Voltage-tunable ferromagnetism in semimagnetic quantum dots with few particles: magnetic polarons and electrical capacitance

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Abstract

Magnetic semiconductor quantum dots with a few carriers represent an interesting model system where ferromagnetic interactions can be tuned by voltage. By designing the geometry of a doped quantum dot, one can tailor the anisotropic quantum states of magnetic polarons. The strong anisotropy of magnetic polaron states in disk-like quantum dots with holes comes from the spin splitting in the valence band. The binding energy and spontaneous magnetization of quantum dots oscillate with the number of particles and reflect the shell structure. Due to the Coulomb interaction, the maximum binding energy and spin polarization of magnetic polarons occur in the regime of Hund’s rule when the total spin of holes in a quantum dot is maximum. With increasing number of particles in a quantum dot and for certain orbital configurations, the ferromagnetic state becomes especially stable or may have broken symmetry. In quantum dots with a strong ferromagnetic interaction, the ground state can undergo a transition from a magnetic to a nonmagnetic state with increasing temperature or decreasing exchange interaction. The characteristic temperature and fluctuations of magnetic polarons depend on the binding energy and degeneracy of the shell. The capacitance spectra of magnetic quantum dots with few particles reveal the formation of polaron states.

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I. INTRODUCTION

Diluted magnetic semiconductors combine high-quality crystal structures with the magnetic properties of impurities\(^1\) and represent an important class of materials for spintronics and quantum information\(^2\). The ferromagnetic ordering in diluted magnetic semiconductors can come from the carrier-mediated interaction between magnetic ions\(^3\)\(^4\)\(^5\)\(^6\). Since the carrier density in semiconductor field-effect transistors is a voltage-tunable parameter, the ferromagnetic state of the impurities coupled to the carriers also becomes controlled by the voltage\(^6\). Voltage-control of the ferromagnetic phase transition has been already demonstrated for the Si/Ge and A\(_3\)B\(_5\) material systems\(^4\). This ability to externally control the properties of magnetic crystals with means other than the external magnetic field may have important device applications.

An important feature of modern nanotechnology is the ability to shape semiconductor crystals, designing their quantum properties. Quantum confinement of carriers is expected to strongly affect the magnetic properties of crystals since the quantum-confined structures can be designed in a way to strongly localize carriers near magnetic impurities. The first step toward quantum confinement has been made with the quasi-two-dimensional structures where unusual ferromagnetic properties have been described\(^6\)\(^7\)\(^8\)\(^9\). For one-dimensional lithographic structures, it was found that their transport properties are controlled by the domain walls\(^10\). In parallel, self-organization growth technology suggests zero-dimensional nano-size quantum dots (QDs)\(^11\)\(^12\) which can locally store carriers. Moreover, it has been demonstrated in many experiments\(^13\)\(^14\)\(^15\)\(^16\) that the numbers of carriers and wave function of a QD can be changed by a voltage applied to specially-designed metal contacts; this may permit manipulation of ferromagnetic states in QDs by voltage. Therefore, the combination of semiconductor QDs with magnetic impurities looks particularly interesting; information in such magnetic QDs can be stored not only in the number of carriers but also in the form of the Mn magnetization. Currently, magnetic QDs are a hot topic\(^17\)\(^18\)\(^19\)\(^20\)\(^21\)\(^22\)\(^23\)\(^24\)\(^25\)\(^26\). One important property of Mn-doped nanostructures is that a single particle (electron or hole) can strongly alter the ground state of the system, leading to formation of a magnetic polaron (MP)\(^9\)\(^25\)\(^26\). In the case of a QD with a single Mn impurity, single carriers lead to the formation of hybrid electron-Mn states\(^18\)\(^21\)\(^22\)\(^24\).

In a semi-magnetic QD, a localized MP is formed due to the exchange interaction between
the spins of Mn ions and a carrier trapped in a QD. The MP localized inside a QD resembles a localized acceptor-bound exciton in a bulk semiconductor doped by magnetic impurities. However, self-assembled QDs have important differences: (1) a single QD can trap several electrons (holes), (2) the number of particles in a QD can be tuned with the voltage applied to a metallic contact, and (3) the confining potential of QDs typically is very different from the Coulomb potential.

Here we describe anisotropic MP states of doped QDs of cylindrical symmetry with few holes. The binding energy and magnetization of MP states demonstrate oscillations as a function of the number of holes due to the shell structure. The maximum binding energy and magnetization occur in the regime of the maximum total spin of the hole subsystem when Hund’s rule is applied. Note that a recent paper describes only the odd-even parity oscillations in magnetic QDs (these oscillations come from the spin susceptibility of a system with a discrete spectrum). The enhanced Mn polarization and strong MP binding in a QD with cylindrical symmetry described here originate from the symmetry of the system and the Coulomb interaction. For QDs with few particles we also predict transitions from magnetic to non-magnetic states when the temperature or exchange interaction varies. In addition, we focus here on the hole-mediated ferromagnetism in disk-shaped QDs where the MP state is strongly anisotropic: the spontaneous magnetization appears preferentially in the growth direction.

Another interesting question related to nano-scale ferromagnetism is how the coupled Mn-hole system develops from the MP behavior toward the Zener ferromagnetic phase transition regime in the limit of a large number of particles. To address this question, we will also consider spin fluctuations in a QD as a function of the hole number and a formal self-consistent solution with critical behavior. These results may help to answer the above question. The crossover from the MP regime toward the phase transition behavior can also be important from the point of view of device applications. For example, it is essential to estimate the minimal number of carries needed to achieve a stable ferromagnetic state in a single QD.

The paper is organized as follows: Section II presents a model of a self-assembled QD, Sections III and IV describe the anisotropic MP state with one hole, Section V, VI, and VII contain the results on few-hole QDs, and Sections VIII, IX, and X discuss critical phenomena, fluctuations, and electrical capacitance of magnetic QDs.
II. MODEL

We model the hole-Mn complex in a self-assembled diluted semiconductor QD with the following Hamiltonian:

\[ \hat{H}_{hh} = \frac{\hat{p}^2}{2m_{hh}} + U(R) - \frac{\beta}{3} \hat{j}_z \hat{S}_z, \]  

(1)

where \( \mathbf{R} = (r, z) \) is the radius vector, \( z \) is the vertical coordinate, \( \mathbf{r} = (x, y) \), \( \hat{p} \) is the momentum, and \( \hat{j}_z \) and \( \hat{S}_z \) are the z-components of the hole and Mn momentums, respectively.

\[ \hat{S}_z = \sum_i \hat{S}_{i,z} \delta(\mathbf{R} - \mathbf{R}_i), \]

where \( \hat{S}_{i,z} \) and \( \mathbf{R}_i \) are the spin and position of the \( i \)-impurity, respectively. It is convenient to model the in-plane motion of a hole by the parabolic potential and the vertical motion with a square well. So, we write: \( U(R) = u(z) + m\omega_0^2 r^2 / 2 \), where \( u(z) \) is the z-confinement potential and \( \omega_0 \) is the in-plane frequency. The anisotropic exchange interaction in eq. (1) implies that the QD is disk shaped and the vertical size of QD, \( L \), is much smaller than the in-plane wave function dimension, \( l = \sqrt{\hbar / m_{hh}\omega_0} \), i.e. \( L \ll l \). Since we consider the only heavy-hole states, the exchange interaction (the last term in eq. (1)) becomes strongly anisotropic. In our model, the light-hole states are assumed to be strongly split from the lowest heavy-hole states in the QD. The single-particle spatial hole wave functions and their energies in the absence of Mn impurities are given by \( \psi_{n,m} = f_0(z) \chi_{n_x,n_y}(\mathbf{r}) \) and \( \epsilon_{n,m} = \hbar \omega_0(n_x + n_y + 1) \), respectively. Here, the wave function \( f_0(z) \) corresponds to the lowest state in a square potential well in the z-direction, and \( \chi_{n_x,n_y}(\mathbf{r}) \) are the usual wave functions of a 2D harmonic oscillator; \( n_{x(y)} = 0, 1, 2, \ldots \) are the quantum numbers.

In the spirit of the mean field theory, we can average the operator (1) over the impurity positions and write:

\[ \hat{H}'_{hh} = \frac{\hat{p}^2}{2m_{hh}} + U(R) - \frac{\beta}{3} x_{Mn}(R) N_0 \hat{j}_z \bar{S}_z(R), \]  

(2)

where \( x_{Mn}(\mathbf{r}) \) is the reduced Mn spatial density in the system, \( N_0 \) is the number of cations per unit volume, and \( \bar{S}_z(R) \) is the locally averaged Mn spin:

\[ \bar{S}_z(R) = S B_S \left( \frac{\beta/3 \hat{j}_z(R)}{k_B(T + T_0)} \right), \]  

(3)

4
FIG. 1: (a) Model of a Mn-doped self-assembled QD. (b) Transistor structure with magnetic QDs; the gate voltage controls the number of holes in the QD layer and therefore the magnetic state of QDs. Capacitance of this structure can reveal the magnetic state of QDs.

where $S = 5/2$, $B_S$ is the Brillouin function, $\hat{j}_z(R) = \langle \Psi(R',\chi)|\hat{j}_z\delta(R - R')|\Psi(R',\chi) \rangle$ is the averaged momentum of the hole at the Mn position, and $|\Psi(R,\chi)\rangle$ is the wave function of a hole which depends on the spatial and spin coordinates, $R$ and $\chi$, respectively; $\chi = \pm 3/2$; $T_0$ in eq. 3 appears due to the anti-ferromagnetic interaction between Mn ions.

III. MEAN FIELD SOLUTION

According to eq. 2, the hole in a magnetic QD moves in the presence of the effective spin-dependent potential:

$$U_{eff} = U(R) - \frac{\beta}{3}x_{Mn}(R)N_0\hat{j}_z\hat{S}_z(R).$$

In a QD with strong spatial confinement, we can neglect the effect of the second term in eq. 4 on the spatial wave function. At the same time, we should keep it for the spin part of
the wave function. The ground state of a magnetic QD with a single hole has a simple form:

\[ \Psi = \psi_{0,0}|\uparrow>, \quad \vec{S}_z(R) = SB_S\left(\frac{\beta/3\vec{j}_z(R)}{k_B(T + T_0)}\right), \quad \vec{J}_z(R) = \frac{3}{2}\psi_{0,0}(R)^2, \]  

(5)

where \(|\uparrow>\) is the hole state with \(J_z = +3/2\). Since \(\beta < 0\) for the Mn-hole interaction, \(\vec{S}_z(R) < 0\): at low \(T\), the spins of the hole and Mn ions inside the QD are anti-parallel. This is a state of magnetic polaron with the energy: \(E_0(T) = \hbar\omega_0 - E_b(T)\). The second term in the above equation plays the role of the MP binding energy:

\[ E_b(T) = -\beta/3 \int_R d^3R [\psi_{0,0}^2 x_{Mn}(R) N_0 \frac{3}{2} SB_S(R, T)]. \]  

(6)

In our definition, the binding energy \(E_b(T) < 0\). The ground state of a MP is two fold degenerate since the states \(J_z = \pm 3/2\) have the same binding energy.

The total Mn polarization is calculated as

\[ S_{tot}(T) = \int_R d^3R [x_{Mn} N_0 \beta/3 \vec{j}_z(R) k_B(T + T_0)]. \]  

(7)

The corrections to the wave function (5) and the energy (6) can be found by perturbation theory, in which \(\delta U_{eff} = -\beta/3 x_{Mn}(R) N_0 (3/2) \vec{S}_z(R)\) is a perturbation:

\[ \delta \Psi(R, \chi) = \sum_{(n_x, n_y) \neq (0,0)} \frac{<\psi_{n,m}(R)|\delta U_{eff}|\psi_{0,0}>}{\epsilon_{0,0} - \epsilon_{n,m}} \psi_{n,m}(R)|\uparrow>. \]  

\[ \delta E_0(T) = \sum_{(n_x, n_y) \neq (0,0)} \frac{|<\psi_{0,0}(R)|\delta U_{eff}|\psi_{n,m}>|^2}{\epsilon_{0,0} - \epsilon_{n,m}}. \]  

(8)

Now we can estimate the precision of the perturbation theory: \(\delta E_0/E_b\) is about a few % for the typical parameters of the problem, which will be specified below. The precision is high because of the orthogonality of the spatial harmonic-oscillator functions \(\psi_{n,m}(r)\). For example, the nonzero matrix element \(<\psi_{2,0}(R)|\delta U_{eff}|\psi_{0,0}>\) at zero temperature is about 1 meV and the perturbation-theory parameter \(<\psi_{n,m}(R)|\delta U_{eff}|\psi_{0,0}>/(\hbar\omega_0) \sim 0.02\). With increasing temperature the parameter \(<\psi_{n,m}(R)|\delta U_{eff}|\psi_{0,0}>/(\hbar\omega_0)\) decreases and
the precision of the perturbation theory becomes further improved. The above estimates tell us that the perturbation theory with respect to the potential \( \delta U_{\text{eff}} \) provides us with reliable results. We also note that our results are in agreement with ref.\textsuperscript{17}.

The ferromagnetic state of a QD depends on the spatial distribution of Mn ions, which is given by the crystal growth technology. Diffusion of Mn ions during QD growth is driven by strain and composition inhomogeneity and can lead to a strongly non-uniform distribution of Mn ions in the system. For example, a study of the \( \text{InMnAs} \) system\textsuperscript{31} has shown that the Mn ions during the growth process mostly substitute for the In atoms inside a QD. Here we are going to use a simple model (fig. 1) in which the Mn-doped region forms a disk of radius \( R_{\text{Mn}} \) inside a QD: \( x_{\text{Mn}}(\rho) = x_{\text{Mn}} \) for \( \rho < R_{\text{Mn}} \) and 0 otherwise. Here \( \rho \) is the distance to the center of a QD in the 2D plane. It follows from eq. 6 that the binding energy of a MP and the "robustness" of a magnetic state depend on the overlap between the Mn distribution \( x_{\text{Mn}}(R) \) and the wave function of the hole. Since both of them can be tailored and controlled by the growth process the ferromagnetic state can be artificially designed.

Fig. 2 shows the calculated energy of a MP with one hole for different Mn distributions, \( R_{\text{Mn}} = 2, 3 \) nm and \( \infty \). The corresponding Mn magnetization is shown in fig. 3. The following parameters of a magnetic semiconductor have been used: \( x_{\text{Mn}} = 0.04, \beta N_0 = -1.3 \) eV, \( T_0 = 3.6 \) K, and \( N_0 = 15 \) nm\(^{-3} \). The QD geometrical parameters were chosen as follows: \( l_0 = 4 \) nm and \( L = 2.5 \) nm. The above material parameters represent a \( \text{CdMnTe} \) QD.

IV. ANISOTROPY OF BINDING ENERGY AND MAGNETIZATION

The MP state of a hole in a disk-shaped QD is strongly anisotropic due to the valence band structure\textsuperscript{30}. This magnetic anisotropy comes from the heavy-light hole splitting in the valence band and reveals itself in the last term in the operator (1). The general solution of the one-hole problem can be written as

\[
\Psi = \psi_{0,0}(a|\uparrow> + b|\downarrow>),
\]

where \( |a|^2 + |b|^2 = 1 \). Then the MP binding energy takes the form:
FIG. 2: Calculated energy of a MP with one hole for different Mn distributions; \( x_{Mn} = 0.04 \) and \( R_{Mn} = 2, 3 \text{ nm}, \) and \( \infty \).

\[
E_b(T) = -\frac{\beta}{3} \int_R d^3R [\psi_{0,0}^2(R)x_{Mn}(R)N_0^3 \frac{3}{2}(2|a|^2 - 1)SBS(\frac{\beta/3\psi_{0,0}(R)^2}{k_B(T + T_0)}|2|a|^2 - 1)]
\]

where \( 0 \leq |a| \leq 1 \). Figure 4 shows the MP binding energy as a function of \( |a| \). For the cases \( |a| = 1 \) and \( |b| = 1 \), the binding energy magnitude is maximum; for \( |a| = 1/\sqrt{2} \) it equals to zero. Therefore, the ground state of a MP corresponds to the pure states \(|\uparrow\rangle\) or \(|\downarrow\rangle\). This anisotropy likely plays an important role in optical experiments with excitons trapped in semi-magnetic QDs. In such experiments, an optically-created electron-hole pair rapidly relaxes to its ground state resulting in the formation of a MP with the spin parallel (or antiparallel) to the growth axis.

V. FEW PARTICLE STATES

In the next step, we study the QD magnetization in the presence of few carriers which can be loaded from the metal back contact in a voltage-tunable transistor structure (fig. 1b). We
FIG. 3: Calculated magnetization of Mn ions as a function of temperature for the QD with one hole and different Mn distributions; \( x_{Mn} = 0.04 \) and \( R_{Mn} = 2, 3 \) nm, and \( \infty \).

will sequentially consider the few first charged states of a QD starting from \( n_h = 1 \) (fig. 5).

The Coulomb interaction will be treated within perturbation theory which is valid for QDs with a strong confinement; namely, we will assume that the quantization energy of QD, \( \hbar \omega_0 \), is larger than the characteristic parameter of Coulomb interaction between particles, \( E_{Coul} = e^2/(\epsilon l) \). For the QD parameters specified above, we obtain: \( \hbar \omega_0 \approx 47 \) meV and \( E_{Coul} \approx 29 \) meV. This simplified perturbation approach is very convenient and was successfully used for description of experimental data in several publications\(^{13,15,32,33}\). Note that the parameters of the QDs studied in\(^{13,15}\) are close to those used here.

In the perturbation approach, we will neglect the Coulomb-induced mixing between shells; at the same time, we will calculate exactly the Coulomb-induced mixing within shells by diagonalizing the corresponding matrix. Thus, the Coulomb correlations will play a very important role for certain states, such as the Hund’s states with two particles in the \( p \)-shell \( (n_h = 4) \) and three holes in the \( d \)-shell \( (n=9) \).

For some derivations, it will be convenient to treat this problem using the second quantization approach. In this approach, the \( z \)-component of the total angular momentum of the
FIG. 4: Spin anisotropy of the MP energy as a function of the probability to find the hole in the state \( j_z = +3/2; \) \( x_{ Mn} = 0.04 \) and \( R_{ Mn} = \infty. \)

The hole subsystem is given by

\[
\hat{j}_{z,tot}(R) = \sum_{\gamma,j_z=\mp3/2} j_z \vert \psi(\gamma)(R) \vert^2 \hat{c}_{\gamma,j_z}^+ \hat{c}_{\gamma,j_z},
\]

where \( \hat{c}_{\gamma,j_z}^+ \) is the creation operator for the single-particle state (\( \gamma, j_z \)). Here \( \gamma \) stands for the pair of orbital quantum numbers; \( \gamma \) may be \( n_x, n_y \) or \( n, m_z \) depending on the choice of wave functions; here \( n \) and \( m_z \) are the radial quantum number and the orbital angular momentum, respectively. For the few-particle wave functions, we will employ the following notations: \( \vert s_\uparrow, s_\downarrow; p_{1\uparrow}, p_{1\downarrow}; p_{2\uparrow}, p_{2\downarrow}; d_{1\uparrow}, d_{1\downarrow}; d_{2\uparrow}, d_{2\downarrow}; d_{3\uparrow}, d_{3\downarrow}; \ldots \rangle \), where \( s_{\uparrow(\downarrow)}, p_{i\uparrow(\downarrow)} \), and \( d_{k\uparrow(\downarrow)} \) are the occupation numbers for the s-, p-, and d- states with the corresponding spins. These occupation numbers can be either 0 or 1. For the indices \( i \) and \( k \), \( i = 1, 2 \) and \( k = 1, 2, 3 \).

VI. QUANTUM DOT WITH TWO HOLES

If the QD is occupied by two particles (\( n_h = 2 \)), the spin and spatial variables in the two-hole wave function can be separated. Then, for a QD with strong confinement, it becomes obvious that the z-component of the total hole spin in the ground state is zero (\( j_{z,tot} = 0 \)) (fig. 6a) and therefore the exchange interaction with the Mn subsystem vanishes. However,
FIG. 5: (a) Ground-state configuration of a cylindrically symmetric QD with few holes and without Mn ions; the state with \( n_h = 4 \) is constructed according to Hund’s rule. These configurations remain ground states if the Mn-hole interaction is weak or the QD confinement is strong enough. (b) Ground-state configuration for \( n_h = 9 \) in the regime of Hund’s rule; again this configuration represents a ground state if the Mn-hole interaction is not very strong.

if the confinement is not strong enough, the ground state can change with temperature. Figure 6b shows the hole configuration which can become a ground state if the Mn-hole interaction is strong enough. This state has the total spin \( j_{z,tot} = 2 \cdot (3/2) \) and a non-zero binding energy in the presence of the Mn subsystem. The ground state transition occurs when

\[
E_1 - E_0 = |E_{b}^{n_h=2}|. \tag{12}
\]

Here \( E_0 \) and \( E_1 \) are the hole energies of the states (a) and (b) in fig 6. The hole energies \( E_{0(1)} \) also include the contributions from the Coulomb interaction. \( E_{b}^{n_h=2} \) is the binding energy for the configuration (b) in fig 6.

The Hamiltonian of the system is now written as:
FIG. 6: Ground-state configurations of a QD with cylindrical symmetry and two holes. The configuration (a) is realized in the case of a relatively weak Mn-hole interaction (high temperature or small Mn density) and the state (b) corresponds to the system with a strong Mn-hole interaction (low temperature or high Mn density).

\[ \hat{H}_{n_{h}=2} = \sum_{i=1,2} [\hat{T}_{i} + U(R_{i})] - \frac{\beta}{3} x_{Mn}(R) N_{0} \hat{j}_{z,tot} S_{z}(R_{i}) + U_{Coul}, \]

where \( \hat{j}_{z,tot} \) is the z-component of the total momentum (spin) of holes, \( \hat{T}_{i} \) is the kinetic energy of the \( i \)-hole, and \( U_{Coul} \) is the Coulomb interaction. For the Coulomb potential, we use the usual formula: \( e^{2}/\epsilon |R_{1} - R_{2}| \), where \( \epsilon \) is the dielectric constant.

Regarding the Coulomb interaction in eq. 13 we will treat it again as a perturbation assuming a strongly-confined QD\textsuperscript{13,15,32,33}. Then, we obtain for the energies of the states: \( E_{0} = 2\hbar \omega_{0} + U_{ss}^{dir} \) and \( E_{1} = 3\hbar \omega_{0} + U_{sp}^{dir} - U_{sp}^{exc} \), where \( U_{\alpha,\alpha'}^{dir} \) and \( U_{\alpha,\alpha'}^{exc} \) are the direct and exchange Coulomb elements, respectively. The Coulomb matrix elements \( U_{sp}^{dir(exc)} \) do not depend on a particular choice of the single-particle wave functions for the p-shell. For the state (b) in fig. 6 the operator \( \hat{j}_{z,tot} \) includes two terms:

\[ \hat{j}_{z,tot}(R) = \frac{3}{2} (|\psi_{0,0}(R)|^{2} \hat{c}_{s,\uparrow}^{+} \hat{c}_{s,\uparrow} + |\psi_{p}(R)|^{2} \hat{c}_{p,\uparrow}^{+} \hat{c}_{p,\uparrow}), \]

where the p-state orbital wave function \( \psi_{p} \) can be written as:

\[ \psi_{p} = \alpha \psi_{+} + \beta \psi_{-}; \]

here \( \psi_{+(-)}(\phi) \propto e^{\pm \phi} \) and \( |\alpha|^{2} + |\beta|^{2} = 1 \), where \( r = (\rho, \phi) \). The indices \(+(-)\) correspond
FIG. 7: (a) Orbital anisotropy of the MP energy with $n_h = 2$ and $j_{z,\text{tot}} = 3$. The minimum energy corresponds to the state with $|\alpha|^2 = 1/2$ in which the angular part of the spatial probability distribution is most inhomogeneous, i.e. $\psi_p^2(\phi) \propto \cos(\phi + \phi_0)^2$. (b) The energy of the states $j_{z,\text{tot}} = 3$ and $j_{z,\text{tot}} = 0$ (figs. 6a and b) as a function of temperature; the ground state of the system changes with temperature. $R_{\text{Mn}} = \infty$ and $x_{\text{eff}} = 0.04$

to the wave functions with the orbital angular momentum $m_z = +1(-1)$, respectively. The binding energy of the MP with two holes

$$E_b(T, \alpha) = -\frac{\beta}{3} \int d^3R [F_2(R)x_{\text{Mn}}(R)N_0\frac{3}{2}\overline{S}B_0\left(\frac{\beta/3F_2(R)^2}{k_B(T + T_0)}\right)], \quad (16)$$

where $F_2(R) = |\psi_{0,0}(R)|^2 + |\psi_p(R)|^2$ is the particle density in the system. The binding energy now depends on the parameter $\alpha$. We find numerically that the ground state is realized for $|\alpha|^2 = 1/2$. This ground state corresponds to the wave function with the most inhomogeneous p-orbital as a function of $\phi$: $\psi_p \propto \cos(\phi + \phi_0)$, where $\phi_0$ is an arbitrary phase. It can be understood as follows: in the ground state, the spatial wave function of holes should be most localized because a strongly localized hole can better control the Mn spins and more strongly lower the total energy. This case resembles somewhat a self-trapped
MP in the systems with translation invariance. We note that the ground state of MP is degenerate since the phase $\phi_0$ is arbitrary in a system with cylindric symmetry. However, if the QD confinement is anisotropic, the phase $\phi_0$ will be fixed by the anisotropy of the QD potential.

In fig. 7a, we show the MP binding energy as a function of $|\alpha|^2$. When $|\alpha|^2 = 1$ or 0, the p-hole state has the orbital angular momentum $m_z = +1$ or $m_z = -1$, when $|\alpha|^2 = 1/2$, it has linear polarization. For a QD with the parameters $m_{hh} = 0.1m_0$, $\hbar \omega_0 = 47.3 \text{ meV}$, and $\epsilon = 12.5$, the Mn-hole system demonstrates a critical temperature at which the ground state changes from the state $j_{z,\text{tot}} = 3$ to the state with $j_{z,\text{tot}} = 0$ (fig. 7b). This critical temperature corresponds to the solution of eq. 12. The existence of this transition depends on the particular choice of parameters of a QD. For example, if the density of Mn impurities is small enough, the ground state will be always $j_{z,\text{tot}} = 0$ and the transition will not occur at all.

VII. QUANTUM DOT WITH A SMALLER Mn DENSITY AND $n_h = 1 - 6$

We now consider a QD with $n_h$ ranging from 1 to 6. As was pointed out above Coulomb correlations will become very important for certain states. In addition, we will assume that the Mn density is a few times smaller than in the previous calculations and the ground-state transitions of the type shown in fig. 7 do not occur. Therefore, the ground state configurations coincide with those in a non-magnetic QD (fig. 5). The calculated binding energies for the lower doping $x_{\text{eff}} = 0.01$ are shown in fig. 8.

We now proceed to the case of three holes ($n_h = 3$). In this case, the ground state has two holes in the s-shell and one in the p-shell. The closest excited state has one hole in the s-shell and two spin-polarized holes in the p-shell ($j_{z,\text{tot}} = 3 \times (3/2)$). Despite the Mn-hole interaction, the configuration $j_{z,\text{tot}} = 3 \times (3/2)$ remains an excited state since the additional MP binding energy in this state is about 20 meV and significantly lower than the quantization energy $\hbar \omega_0$ for our parameters. In the ground state with $n_h = 3$, a nonzero angular momentum of the hole system comes from the unpaired hole in the p-state. The result for the MP binding energy is given by eq. 16 with $F_2(R) = |\psi_p(R)|^2$. Again the ground state is anisotropic due to the degeneracy of the p-shell.

To describe the ground state of a QD with $n_h = 4$, we should apply Hund’s rule. In the
FIG. 8: Calculated binding energy of a MP as a function of temperature for various charged states of the QD: $n_h = 1, 3, 4$ and 5. The binding for the states with completely filled shells ($n_h = 2, 6$) is zero in our approach; $R_{Mn} = \infty$.

state $n_h = 4$, the s-shell is completely filled, whereas the p-shell is occupied by two holes. According to Hund’s rule, the ground state of the many-particle system without the Mn subsystem should have two particles with parallel spins in the upper shell and the maximum spin 3. The corresponding wave function should be found by mixing the Slater determinants related to the p-shell and diagonalizing the Coulomb matrix. The states with the smallest energy form a triplet:

$$
|1, 1; 1, 0; 1, 0 >, \ |1, 1; 0, 1; 0, 1 >, \ \frac{|1, 1; 0, 1; 1, 0 > +|1, 1; 1, 0; 0, 1 >}{\sqrt{2}}.
$$

(17)

For the case $n_h = 4$, the first excited states within the p-shell are $|1, 1; 1, 1; 0, 0 >$ and $|1, 1; 0, 0; 1, 1 >$. The above wave functions should be represented by Slater determinants composed of the orbitals $\psi_s$ and $\psi_{\pm}$. The state with the largest energy has the configuration:

$$
\frac{|1, 1; 0, 1; 0 > -|1, 1; 1, 0; 1 >}{\sqrt{2}}.
$$

The energies of the above states are given by: $E_0 - U_{pp}^{exc}$, $E_0$, and $E_0 + U_{pp}^{exc}$, where $U_{pp}^{exc}$ is the exchange integral and $E_0$ involves single-particle energies and
some Coulomb interactions. In the absence of the Mn subsystem, the difference of energy between the ground state \((j_{z,\text{tot}} = 3)\) and the first excited states \((j_{z,\text{tot}} = 0)\) is equal to the exchange energy between p-states: 

\[
U_{pp}^{\text{exc}} = \left(\frac{e^2}{\epsilon}\right) \int dR_1 dR_2 \frac{\psi_+^*(R_1) \psi_+(R_2) \psi_+(R_2) \psi_-^*(R_1)}{|R_1 - R_2|}. 
\]

Moreover, the energy of the state \(\Psi_0 = |1, 1; 1, 0; 1, 0\rangle\) will be lowered due to interaction with Mn spins and this lowering can be very significant. The reason is the 2-fold increase of the total hole spin:

\[
< \Psi_0 | \hat{j}_{z,\text{tot}}(R) | \Psi_0 > = \frac{3}{2} |\psi_+(R)|^2. 
\]

The MP binding energy for this state is

\[
E_{b,n_h=4}(T) = -\frac{\beta}{3} \int d^3R [|\psi_+(R)|^2 x_{Mn}(R) 3N_0SB \frac{\beta |\psi_+(R)|^2}{k_B(T + T_0)}]. (19)
\]

We note that the increase of the binding in eq. (19) comes from the factors 2 before and inside the Brillouin function. Also, the MP energy (eq. (19)) in the case of \(n_h = 4\) does not depend on the particular choice of the single particle functions; in the coordinate representation, the p-electrons are described with the anti-symmetric wave function: 

\[
\Psi_0 \propto sin(\phi_1 - \phi_2) |\uparrow_1 > | \uparrow_2 >.
\]

The magnitude of the calculated MP binding energy demonstrates a strong increase (about two times) (fig. 3). The Mn magnetization also increases for the case \(n_h = 4\) (fig. 3).

The ground state \(n_h = 5\) is again non-uniform as a function of angle \(\phi\):

\[
\Psi_0^{n_h=5} = \alpha |1, 1; 1, 1, 0 > + \beta |1, 1; 1, 0; 1, 1 > . (20)
\]

When we ignore inter-shell mixing, the results for \(n_h = 5\) become similar to those for \(n_h = 3\). The magnetization comes from one unpaired p-hole:

\[
< \Psi_0 | \hat{j}_{z,\text{tot}}(R) | \Psi_0 > = \frac{3}{2} |\alpha \psi_+ + \beta \psi_-|^2. 
\]

The lowest MP energy is obtained for the "linearly-polarized" state of holes \((\alpha = 1/\sqrt{2})\) as in the case of \(n_h = 3\) (fig. 3).

The state \(n_h = 6\) has completely filled s- and p-shells and spontaneous Mn magnetization does not appear for QDs with a relatively weak Mn-hole interaction (the inter-shell mixing is neglected).
FIG. 9: Calculated magnetization of Mn ions as a function of temperature for various charged states of QD: $n_h = 1, 3, 4$ and $5$. The magnetization for the states with completely filled shells ($n_h = 2, 6$) is zero in our approach; $x_{eff} = 0.01$ and $R_{Mn} = \infty$ nm.

VIII. FORMAL SELF-CONSISTENT SOLUTION FOR A FEW PARTICLE QUANTUM DOT: TOWARD THE FERROMAGNETIC PHASE TRANSITION.

The system of mobile carriers can undergo a ferromagnetic transition if the Mn carrier interaction is strong enough and exceeds the anti-ferromagnetic interaction between ions. The Curie temperature of Zener ferromagnetism is given by the density of states of mobile carriers, the density of Mn ions, and the Mn-hole interaction constant $x_{eff}$:

$$T_{Curie} \propto S(S+1)x_{eff}N_0\rho(E_F)\beta^2/k_B,$$

where $\rho(E_F)$ is the density of states at the Fermi level. It is known that the MP state in a QD does not undergo the phase transition: the spontaneous polarization in the MP state simply decreases with temperature $^{27}$. This is due to the fact that the system contains just one or few carriers.
We now consider a formal self-consistent solution of the mean field theory in a manner similar to the Zener theory. For \( n_h = 1 \), we employ eq. 3 and obtain a self-consistent integral equation

\[
\bar{S}_z(R) = S B_S \frac{\beta/3 < j_z >_T}{k_B(T + T_0)}, \quad (23)
\]

where the \( < j_z >_T \) is now averaged over the states of a hole:

\[
< j_z >_T = \frac{3}{2} \psi_{0,0}^2 e^{-\frac{\Delta_1}{k_B T}} - e^{\frac{\Delta_1}{k_B T}} Z_s(T), \quad (24)
\]

where the quantity

\[
\Delta_1 = -\frac{\beta}{3} \int d^3 R \left[ \psi_{0,0}^2 x_{eff} N_0 \bar{S}_z(R) \right] \quad (25)
\]

plays the role of binding energy for the hole state +3/2; \( Z_s = e^{-\frac{\Delta_1}{k_B T}} + e^{\frac{\Delta_1}{k_B T}} \) is the partition sum for the s-shell. The inter-shell mixing is again ignored. Assuming a homogeneous spatial Mn distribution and integrating eq. 23, we obtain:

\[
\Delta_1 = -\frac{\beta}{3} \int d^3 R \psi_{0,0}^2 N_0 f_1(\Delta_1), \quad (26)
\]

where \( f_1(\Delta_1) = \int B_S \frac{\beta/3 < j_z >_T}{k_B(T + T_0)} d^3 R \). This simple equation leads to the critical behavior: a solution with nonzero magnetization \( (\Delta_1 > 0) \) exists if \( T < T_{crit} \) where

\[
k_B T_{crit} = \sqrt{\left( \frac{\beta}{3} \right)^2 j_h^2 S(S+1) x_{eff} N_0 \int d^3 R \psi_{0,0}^4 - k_B T_0} \quad (27)
\]

This critical behavior occurs in the regime of strong fluctuations in the MP state and obviously is incorrect. It is known that MPs do not exhibit a critical behavior. However, it seems to be interesting to compute how this formal self-consistent solution develops with increasing number of holes in the QD. First, the self-consistent approach becomes more reliable with increasing the number of particles. Second, eq. 27 gives a useful estimate of the characteristic temperature of spontaneous magnetization for the MP states in a QD.

For the cases of \( n_h = 4 \) and \( n_h = 9 \), we have found similar self-consistent solutions for the energy and magnetization by solving the corresponding nonlinear equations. For example,
in the case of \( n_h = 9 \), the ground-state configuration is constructed according to Hund’s rule (fig. 5b) and the self-consistent problem is reduced to a system of two nonlinear equations of two variables: 
\[
x_1 = \int d^3 \mathbf{R} [\psi_{d,0}^2 \bar{S}_z(\mathbf{R})] \quad \text{and} \quad x_2 = \int d^3 \mathbf{R} [\psi_{d,+}^2 \bar{S}_z(\mathbf{R})],
\]
where \( \psi_{d,0} \) and \( \psi_{d,+} \) are the d-orbitals with \( m_z = 0 \) and \( +2 \), respectively. Again we ignored inter-shell mixing and diagonalized the Coulomb matrix inside the p- and d-shells. Then, using the obtained energies, we constructed the partition sums for the p- and d-shell. In figure 10b, we show by dashed lines the averaged energies of MPs calculated within the self-consistent approach. In the same figure, the solutions without the self-consistent averaging of the spin inside the Brillouin function are shown as solid curves.

Equation (27) gives a characteristic temperature at which spontaneous Mn polarization exists in a QD. From fig. 10, we see that the critical temperature increases with number of holes. However, this increase is not very strong. The reason is that the p- and d-shells have several states and some of these states have opposite spins or no spin. These states contribute to the partition sum and therefore effectively reduce the magnetization.

It is interesting to compare the typical temperature of MPs with that of ferromagnetic phase transitions in bulk. For CdMnTe, it was suggested that the Curie temperature for the highest hole densities can be as high as a few Kelvins\(^3\). In a QD with about 20 Mn ions considered here \( (x_{\text{eff}} = 0.01, l = 4 \text{ nm, and } L_z = 2.5 \text{ nm}) \), the typical temperature given by eq. 27 is about 22 K, an order of magnitude larger than that in bulk. This is also consistent with previous papers on MPs\(^{19,20}\). We can also note the important differences between the above equations for \( T_{\text{Curie}} \) and \( T_{\text{crit}} \). The critical temperature is proportional to the first power of the interaction \( \beta \) and depends on the QD localization length \( (T_{\text{crit}} \sim l^{-3/2}) \).

IX. FLUCTUATIONS AS A FUNCTION OF \( n_h \)

To better understand the behavior of MPs at high temperature, we now compute the relative fluctuations of the hole subsystem:

\[
\delta j_{z,\text{tot}}(T, n_h) = \sqrt{\frac{\langle (\hat{j}_{z,\text{tot}} - \langle \hat{j}_{z,\text{tot}} \rangle_T)^2 \rangle_T}{\langle \hat{j}_{z,\text{tot}} \rangle_T}}, \tag{28}
\]

where \( \langle ... \rangle_T \) means thermal averaging over the many-particle states assuming a given Mn spin distribution. For the Mn spin distribution we will use eq. 3 calculated for a certain
quantum state of the hole subsystem.

In the low-temperature regime, the relative fluctuations are much weaker than the average spin of holes (fig. 10). With increasing temperature, the fluctuations grow and exceed $<\hat{j}_{z,\text{tot}}>_T$. Figure 10 shows clearly the tendency of stabilization of the MP state with increasing number of holes for the states $n_h = 1, 3, 9$. This stabilization is consistent with the increase of binding. Clearly, methods beyond the mean-field theory are required to understand the mechanism of crossover from the MP behavior to the Zener ferromagnetic
FIG. 11: (a) Calculated energies of different charged states in the layer of semi-magnetic quantum dots with $x_{Mn} = 0.01$; $R_{Mn} = \infty$. The labels show the charged states of the system.

Phase transition\textsuperscript{28,36}.

X. CAPACITANCE OF QD SYSTEMS WITH MAGNETIC POLARONS

One efficient method to study quantum states of QDs is capacitance spectroscopy. Such spectroscopy is typically performed at a nonzero frequency $\omega$. The capacitance of the QD structure (fig. 1b) includes the contribution of charges trapped inside the QD layer\textsuperscript{15}:

\begin{equation}
\delta C = e^2 D(V_g),
\end{equation}

where $V_g$ is the gate voltage and $D(V_g)$ is the effective density of states in the QD layer. The latter is defined as

\begin{equation}
D(V_g) = N_{dot} \frac{dn_h}{|e|dV_g},
\end{equation}
FIG. 12: (a) Calculated capacitance spectra for non-magnetic (a) and magnetic (b) QDs; $x_{\text{eff}} = 0.01$. The broadening of the peaks was taken as 16 meV.

where $n_h$ is the number of holes trapped in a single QD and $N_{\text{dot}}$ is the 2D density of QDs. This approach to the capacitance is valid at low frequencies $\omega \tau \ll 1$, where $\tau$ represents both the tunneling time and the relaxation time to form the ground MP state. In other words, this approach assumes that the weakly coupled system "QD + metal contact" has a short relaxation time and always remains in its ground state while the gate voltage changes in time as $V_g + \delta V_g \cos(\omega t)$ ($V_g \gg \delta V_g$).

According to the simple model of a field-effect structure with a QD layer, the energy of a single particle in the QD is written as: $E_0(V_g) = E_{\text{sp}} + |e|\gamma V_g$, where $E_{\text{sp}}$ is the single-particle energy inside the QD, $\gamma = d_1/d_2$ is the lever arm coefficient, and $d_1$ and $d_2$ are the dimensions of the structure (fig. 1). For example, $\gamma \sim 1/6$ in ref. 15. For convenience, we assume that the Fermi energy of the metal contact is zero and loading of the first hole to the QD occurs at zero bias. Then, the first charged states of the QD ($n_h = 1$ and 2) have the energies:
\[ E_1(V_g) = \gamma |e| V_g + E_b^{n_h=1}, \]
\[ E_2(V_g) = 2\gamma |e| V_g + U_{\text{dir}}. \]  
(31)

The energies of the next charged states \( n_h = 3 \) and \( 4 \) are

\[ E_3(V_g) = \hbar \omega_0 + 3\gamma |e| V_g + U_{\text{coul},3} + E_b^{n_h=3}, \]
\[ E_4(V_g) = 2\hbar \omega_0 + 4\gamma |e| V_g + U_{\text{coul},4} + E_b^{n_h=4}. \]  
(32)

The Coulomb energies \( U_{\text{Coul},n_h} \) in the above equations should be calculated for the ground-state configurations shown in fig. 5a. Figure 11 shows the calculated energies of the first changed states related to the s- and p-shells for non-magnetic QDs. As was realized in several experiments, the ground state of the system changes with voltage: a QD sequentially traps \( 1, 2, 3, ... \) particles. Then, the quantity \( D(V_g) \) and the capacitance demonstrate peaks at the voltages of the ground-state transitions. In real QD systems these peaks are broadened due to the nonzero size dispersion in a QD ensemble. The effects of the ferromagnetic interaction are clearly seen in the calculated capacitance spectra (fig. 12). The spacing between the p-orbital peaks \( n_h = 4 \) and \( 5 \) becomes strongly increased. It comes from the strong ferromagnetic coupling in the regime of Hund’s rule for the state \( n_h = 4 \). The spacing between the two s-orbital peaks is also increased due to the MP effect. At the same time, the voltage interval between the s- and p-related structures becomes reduced. This is again due to the exchange interaction.

According to fig. 12, the magnetic and non-magnetic QDs with the parameters chosen in this paper show the same order of peaks in the capacitance spectra. With increasing Mn-hole interaction, the situation can change and the sequence of peaks can became different for the magnetic and non-magnetic systems. The reason is that, in magnetic QDs, more carriers can be trapped to achieve the minimum energy.

In addition to the characteristic behavior of the inter-peak spacings, the MP effect in the capacitance spectra can be recognized by varying temperature or by applying an external magnetic field. With temperature, the peaks related to the most bound MP states \( n_h = 1, 3, 4, 5 \) will have the strongest temperature dispersion. As for the magnetic fields, one possibility is to use the in-plane field and to suppress the Mn-hole interaction. Then, the
characteristic magnetic dispersion of the peak positions will reveal the MP binding energy. The behavior of the peaks in a perpendicular magnetic field can also be revealing as was shown in other publications.

XI. DISCUSSION

The model with exchange interaction of the type $\hat{j}_z \hat{S}_{z,i} \delta(R_h - R_i)$ assumes that the magnetic impurity does not create any spin-independent potential. This model is widely applied for II-VI semiconductors where individual impurities do not form bound acceptor states. This is in contrast to the GaAs system. In the GaAs crystal, a magnetic Mn impurity forms a deep acceptor state (about 110 meV above the top of the valence band) and therefore the model of impurity in bulk GaAs should incorporate the effect of the spin-independent attracting potential. Treating the typical III–V QDs realized in the InGaAs system we should take into account two factors: (1) the possible lower binding energy in the InGaAs system for the Mn acceptor state and (2) the spatial confinement in a QD. To understand the importance of the acceptor potential, we should compare the localization length of the Mn-acceptor in bulk with the size of the QD. If the QD dimension is smaller than the Mn-acceptor size, the QD can be treated without the acceptor potential. Simultaneously, the spin-dependent exchange interaction should remain in the model since it leads to the formation of the MP state. In the opposite limit of a weak QD confinement, the QD potential can be treated as a perturbation; this case was recently analyzed in ref. To summarize, if the electronic size of a QD becomes smaller than the dimension of the acceptor states of the Mn impurities inside a QD, the simple model of contact exchange interaction becomes applicable. This suggests that, under certain conditions, InGaAs QDs can also be treated with the simple model used in this paper.

To conclude, we have calculated the MP energies and associated capacitance spectra of QDs with a few holes in the presence of the Mn-hole exchange interaction. The system studied in this paper exhibits several features coming from the joint action of the Mn-hole exchange coupling, quantum confinement, and Coulomb interaction.

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