Ferromagnetism Mediated by Few Electrons in a Semimagnetic Quantum Dot

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A (II,Mn)VI diluted magnetic semiconductor quantum dot with an integer number of electrons controlled with a gate voltage is considered. We show that a single electron is able to induce a collective spontaneous magnetization of the Mn spins, overcoming the short range antiferromagnetic interactions, at a temperature order of 1 K, 2 orders of magnitude above the ordering temperature in bulk. The magnetic behavior of the dot depends dramatically on the parity of the number of electrons in the dot.

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Introduction.—The range, strength, and sign of exchange interactions between magnetic impurities in diluted magnetic semiconductors (DMS) depend on the density and nature of the states at the Fermi level. Mn doped semiconductors of the families (II,Mn)VI and (III,Mn)V order ferromagnetically in the presence of carriers that mediate indirect exchange interactions between Mn. In the case of (III,Mn)V compounds such as GaAsMn, Mn acts as an acceptor supplying holes responsible for the ferromagnetism below a transition temperature which depends on both Mn and hole densities [1] and can reach 160 K [2]. In contrast, Mn does not supply itinerant carriers in (II,Mn)VI compounds. These materials do not order ferromagnetically [3] unless further doping with acceptors provides holes which produce ferromagnetism below a carrier density dependent Curie temperature (T_C) of approximately 2 K [4].

Electrical control of the carrier density, in contrast with chemical doping, has been demonstrated in a number of DMS heterostructures, making it possible to alter reversibly properties of the material such as the T_C [5,6] and the coercive field [7] of these systems. The fact that the carrier density is much higher than the relative change achieved artificially sets limits to the control. In contrast, the number of electrons in a GaAs quantum dot can be varied one by one, starting from zero, in single electron transistors [8]. Single electron transistors with nonmagnetic II-VI (CdSe) quantum dots of lateral size smaller than 10 nm and energy level spacings of tens of meV have also been fabricated [9]. (II,Mn)VI quantum dots of similar size have also been grown and studied magneto-optically by several groups [10]. Therefore, the fabrication of a single electron transistor with (II,Mn)VI quantum dots seems feasible and motivates this work.

We study the magnetic properties of a (II,Mn)VI quantum dot with an integer number of electrons. The lateral dimensions of the dot are smaller than 10 nm and, for the range of Mn concentration considered, $x < 0.05$, the number of Mn atoms in the dot is of the order of 100 and the number of electrically injected electrons ranges from zero to 10. We find compelling theoretical and numerical evidence that the addition of a single electron to an otherwise paramagnetic DMS dot is enough to couple most of the Mn spins so that a net total magnetic moment emerges spontaneously. Therefore, a single electron transistor with a DMS quantum dot would switch on and off the total magnetic moment *completely* in a controlled and reversible manner.

Hamiltonian.—The Hamiltonian describing the system is the zero dimensional version of the standard exchange Hamiltonian used for both bulk [3,11] and two dimensional systems [12]. For simplicity we consider only conduction band electrons, for which we can ignore spin orbit interaction, and we ignore Coulomb repulsion. The latter tends to enhance carrier mediated exchange interactions so that our results do not change qualitatively if this approximation is abandoned. Conduction band electrons (creation operator $c^\dagger_{n,\sigma}$) occupy confined levels ϵ^0_n of the dot. The Mn spins are described with $S = 5/2$ operators $\vec{M}_I$. The spin of the quantum dot electrons and the Mn interact via a zero range exchange interaction. The Mn spins interact also with each other via a short range antiferromagnetic superexchange interaction [3] $J_{I,I'}$, which competes with the ferromagnetic carrier mediated coupling. The Hamiltonian reads

$$\mathcal{H} = \sum_{n,\sigma} \epsilon^0_n c^\dagger_{n,\sigma} c_{n,\sigma} - J_c \sum_I \vec{M}_I \cdot \vec{S}_e(\vec{x}_I) + \mathcal{H}_{\mathrm{AF}}, \quad (1)$$

where $\mathcal{H}_{\mathrm{AF}} = \frac{1}{2}\sum_{I,I'} J_{II'} \vec{M}_I \cdot \vec{M}_{I'}$,

$$\vec{S}(\vec{x}_I) \equiv \sum_{\sigma,\sigma',n,n'} \phi^*_n(\vec{x}_I)\phi_{n'}(\vec{x}_I)\frac{1}{2}\vec{\tau}_{\sigma,\sigma'} c^\dagger_{n,\sigma} c_{n',\sigma'} \quad (2)$$

is the quantum dot electron local spin density, and $\phi_n(\vec{x}_I)$ is the nth orbital wave functions of the dot. Exchange interaction produces transitions between different levels of the dot. For the dots considered here, the typical interlevel spacing δ of the order of tens of meV [9], except for the degeneracies that some of the dots might have. The exchange energy of a quantum dot electron with a single Mn is approximately given by $j \equiv J_c/\Omega_D$, where Ω_D is the quantum dot volume. For $Cd_{1-x}Mn_xTe$ quantum dots

with 10 nm of lateral dimension, we have $j \simeq 1.5 \times 10^{-2}$ meV, much smaller than the level spacing.

Effective spin Hamiltonian.—In the following we derive analytically the effective interaction between the Mn spins, which are treated classically. The canonical ensemble equilibrium partition function for the dot with a fixed number of electrons N_e is given by

$$\mathcal{Z}_N = \int d\vec{M}_1 \cdots d\vec{M}_N e^{-\beta \mathcal{H}_{\rm AF}} \sum_\alpha e^{-\beta E_{\alpha,N}(\vec{M}_I)}, \quad (3)$$

where $\beta = 1/k_B T$ and $E_{\alpha,N_e}(\vec{M}_I)$ is the energy of the Slater determinant labeled with α, for a given Mn spin configuration, $(\vec{M}_I)$. For $k_B T \ll \delta$ we can safely neglect in (3) all the higher energy configurations α, except the ground state Slater, whose energy we denote with $\mathcal{E}_N(\vec{M}_I)$. In this approximation we have $\mathcal{Z}_N = \int d\vec{M}_1 \cdots d\vec{M}_N e^{-\beta \mathcal{H}_{\rm eff}}$ where the effective Mn-Mn coupling is

$$\mathcal{H}_{\rm eff} = \mathcal{H}_{\rm AF} + \mathcal{E}_N(\vec{M}_I). \quad (4)$$

Let $\lambda_l(\vec{M}_I)$ be the eigenstates of the Hamiltonian matrix

$$\mathcal{H}_{n\sigma,n'\sigma'} = \epsilon_n^0 - \frac{J_c}{2} \sum_I \vec{M}_I \cdot \vec{\tau}_{\sigma,\sigma'} \phi_n^*(\vec{x}_I) \phi_{n'}(\vec{x}_I) \quad (5)$$

associated with (1) for a given configuration of classical spins, $\vec{M}_I$. We have $\mathcal{E}_N(\vec{M}_I) = \sum_{l=1,N} \lambda_l(\vec{M}_I)$. Using the fact that δ is the largest energy scale, we treat the $n \neq n'$ terms as a perturbation and diagonalize each of the n intralevel 2×2 boxes. To linear order in J_c we have $\lambda_{n,\pm} \simeq \epsilon_n^0 \pm |\frac{J_c}{2} \sum_I |\phi_n(\vec{x}_I)|^2 \vec{M}_I| + \mathcal{O}(\frac{(J_c c_{\rm Mn})^2}{\delta})$. For an odd number of electrons, $N_e = 2N + 1$, all the contributions linear in J_c coming from the first $2N$ electrons vanish identically and the only contribution comes from the most external electron. Modulo an irrelevant constant, the ground state electronic energy for a dot with $2N + 1$ electrons is $\mathcal{E}_{2N+1} \simeq \lambda_{N-}$ or

$$\mathcal{E}_o = -\frac{|J_c|}{2} \sqrt{\sum_{I,I'} |\phi_{N+1}(\vec{x}_I)|^2 |\phi_{N+1}(\vec{x}_{I'})|^2 \vec{M}_I \cdot \vec{M}_{I'}}. \quad (6)$$

Equations (4) and (6) define the carrier mediated Mn-Mn coupling which is one of the important results of this Letter. The Mn-Mn interaction mediated by an odd number of electrons, including a single electron, always favors ferromagnetic couplings and it scales with J_c. These features are in contrast with the standard bulk RKKY coupling, which scales with J_c^2, and it can be either positive or negative. For these reasons the effective coupling (6) is stronger than its bulk counterpart.

For an even number of electrons all the contributions linear in J_c cancel. The leading order contribution to $\mathcal{E}_e$ comes from interlevel exchange coupling. We calculate $\mathcal{E}_e$ doing perturbation theory around $J_c = 0$. The magnetic part of the ground state electronic so obtained reads

$$\mathcal{E}_e = \sum_{I,I'} \left[\frac{J_c^2}{2} \sum_{n,n'} \gamma_{n,n'}(I, I') \frac{f_n - f_{n'}}{\epsilon_n^0 - \epsilon_{n'}^0} \right] \vec{M}_I \cdot \vec{M}_{I'}, \quad (7)$$

where $\gamma_{n,n'}(I, I') \equiv \phi_n^*(\vec{x}_I) \phi_{n'}(\vec{x}_I) \phi_n^*(\vec{x}_{I'}) \phi_{n'}(\vec{x}_I')$ and $f_n = 0, 1$ are the occupation of the unperturbed dot in the ground state electronic configuration. The effective couplings (7) are weaker than (6) and can be both positive and negative (ferromagnetic or antiferromagnetic) for a given dot and different Mn couples. The striking differences between effective interactions (6) and (7) permit us to anticipate very different behavior for dots with open and closed shell electronic structure.

Local mean field theory.—Effective interactions (6) and (7) between the spins result from integrating out the conduction electrons in some limits and treating the Mn spins classically. We now do a mean field theory for Hamiltonian (1), keeping track of both Mn and electrons. Quantum dot electrons interact with an effective exchange field provided by the expectation value of the Mn spin operators. The latter is calculated assuming that each Mn spin interacts with an effective field provided by the quantum dot spin density *and* its neighboring Mn spins, via antiferromagnetic superexchange interaction. Since both the electron spin density and the Mn neighborhood are different for each Mn, each Mn has a different expectation value, in contrast with the homogeneous models used for bulk [3,11]. The local mean field felt by the Mn at $\vec{x}_I$ reads

$$g \mu_B \langle \vec{\mathcal{B}}_I \rangle = J_c \langle \vec{S}(\vec{x}_I) \rangle - \frac{1}{2} \sum_{I'} J_{II'} \langle \vec{M}_{I'} \rangle. \quad (8)$$

The expectation value for the magnetization of a spin $S = 5/2$ in the effective field of Eq. (8) reads

$$\langle \vec{M}_I \rangle = S \vec{u}_I B_S \left(\frac{S}{k_B T} g \mu_B \langle \vec{\mathcal{B}}_I \rangle \right), \quad (9)$$

where $\vec{u}_I$ is the unitary vector parallel to $\langle \vec{\mathcal{B}}_I \rangle$ and B_S is the Brillouin function [3]. Finally, the equation for the average conduction electron local spin density is

$$\langle \vec{S}(\vec{x}_I) \rangle = \frac{1}{Z_{el}} \sum_\alpha e^{-\beta E_\alpha} \sum_l \langle \Phi_l | \vec{S}(\vec{x}_I) | \Phi_l \rangle f_l(\alpha), \quad (10)$$

where $Z_{el} = \sum_\alpha e^{-\beta E_\alpha}$, $|\Phi_l\rangle$ is the eigenvector associated with the λ_l eigenvalue of Hamiltonian (5) with $\vec{M}_I$ replaced by $\langle \vec{M}_I \rangle$ given by Eq. (9), and $f_l(\alpha)$ is the occupation (0 or 1) level in the many electron configuration α.

In the following we show results for $Cd_{1-x}Mn_xTe$ hard wall (HW) cubic quantum dots of dimensions L_x, L_y, and L_z for which ϵ_n^0 and ϕ_n are known analytically. The effective mass of (Cd,Mn)Te is approximately [13] $m^* = 0.11 m_0$. In the HW model, the only role of m^* is to scale ϵ_n^0. In order to compensate for the overestimation of the energy level spacings of the HW model, we use $m^* = 0.3 m_0$ everywhere except in Fig. 1 (right). Unless stated

otherwise, we take [3] $J_c = 15$ eV Å^3. Superexchange antiferromagnetic coupling $J_{II'}$ decays exponentially with distance and we consider coupling only to first neighbors, $J_{\rm FN} = 0.5$ meV [3,14].

The positions of the Mn atoms are randomly chosen in the cation sites of the zinc blende lattice with lattice constant, $a = 6.4$ Å. The initial value of $\langle \vec{M}_I \rangle$ is also chosen randomly. A self-consistent numerical solution of Eqs. (8)–(10) yields converged values of $\langle \vec{M}_I \rangle$ which are stable, independent of all initial conditions, convergence procedure, and small variations of the energy scales of the problem. We characterize the collective magnetic order with the average Mn magnetization per Mn: $\langle M \rangle \equiv |\frac{1}{N}\sum_I \langle \vec{M}_I \rangle|$, which is bounded between 0 and $S = 5/2$. Different realizations of the Mn positional configurations, $\{\vec{r}_I\}$, for otherwise identical dots give different values of $\langle M \rangle$. To make sure that general results are independent of a particular Mn realization we perform averages over different realizations of $\{\vec{r}_I\}$. The net magnetization of a dot averaged over configurations is denoted by $\langle\langle M \rangle\rangle$. The mean standard deviation among different realizations of $\{\vec{r}_I\}$ is denoted by σ_M.

In Fig. 1 we plot $\langle\langle M \rangle\rangle$ and σ_M (vertical bars) as a function of N_e for two different dots. In Fig. 1 (left) we consider two different temperatures, $k_B T = 0.1$ and 2.0 K, whereas in Fig. 2 we consider three different values of the effective mass, $0.1m_0$, $0.2m_0$, and $0.3m_0$. Dots 1 and 2 (left and right panels) have the same Mn concentration and the same L_x ($x = 0.01$, $L_x = 4$ nm) but different dimensions. Dot 1 has $L_y = 6$ nm, $L_z = 7$ nm and dot 2 has $L_y = 8.5$ nm, $L_z = 9$ nm. The number of Mn impurities is 25 and 46, respectively. We find the following:

(i) A single electron is enough to induce a spontaneous collective magnetization different from zero in the absence of applied field. The collective magnetization survives at temperatures of 1 K and higher. The bulk mean field Curie temperature for parabolic bands yields, for $Cd_{0.99}Mn_{0.01}Te$, $k_B T_C|_{\rm bulk} = \frac{S(S+1)}{3}\frac{3}{2} c_{\rm Mn}\frac{n}{\epsilon_F} \simeq 20$ mK, where n and $c_{\rm Mn}$ are the electron and Mn density, respectively, corresponding to the dot of Fig. 1. Therefore, carrier mediated spin correlations in the quantum dots survive at temperatures 2 orders of magnitude larger than T_C in bulk.

(ii) The addition or the removal of a single electron produces a dramatic change in the magnetization of the dots.

(iii) The spontaneous magnetization is larger, in general, for open shell configurations (odd number of electrons) than for closed shell configurations (even number of electrons). Closed shell configurations with a small gap can yield larger interlevel couplings, as in the case of $N_e = 4$ in the right panel of Fig. 1. In the right panel of Fig. 1, we see how the relative change of $\langle\langle M \rangle\rangle$ as a function of m^* is much smaller for odd N_e than for even N_e. This reflects the intralevel origin of the effective interaction (6) and the interlevel origin of the effective interaction (7).

In Fig. 2 we analyze how the magnetization changes as a function of temperature and how these curves scale with the value of the exchange constant which we take $J_c = a \times 15$ eV Å^3, with $a = 1$ (solid symbols) and

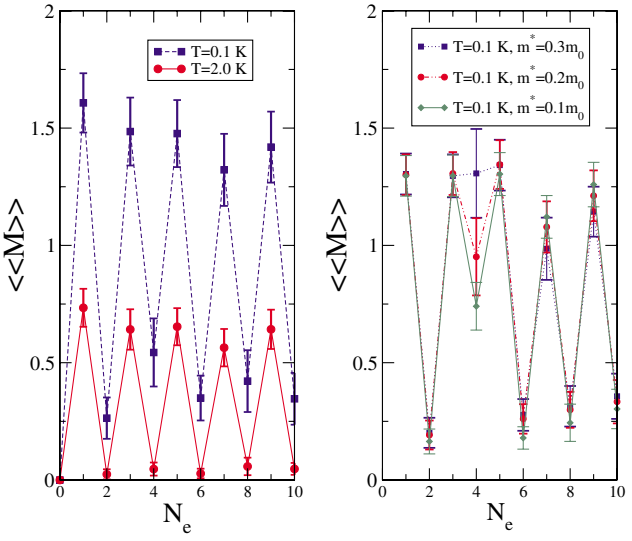

FIG. 1 (color online). Magnetization per Mn versus N_e, averaged over disorder configuration for two different dots.

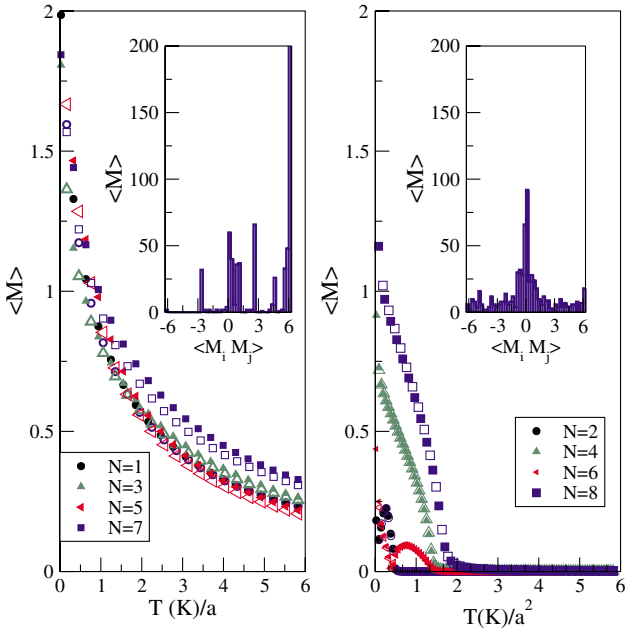

FIG. 2 (color online). Magnetization per Mn vs temperature for odd (left) and even (right) number of electrons for dot 1 (see the text). Exchange $J = a \times 15$ eV Å^3. Filled symbols: $a = 1$; empty symbols: $a = 0.5$. In the inset we show the histograms of $\langle \vec{M}_i \cdot \vec{M}_j \rangle$ at $k_b T = 50$ mK for $N = 1$ (left panel) and $N = 2$ (right panel).

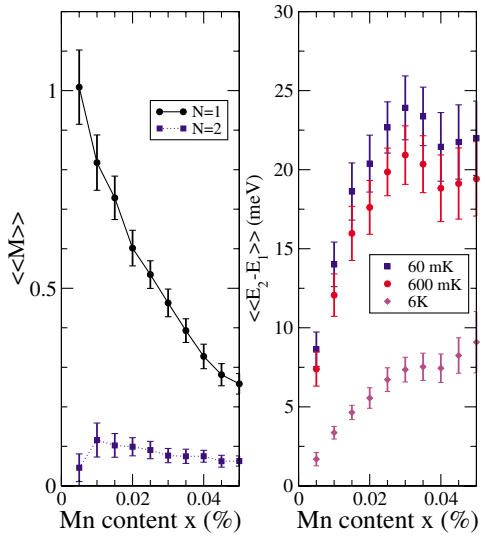

FIG. 3 (color online). Left panel: $\langle\langle M\rangle\rangle$ per Mn as a function of Mn content, x at $k_B T = 0.6$ K. Right panel: spontaneous spin splitting of the lowest level of the dot, at three temperatures, as a function of Mn content x.

$a = 0.5$ (open symbols). In the left [right] panel of Fig. 2 we show $M(T/a)$ [$M(T/a^2)$] for an odd [even] number of electrons in dot 1. The shapes of the curves for N_e odd are all very similar to each other and resemble the spontaneous magnetization of a confined exciton polaron [15]. Notice that, in agreement with Eq. (6), the curves scale with T/J_c. In the case of even N_e the $M(T)$ curves scale with T/J_c^2, in agreement with (7), and their shape varies from case to case, as a result of the complicated competition of carrier mediated ferromagnetic and antiferromagnetic couplings and superexchange. To quantify this, we plot in the insets histograms of the values of the correlation matrix $\langle \vec{M}_I \cdot \vec{M}_J \rangle$ for $N_e = 1$ (left) and $N_e = 2$ (right) at very low temperatures (50 mK). The $N_e = 1$ histogram is biased towards positive values (ferromagnetic couplings), whereas the $N_e = 2$ histogram displays a rather symmetric distribution of ferromagnetic and antiferromagnetic couplings.

The competition between carrier mediated and superexchange couplings evolves as the Mn concentration increases. In the left panel of Fig. 3 we show $\langle\langle M\rangle\rangle$ and σ_M (vertical bars) as a function of Mn concentration, x, for dot 2. We see that, for $N_e = 1$, $\langle\langle M\rangle\rangle$ decreases monotonically as a function of x as a result of the increase of the number of first neighbor pairs coupled antiferromagnetically. The curve for $N_e = 2$ has a maximum around $x = 0.01$ and decreases at higher concentrations.

The effects related to spontaneous magnetization can be observed provided that the collective magnetization is static during the time scale of the probe. A small source of spin anisotropy, like an applied field or Rashba spin orbit interaction, can be very efficient in slowing down the magnetization dynamics. The spontaneous magnetization results in a splitting of the quantum dot energy levels which could be measured in transport [8]. In the right panel of Fig. 3 we plot the energy splitting of the lowest quantum dot doublet as a function of x, for $N_e = 1$ at three different temperatures for dot 2. The splitting is a decreasing function of temperature and an increasing function of the Mn content up to $x = 0.03$, declining for higher x, due to the increase of first neighbors pairs.

In summary, our results indicate that a very large control of carrier mediated interactions can be achieved in quantum dots in which the number of electrons can be changed one by one. We claim that a few conduction band electrons couple the spin of several tens of Mn atoms in a (II,Mn)VI semiconductor quantum dot. An odd number of electrons, including just one, yields a ferromagnetic coupling [Eq. (6) and Fig. 1 and the left panel of Fig. 2)], whereas an even number of electrons give both ferromagnetic and antiferromagnetic carrier mediated couplings [Eq. (7) and Fig. 1 and the right panel of Fig. 2]. The result of the competition between carrier mediated interactions and short range antiferromagnetic superexchange is a spontaneous collective magnetization which survives at temperatures of the order of 1 K, 2 orders of magnitude higher than the mean field prediction for n-doped bulk material.

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