

Magnetic Exchange Interactions in Quantum Dots Containing Electrons and Magnetic Ions

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We present a theory of magnetic exchange interactions in quantum dots containing electrons and magnetic ions. We find the interaction between the electron and Mn ion to depend strongly on the number of electrons. It can be switched off for closed shell configurations and maximized for partially filled shells. However, unlike the total electron spin S which is maximized for half-filled shells, we predict the exchange interaction to be independent of the filling of the electronic shell. We show how this unusual effect manifests itself in quantum dot addition and excitation spectrum.

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There is currently interest in nanospintronics, i.e., controlling spin-related phenomena on the nanoscale [1–16]. Quantum dots with a single magnetic ion have been recently realized by Besombes *et al.* [13]. A single magnetic ion on a solid state surface has been probed by a spin-flip STM [9], by tunneling in a break junction [1], by STM in bulk GaAs [11], and both Mn-doped nanocrystals [5,6] as well as quantum dot tunneling devices embedded in a magnetic barrier have been investigated [7]. Theoretically, interaction of carriers with magnetic impurities in quantum wells [4], nanocrystals [6–8], and quantum dots [5,14–16] has been investigated, including a mean field theory of noninteracting electrons interacting with large number of Mn ions [14]. In this Letter we present a theory of a hybrid nanoscale system consisting of a single magnetic ion placed in a controlled electronic environment provided by a quantum dot (QD) filled with N electrons, an artificial atom.

We consider a quasi-two-dimensional quantum dot with parabolic confinement, a model suitable for self-assembled quantum dots [17], containing N interacting electrons, and a single magnetic Mn ion, described by the Hamiltonian:

$$\hat{H} = \sum_{i=1}^N \left[\frac{\vec{P}_i^2}{2m^*} + \frac{1}{2} m^* \omega_0^2 \vec{r}_i^2 \right] + \sum_{i<j} V(\vec{r}_i - \vec{r}_j) - J_C^{2D} \sum_{i=1}^N \vec{M} \cdot \vec{S}_i \delta(\vec{r}_i - \vec{R}). \quad (1)$$

The first term in Eq. (1) corresponds to the sum of the one-electron Hamiltonians, the second term describes electron-electron Coulomb interactions, and the third term describes $sp-d$ contact exchange interaction between Mn ion at position \vec{R} and N electrons at positions $\vec{r}_i = (x_i, y_i)$. The single-particle states and energies of an electron in a parabolic QD correspond to two coupled harmonic oscillators with quantum numbers m and n , i.e., $\varphi_{mn}(x, y) = \varphi_m(x)\varphi_n(y)$, where $\varphi_m(x)$ is the wave function of a one-dimensional harmonic oscillator. The first three states are listed here: $\varphi_0(x) = e^{-x^2/4}/(2\pi l_0^2)^{1/4}$, $\varphi_1(x) = xe^{-x^2/4}/$

$(2\pi l_0^2)^{1/4}$, and $\varphi_2(x) = (x^2 - 1)e^{-x^2/4}/(8\pi l_0^2)^{1/4}$, with $x = x/\ell_0$ and $\ell_0 = 1/\sqrt{\omega_0}$. Here ω_0 is shell spacing, and length and energy are measured in effective Bohr radius a_B and effective Rydberg Ry. The corresponding single-particle energies are $E_{nm} = (n + m + 1)\omega_0$, with degenerate electronic shells with degeneracy $g_s = (n + m + 1)$ and shell spacing ω_0 . The electron-electron interaction $V(\vec{r}_i - \vec{r}_j) = e^2/(\epsilon|\vec{r}_i - \vec{r}_j|)$ is approximated by 2D Coulomb interaction, with e being electron charge and ϵ the dielectric constant. The finite thickness d of quantum dot is known to change the quantitative but not the qualitative effects of $e-e$ interaction [18].

The short ranged electron-Mn $sp-d$ exchange interaction is modeled here by contact interaction, with strength $J_C^{2D} = J_c 2/d$, where J_c is bulk exchange constant. In what follows we adopt $J_c = 15 \text{ eV } \text{\AA}^3$ [14], $d = 2 \text{ nm}$, $\epsilon = 10.6$, $m^* = 0.106$, Bohr radius $a_B = 52.9 \text{ \AA}$, Ry = 12.8 meV, with typical $\omega_0 = 4 \text{ Ry}$ and effective width $l_0 = 26.45 \text{ \AA}$, applicable to II-VI (Cd, Mn)Te semiconductor QDs.

For computational purposes, we denote single-particle states $\{n, m\} = \{i\}$, and transform the Hamiltonian into second quantization form by expanding the electron field operators $\hat{\psi} = \sum_{i,\sigma} \varphi_i(x, y) \chi_{\sigma} c_{i,\sigma}$ in orbital and spin eigenstates and annihilation (creation) $c_{i,\sigma}$ ($c_{i,\sigma}^\dagger$) operators. The key novelty is the electron-magnetic ion scattering term, known from the Kondo and Anderson models, given by $H_{\text{el-Mn}} = -J_C^{2D} \sum_{i,j,\sigma,\sigma'} (\int d\vec{r} \varphi_i^*(\vec{r}) \delta(\vec{r} - \vec{R}) \varphi_j(\vec{r})) \times \langle \chi_{\sigma'} \vec{S} \chi_{\sigma} \rangle \vec{M} c_{i,\sigma}^\dagger c_{j,\sigma'}$. Integrating out position and spin degrees of freedom results in a Hamiltonian $H_{\text{el-Mn}} = -\sum_{i,j,\sigma,\sigma'} J_{ij}(R) \vec{S}_{\sigma\sigma'} \vec{M} c_{i,\sigma}^\dagger c_{j,\sigma'}$ which involves exchange matrix elements $J_{ij}(R) = J_C^{2D} \varphi_i^*(R) \varphi_j(R)$, Pauli spin matrices $\vec{S}_{\sigma,\sigma'}$, and Mn spin operators. The exchange matrix elements $J_{ij}(R)$ are determined by the wave function of the two states (i, j) at the position R of the Mn ion. The electron-Mn exchange interaction combines two effects: flipping of electron spin simultaneous with flipping of Mn ion spin, and scattering of electrons between different

orbitals, i.e., disorder. We can bring these effects out by rewriting the exchange Hamiltonian in terms of Mn spin rising and lowering operators M^+ , M^- , with the final form of the Hamiltonian:

$$\begin{aligned} \hat{H} = & \sum_{i\sigma} E_{i,\sigma} c_{i,\sigma}^+ c_{i,\sigma} + \frac{1}{2} \sum_{ijkl} \langle i, j | V_{ee} | k, l \rangle c_{i,\sigma}^+ c_{j,\sigma'}^+ c_{k,\sigma'} c_{l,\sigma} \\ & - \sum_{i,j} \frac{J_{ij}(R)}{2} [(c_{i,\uparrow}^+ c_{j,\uparrow} - c_{i,\downarrow}^+ c_{j,\downarrow}) M_z + c_{i,\downarrow}^+ c_{j,\uparrow} M^+ \\ & + c_{i,\uparrow}^+ c_{j,\downarrow} M^-], \end{aligned} \quad (2)$$

where $E_{i,\sigma}$ is energy of an electron on the single-particle orbital $|i\rangle$ with spin σ_i . The two-body Coulomb matrix elements $\langle i, j | V_{ee} | k, l \rangle$ [18] scatter electrons from states $|k\sigma', l\sigma\rangle$ to states $|i\sigma, j\sigma'\rangle$. The last term in Eq. (2) is the electron-Mn Hamiltonian which consists of three terms. The first term measures difference in spin-up and -down population and acts as Zeeman energy while the second and third term involves flipping of electron spin compensated by the flipping of Mn spin.

To calculate the electronic properties of the interacting electron-Mn system we expand the wave function in the following basis: $|i_1, i_2, \dots, i_{N\uparrow}\rangle |j_1, j_2, \dots, j_{N\downarrow}\rangle |M_z\rangle$, where $|i_1, i_2, \dots, i_{N\uparrow}\rangle = c_{i_1,\uparrow}^+ c_{i_2,\uparrow}^+ \dots c_{i_{N\uparrow},\uparrow}^+ |0\rangle$, $|0\rangle$ is the vacuum, and $N\uparrow$ ($N\downarrow$) is the number of spin-up (-down) electrons, $N\uparrow + N\downarrow = N$. The basis states are grouped into spin-up and spin-down electron states for each state of the Mn ion $|M_z\rangle$, with $M_z = \pm 5/2, \pm 3/2, \pm 1/2$. The number of possible configurations is determined by the number of single-particle orbitals N_S , the number of electrons, and the size M of the magnetic ion spin $Nc = (2M + 1) \sum_{N\downarrow=0}^N \binom{N_S}{N\downarrow} \binom{N_S}{N\uparrow}$. Using the basis states we calculate the Hamiltonian matrix which upon diagonalization gives the eigenenergies and eigenstates of a single Mn and N interacting electron complex. Then, the strength of magnetic interaction can be investigated in terms of energy shift Δ , defined by the difference between the ground state energy of single Mn-doped (E_c) and undoped (E_e) QDs. Because this energy splitting can be thought of as induced by a local magnetic field produced by the magnetic moment of Mn ion, we call it the local Zeeman spin splitting.

Note that in the absence of electron-Mn exchange interaction, Coulomb interaction conserves the total angular momentum and z component of the total spin of electrons. Hence the diagonalization of the Hamiltonian can be performed separately for each (L_e, S_{Tz}) subspace. In a Mn ion doped QD, however, electron-Mn exchange interaction induces coupling between electronic configurations with different total spin of electrons and/or different total angular momentum, breaking down the symmetry. As a result, the diagonalization has to be performed in the entire Hilbert space, considerably increasing computational effort in comparison with that needed for an undoped QD.

The effect of the Mn ion is determined by the electron-Mn exchange interaction matrix elements. The scattering

involves occupied and empty states, and hence predominantly states in the vicinity of the Fermi level are involved. This opens up the possibility of engineering matrix elements by moving the Fermi level with increasing the number of electrons, and by moving the Mn ion position with respect to the center of the quantum dot. Figure 1 shows the exchange parameter $\langle m', n' | J(R) | n, m \rangle = J_C^{2D} \varphi_{m',n'}^*(R) \varphi_{m,n}(R)$ as a function of Mn position R evaluated using harmonic oscillator states in Cartesian coordinates for s shell [Fig. 1(a)], p shell [Fig. 1(b) and 1(c)], and d shell [Fig. 1(d)–1(f)]. In the s shell the $J_{ss} = \langle 0, 0 | J | 0, 0 \rangle$ matrix element decays as we move Mn away from the center of the quantum dot. For the p shell, the two matrix elements $\langle 1, 0 | J | 0, 1 \rangle$ and $\langle 0, 1 | J | 1, 0 \rangle$ are zero at $R = 0$, have a maximum at finite R , and zeros along the X or Y axis. By choosing our coordinate system in such a way that $R = (X, 0)$ we see that Mn can be coupled to one of the two p orbitals. The situation is similar in a d shell. There are 3 orbitals shown. For a position of Mn $X = l_0$ we have coupling to only one of the d orbitals as one of the orbitals has always a node in this direction, and a second one has a node at this distance from the center of the dot. Hence we see that we can engineer Mn position so it will be coupled

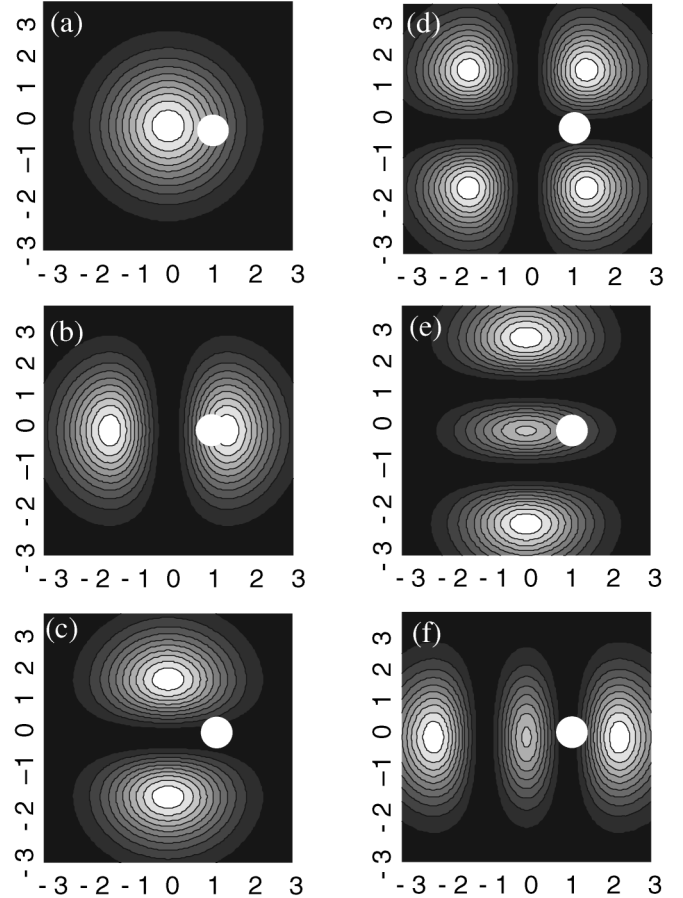


FIG. 1. Exchange matrix elements as a function of Mn position R for a QD with $\omega_0 = 4$ Ry, $d = 20$ Å for s shell (a), p shell (b) and (c), and d shell (d), (e), and (f). Empty circle indicates the position of Mn ion used in our calculation.

to only one of the three states of the d shell. Figure 1 clearly illustrates the potential for using Mn ion position as a tool in engineering magnetic interactions.

We now choose the position of Mn $R = (l_0, 0)$, and calculate the ground state energy of the full interacting system as a function of the number of electrons N . Figure 2 shows the calculated energy shift Δ , $\Delta = |E_c(R) - E_e|$. The calculations are done in a limited Hilbert space of each partially filled shell. The solid line shows schematically the total spin of the ground state of the dot as a function of the number of electrons N . The total spin is zero for closed shells at $N = 2, 6, 12, \dots$ and reaches maximum for a half-filled shell, i.e., $N = 1$ for s shell, $N = 4$ for a p shell, and $N = 9$ for a d shell [18,19]. One might expect that Δ is proportional to the total spin S of electrons. However, Fig. 2 shows a very different and unexpected behavior of Δ with increasing number of electrons. We find for closed shells $\Delta = 0$, which one expects for total electron spin $S = 0$ from effective exchange interaction of the form $H_{\text{el-Mn}} = -\vec{J}\vec{S}\cdot\vec{M}$, with some effective exchange constant \vec{J} . However, for partially filled shells we find Δ to be independent of the filling of the shell despite the fact that the total calculated spin of the ground state, shown in Fig. 2, varies with shell filling. This can be understood in two ways. One is to think of total spin S and effective exchange coupling \vec{J} . When S increases with the filling up of a degenerate shell, effective \vec{J} decreases. An alternative point of view, supported by Fig. 1, is that Mn spin couples effectively to only one electron of the electronic shell, irrespective of the shell

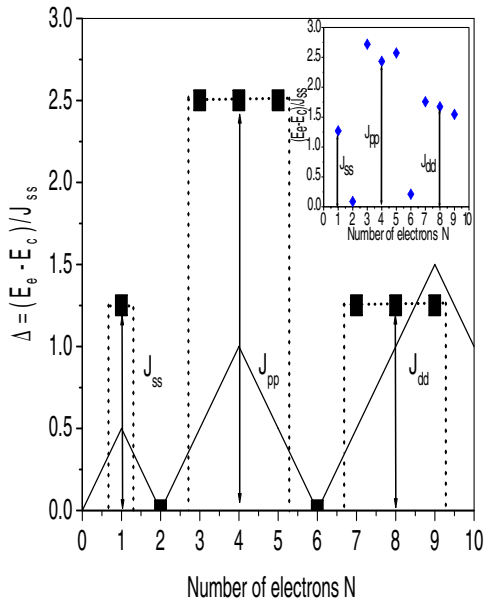


FIG. 2 (color online). Local Zeeman spin splitting $\Delta = |(E_c - E_e)|$ vs number of electrons N for a QD with $\omega_0 = 4$ Ry, $d = 20$ Å, and $R = l_0$, calculated using a limited Hilbert space. The corresponding exchange parameters are indicated by J_{ss} , J_{pp} , and J_{dd} , respectively. Inset shows results of numerical calculation including all configurations of s , p , and d shells.

filling, canceling all many-particle contributions. This cancellation is no longer perfect when higher shells and excitations from filled shells are included. The inset in Fig. 2 shows the calculated shift Δ including all configurations of the s , p , and d shells. We find that Δ is no longer exactly zero for closed shells, and it shows small oscillation across the d shell. The important point is that the results presented here are independent of Mn position for up to $N = 6$ electrons, as clearly shown in Fig. 1. For higher electron numbers $N > 6$ we find the cancellation of many-particle contribution to become more sensitive to Mn position other than the one shown in Figs. 1 and 2, but the general trend remains.

One of the methods to observe this unusual behavior is either capacitance [20] or Coulomb blockade spectroscopies [2,19–21]. In both cases one measures addition energy equal to the chemical potential of the dot. The chemical potential is defined as the difference of total energies $\mu(N) = E(N) - E(N - 1)$. The effect of Mn ion on the chemical potential of N electron QD can be extracted from the difference of chemical potentials of Mn ion doped and undoped dots. In Fig. 3(a) we show the difference of the chemical potential $\Delta\mu = \mu_c - \mu_e$ of the QD with and without Mn ion as a function of N . This change of the chemical potential shows a characteristic behavior related

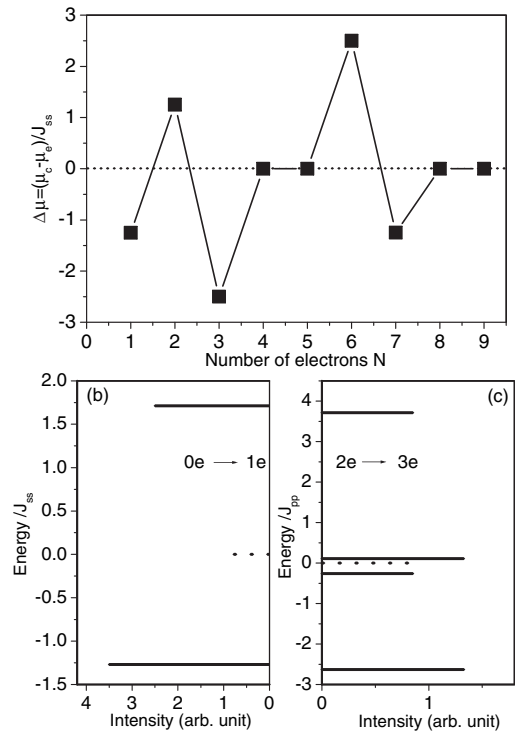


FIG. 3. (a) Difference of the chemical potential $\Delta\mu = \mu_c - \mu_e$ between a single Mn ion doped and undoped quantum dot as a function of the number of electrons N . Spectral function for adding an electron with spin-down to s level of an empty QD (b), and to p shell of $N = 2$ QD (c) with $\omega_0 = 4$ Ry and $d = 20$ Å. For comparison, the spectral function of QD without Mn ion is shown as a dotted line in (b) and (c).

to Fig. 2. We see a negative shift for $N = 1$ and a positive and equal shift for $N = 2$. The $N = 1$ shift measures $-\Delta$. For $N = 2$ the two-electron dot has a closed shell, its energy is not modified by the presence of Mn ion, and the shift is now $+\Delta$. For $N = 3$ we measure $-\Delta$ for a 3 electron dot. For $N = 4$, i.e., in the half-filled shell, the shift in the energy of the N electron and $N - 1$ electron dot is identical and cancels in the chemical potential. Hence in the vicinity of half-filled shells we see no effect of Mn ion on addition energy, the effects are only visible when electron number N crosses from one shell to the next one. Even more dramatic manifestation of the presence of Mn ion can be obtained from the high source and drain spectroscopy [2,21] which measures the spectral function $A(m, \sigma, \omega) = \sum_i P_i \sum_f | \langle f, N + 1 | c_{m,n\sigma}^+ | i, N \rangle |^2 \delta(E_f - E_i - \omega)$ of the quantum dot in state “ i ” and probability P_i . In lower part of Fig. 3 we show examples of spectral functions. Figure 3(b) shows the spectral function for adding an electron with spin-down to s level of an empty quantum dot containing Mn ion. In the absence of Mn ion the spectral function, shown as a dotted line, is a single peak $A(s, \downarrow, \omega) = \delta(\omega_0 - \omega)$. However, for a dot with Mn ion the spectral function (solid lines) breaks into two pieces, $A(s, \downarrow, \omega) = A_- \delta(\omega_0 - J_{ss} - \omega) + A_+ \delta(\omega_0 + 2J_{ss} - \omega)$, separated by $3J_{ss}$. This can be understood by examining the effective exchange Hamiltonian $H_{\text{el-Mn}} = -\tilde{J} \tilde{S} \tilde{M}$. For $S = 1/2$ there are two possible values of total spin $J_{\pm} = M \pm 1/2$. The two total spin values generate two degenerate groups, with degeneracy $2J_{\pm} + 1$, and the two groups of degenerate states are probed by the added electron. When magnetic field is applied, the degeneracies are removed and peaks split into groups of five and seven, a direct manifestation of the presence of Mn ion. A similar situation is encountered for a dot with $N = 2$ electrons and Mn ion when adding extra electron to a p shell, as shown in Fig. 3(c). The spectral function now splits into 3 pieces. The origin of the two extreme pieces, separated by $\sim 6J_{pp}$, is analogous to the s shell, and a third central piece represents the orbital not directly coupled to Mn ion. The indirect coupling with closed s shell electrons does lead to a small splitting of degenerate Mn levels. The application of weak magnetic field would reveal proper degeneracies associated with Mn ion.

We can now compare the effect of Mn ion and quantum dot confinement on electron properties. Quantum confinement leads to quantization of energy levels with large spacing ω_0 but degenerate with respect to spin. The effect of Mn ion is to remove the spin degeneracy of energy levels of electron and of Mn ion. The scale of the effect presented here is measured in terms of exchange coupling J_{ss} . The scale of this coupling is proportional to level spacing, $J_{ss} \sim \omega_0$. The smaller the dot, the larger the spacing. Hence the coupling of an individual electron to Mn ion in a quantum dot is enhanced due to electron confinement. For a model investigated here J_{ss} is found to be 0.21 meV for $\omega_0 = 51.32$ meV.

In summary, we have presented a theory of coupled system of interacting electrons and a single Mn ion in II-VI parabolic quantum dots (QDs). Tuning the number of electrons leads to the variation of total spin of electrons and their exchange coupling with the Mn ion. The exchange coupling can be engineered by the choice of the electronic shell and Mn ion position. It can be switched off for closed electronic shells and maximized for partially filled shells. Unlike the total spin which is maximized for half-filled shells, the exchange interaction does not depend on shell filling. We show how this unexpected effect leads to a characteristic addition and excitation spectrum. Therefore, we demonstrate that the exchange interaction of Mn ions with controlled electronic environment can be engineered in a quantum dot by optimizing quantum confinement, precise control of electron number, and proper choice of Mn position.

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