Tailoring Magnetism in Quantum Dots

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We study magnetism in magnetically doped quantum dots as a function of confining potential, particle numbers, temperature, and strength of Coulomb interactions. We explore possibility of tailoring magnetism by controlling the electron-electron Coulomb interaction, without changing the number of particles. The interplay of strong Coulomb interactions and quantum confinement leads to enhanced inhomogeneous magnetization which persist at higher temperatures than in the non-interacting case. The temperature of the onset of magnetization can be controlled by changing the number of particles as well as by modifying the quantum confinement and the strength of Coulomb interactions. We predict a series of electronic spin transitions which arise from the competition between the many-body gap and magnetic thermal fluctuations.

Magnetic doping of semiconductor quantum dots (QDs) provides an interesting interplay of interaction effects in confined geometries and potential spintronic applications. In the bulk-like dilute magnetic semiconductors the carrier-mediated ferromagnetism can be photoduced and electrically controlled by gate electrodes, suggesting possible nonvolatile devices with tunable optical, electrical, and magnetic properties. QDs allow for a versatile control of the number of carriers, spin, and the effects of quantum confinement which can have important ramifications. An extra carrier can both strongly change the total carrier spin and the temperature of the onset of magnetization which we show can be further controlled by modifying the quantum confinement and the strength of Coulomb interactions.

We study the magnetic ordering of carrier spin and magnetic impurities in (II,Mn)VI QDs identified as a versatile system to demonstrate interplay of quantum confinement and magnetism. Because Mn is isoelectronic with group-II elements it does not interact with the strain and the strength of Coulomb interactions. We reveal that the interplay of strong Coulomb interactions and quantum confinement leads to enhanced inhomogeneous magnetization which persist at higher temperatures than in the non-interacting case and the bulk structures. We refer to such a spin-polarized state in QD at zero applied magnetic field as “ferromagnetic” state.

Here we focus on magnetic QD in zero applied magnetic field described by the Hamiltonian $H = H_e + H_m + H_{ex}$, with the electron contribution $H_e = \sum_i \left[-\frac{\hbar^2}{2m^*} \nabla_i^2 + U_{QD}(\mathbf{r}_i)\right] + \frac{2}{\epsilon} \sum_{i \neq j} \frac{\mathbf{s}_i^2}{|\mathbf{r}_i - \mathbf{r}_j|}$, where $\hbar$ is the Planck constant, $m^*$ is the effective electron mass, and $U_{QD}(\mathbf{r})$ is the confining potential of a three-dimensional QD. The last term in the equation is the repulsive electron-electron (e-e) Coulomb interaction screened by the dielectric constant $\epsilon$, $-e$ is electron charge, and $\gamma$ accounts for reduction of Coulomb strength due to screening effects of the gate electrodes.

The Hamiltonian is $H_m = \sum_{I,I'} J_{I,I'}^{AF} \tilde{M}_I \cdot \tilde{M}_{I'}$, where $J_{I,I'}^{AF}$ is the direct Mn-Mn antiferromagnetic coupling. The z-component of $\tilde{M}_I$ of impurity spin satisfies $M_z = -M, -M+1, \ldots, M$, where we choose $\hat{z}$ as the quantization axis and $M = 5/2$ for Mn. The electron-Mn (e-Mn) exchange Hamiltonian is $H_{ex} = -J_{sd} \sum_{i,I} \mathbf{\hat{s}}_i \cdot \tilde{M}_I \delta(\mathbf{r}_i - \mathbf{R}_I)$, where $J_{sd}$ is the exchange coupling between electron spin $\mathbf{\hat{s}}_i$, at $\mathbf{r}_i = (\mathbf{\hat{r}}_i, z_i)$, and impurity spin $\tilde{M}_I$, at $\mathbf{R}_I$. An effective mean field Hamiltonian describing electrons can be obtained by replacing the Mn spins, that are randomly distributed, with a classical continuous field $H_{ef} = H_e - J_{sd} \sum_{i,I} n_m \frac{\gamma}{2} M_z(\mathbf{r}_i)$, where $n_m$ is the averaged density of Mn, and $\sigma = \pm 1$ for spin up $(\uparrow)$, and down $(\downarrow)$. The effective magnetic field seen by the electrons is the mean field induced by Mn. Assuming that impurities are in equilibrium with thermal bath it follows $M_z(\mathbf{r}_i) = MB_{BZ}(mb(\mathbf{r}_i)/kB T)$. The Brillouin function $\gamma \equiv MB_{BZ}(mb(\mathbf{r}_i)/kB T)$ is the Brillouin function. Here $b(\mathbf{r}_i) = -Z_{M_n} J_{app}(M_z(\mathbf{r}_i)) + J_{sd}[n_\uparrow(\mathbf{r}_i) - n_\downarrow(\mathbf{r}_i)]/2$.
is the effective field seen by the Mn. The first term in $b(r)$ describes the mean field of the direct Mn-Mn antiferromagnetic coupling. $Z_{Mn}$ is the averaged Mn coordination number, and $n_{\sigma}(r)$ is spin-resolved electron density. We decompose the planar and perpendicular components of the confining potential of a single QD, and fit it to a realistic QD potential. The resulting potential, $U_{QD}$, is a sum of a two-dimensional (2D) Gaussian $V_{QD} = V_0 \exp(-\rho^2/\Delta^2)$ and one-dimensional parabolic potential $V_{QD}^2 = m^*\Omega^2z^2/2$, where $\rho \equiv (x,y)$. For $V_{QD}$ we find that Gaussian potential is more realistic than usually studied parabolic potential. Here $V_0$ and $\Omega$ are the planar depth of the QD minimum, and the characteristic subband energy associated with the perpendicular confinement. In typical disk-shaped QDs, and low density of electrons, only the first subband is filled. After expanding the QD wave functions in terms of its planar $\psi_{\sigma}(\rho)$ and subband wave function $\xi(z)$, we project $H_{QD}$ into a two-dimensional Hamiltonian by integrating out $\xi(z)$. In LSDA the two-body Coulomb interaction can be written as sum of Hartree potential $V_H$ and spin dependent exchange-correlation potential $V_{XC}$. We use Vosko-Wilk-Nusair exchange-correlation functional, and express the Kohn-Sham Hamiltonian as

$$H_{KS} = -\hbar^2/2m^* \nabla^2 + V_{QD} + \gamma V_H + \gamma V_{XC} - \hbar^2/2 h_{sd}(\rho),$$

where

$$h_{sd}(\rho) = J_{em} \int dz |\xi(z)|^2 B_M \left( \frac{M b(\rho,z)}{k_B T} \right),$$

and $J_{em} = J_{sd} n_m M$ is the e-Mn exchange coupling. The Kohn-Sham eigenvectors and eigenvalues of Eq. (1), $\psi_{\sigma}(\rho)$, and $\epsilon_{\sigma\alpha}$ are calculated numerically.

We illustrate our findings and the iterative solutions of Eq. (1) for (Cd,Mn)Te QD. The material parameters are $J_{sd} = 0.015$ eV nm$^3$, $m^* = 0.106$, $\epsilon = 10.6$, and we choose $Z_{Mn} J_{AF} = 0.02$ meV. The planar $(x,y)$, and perpendicular $(z)$, dimensions of the QD are taken as 42 nm and 1 nm with $n_m = 0$, 0.025, 0.1 nm$^{-3}$. In the central region of QD of area $4a_B^2$, where $a_B = 5.29$ nm is the effective Bohr radius in CdTe, $n_m = 0.1$ nm$^{-3}$ corresponds to $\approx 10$ Mn atoms. For a planar confinement, $V_{QD}$, we consider a Gaussian potential with $V_0 = -128$ meV, and $\Delta = 38.4$ meV, corresponding to $\omega_0 = 27$ meV. Here $\omega_0$ is calculated by expanding $V_{QD}$ in the vicinity of the minimum which yields $V_{QD} = V_0 + m^* \omega_0^2 \rho^2/2 + \ldots$, with the strength $\omega_0 = \sqrt{2|V_0|/m^*}/\Delta$.

In QDs electron density is inhomogeneous, implying that both the electron spin density, $n_{\uparrow}(\rho) - n_{\downarrow}(\rho)$, and Mn-magnetization density $(M_z(\rho))/J_{em}$ are inhomogeneous. For $N = 8$ and $n_m = 0.1$ nm$^{-3}$, we show the self-consistent spin density in Fig. (1a), and Mn-magnetization density in Fig. (1b). Outside the QD, $n_{\sigma}(\rho)$ decays exponentially, and an effective field $b(\rho)$ seen by Mn becomes negligible. This is consistent with vanishing $M_z(\rho)$ at the QD boundary [Fig. (1b)].

We next turn to spatially-averaged quantities such as Mn-magnetization per unit area $A$, $(M_z) = \frac{1}{A} \int d^2 \rho \langle M_z(\rho) \rangle$, electron (spin) polarization $P = (N_{\uparrow} - N_{\downarrow})/N$, and the z-component of the total spin of electrons, $s_z = (N_{\uparrow} - N_{\downarrow})/2$. In Fig. (2) we show $\langle M_z \rangle$ as a function of $N$ for $n_m = 0.025$ nm$^{-3}$, and both non-interacting ($\gamma = 0$), and interacting ($\gamma = 1$) electrons. The magnetic behavior of QD can be described based on the interplay of the many-body spectrum (determined by the shell structure of the confining potential, and e-e Coulomb interaction) and the strength of e-Mn exchange coupling $J_{em}$. In the following we summarize the spin structure of the QD in the absence and presence of $J_{em}$ with $\gamma = 0, 1$.

i) $J_{em} = 0$: The shell structure of the 2D Gaussian potential is shown in Fig. (2a). The energy gap between s-, p-, and d-orbitals is characterized by $\omega_0$. In contrast to 2D parabolic potential [1,2], d-shell levels are not completely degenerate, and therefore we focus on $N > 6$ states. Degenerate levels $d_+ \ldots$ and $d_-$ are separated by an energy gap (1.5 meV) from $d_0$-level, where $\pm 0$ refer to angular momentum $J = \pm 1,0$. However, e-e interaction changes the structure of d-shell as it over-turns the ordering of the d-orbitals, e.g., the Kohn-Sham energies of pair of degenerate $d_+$, and $d_-$ are below $d_0$ (with energy gap $\approx 1$ meV). Because of d-shell over-turning, $N = 10,12$
form closed shells with \( s_z = 0 \), and \( N = 7, 9, 11 \) form open shells with \( s_z = 1/2 \). The \( N = 8 \) corresponds to half-filled shell with \( s_z = 1 \), and electron polarization, \( P = 2/8 \). The evolution of \( s_z \) as a function of \( N \) for \( \gamma = 1 \) is shown in Fig. 2(b) (empty circles).

ii) \( J_{em} \neq 0 \): \( \gamma = 0 \) and increasing e-Mn coupling to \( J_{em} = 3.75 \text{ meV} \), leads to transitions \( P = 0/8 \rightarrow 2/8 \), and \( P = 1/9 \rightarrow 3/9 \) for \( N = 8 \), and 9, whereas \( N = 10 \) shows \( P = 2/10 \). For \( \gamma = 1 \), the dependence of \( P \) on \( J_{em} \) is negligible in \( s_- \) and \( p_- \) shells. In contrast, in \( d_- \)-shell, we find transitions \( P = 1/9 \rightarrow 3/9 \), and \( P = 0/10 \rightarrow 2/10 \) for \( N = 9 \) and \( N = 10 \) at low T. However, we find no change in \( P \) for \( N = 7, 8, 11 \), and 12. Figure 2(b) (filled circles) show \( s_z \) as a function of \( N \) for \( n_m = 0.025 \) nm\(^{-3} \) (\( J_{em} = 0.94 \text{ meV} \)). Increasing the density of Mn to \( n_m = 0.1 \) nm\(^{-3} \) (\( J_{em} = 3.75 \text{ meV} \)) does not change \( s_z \).

In Fig. 2 we observe that \( \langle M_z \rangle = 0 \) in closed shells \( (N = 2, 6, 12) \) for both \( \gamma = 0 \) and \( \gamma = 1 \), because of well separated \( s_-, p_-, \) and \( d_- \) orbitals due to large \( \omega_0 (\approx 30 J_{em}) \). Comparing \( \langle M_z \rangle \) between \( \gamma = 0 \) and \( \gamma = 1 \) one can observe that the e-e interaction stabilizes the ferromagnetic state due to the spin Hund’s rule. This condition is easily satisfied for open shells where the maximum electron polarization is obtained in half-filled shell with \( N = 4 \). The

\( N = 8 \) state (recall Fig. 1) is more interesting. At \( \gamma = 0 \) electrons fill single particle levels following the Pauli exclusion principle. Even with \( J_{em} = 0.94 \text{ meV} \) (smaller than single particle e-h excitation gap), \( N = 8 \) forms closed shell and \( P = \langle M_z \rangle = 0 \). In the case of \( \gamma = 1 \), and because of \( d_- \)-shell overturning, polarized electrons in \( d_+ \) and \( d_- \) give \( P = 2/8 \), and finite \( \langle M_z \rangle \). We also see that the maximum \( \langle M_z \rangle \) occurs at \( N = 9 \) because \( J_{em} = 0.94 \text{ meV} \) induces three polarized electrons in \( d_- \) levels. Figure 2(c) reveals the dependence of magnetic transitions on \( \gamma \), and \( n_m \). With increasing \( Z_{Mn} J^F \), the transition to ferromagnetic state occurs at larger \( \gamma \). Our findings clearly demonstrate that the magnetism induced by strong Coulomb interaction can be controlled by the electric gates or by changing the semiconductor host (and thus changing \( \epsilon \)) without changing the number of carriers confined in QD. We also suggest that because of the sensitivity of the \( \langle M_z \rangle \) to the electronic spin transitions, the former can be used to infer the spin of electrons, and could be potentially applied to manipulation of spin qubits in semiconductor nanostructures [4].

![FIG. 2: The averaged magnetization per unit area \( \langle M_z \rangle \) as a function of number of electrons \( N \) at \( T = 1 K \) and Mn-density \( n_m = 0.025 \) nm\(^{-3} \) for non-interacting (\( \gamma = 0 \), empty triangles) and interacting (\( \gamma = 1 \), filled triangles) electrons. The ground state of the QD switches between ferromagnetic and antiferromagnetic states as function of \( N \). Coulomb interaction changes the state of \( N = 8 \) from antiferromagnetic to ferromagnetic state. (a) The schematic single particle levels of a 2D Gaussian confining potential. (b) The \( z \)-component of the total spin of electrons, \( s_z \), as a function of \( N \) for \( \gamma = 1 \). (c) Antiferromagnetic-ferromagnetic transitions for \( N = 8 \) as function of \( n_m \) and \( \gamma \).](image)

![FIG. 3: Temperature evolution of Mn-magnetization per unit area \( \langle M_z \rangle \) (a), the electron polarization \( P \) (b), and the free energy difference \( \Delta F \) between ferromagnetic and antiferromagnetic QD (c). At low \( T \), \( N = 4, 8 \) form half-filled shells with \( P = 2/4, 2/8 \). \( T = T^* \), characterizes vanishing of \( \langle M_z \rangle \), \( P \), and \( \Delta F \).](image)

We next examine the temperature dependence of magnetism in QDs. In Fig. 3 we show \( \langle M_z \rangle \) (a), \( P \) (b), and the free energy difference \( \Delta F \) between ferromagnetic and antiferromagnetic states (c) for \( N = 4 \) and \( N = 8 \). The suppression of \( \langle M_z \rangle \), shown in Fig. 3(a) is accompanied by a series of spin transitions in electronic states and sup-
pression of $P$. At low $T$, the spin triplet is realized as the ground state of the $N = 4$, and $N = 8$ open $p$-, and $d$-shells (due to Hund’s rule). We define a characteristic temperature, $T^*$, at which $\langle M_z \rangle = P = \Delta F = 0$.

In Fig. 1 we plot $T^*(N)$ for $\omega_0 = 27$ meV and $\gamma = 1$ which decreases non-monotonically with $N$. The inset shows $T^*(\omega_0)$ for $N = 1$, and $N = 4$ ($\gamma = 0, 1$). At low $\omega_0$, the e-e interaction strongly enhances $T^*$, while at large $\omega_0$ the effect of confinement potential is dominant. Thus we find $T^*(\gamma = 1) \to T^*(\gamma = 0)$ with increasing $\omega_0$, which in turn gives rise to a peak in $T^*$. Several trends in calculated $T^*(N, \omega_0)$ can be obtained from a perturbative approach by approximating 2D Gaussian with 2D parabolic potential. Near $\langle M_z \rangle = P = 0$ for QD with one valence electron in $s$-, $p$-, or $d$-shells, we find $T^* = J_{em} \sqrt{\frac{M+1}{8\pi n_{萎}} \left[ \int d^3r |\psi_f(r)|^4 \right]^{1/2}}$, where $\psi_f$ is the wave-function of the highest occupied orbital, and $J^{AF} = 0$. For a given $\omega_0$, $T^*$ decreases with $N$, e.g., $T^*_N = 0.7T^*_{N-1}$ and $T^*_N = 0.6T^*_{N-1}$. One can also show that $T^* \propto \sqrt{\omega_0}$, consistent with bound magnetic polarons [23].

In conclusion we have investigated the existence of magnetism in magnetically doped QDs, as function of particle numbers, confining potential, temperature and strength of Coulomb interactions, using finite-temperature LSDA. Our results show that QDs embedded in magnetic semiconductor host can be considered as ferromagnetic centers which exhibit spatial ordering in spin density and magnetization, even at elevated temperatures where no such ordering exist in the host material [26]. In the limit of small $\omega_0$, where the Coulomb interaction among particles is the largest characteristic parameter of the QDs, we find magnetism substantially stronger than predicted from the non-interacting picture. In contrast to the carrier-controlled ferromagnetism in the bulk-like structures [10, 11, 12], we reveal that magnetism in QDs, can be tuned even at the fixed number of carriers by gate voltage which controls the inter-particle Coulomb interaction screening. For potential spintronic applications based on II-VI magnetic QDs, we anticipate that it is possible to further increase the magnetization and the temperature at which it vanishes. In addition to exploring a larger hole-Mn exchange coupling [22] in (II, Mn)VI QDs, it would also be advantageous to consider (II, Cr)VI QDs as there is a support for the room-temperature ferromagnetism in their bulk counterparts [27].

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