Invited Paper

Magnetism of magnetic ion doped semiconductor nanocrystals

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ABSTRACT

Magnetic ion-doped semiconductor nanocrystals (NCs) have recently drawn a great deal of interest because of the intriguing physical properties and the application potential in spintronics and magneto-electronics. In this work, we report on theoretical studies of magnetism in colloidal CdSe NCs doped with Mn^{2+} ions. Numerically, the exact diagonalization (ED) technique is employed to calculate the electronic structures and magnetizations of singly changed CdSe NCs doped with four Mn ions in various spatial distributions. The numerical results show that the magnetism in a few-Mn doped NC is not only determined by the total number of Mn ions, but also sensitively depends on the individual locations, which are however hardly considered by widely used mean field theory. Remarkably, the formation of Mn clusters in a NC leads to the significant deviation of the magnetization from the standard Brillouin function description for an ideal paramagnet. The quantum size effect is shown to enhance the magnetizations of magnetic NCs via the interactions between the quantum confined carriers and Mn-clusters. A solvable constant interaction model (CIM) with the consideration of individual Mn spins is presented for the explanation of the numerical data.

Keywords: Nanocrystals, quantum dots, magnetic semiconductors, paramagnetism, spin physics

1. INTRODUCTION

Since Bhargava *et al.*[1] for the first time successfully incorporated magnetic ion (Mn^{2+}) dopants into colloidal semiconductor (ZnS) nanocrystals in 1994, magnetic ion doped semiconductor nanostructures have persistently received the increasing attention from the investigators in both fields of applied and fundamental physics. To date, magnetic ion doped colloidal semiconductor nanocrystals have been successfully synthesized for a variety of semiconductor materials, including ZnO, ZnS, ZnSe, CdS, CdSe, and PbSe [2-5]. Besides the attractive application potential,[6-9] those magnetic semiconductor nanostructures with the hybrid nature of semiconducting, magnetic and quantum properties serve as an unique test bed for studying fundamental physics of magnetism [10-13].

Perhaps one of the most intriguing physics in magnetic NCs is the competitive interplay between the spin interactions between magnetic ions and those between magnetic ions and quantum confined carriers. While the Mn-Mn spin interactions are anti-ferromagnetic (AFM) and short-ranged, the carrier-Mn interactions are ferromagnetic (FM) and depend on the carrier wave functions that basically spread over the interior of nanocrystals. The contrast between the two types of the existing spin interactions in charged or photon excited magnetic NCs makes the magnetic properties rich but also some complications hindering detailed theoretical investigations. [14-15].

Theoretically, the magnetism of magnetic semiconductor bulk and thin film systems have been studied widely using the mean field theory (MFT).[16-18] In the MFT, the couplings of the total Mn spin to a carrier are modeled as a continuous field following the standard paramagnetic behavior described by the Brillouin function.[16] In a magnetic NC, the number of magnetic ions is however quite small (typically few or tens of magnetic ions only), and if whether the MFT is still valid for few-Mn doped NCs remains an open question. In this work, we employ the numerical exact diagonalization (ED) approach to study the magnetism in singly charged NC with the small number of Mn ions up to four. As presented in our previous works, the discreteness of the spatial distribution of few Mn ions in a NC leads to the significant deviation of the magnetization from the standard Brillouin function description and various magnetic anisotropies in symmetric NCs, both of which are beyond the expectation from the MFT. [19-21]

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Spintronics VI, edited by Henri-Jean Drouhin, Jean-Eric Wegrowe, Manijeh Razeghi, Proc. of SPIE Vol. 8813, 88130H · © 2013 SPIE
CCC code: 0277-786X/13/$18 · doi: 10.1117/12.2023623

However, the restriction of using the exact diagonalization approach lies in the high demand of numerical cost. Since the total number of many-Mn configurations $N_c \propto 6^{N_{Mn}}$ rapidly increases with increasing number of Mn, the ED technique works well only for the system with small number of Mn ions. For more physical analysis and also the possible extension to the cases of more Mn ions, a simplified constant interaction model (CIM) is presented. Via the comparison between the model analysis and numerical results, the validity of the CIM is discussed.

1.1 Theoretical model

The Hamiltonian for a singly charged semiconductor NC coupled to magnetic ions subject to an external magnetic field is written as,

$$H = H_e + H_{e-Mn} + H_{Zeeman} + H_{Mn-Mn}, \tag{1}$$

consisting of the Hamiltonians of the single electron, the electron-Mn interactions, the Mn-Mn interactions, and the spin Zeeman energies. Generally, the single-electron Hamiltonian is written as $H_e = \sum_{i\sigma} E_i c_{i\sigma}^{\dagger} c_{i\sigma}$ in a second quantized form, where the subscript *i* labels the single-electron orbital states, $\sigma = \uparrow$ ($\downarrow$) denotes the up (down) spin of electron with the *z*-component, $s_z = +\frac{1}{2}$ ($s_z = -\frac{1}{2}$), $c_{i\sigma}^{\dagger}$ ($c_{i\sigma}$) is defined as the creation (annihilation) operator, and E_i the eigen-energy of a single electron in state $|i\rangle$. Within the hard wall spherical model, the eigen-energies and wave functions of a single-electron in a spherical NC are given by $E_{nlm} = \frac{2\hbar\alpha_{nl}^2}{m^* d^2}$ and $\psi_{nlm}(\vec{r}) = \langle \vec{r} \mid nlm \rangle = \frac{4}{d^{3/2}} \frac{J_l\left(\frac{2\alpha_{nl}}{d} r\right)}{J_{l+1}(\alpha_{nl})} Y_{lm}(\theta, \varphi)$, respectively, where $\vec{r} = (r, \theta, \varphi)$ is the position of electron in polar coordinate, d the diameter of spherical NC, m^* the effective mass of electron, $J_l(r)$ the spherical Bessel function, α_{nl} the *n*-th zero of $J_l(r)$, $Y_{lm}(\theta, \varphi)$ the spherical Harmonic function, and $m^* = 0.15\ m_0$ is the effective mass of electron for CdSe.

The contact ferromagnetic interaction between electrons and magnetic ions is expressed as

$$H_{e-Mn} = -\sum_{i,i',I} \frac{J_{ii'}^{eM}(\vec{R}_I)}{2} \left[\left(c_{i'\uparrow}^{\dagger} c_{i\uparrow} - c_{i'\downarrow}^{\dagger} c_{i\downarrow} \right) M_I^z + c_{i'\downarrow}^{\dagger} c_{i\uparrow} M_I^+ + c_{i'\uparrow}^{\dagger} c_{i\downarrow} M_I^- \right] \tag{2}$$

where the first term on the right-hand side (rhs) describes that the *z* components of electron spins act as an effective field acting on Mn spins M_I^z, and the last two terms involving operators $M_I^{\pm} = M_I^x \pm i M_I^y$ describe the electron spin flip accompanied by the change of Mn spin. The strength of the e-Mn interaction is given by $J_{ii'}^{eM}(\vec{R}_I) \equiv J_{eM}^{(0)} \psi_i^*(\vec{R}_I) \psi_{i'}(\vec{R}_I) \propto d^{-3}$ with $J_{eM}^{(0)} = 10.8$ meV nm^3, depending on the positions of Mn ions and the NC size. The spin Zeeman term is written as

$$H_{Zeeman} = -\sum_i \left(g_e \mu_B B s_i^z \right) c_{i\sigma}^{\dagger} c_{i\sigma} - \sum_I \left(g_{Mn} \mu_B B \right) M_I^z \tag{3}$$

for the external magnetic field $\vec{B}$ in the *z*-direction, where $g_e = 1.2$ ($g_{Mn} = 2.0$) is the g factor of electron (Mn) in CdSe NCs.

The AFM interaction between magnetic ions H_{Mn-Mn} is described by

$$H_{Mn-Mn} = -\frac{1}{2}\sum_{I\neq J} J_{MM}\left(\vec{R}_{IJ}\right)\vec{M}_I \cdot \vec{M}_J \tag{4}$$

where $\vec{M}_I$ is the spin of the *I*-th magnetic impurities Mn^{2+} at position $\vec{R}_I$ and $J_{MM}\left(R_{IJ}\right) = J_{MM}^{(0)} \exp\left\{-\lambda\left[\left(R_{IJ}/a_0\right)-1\right]\right\} < 0$ is the short ranged AFM coupling between Mn ions, rapidly decreasing with increasing the Mn-Mn distance $R_{IJ} = \left|\vec{R}_I - \vec{R}_J\right|$. Following previous works in Refs.[15,20], we take $J_{MM}^{(0)} = -0.5$ meV, $a_0 = 0.55$ nm, and $\lambda = 5.1$ for CdSe:Mn.

The second quantized forms of Eqs (1)-(3) allow for straightforward implementation of exact diagonalization and extension for more number of electrons. For nanocrystals, the energy quantization is so high (typically greater than 10^2 meV, two order of magnitude higher than the spin interactions) that the electron scattering to the high orbitals are negligible, and the electron in the ground state can be reasonably assumed fixed onto the lowest *s*-orbital. Accordingly, one can reformulated Eq.(1) as,

$$H_{eff} = \sum_I J_{ss}^{eM}\left(\vec{R}_I\right)\vec{s}\cdot\vec{M}_I - \frac{1}{2}\sum_{I\neq J} J_{MM}\left(\vec{R}_{IJ}\right)\vec{M}_I\cdot\vec{M}_J - \left(g_e\mu_B s_z + g_{Mn}\mu_B\sum_I M_I^z\right)B \tag{5}$$

where the constant kinetic energy of electron as an energy offset is removed for brevity. Employing the ED technique the energy spectra of single-electron-few-Mn complexes in magnetic NCs can be numerically calculated at high accuracy. Firstly, we take the all possible electron-Mn configurations $\left|s_z, M_1^z, M_1^z, ..., M_N^z\right\rangle$ classified by the *z* component of electron spin s_z, and the *z* component of the spin of the *I*-th Mn ion M_I^z =−5/2, −3/2,...,5/2, as the basis for expanding the eigen states of the magnetic system. Accordingly, we build up the corresponding Hamiltonian matrix, carry out direct diagonalization for it, and finally find the eigen-energies and -states. Notably, the both of the e-Mn and Mn-Mn interactions depend on the locations $\left\{\vec{R}_I\right\}$ of the Mn ions in the NC, so does the resulting energy spectrum (see Fig.1).

The magnetization of a magnetic NC at temperature *T* is calculated according to the definition, $\mathbf{M} = k_B T\left(\frac{\partial \ln Z}{\partial B}\right)_T$,

in terms of the partition function $Z = \sum_i d_i e^{-\frac{E_i}{k_B T}}$, where E_i (d_i) is the energy (degeneracy) of the *i*-th energy level of the magnetic NC. The magnetic susceptibility $\chi = \frac{\partial \mathbf{M}}{\partial B}$ is defined as the partial derivative of magnetization with respect to the magnetic field.

2. RESULTS AND ANALYSIS

2.1 Numerical results

Figure 1 shows the energy spectra, magnetizations, and magnetic susceptibilities of singly charged CdSe NCs of diameter $d = 8$ nm doped with four Mn ions (distributed in three different ways in the NCs) with the magnetic fields up to 10 Tesla. For the NC with four distant Mn ions as considered in Fig.1(a)-(c), since the AFM interactions between the distant Mn ions are negligible, the magnetization and magnetic susceptibility as functions of B shows a typical behavior of paramagnetism that can be well described by the Brillouin function $\mathbf{M}_J = g_J\mu_B J\, B_J\left(g_J\mu_B B/kT\right)$ with the total angular momentum of single-electron-four-Mn complex, $J = 4\times\frac{5}{2}+\frac{1}{2} = 10\frac{1}{2}$ [see Fig.1 (b) and (c)]. Figure 1 (d)-(f)

shows the numerical results for the singly changed NC with two distant Mn ions and one two-Mn cluster. In the presence of short-range AFM interaction in the Mn clusters, the magnetization of the NC is diminished at low magnetic field because of the anti-parallel spin of Mn's in the cluster in the ground states. With further increasing the magnetic field, the Mn spins in the cluster turn out to be towards the direction of the magnetic field and the ground state transitions happen with the increment of spin at certain finite magnetic fields. As a result, the increasing magnetization shows a staircase feature and magnetic susceptibility oscillates with magnetic field. Those magnetic features deviated from the standard Brillouin function description could be varied significantly by changing the Mn-locations or the number of Mn ions in a cluster. Figure.1(g)-(i) shows the results for the same charged NC but with four Mn ions clustered together. With the four Mn ions all of which are AFM interacting with each other, the magnetization of the NC with a Mn-cluster is significantly suppressed at low magnetic field, and then increases with increasing the magnetic field. The diminished magnetization at the low magnetic field, say $B = 1T$, results from the strong AFM in the Mn-cluster that overwhelms the weak FM e-Mn interaction in the NC of diameter $d = 8$ nm.

2.2 A solvable model and analysis

From the previous discussion, we realize that the magnetic behavior of a small NC doped with small number of Mn ions is not determined by the total number of Mn ions but also sensitively depends on their locations, which is however hardly considered appropriately using the MFT. On the other hand, the use of the ED technique to study magnetic ion doped NCs is however limited by the high demand of numerical resources, rapidly increasing with a slight increase of the number of Mn ions. Therefore, a valid simplified model that has no need of heavy numerical computation but still take into account the discreteness of Mn spin distribution would be very useful.

For the purpose, a constant interaction model has been proposed. While in the MFT the spins of individual Mn ions are smeared out and modeled as continuous fields, one treats the e-Mn and Mn-Mn interactions as constant values but still preserve the individual spins of Mn's in the CIM. Under the simplification, the effective Hamiltonian for a singly charged magnetic NC in the CIM reads

$$H_{\text{eff}} = -J_c \sum_{I=1}^{N} \vec{s_e} \cdot \vec{M}_I + J_M \sum_{I \neq J; I,J=1}^{Q} \vec{M}_I \cdot \vec{M}_J + \hat{J}_z g_J \mu_B B \tag{6}$$

where $g_J = \frac{(g_s + g_{Mn})}{2} + (g_{Mn} - g_s) \frac{M_N(M_N+1) - s(s+1)}{2J(J+1)}$, the effective FM coupling constant is estimated by $J_c = 6J_{eM}^{(0)} / \pi d^3$ meV, and the effective AFM coupling constant J_M is the average AFM Mn-Mn interaction over the all short range interacting Mn ions. Although the Mn-position dependences of the spin interactions in the CIM are neglected, the two types of the interactions, the FM ones between electron and all Mn ions and the AFM ones between Mn's in clusters, are distinguished. One can show that the effective Hamiltonian of Eq.(6) commutes with the total spin M_N of all N Mn ions and also the total spin M_Q of the AFM interacting Q Mn ions. It turns out that M_N, M_Q can be used as good quantum numbers for the single-electron-many-Mn eigen states. In addition to the total spin and its magnetic quantum number of the entire charged magnetic NC, $J = M_N \pm 1/2$ and m_J, the eigen solutions for Eq.(6) are explicitly given by

$$E\left(J = M_N \pm \frac{1}{2}, M_N, M_Q, m_J\right) = -\frac{J_c}{2}\left(\frac{1}{2} \pm \left(M_N - \frac{1}{2}\right)\right) + \frac{J_M}{2}\left(M_Q(M_Q+1) - \frac{35}{4}Q\right) - m_J g_J \mu_B B \tag{7}$$

According to Eq.(6), the spin of the ground state is given by

$$J^{(GS)} = M_N^{(GS)} + 1/2, \tag{8a}$$

with

$$M_N^{(GS)} = M_Q^{(GS)} + \frac{5}{2}(N-Q) \qquad (8b)$$

and

$$M_Q^{(GS)} = \left[\frac{J_c}{2J_M} + \frac{g\mu_B B}{J_M}\right] + c, \qquad (8c)$$

where $c = 0$ $\left(1/2\right)$ for even (odd) Q, and $[y]$ is the Gauss symbol which take the integer part of y. Apparently, one sees that the total spin of a magnetically doped NC explicitly depend on the numbers of the $P(=N-Q)$ distant and the Q short range interacting Mn ions and the strengths of the relevant FM and AFM interactions, J_c and J_M ,as well. An impressive feature revealed by the CIM is that a NC with AFM interacting Mn clusters might undergoes ground state transitions with the increment of spins $M_Q^{(GS)}$, $M_N^{(GS)}$ and J as the increasing magnetic field coincidences $B = B_c^{(n)} = \dfrac{nJ_M - J_c/2}{g\mu_B}$ for $n = 1,2,3\ldots$. The magnetic state transitions result in the staircase feature of magnetization as shown in Fig.1 (e) and (h), and can be viewed as a direct signature of anti-ferromagnetic interactions in a NC with Mn-clusters. According to Eqs. (8a)-(8c), the total spins of the ground states of the NCs considered in Fig.1 are predicted. For instance, with the coupling constants, $J_c = 0.107$ meV and $J_M = J_{MM}^{(0)} = 0.5$ meV , the NC of diameter $d = 8nm$ with P=2 and Q=2 is predicted as $J = 11/2$ ($J = 15/2$) at $B = 0$ ($B = 10T$), and a ground transition happens at $B_c = 3.85\ T$, consistent with the numerical results. Figure 2 compares the numerical results calculated using ED method and those using the CIM. In general, the CIM yields the results in excellent agreement with the NCs containing P distant Mn ions and a single Q-Mn cluster. The CIM however underestimates the magnetization for the magnetic NCs containing more Mn-clusters, as shown for the NCs with two Mn-dimers in Fig.2(b), since the AFM couplings between distant dimmers are actually nearly vanishing but still taken into accounted by CIM. By reducing the size of NC, the FM e-Mn interaction might become competitive to the AFM interaction in the Mn-cluster and the magnetization becomes finite at the same low magnetic field. Fig. 3 (a) shows the magnetizations of the singly charged NCs of different sizes with the same four-Mn cluster as functions of magnetic field. As shown in Fig.3 (a) and (b), the low-field magnetization of a small NC is increased by reducing the NC size due to the increasing strength of the FM e-Mn interaction which is inversely proportional to the volume of NC, $J_{ss}^{eM}\left(\vec{R}_I\right) \equiv J_{eM}^{(0)}\left|\psi_{100}^{*}\left(\vec{R}_I\right)\right|^2 \propto d^{-3}$.

3. CONCLUSION

In summary, a numerical exact diagonalization study of the magnetic properties of singly charged II-VI CdSe nanocrystal doped with four Mn^{2+} ions is presented. The numerical results show that the magnetism in a few-Mn doped NC sensitively depends on the locations of individual distant Mn ions, the number of Mn-clusters. With increasing magnetic fields, the formation of AFM interacting Mn clusters lead to intriguing staircase-like increasing magnetization significantly deviated from an ideal described by the Brillouin-function description, resulting from the competitive interplay between the AFM Mn-Mn interactions and the FM carrier-Mn ones. The quantum size effect is shown to enhance the magnetizations of magnetic NCs via the interactions between the quantum confined carriers and Mn-clusters. By contrast to the widely used mean field theory, a solvable constant interaction model that allows us to take into account the individual Mn spins in a magnetically doped NC is employed for physical explanations.

4. ACKNOWLEDGEMENTS

The authors acknowledge support from the National Science Council of Taiwan (Contract No. NSC-100-2112-M-009-013-MY2), and the National Center of Theoretical Sciences.

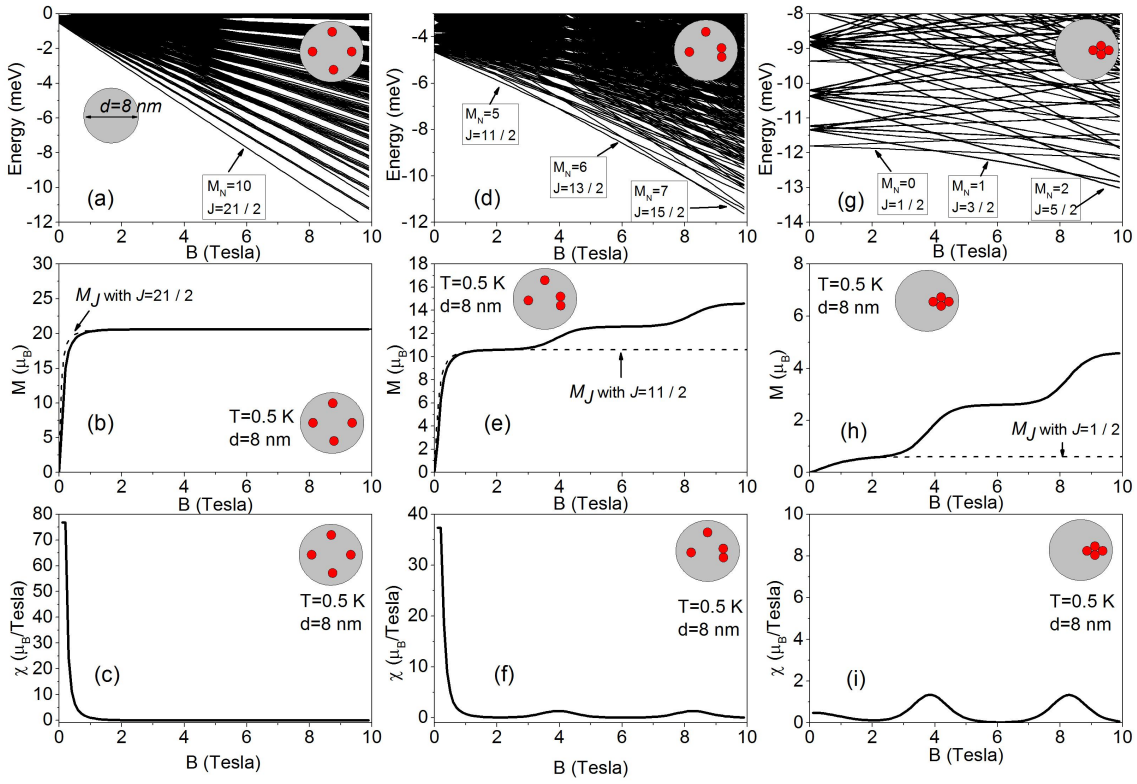

Figure 1. The calculated energy spectra, magnetizations, and magnetic susceptibilities of singly charged CdSe NCs doped with four Mn ions in different distributions with the magnetic fields up to 10 Tesla using the exact diagonalization approach. In (b), (e) and (h), the dash lines show the magnetizations for ideal paramagnets of angular momenta J, which are explicitly described by the Brillouin function, $M_J = g_J \mu_B J \, B_J\left(g_J \mu_B B / kT\right)$.

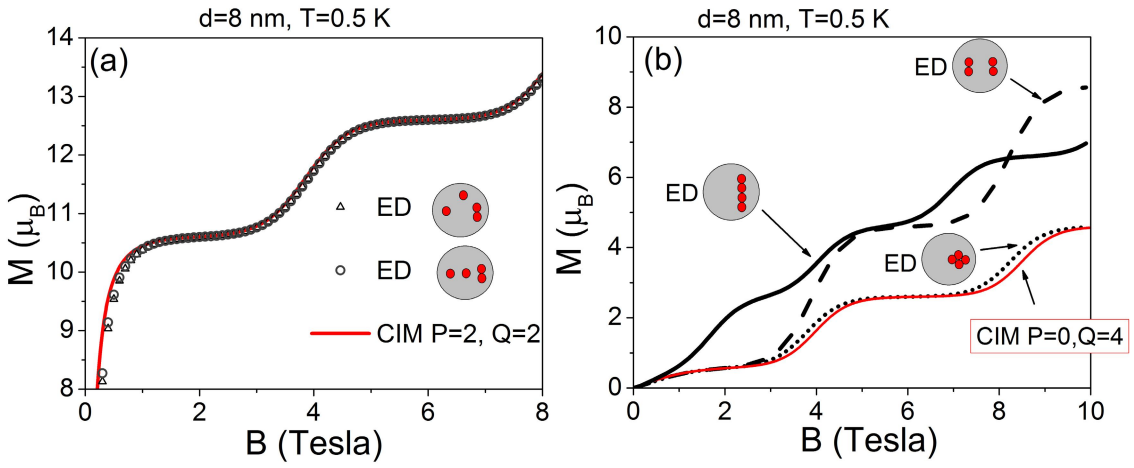

Figure 2. The magnetism for singly charged CdSe NCs of diameter $d = 8$ nm, doped with four Mn ions in different spatial distributions. The left panel (a) shows the numerical results of ED and CIM for the cases with two distant Mn ions (P=2) and two clustered ones (Q=2).The right panel (b) shows the numerical result of ED with different Mn ion distributions and CIM for different P=0 and Q=4 cases.

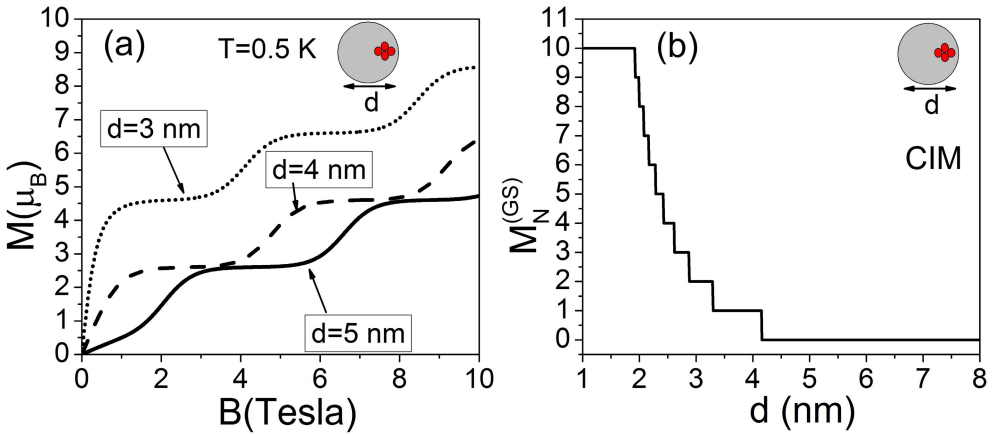

Figure 3. (a) The magnetizations of singly charged NCs of different sizes, containing a four-Mn cluster, as functions of the magnetic field. (b) The increasing total spin of four clustered Mn ion in the ground state of a magnetic NC with reducing the size of the NC.

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