

Electronic states in a magnetic quantum-dot molecule: Instabilities and spontaneous symmetry breaking

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We show that a double quantum-dot system made of diluted magnetic semiconductor behaves unlike the usual molecules. In a semiconductor double quantum dot or in a diatomic molecule, the ground state of a single carrier is described by a symmetric orbital. In a magnetic material molecule, new ground states with broken symmetry can appear due to the competition between the tunneling and magnetic polaron energy. With decreasing temperature, the ground state changes from the normal symmetric state to a state with spontaneously broken symmetry. Interestingly, the symmetry of a magnetic molecule is recovered at very low temperatures. A magnetic double quantum dot with broken-symmetry phases can be used as a voltage-controlled nanoscale memory cell.

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In diluted magnetic semiconductors, the spins of static impurities interact through mobile carriers and can form a ferromagnetic state.^{1,2} Since the carrier density in semiconductors is a voltage-tunable parameter, the ferromagnetic state also becomes controlled by the voltage.³ For technology, this may give an important advantage compared to the conventional memories based on ferromagnetic metals and controlled by the magnetic field. When a single confined carrier interacts with magnetic impurities, it forms a new stable state, called a magnetic polaron.⁴ One important class of confined nanostructures is the quantum dots (QDs) where the number of trapped carriers can be easily changed by the voltage applied to the top gate.⁵ Information in a single QD made of magnetic semiconductor can be stored in the form of spin polarization and therefore such a QD can be viewed as a nanoscale magnetic memory element. Magnetic QDs as memory elements have some properties, that look very attractive for the technology: (1) small sizes, (2) small number of carriers, and (3) voltage control of the number of carriers. Currently, the physics of electronic states in magnetic QDs is an active field of research.⁶⁻¹⁰ Here we make a logical step from a single magnetic nanocrystals toward QD molecules and show that a magnetic QD pair has unique physical properties that may also be useful for device applications.

In this paper, we consider a magnetic double QD with one hole. Using the mean field theory, we calculate the physical properties of magnetic polarons formed due to the Mn-hole exchange interaction. We find that this system undergoes two transitions (Fig. 1). At high temperatures, the kinetic tunneling energy of the hole is larger than the magnetic polaron binding energy and a symmetric state (with equal hole probabilities for both dots) is realized. With decreasing temperature (T), the local magnetic energy becomes large enough to trap the hole in one of the dots. This self-trapped process spontaneously breaks the symmetry of the system. At very low temperature, the local Mn-spin polarization can become so strong that the symmetry is recovered. Our broken-symmetry polaron has several unique properties: it appears in the confined geometry, is voltage controllable, and may vanish at very low T . The above transitions occur also with

changing the energy barrier between the dots. Since a double QD can be controlled by the gate voltage, the above phases can be prepared and read using electrical means.

Model. The Hamiltonian of the system composed of two magnetic QDs and one hole has a form

$$H_{hh} = \frac{\hat{\mathbf{p}}^2}{2m_{hh}} + U_0(\mathbf{R}) - \frac{\beta}{3} \hat{j}_z \cdot \hat{S}_z, \quad (1)$$

where $\mathbf{R}=(\mathbf{r}, z)$, \mathbf{r} and z are the in-plane vector and the vertical coordinate, respectively; \hat{j}_z is the z component of the hole spin and $j_z = \pm 3/2$; the operator $\hat{S}_z = \sum_i \hat{S}_{i,z} \delta(\mathbf{R} - \mathbf{R}_i)$ is the spin operator associated with the Mn subsystem; $\hat{S}_{i,z}$ and \mathbf{R}_i are the single-impurity spin and position, respectively; i is the Mn-impurity index. In this study, we will model the in-plane hole motion by a parabolic potential and the vertical motion with a square double well,¹¹ i.e., $U_0(\mathbf{R})$

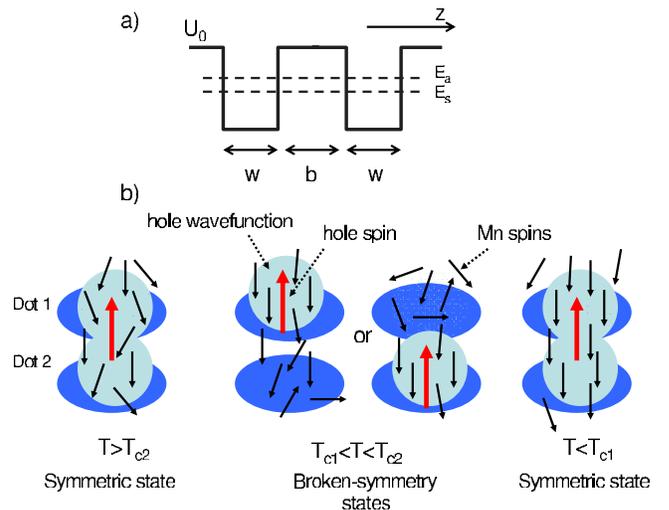


FIG. 1. (Color online) (a) The potential $u(z)$. (b) Schematics of the magnetic QD molecule and three magnetic-polaron phases with a single hole.

$=u(z)+m_{hh}\omega_0^2r^2/2$, where $u(z)$ is a double well potential in the z direction [Fig. 1(a)] and ω_0 is the in-plane frequency, $m_{hh}=m^*$ is the mass of heavy holes. In our model describing disklike self-assembled QDs, the light-hole states are assumed to be split from the lowest heavy-hole states. In the absence of Mn impurities, the hole wave function can be written as $\psi_J=\phi_j(z)\varphi_{n_x,n_y}(r)$, where $J=(j,n_x,n_y)$, $n_x(n_y)=0,1,2,\dots$, and $j=(s,a,k_z)$ label the discrete (s and a) and continuous states (k_z) related to the z motion. For the QD parameters chosen in this paper, there are only two bound states, symmetric (s) and antisymmetric (a). The single-particle wave functions were calculated numerically and the size of a hole wave function is comparable with the size of the QD pair. The exchange interaction between the hole and Mn impurities may lead to additional localization in one of the dots (i.e., to the broken-symmetry phase).

Within the mean field theory, the Hamiltonian becomes

$$\bar{H}_{hh}=\frac{\hat{\mathbf{P}}^2}{2m^*}+U_0(\mathbf{R})-\frac{\beta}{3}x_{\text{Mn}}N_0\hat{j}_z\cdot\bar{S}_z, \quad (2)$$

where x_{Mn} is the reduced Mn concentration, N_0 is the number of cations per unit volume, and $\bar{S}_z(\mathbf{R})$ is the local average Mn spin

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

where $S=5/2$, B_S is the Brillouin function, T_0 describes the effect of antiferromagnetic interaction between Mn impurities, $\bar{j}_z(\mathbf{R})=\langle\psi(\mathbf{R}')|\hat{j}_z\delta(\mathbf{R}-\mathbf{R}')|\psi(\mathbf{R}')\rangle$, where ψ is the hole wave function.

In the spirit of mean field theory, the effective spin-dependent potential for a hole is

$$\bar{U}=U_0+\delta U, \quad \delta U(\mathbf{R})=-\frac{\beta}{3}x_{\text{Mn}}(\mathbf{R})N_0\hat{j}_z\cdot\bar{S}_z. \quad (4)$$

The corresponding ground state wave function has the form $\psi=\psi_G|\uparrow\rangle$, where $|\uparrow\rangle$ is the hole state with $j_z=+3/2$. The spatial wave function ψ_G should be now determined. To the lowest order perturbation theory (PT),¹³ the ground-state wave function can be written as $\psi_G^0=\phi_G(z)\varphi_{0,0}(r)$, with

$$\phi_G=a\phi_s+\sqrt{1-a^2}\phi_a, \quad (5)$$

where ϕ_s and ϕ_a are the bound states in the double well. Then, the real parameter a should be determined by the variational method. From Eqs. (3), (4), and $\bar{j}_z(\mathbf{R})=\frac{3}{2}\psi_G^2(\mathbf{R})$ we can obtain the polaron binding energy and the total energy

$$E_b(T)=-\frac{\beta}{2}x_{\text{Mn}}N_0S\int d^3R\left[(\psi_G^0)^2B_S\left(\frac{\beta/3\bar{j}_z(\mathbf{R})S}{k_B(T+T_0)}\right)\right],$$

$$E_{\text{tot}}^1(a)=a^2E_s+(1-a^2)E_a+E_b. \quad (6)$$

To solve the problem at nonzero temperatures, we should minimize the free energy functional¹⁴

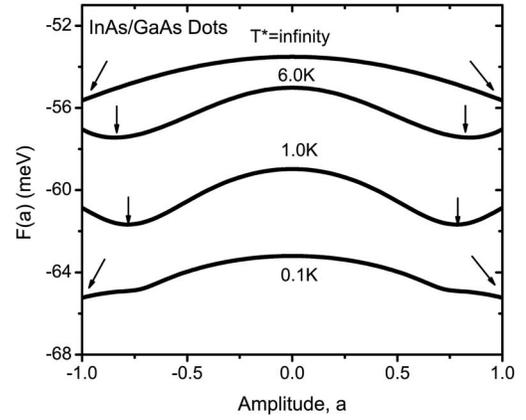


FIG. 2. The total energy of magnetic polaron as a function of the wave function parameter a at different T^* . The arrows show the ground states of the system.

$$V_b(T)=-\frac{\beta}{2}x_{\text{Mn}}N_0S\int d^3R\left[(\psi_G^0)^2F_S\left(\frac{\beta/3\bar{j}_z(\mathbf{R})S}{k_B(T+T_0)}\right)\right],$$

$$F^1(a)=a^2E_s+(1-a^2)E_a+V_b, \quad (7)$$

where the appropriate exchange contribution $F_S(u)=\ln\{\sinh[(2S+1)u/2S]/(2S+1)\sinh(u/2S)\}/u$, $u=\beta\psi_G^2(\mathbf{R})S/2k_B(T+T_0)$; the function $F_S(u)$ can be expressed through the Brillouin function $B_S(u)=d[uF_S(u)]/du$.^{14,15} The general variational wave function can be written as $\psi_G=\phi_G(z)\varphi_{0,0}(\mathbf{r})+\sum_J a_J\psi_J$, where the second term will be considered as perturbation. The zero-order degenerate PT provides us with two orthogonal wave functions ψ_G^0 and ψ_E^0 .¹³ The corresponding first-order PT contribution to the free energy is given by Eq. (7). Then, the second-order correction to the free energy becomes

$$\delta F=\sum_{\Gamma}\frac{\left|\delta V_{G,\Gamma}(\psi_G^0)+\frac{1}{2}\left(\frac{\partial\delta V(\psi_G^0)}{\partial a_{\Gamma}}\right)_{G,G}\right|^2}{E_{\text{tot}}^1-E_{\Gamma}}, \quad (8)$$

where $\delta V_{A,B}=-\frac{\beta}{2}x_{\text{Mn}}N_0S\int d^3R[(\psi_A^*\psi_B)F_S(u)]$, $\{A,B\}=\{\Gamma,G\}$, $\Gamma=\{\gamma,n_x,n_y\}$, $\gamma=(G,E,k_z)$, $n_x,n_y=\{0,1,2,\dots\}$, $\Gamma\neq\{G,0,0\},\{E,0,0\}$; here E_{Γ}^1 and ψ_G^0 describe the ground state calculated within the lowest-order PT and k_z labels the delocalized states for the z motion.

The approach to solve the nonlinear Schrödinger equation (2) at finite temperature is to minimize the free-energy functional $F(a)$. At low temperatures ($T\ll T_0$), we also can try energy functional $E_{\text{tot}}(a)$. In our approach, $F(a)\neq E_{\text{tot}}(a)$ even at $T\rightarrow 0$. Therefore, the energy functional will give lower ground-state at $T=0$ (or at $T\ll T_0$). In the following, we use $F(a)$ for arbitrary T and $E_{\text{tot}}(a)$ for low $T\ll T_0$.

Transition as a function of temperature. To find the variational ground state, we calculate the total free energy $F_{\text{tot}}(a)=F^1(a)$ as a function of a at various temperatures (Fig. 2). The quantity a is the hole amplitude related to the symmetric state. The material parameters are the following: $\beta N_0=-1.8$ eV, $N_0=23$ nm⁻³, $l_0=\sqrt{\hbar/m^*}\omega_0=2.5$ nm, $b=4$ nm, $w=3$ nm, $U_0=90$ meV, $m=0.38m_0$, $x_{\text{Mn}}=0.005$, and

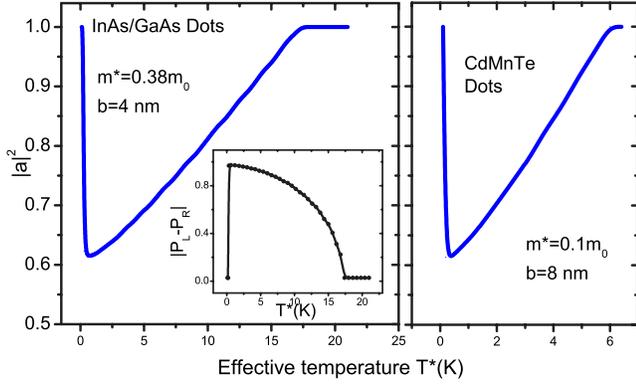


FIG. 3. (Color online) The wave function factor a^2 as a function of the effective temperature; the function $a^2(T^*)$ shows the appearance of broken-symmetry phase ($a^2 < 1$). Inset: The effective order parameter (degree of asymmetry) as a function of T^* .

$R_{\text{Mn}} = 15.0$ nm. This parameter set represents a InAs/GaAs QD doped with Mn impurities. One can clearly see three different phases. For high T , the ground state corresponds to $a^2 = 1$, i.e., the ground state is symmetric. With decreasing T , the ground changes from $a^2 = 1$ to almost $a^2 = 0.5$. It is a state of hole staying mostly in one dot. Thus, the symmetry of the system becomes broken. For very low T , the symmetry is recovered.

The probability a^2 (Fig. 3) as a function of the effective temperature $T^* = T + T_0$ clearly shows the existence of three phases. We may consider the difference of hole probabilities of being inside the dots $\delta = |P_{\text{upper}} - P_{\text{lower}}| = 2a\sqrt{1-a^2}$ as an order parameter (as shown in the inset of Fig. 3). The order parameter δ is nonzero for the broken-symmetry phase at intermediate T^* and is zero for the symmetric state at high and low T^* .

Basically, the physical picture is the following: the interaction between the hole and Mn impurities leads to the ferromagnetic ordering of Mn-spins inside the double QD. The resulting polaron state tends to be as localized as possible because the magnetic binding energy increases with decreasing the localization strength of hole. The latter can be seen from Eq. (6): the Brillouin function strongly increases with increasing the hole wave function amplitude ψ^2 . Therefore, in our system there is a competition between the kinetic energy (tunnel splitting) and the magnetic energy. At high T , due to the thermal fluctuations, the interaction between the hole and Mn impurities is weak and the kinetic energy wins. In this case, a symmetric ground state is realized. With decreasing T , the Mn-hole interaction becomes stronger and, below some critical temperature T_{c2} , the magnetic localization effect overcomes the tunneling. The hole is now trapped in one of the dots since such a spatial configuration lowers the total energy. However, for very low T^* , the hole is able to polarize the Mn spins inside the QD pair much stronger and the magnetic potential δU becomes almost flat. Therefore, the symmetric orbital state is recovered. There is an obvious condition for the existence of the low- T phase: $T_{c1}^* > T_0$ ($T_{c1} > 0$). In other words, for the existence of the low- T phase, the Mn-hole interaction must be strong enough to overcome the antiferromagnetic interaction. Figure 1(b) shows schematically the three phases.

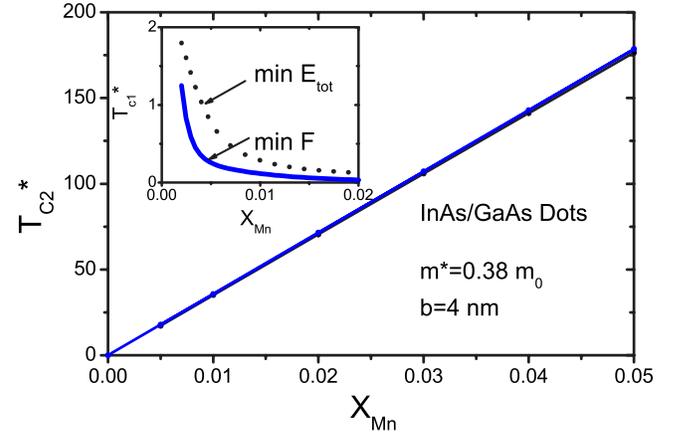


FIG. 4. (Color online) Temperatures T_{c2}^* and T_{c1}^* as functions of the Mn concentration. The temperature T_{c1}^* was calculated by two methods [$F(a) = \min$ and $E_{\text{tot}}(a) = \min$].

We should point out that the symmetric polaron at low T has a much stronger Mn spin polarization than the symmetric state at high T . The temperature T_{c2}^* corresponds to a smooth, type-II phase transition, while the low- T transition is sharp and can be classified as a type-I phase transition. The low- T transition occurs as a jump between the broken-symmetry phase ($a \approx \pm 1/\sqrt{2}$) and the symmetric state ($|a| = 1$).

The temperature T_{c2}^* can be found analytically by expanding the total energy in the vicinity of the type-II phase transition

$$k_B T_{c2}^* = \frac{\beta^2 N_0 S(S+1)}{12(E_a - E_s)} \int d^3 R x_{\text{Mn}} (3\phi_s^2 \phi_a^2 - \phi_s^4). \quad (9)$$

Figure 4 shows numerically calculated $T_{c2}^*(x_{\text{Mn}})$ (dots) and the data obtained from Eq. (9) (line). Our numerical and analytical results agree well. The temperature $T_{c2}^* \propto x_{\text{Mn}}$. This linear dependence may be understood qualitatively. For a higher Mn concentration, stronger thermal fluctuations (higher T) are needed to destroy the broken-symmetry polaron phase. In contrast to T_{c2}^* , the temperature T_{c1}^* decreases with x_{Mn} (approximately as $1/x_{\text{Mn}}$) (Fig. 4). Qualitatively, such a dependence can be explained as follows. The low- T symmetric phase appears when the Mn spins are strongly polarized. Therefore, a lower T_{c1}^* is needed to polarize a larger number of Mn spins. Figure 4 shows the results for both methods [$E_{\text{tot}}(a) = \min$ and $F(a) = \min$]. The results obtained from both methods are not very different. However, T_{c1}^* obtained from the equation $E_{\text{tot}}(a) = \min$ is somewhat higher. It may be that the energy minimum principle at low T gives a better approximation (since this method does not involve the nonlinear Schrödinger equation). Simultaneously, the results obtained from $E_{\text{tot}}(a) = \min$ are only valid in the low- T limit ($T \ll T^*$ or $T \ll T_0$).

As discussed above, the transition temperature T_{c2}^* depends on the competition between the kinetic and magnetic energies and qualitatively corresponds to the condition $|E_a - E_s| \sim \delta U$. T_{c2}^* depends on a material. In Fig. 3 we show the results for a CdMnTe QD. For the parameters, we use $\beta N_0 = -1.3$ eV, $N_0 = 15$ nm⁻³, $l_0 = 4$ nm, $b = 8$ nm, $w = 5$ nm,

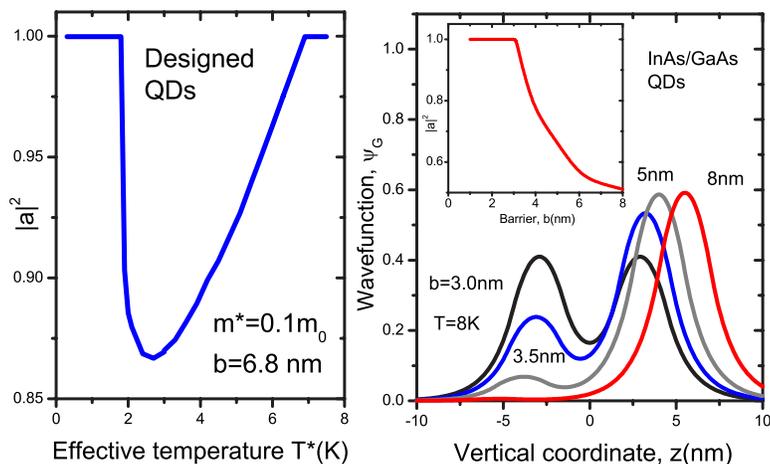


FIG. 5. (Color online) Left: Transitions in a specially designed magnetic QD pair with a larger T_{c1}^* . Right: The ground-state wave function for different barrier widths for the Mn-doped InAs/GaAs QD pair. Inset: Transition as a function of the barrier width.

$U_0 = 150$ meV, $m = 0.1m_0$, $x_{\text{Mn}} = 0.005$, and $R_{\text{Mn}} = 15.0$ nm. The effective transition temperature is 14 K and larger than the typical antiferromagnetic CdMnTe temperature $T_0 = 3.6$ K.

One may notice that the transition temperature T_{c1}^* can be very small (Fig. 4, inset). It may be impossible to observe the third phase ($T_{c1}^* < T_0$), if some antiferromagnetic interactions are present. However, due to the tunability of QD systems, we may design a system with a relatively high T_{c1}^* . Figure 5 shows the transitions in a designed system with $T_{c1}^* \sim 2$ K. For the parameters, we took $\beta N_0 = -2.5$ eV, $N_0 = 10$ nm $^{-3}$, $l_0 = 4$ nm, $B = 6.8$ nm, $w = 5$ nm, $U_0 = 150$ meV, $m = 0.1m_0$, $x_{\text{Mn}} = 0.005$, and $R_{\text{Mn}} = 2.0$ nm.

In recent experiments,¹² self-assembled double QDs were built into transistor structures with top and back contacts. By using applied voltage in these structures one can change the number of trapped carriers and also modify their wave functions. Two stable magnetic states in our system [Fig. 1(b)] can be considered as a bit of classical information. In these states, the hole mostly resides either in the upper dot or in the lower dot. Using the gate voltage, our system can be prepared in the desired state. The following electrical readout of the polaron state can be done, for example, using capacitance spectroscopy.⁵ Note that this paper considers a QD pair with a single carrier. We expect that a double QD with few holes may also demonstrate some of the effects predicted here. In particular, for a QD pair with few holes, there will be additional effects coming from the Coulomb interaction. In addition, in this study, there are only two bound states (s and a) in a QD pair. For deeper QDs, there may be more bound states. However, we expect the same behavior for the magnetic QD pair (transitions). Finally, we also should mention about likely asymmetry of a QD pair in experiments. Individual self-assembled dots in a QD pair are never the same. However, the QD asymmetry parameter ($|P_{\text{upper}} - P_{\text{lower}}|$) can be tuned to zero using the voltage applied between the top and back electrons.¹² First, by adjusting the voltage, one can tune the QD pair in the symmetric state ($|P_{\text{upper}} - P_{\text{lower}}| = 0$) at $T^* > T_{c2}^*$. Then, by lowering the temperature, one can reach the predicted state with broken symmetry.

It is interesting to compare the transitions in our system

with those described in Ref. 16 for the case of spatially non-uniform magnetic medium. Spontaneous symmetry breaking in Ref. 16 comes from shift of the particle wave function from a nonmagnetic layer toward a magnetic barrier. In our case, the magnetic medium is uniform and the transitions originate from a competition between the tunneling and magnetic interaction. We also note that the paper¹⁶ does not describe the low-temperature transition (T_{c1}^*). Another type of bistability was predicted for an exciton in a multiwell structure;¹⁷ this bistability may appear due to the competition between the Coulomb interaction and nanostructure potential.

Quantum phase transition. We may look at the physics from another point of view. The kinetic energy of the hole, i.e., the hopping between the dots, is related to the width of the barrier between QDs and thus can be tuned by appropriate growth process. In Fig. 5, we show the ground-state wave functions for various widths of the barrier. With increasing of the width, the ground states become more asymmetric. It is clear that there is a quantum phase transition with the increasing of barrier width. The critical width is around 3 nm (see inset of Fig. 5).

Second order corrections to energy. All the previous discussions are based on the perturbation theory applied to the nonlinear Schrödinger equation. The second-order correction to the energy can be calculated by Eq. (8). For the sets of parameters used above, it is of a few percent of the first-order result. We also found numerically that, in the vicinity of transition temperatures T_{c1}^* and T_{c2}^* , the second-order corrections are even smaller.¹⁸ This allows us to give the results for T_{c1}^* and T_{c2}^* in a wider range of x_{Mn} (see Fig. 4).

In conclusion, we have studied a magnetic QD molecule with a single hole. With decreasing temperature, the magnetic polaron ground state undergoes two transitions. At a higher temperature, the normal symmetric state turns into the polaron state with broken symmetry. At lower temperatures, a symmetric polaron state can be recovered. Our results suggest that a magnetic QD molecule can be used as a nanoscale magnetic memory cell controlled electrically.

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- ¹⁸For $T \sim T_{c2}$, the magnetic potential δU is small because of relatively high temperature. For the temperature interval $T \sim T_{c1}$, the matrix elements in Eq. (6) becomes small due to the orthogonality of wave functions.