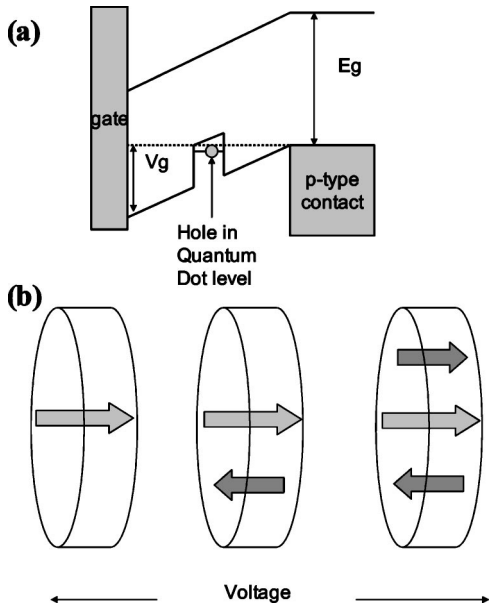

FIG. 1. (a) Schematic illustration of the device structure allowing voltage-control of the number of holes in a quantum dot. (b) Interacting spins in a III-V semiconductor quantum disk with one Mn ion under positive bias (left), zero bias (center), and negative bias (right). Light-shaded arrow corresponds to the Mn ion spin and dark arrows to valence holes spins.

electrons, and $\mathcal{J}(r)$ is a function rapidly decreasing as the distance from the Mn center increases.[8] The third term stands for the hole orbital Hamiltonian,

$$H_h = H_{\mathrm{LK}} - \frac{e^2}{\epsilon^* \sqrt{\rho^2 + z^2}} + V_c(\rho, z). \tag{2}$$

Due to spin-orbit interaction, hole states are described not by spin but by total angular momentum through the Luttinger-Kohn Hamiltonian H_{LK}.[9,19–21] The second term in Eq. (2) describes the Coulomb interaction between the positively charged hole and the negatively charged Mn acceptor, for simplicity placed in the center of the disk. $V_c(\rho, z)$ is the confinement potential, e is the hole charge, and ϵ^* is the effective dielectric constant of the disk material. No central cell correction for the acceptor potential is considered in this qualitative study.[22] The singly and doubly charged acceptor states in SiGe quantum wells and dots have been studied in detail by Rego *et al.*[19] InAs self-assembled quantum dots with shape resembling quantum disks can be fabricated using In-flush technique.[23] Following Ref. 19, we treat our quantum dot as a flat disk with two lowest subbands $\xi_0(z) = \sqrt{(2/W)}\cos(\pi z/W)$ and $\xi_1(z) = \sqrt{(2/W)}\sin(2\pi z/W)$, with z as the growth direction.

The hole wave function corresponding to eigenvalue[24] k, "parity up" (down), and total angular momentum z-projection F_z can be written as a four-component Luttinger spinor,[21]

$$|F_z, \nu = \uparrow(\downarrow), k\rangle = \sum_n \begin{pmatrix} A^{F_z,\nu=\uparrow(\downarrow),k}_{J_z=+3/2,n} f_{F_z-3/2,n}(\rho,\theta)\xi_0(z)(\xi_1(z)) \left| J_z = \frac{+3}{2} \right\rangle \\ A^{F_z,\nu=\uparrow(\downarrow),k}_{J_z=-1/2,n} f_{F_z+1/2,n}(\rho,\theta)\xi_0(z)(\xi_1(z)) \left| J_z = \frac{-1}{2} \right\rangle \\ A^{F_z,\nu=\uparrow(\downarrow),k}_{J_z=+1/2,n} f_{F_z-1/2,n}(\rho,\theta)\xi_1(z)(\xi_0(z)) \left| J_z = \frac{+1}{2} \right\rangle \\ A^{F_z,\nu=\uparrow(\downarrow),k}_{J_z=-3/2,n} f_{F_z+3/2,n}(\rho,\theta)\xi_1(z)(\xi_0(z)) \left| J_z = \frac{-3}{2} \right\rangle \end{pmatrix}. \tag{3}$$

Here $|J_z\rangle$ is the periodic part of the Bloch functions and $f_{F_z-J_z,n}(\rho,\theta)$ is the in-plane envelope part with zth component of the envelope angular momentum $m_z = F_z - J_z$,

$$f_{F_z-J_z,n}(\rho,\theta) = \frac{e^{im_z\theta}}{\sqrt{2\pi}} \frac{\sqrt{2}}{R} \frac{J_{m_z}(k_n^{m_z}\rho)}{|J_{m_z+1}(k_n^{m_z}R)|}. \tag{4}$$

$J_{m_z}(k_n^{m_z}\rho)$ is the Bessel function of order m_z and radial quantum number n. $k_n^{m_z}$ represents the hole wave vector, defined in terms of the Bessel function roots $(\alpha_n^{m_z})$ as $k_n^{m_z} = \alpha_n^{m_z}/R$.

The parity ν quantum number has two possible configurations, $\nu = \uparrow$ and $\nu = \downarrow$. States of opposite parities are orthogonal and the two parity configurations are degenerate in the absence of magnetic field. Under a suitable definition of parity operators, the parity quantum number is isomorphic to the electronic spin quantum number.[20,21]

To derive an effective Mn Hamiltonian for small quantum dots, we only retain the two lowest hole states. Moreover, due to short range of the exchange interaction on a quantum dot length scale, we can approximate $\mathcal{J}(r)$ by $\delta(r)$. These two approximations, together with the circular symmetry of the system, lead to matrix elements of the *sp-d* exchange interaction in the hole states $|i\rangle = |F_z, \nu, k\rangle$,

$$\langle j|H_{\mathrm{ex}}|i\rangle = -\sum_{n,J_z}\sum_{n',J_z'} A^i_{J_z,n} A^j_{J_z',n'} \langle f_{F_z'-J_z',n'}(\rho,\phi)\xi^{\nu'}_{J_z'}(z)| \times \langle J_z'|\delta(r)\sigma_z S_z|J_z\rangle |f_{F_z-J_z,n}(\rho,\phi)\xi^{\nu}_{J_z}(z)\rangle. \tag{5}$$

These matrix elements are diagonal in the basis of the lowest lying parity up and parity down hole states, and result in an effective Ising Mn^0 Hamiltonian,

$$H^{\mathrm{QM}}_{\mathrm{Mn}^0}(i, S_z) = E_i + [g\mu_B B + \beta B^0_{\mathrm{eff}}(i)]S_z. \tag{6}$$

Here E_i is the energy of the hole state $|i\rangle$, given by Eq. (2), and the bracketed term is a sum of an external plus an effective internal magnetic field. $\beta = \langle J_z|\delta(r)|J_z\rangle$ is the hole exchange parameter, and $B^0_{\mathrm{eff}}(i)$ is the effective magnetic field,

$$B^0_{\mathrm{eff}}(i) = -\frac{1}{WR^2}\left(\delta_{\nu,\uparrow} \sum_{J_z=\frac{3}{2},-\frac{1}{2}} \mathcal{F}_{i,J_z}(0) + \delta_{\nu,\downarrow} \sum_{J_z=\frac{1}{2},-\frac{3}{2}} \mathcal{F}_{i,J_z}(0) \right), \tag{7}$$

with $\mathcal{F}_{i,J_z}(0)$ being a form factor related to the charge density of the neutral hole state $|i\rangle$ at the center of the disk,

$$\mathcal{F}_{i,J_z}(0) = \delta_{mz,0} \frac{J_z}{|J_z|} \left(\sum_n \frac{A^i_{J_z,n}}{|J_1(\alpha_n^0)|} \right)^2. \tag{8}$$

The spectrum of the interacting hole and Mn complex calculated by $H^{\mathrm{QM}}_{\mathrm{Mn}^0}$ is limited to $T = 0$ K. At finite temperature one often tries to replace the full quantum mechanical problem by an effective mean field problem of either the Mn ion in the effective field of a hole or a hole in the effective field of the Mn ion. In this mean-field approach, we use the thermodynamical average of the hole spin to derive an effective (mean-field) Mn^0 Hamiltonian which accounts for the influence of finite temperature,

$$H^{\mathrm{MF}}_{\mathrm{Mn}^0}(S_z) = g\mu_B B S_z + \sum_i \varrho(E_i)[E_i + \beta B^0_{\mathrm{eff}}(i) S_z], \tag{9}$$

where $\varrho(E_i) = Ze^{-(E_i - E_{\mathrm{GS}})/k_B T}$ is the density matrix, with Z standing for its normalization constant, E_{GS} for the energy of the hole ground state, k_B for the Boltzmann constant and T for the temperature.

It is inferred from $\delta_{mz,0}$ in Eq. (8), that only one J_z component of each hole state $|i\rangle$ may yield a nonzero contribution to the effective field B^0_{eff}. Moreover, Eq. (7) reveals explicit dependence of the effective magnetic field on the quantum disk size, in agreement with previous work of Efros *et al.*[6,7]

To illustrate the effect of the hole on the Mn ion energy levels, we calculate the Zeeman spectrum of a neutral Mn^0 impurity in a model quantum disk of thickness $W = 2$ nm and radius $R = 5$ nm with an external magnetic field applied along

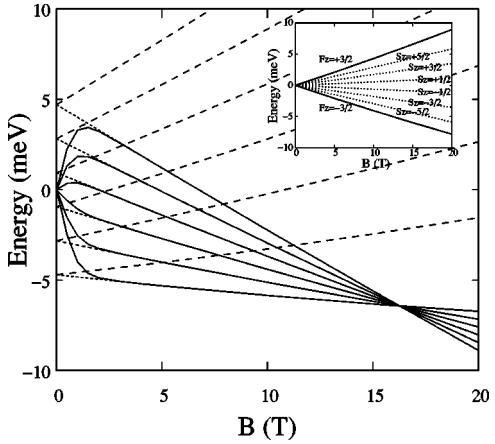

FIG. 2. Energy spectrum vs magnetic field of a Mn ion bound to one valence hole in an InAs quantum disk, calculated with $H_{\mathrm{Mn}^0}^{\mathrm{MF}}$ at $T=4$ K (solid lines) and with $H_{\mathrm{Mn}^0}^{\mathrm{QM}}$ (dashed lines). For dashed lines, long period lines represent the energy levels arising from the hole state $|3/2,\uparrow,1\rangle$, whereas short period lines represent those arising from the state $|-3/2,\downarrow,1\rangle$. These spectra correspond to the zero bias operation mode of our model device. The inset shows the Zeeman splitting of the free hole states $|3/2,\uparrow,1\rangle$ and $|-3/2,\downarrow,1\rangle$ (solid lines) and that of a free Mn ion (dotted lines).

the growth direction. We use both $H_{\mathrm{Mn}^0}^{\mathrm{QM}}$ and $H_{\mathrm{Mn}^0}^{\mathrm{MF}}$. A set of parameters close to InAs material is employed, Landé factor of Mn $g=2.02$,[25] Luttinger parameters $\gamma_1=19.67$, $\gamma_2=8.37$, and $\gamma_3=9.29$, and an effective dielectric constant of 12.4. The low-lying states of the hole are expanded in up to $n=15$ radial wave functions and obtained by diagonalization of the matrix representation of the Hamiltonian H_h, Eq. (2). It is found that, in absence of an external magnetic field, $|-3/2,\downarrow,1\rangle$ and $|3/2,\uparrow,1\rangle$ form a twofold degenerated ground state. When a magnetic field is applied, these two levels split and the ground state is given by $|-3/2,\downarrow,1\rangle$. For the magnetic field region we study, the magnitude of this splitting is much smaller than the energy spacing between $|3/2,\uparrow,1\rangle$ and the next excited state. Therefore, Eq. (5) is a good approximation. The InAs value of β is unknown, however, we have found out that $\beta=-17.1$ eV Å^3 fits experimental observations for the Mn acceptor ground state in GaAs bulk[16] by employing the model of Ref. 8 with $\mathcal{J}(r)$ replaced by $\delta(r)$. One may expect a similar hole exchange parameter for InAs, therefore we choose $\beta=-17$ eV Å^3.

Figure 2 illustrates the Zeeman spectrum of the Mn^0 complex. Dashed lines represent the 12 lowest energy levels given by $H_{\mathrm{Mn}^0}^{\mathrm{QM}}$ and solid lines the mean-field spectrum given by $H_{\mathrm{Mn}^0}^{\mathrm{MF}}$ at $T=4$ K. The inset shows the noninteracting spectra of the Mn ion (dotted lines) and the hole levels $|-3/2,\downarrow,1\rangle$ and $|3/2,\uparrow,1\rangle$ (solid lines) for comparison. The energy scale has been offset in all cases by the energy of the hole states at $B=0$. According to the Ising $H_{\mathrm{Mn}^0}^{\mathrm{QM}}$, at $B=0$ there are six twofold degenerate levels. When an external magnetic field is applied, the degenerate levels split. The levels arising from the parity up hole state (long period lines) increase in energy without ever crossing. However, the levels arising from the parity down hole state (short period lines) decrease in energy, cross parity up states, become degenerate at a special value of the magnetic field B ($B\approx 16.2$ T in Fig. 2), and finally reverse the order. This degeneracy follows from the cancellation of the atomic Zeeman splitting and the effective internal magnetic field,[26] due to the positive Landé factor of Mn and the antiferromagnetic hole exchange parameter of InAs ($\beta<0$). At low magnetic fields, $|-3/2,\downarrow,1\rangle|S_z=+5/2\rangle$ is the Mn^0 center ground state due to the *sp-d* exchange interaction. However, at $B\approx 16.2$ T the atomic Zeeman effect already compensates the effective magnetic field and for larger external magnetic fields $|-3/2,\downarrow,1\rangle|S_z=-5/2\rangle$ replaces $|-3/2,\downarrow,1\rangle|S_z=+5/2\rangle$ as the Mn^0 center ground state.

The results of the mean field treatment of the hole are shown as solid lines in Fig. 2. At $B=0$, there is no net effective magnetic field acting on the Mn ion. This is because the hole levels $|-3/2,\downarrow,1\rangle$ and $|3/2,\uparrow,1\rangle$ are degenerate and cancel each other in Eq. (9). When an external magnetic field is applied, the hole becomes increasingly polarized, $|-3/2,\downarrow,1\rangle$ starts gaining weight in Eq. (9) and therefore the effective magnetic field increases in strength. As a result, in the region of finite but weak external magnetic field ($0<B<2$ T), B_{eff}^0 leads to a large Zeeman splitting. The magnitude of this splitting at $B=1.8$ T is 8.1 meV, and it is expected to be even larger if the central cell correction is included in the model, as the hole density will be localized closer to the Mn impurity. For larger magnetic fields ($B>2$ T), the hole is already strongly polarized and thus the Mn^0 levels converge to those predicted with $H_{\mathrm{Mn}^0}^{\mathrm{QM}}$ for the parity down hole state.

Next we switch our device to negative bias and study the Zeeman spectrum of a Mn ion surrounded by two interacting holes (i.e., the positively charged acceptor) in an InAs quantum disk. The Hamiltonian of the positively charged acceptor is

$$H_{\mathrm{Mn}^+}=g\mu_B BS_z+H_{\mathrm{ex}}(1,2)+H_h(1)+H_h(2)+V_{hh}(1,2), \tag{10}$$

where $H_{\mathrm{ex}}(1,2)=H_{\mathrm{ex}}(1)+H_{\mathrm{ex}}(2)$ is the *sp-d* exchange interaction between the spins of the two holes and that of the Mn ion, H_h is the one-body hole Hamiltonian given by Eq. (2) and V_{hh} is the hole-hole Coulomb interaction term. The above equation is solved in the basis of the hole states $|i\rangle=|F_z,\nu,k\rangle$, using the parity properties of the Coulomb interaction.[21] The wave function of a two-hole complex corresponding to the pth eigenvalue can be written as a Slater determinant

$$\Phi_p(1,2)=\sum_{i,j}C_{i,j}^p\frac{1}{\sqrt{2}}[\Psi_i(1)\Psi_j(2)-\Psi_i(2)\Psi_j(1)], \tag{11}$$

where $C_{i,j}$ are expansion coefficients which can be classified by its total angular momentum z-projection, $F_{zT}=F_{z1}+F_{z2}$, and total parity, $\nu_T=\nu_1+\nu_2$.

In our small quantum dot the energy spacing between the two-hole ground state, $|0\rangle$, and the first excited state is over 200 meV. Therefore, at low temperature the *sp-d* exchange interaction for the Mn center is well described by the expectation value in the state $|0\rangle$,

$$\langle 0|H_{\mathrm{ex}}(1,2)|0\rangle = -\frac{\beta S_z}{WR^2}\Bigg[\sum_{j,F_{z1}}\Bigg(\delta_{\nu_1,\uparrow}\sum_{J_z=\frac{3}{2},-\frac{1}{2}}\mathcal{G}_{F_{z1}J_z,j}(0) + \delta_{\nu_1,\downarrow}\sum_{J_z=-\frac{3}{2},\frac{1}{2}}\mathcal{G}_{F_{z1}J_z,j}(0)\Bigg) + \sum_{i,F_{z2}}\Bigg(\delta_{\nu_2,\uparrow}\sum_{J_z=\frac{3}{2},-\frac{1}{2}}\mathcal{G}_{F_{z2}J_z,i}(0) + \delta_{\nu_2,\downarrow}\sum_{J_z=-\frac{3}{2},\frac{1}{2}}\mathcal{G}_{F_{z2}J_z,i}(0)\Bigg)\Bigg], \tag{12}$$

where $\mathcal{G}_{F_zJ_z,j}(0)$ is the form factor

$$\mathcal{G}_{F_zJ_z,j}(0) = \delta_{mz,0}\frac{J_z}{|J_z|}\Bigg(\sum_k C^0_{F_z\nu k,j}\sum_n \frac{A^{F_z\nu k}_{J_z,n}}{|J_1(\alpha^0_n)|}\Bigg)^2. \tag{13}$$

Thus, the effective Hamiltonian of the Mn ion in the field of two holes can be written as

$$H_{\mathrm{Mn}^+} = E_0 + (g\mu_B B + \beta B^+_{\mathrm{eff}})S_z, \tag{14}$$

where E_0 is the energy of the two-hole ground state and the bracketed term is a sum of the external plus the effective internal magnetic fields, with $B^+_{\mathrm{eff}}=\langle 0|H_{\mathrm{ex}}|0\rangle/\beta S_z$. As in the single hole case, in Eq. (12) each of the one-body wave functions of $|0\rangle$ contributes to the effective field with at most one J_z component, the sign of such a contribution depending on the one-body wave function parity and total angular momentum z-projection. Eventually, this may result in one hole contribution cancelling that of the second hole in Eq. (12). From the analogy between hole parity and electronic spin quantum numbers, one expects this cancellation to occur when the two holes form a closed shell configuration (with equal parity up and parity down weight).

Eigenvalues and eigenvectors of the Hamiltonian H_{Mn^+}, Eq. (10), are obtained by means of a configuration interaction method, taking the two lowest single-particle states of parity up with $F_z=-3/2,\ldots,9/2$ and parity down with $F_z=-9/2,\ldots,3/2$. The $F_{zT}=0$, singlet parity state is found as the ground state for the entire range of external magnetic fields under study. Figure 3 represents the energy spectrum of the Mn^+ complex in the presence of an external magnetic field using the same parameters as in Fig. 2, with and without *sp-d* exchange interaction (solid and dashed lines, respectively). The results show that the effective magnetic field arising from the *sp-d* exchange interaction is indeed zero for an external magnetic field $B=0$. The underlying reason is that, at $B=0$, parity up terms exactly cancel out parity down terms in Eq. (12). When the external magnetic field is applied, the effective internal magnetic field slightly increases, but it remains negligible even for magnetic fields as high as 15 T, where the two-hole ground state still has singlet parity. This is clearly seen in the inset of Fig. 3, where the *sp-d* exchange energy of the Mn^+ level $|0\rangle|S_z=-5/2\rangle$ is depicted vs the external magnetic field. Although the *sp-d* exchange energy increases with increasing external magnetic field, at $B=15$ T the energy splitting due to the effective magnetic field is of the order of tens of μeV. We can compare this splitting range with the few meV splitting of the Mn^0 complex in an external magnetic field (see Fig. 2).

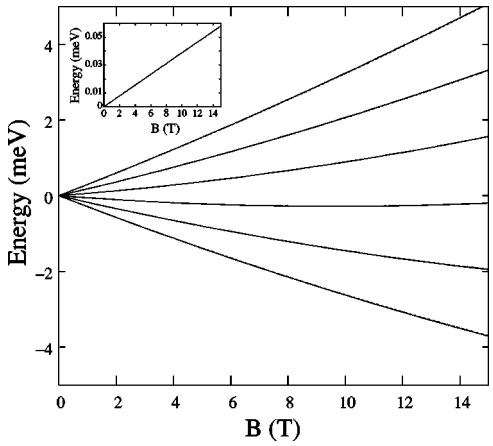

FIG. 3. Energy spectrum vs magnetic field of a Mn ion bound to two valence holes in an InAs quantum disk, including (solid lines) and excluding (dashed lines) *sp-d* exchange interaction. The interacting spectrum corresponds to the negative bias operation mode of our model device. The inset shows the *sp-d* exchange energy corresponding to the Mn^+ level $|0\rangle|S_z=-5/2\rangle$ vs an external magnetic field.

The nonzero contribution of the *sp-d* exchange energy when $B\neq 0$ can be explained in terms of the single hole picture. The external magnetic field increases the band mixing of hole states with parity down more than that of parity up states, causing their effective masses to increase.[21] Therefore, in the presence of an external magnetic field the weight of the parity down component of the ground state increases while that of the parity up component decreases. As a result, the magnitude of contribution of the parity down component to the *sp-d* exchange energy slightly exceeds that of the parity up component and a finite *sp-d* exchange interaction is obtained despite of the singlet parity. For larger external magnetic fields ($B>15$ T), the parity of the ground state is expected to change from singlet to triplet. Then, the effective internal magnetic field will abruptly increase and the spectrum will qualitatively resemble that of the Mn^0 complex.

Our calculations have been performed for the particular case of an impurity sitting in the middle of the disk. In a more realistic situation, the disk may be asymmetric and the impurity off-centered. Nonetheless, the formation of a singlet two-hole state within the dot, which strongly suppresses the effective exchange field acting on the Mn ion, results from Pauli exclusion principle and, therefore, is not restricted to any symmetry. If the dot was asymmetric, the degeneracy of parity up and down states would be removed. Then, the picture would be similar to that we report in the presence of a magnetic field: There would still be a singletlike state, although its effect on the exchange Hamiltonian would not lead to a perfect cancellation. Therefore, an asymmetric situation would only lead to a small departure from our ideal situation.

In summary, we have presented a model system which suggests electrical control of the magnetization of Mn ions in quantum dots by the control of the number of holes in its vicinity. We have calculated the Zeeman spectrum of a single

Mn ion interacting with one and two valence holes in an InAs quantum disk. The single hole in a quantum dot results in an effective Ising like model Hamiltonian. The hole introduces an effective magnetic field on the Mn ion levels yielding a large Zeeman splitting. This splitting can be controlled by the size of the quantum dot. When a second hole is added, a singlet-parity two-hole ground state is formed and the effective magnetic field is strongly suppressed. Therefore, the control over the number of confined holes in a single-ion doped III-V DMS QD provides a mechanism to switch on and off the *sp-d* exchange interaction. Spin resonance experiments are suggested to probe the energy levels of Mn spins in this system.

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