Coherent Manipulation of a Single Magnetic Atom Using Polarized Single Electron Transport in a Double Quantum Dot

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We consider theoretically a magnetic impurity spin driven by polarized electrons tunneling through a double quantum dot system. Spin blockade effect and spin conservation in the system make the magnetic impurity sufficiently interact with each transported electron. This effect yields the nanomagnet coherently driven by a single electron which carries information about spin change of the magnetic impurity. The scheme may develop all electrical manipulation of nanomagnets by means of single electrons, which is significant for the implementation of scalable logical gates in information processing systems.

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Magnetic atoms are critical spin systems which have potential applications in data storage and quantum information processing\(^\text{1,3}\). In particular, using electrons to manipulate magnetic atoms is a natural step toward scalable memory units for future integrated circuits. In dilute II-VI semiconductor quantum dots (QD), interaction between a manganese (Mn) atom and a carrier can be effectively described with the \(sp−d\) exchange interaction\(^\text{4,6}\). Based on the impurity-carrier coupling, electrical control of the single magnetic atom is feasible by injecting different charges in the impurity doped QD\(^\text{4}\).

As shown both in experiment\(^\text{2}\) and theory\(^\text{3}\), electrons can directly tunnel through a Mn atom by taking its energetically separated spin states. These spin states that excited by polarized current can be identified with the conductance, since the ground states and excited states of the Mn atom support different conductance. However, in the direct tunneling from a tip through an individual Mn atom, Mn spin relaxes more frequent then excitation by tunneling electrons at low current\(^\text{2}\). Compared with the case in high dimensional material, the life time of the Mn spin is much longer in a QD\(^\text{8}\). Therefore, coherent electrical manipulation of the Mn spin is possible in low dimensional nanostructure. In a QD doped with a single Mn atom, charge and conductance of the single electron tunneling can be related to the spin state of the Mn\(^\text{9}\). Whereas, it is hard to exactly connect a quantum state of the magnetic atom with single electron in the above QD system, which is a problem needed to solve for quantum information processing.

In this letter, we propose a scheme for coherent driving of the magnetic impurity spin, scaling the number of control electrons down to one. To this end, we consider two coupled semiconductor QDs, in which one QD contains a single magnetic impurity and coupled to a spin polarized electron source. The other QD is localized in a homogeneous magnetic field and connected to a normal conductance, playing a role of a spin filter. In the previous study\(^\text{2,4,7,8}\), electrons move through the magnetic atom without any qualification to their spins, as a result, the interaction between the magnetic atom and electrons are very weak. In contrast, the QD spin filter induces spin dependent tunneling, which makes sure each electron would be completely flipped by the magnetic atom before it passes through the filter. Therefore, each collected electron can be correlated with the change of spin state in the magnetic atom. There are several facts that are very beneficial for the realization of our scheme. First, two coupled QDs can be fabricated, doping a single magnetic ion in one of them\(^\text{10}\). Second, long electron spin life time has been observed in single electron tunneling through QDs in spin blockade and Coulomb blockade regimes\(^\text{11,13}\). Third, Magnetic atom such as Mn impurity with relaxation time from 1 \(\mu s\) to 0.2 \(ms\) is detected in experiment\(^\text{10,14}\), which is longer than driving time of the magnetic atom as shown in the following. Forth, the technique for real time detection of single electron tunneling has been well developed recently\(^\text{15,18}\).

Model and theory.— Our model is illustrated in Fig. 1. The two QDs are denoted by dot 1 and dot 2 with ground orbital levels \(\varepsilon_1\) and \(\varepsilon_2\), respectively. Both electron polarization in the left lead and the external magnetic field \(\vec{B}\) that applied on the dot 2 are assumed to be parallel to the QD growth direction \(z\). Electrons injected from the left lead into the dot 1 are coupled to the magnetic atom by the ferromagnetic Heisenberg type spin exchange interaction. We describe magnetization of the magnetic atom with mean value of its spin along the \(z\) direction \(\langle M_z\rangle\), where \(\hat{M}\) is the magnetic atom spin operator. Bias voltage and the magnetic field is tuned that just the lowest levels of the two QDs fall within the bias window \(\mu_L > \varepsilon_1, \varepsilon_1 (or \varepsilon_2) > \mu_R\), where the indexes \(\downarrow\), \(\uparrow\) indicate electron states with spin up and down, respectively. In addition, the intra-dot Coulomb blockade energies \(U_1, U_2\) corresponding to the dot 1 and the dot 2 are assumed to be much larger than other energy scales which yields only single electron occupation is involved in either of the QDs. It remarkably simplifies the model and our calculation.

The principle of our model is shown in Figs. 1(a)-(d). The magnetic field in this configuration is applied along the \(-z\) direction with value \(\vec{B} = (0, 0, −B)\). It means
The nanomagnet can also be driven reversely, which is presented in Figs. 1(e)-(h). The principle here is the same as that presented above. The difference is that the spin filter only allows spin up electrons tunnel through the dot 2 due to the magnetic field is turned to be in z direction. As a result, to control of the magnetic atom injected electrons are required to be in spin down state and output electrons are expected to be in spin up state. Each transported electron contribute to the spin of the magnetic atom with $-\hbar$. It yields the impurity magnetization orient in the reversal way, that means spin of the magnetic atom would be changed from $\langle M_z \rangle$ to $\langle M_z \rangle - n_t \hbar$ with collection of $n_t$ spin up electrons.

To give a quantitative description to the model, we use the following Hamiltonian [2] [19]:

$$\hat{H} = \hat{H}_{d1} + \hat{H}_{d2} + \hat{H}_{d12} + \hat{H}_{lead} + \hat{H}_{tun},$$

where the Hamiltonian for dot 1 is

$$\hat{H}_{d1} = \varepsilon_1 n_1 + U_1 n_{1\uparrow} n_{1\downarrow} - j_{r} \vec{M} \cdot \vec{S}_1,$n

and the Hamiltonian for dot 2 is

$$\hat{H}_{d2} = \varepsilon_2 n_2 + U_2 n_{2\uparrow} n_{2\downarrow} + g^* \mu_B \vec{B} \cdot \vec{S}_2.$$n

The inter-dot tunneling Hamiltonian reads

$$\hat{H}_{d12} = \hbar \Omega (n_{1\downarrow} n_{1\uparrow} + U \hat{a}_3 \hat{n}_2).$$n

where $\Omega$ is small inter-dot coupling strength. This energy configuration ensures that the spin up electron is forbidden to enter the dot 2. (a) The spin up electron is forbidden to enter the dot 2. (b) The spin up electron is forbidden to enter the dot 2. At the same time, spin state of the magnetic atom is changed. (c) The electron is tunneling through dot 2 and another electron is coming into the dot 1. The process in (e)-(h) is similar to above. In this case, external magnetic field is changed to be up direction and down polarized electrons are injected into the double QD.

FIG. 1: (Color on line) Schematic illustration of single electron tunneling through the two coupled QDs. (a) A spin up electron is injected from the left lead into the dot 1. (b) The spin up electron is forbidden to enter the dot 2. (c) After the electron spin is absolutely flipped, the electron enters the dot 2. At the same time, spin state of the magnetic atom is changed. (d) The electron is tunneling through dot 2 and another electron is coming into the dot 1. The process in (e)-(h) is similar to above. In this case, external magnetic field is changed to be up direction and down polarized electrons are injected into the double QD.
\[
\frac{\partial}{\partial t} \hat{\rho} = \frac{1}{i\hbar} [H_{d1} + H_{d2} + H_{d12}, \hat{\rho}] + \hat{\mathcal{L}}_L \hat{\rho} + \hat{\mathcal{L}}_R \hat{\rho} \quad (7)
\]

The Liouville operators \( \hat{\mathcal{L}}_L \) and \( \hat{\mathcal{L}}_R \hat{\rho} \), describing tunneling on the left and right side of the double dot system, are given by

\[
\hat{\mathcal{L}}_L \hat{\rho} = \frac{1}{2} \sum_{\sigma} \Gamma_L^\sigma (\hat{f}_{L,\sigma} (\hat{c}_{1\sigma}^\dagger \hat{\rho} \hat{c}_{1\sigma} - \hat{c}_{1\sigma} \hat{c}_{1\sigma}^\dagger \hat{\rho}) + (1 - \hat{f}_{L,\sigma}) (\hat{c}_{1\sigma} \hat{\rho} \hat{c}_{1\sigma} - \hat{c}_{1\sigma}^\dagger \hat{c}_{1\sigma} \hat{\rho}) + H.c.),
\]

and

\[
\hat{\mathcal{L}}_R \hat{\rho} = \frac{1}{2} \sum_{\sigma} \Gamma_R^\sigma (\hat{f}_{R,\sigma} (\hat{c}_{2\sigma} \hat{\rho} \hat{c}_{2\sigma} - \hat{c}_{2\sigma}^\dagger \hat{c}_{2\sigma} \hat{\rho}) + (1 - \hat{f}_{R,\sigma}) (\hat{c}_{2\sigma}^\dagger \hat{\rho} \hat{c}_{2\sigma} - \hat{c}_{2\sigma} \hat{c}_{2\sigma}^\dagger \hat{\rho}) + H.c.),
\]

where the Fermi distribution function in the left lead is \( f_{L,\sigma} = (\exp[\varepsilon + U_1 n_{1\sigma} - \mu_L]/k_B T + 1)^{-1} \) and in the right lead is \( f_{R,\sigma} = (\exp[\varepsilon + U_2 n_{2\sigma} - \mu_R]/k_B T + 1)^{-1} \), depending on the charging energy \( U_1 \) conditioned by the occupation, \( n_{i\sigma} = c_{i\sigma}^\dagger c_{i\sigma} \), of electron with spin \( \sigma \). Here, the spin index \( \sigma \) is defined if \( \sigma = \uparrow \) (\( \downarrow \)). The tunneling rates are spin dependent defined by \( \Gamma_\alpha^\uparrow = \Gamma_\alpha (1 + P_0)/2 \) and \( \Gamma_\alpha^\downarrow = \Gamma_\alpha (1 - P_0)/2 \) with the current polarization \( P_0 = (I_0^\uparrow - I_0^\downarrow)/(I_0^\uparrow + I_0^\downarrow) \). Here, \( I_0^\sigma \) is current with pure spin \( \sigma \) on the side of \( \alpha \). The bare tunneling rates can be expressed as \( \Gamma_\alpha = 2\pi \hbar |t_{\alpha}|^2 N_\alpha(\varepsilon) \) with the density of states \( N_\alpha(\varepsilon) \) of electrons at energy \( \varepsilon \).

Results.--- The Hilbert space of the double QD system is generated by the basic vectors \( |i\rangle_1 |m\rangle_1 |j\rangle_2 \), where \( |i\rangle_1 \) and \( |j\rangle_2 \) represent electronic state in dot 1 and dot 2, respectively. Here, \( i, j = \downarrow, \uparrow \) indicates occupation of single electron with spin down and spin up, respectively. \( |m\rangle_1 \) is eigenstate of the impurity spin operator \( \hat{M}_z \) with eigenvalues \( m = -M, -m + 1, ..., M \).

First, we consider a typical single magnetic atom with spin \( M = 5/2 \), such as Fe or Mn. Such magnetic atoms have six quantized spin states \( |m\rangle \) \cite{4}, with corresponding eigenvalues \( m = -5/2, -3/2, -1/2, 1/2, 3/2, 5/2 \). In Fig. 2(a), initial state \( |\psi_M(0)\rangle \) of the magnetic atom is set to be any of the six quantized state. In all cases the magnetic atom is driven to the final state \( |5/2\rangle \). The magnetization is defined by \( \langle M_z \rangle = T \hat{M}_z \hat{\rho} \), in which the trace is taken over the basis vectors \( |i\rangle_1 |m\rangle_1 |j\rangle_2 \). When the spin of magnetic atom and spin of an injected up polarized electron are not parallel, both of them precess relatively due to the coupling between them (see Fig. 1(a)-(d)). As soon as the electron spin flips the electron would tunnels into the dot 2 and spin precession of the magnetic atom would be end with a spin change of \( \hbar \). The variation of the spin state is complex as shown in

\textbf{FIG. 2.:} (Color on line) (a) Magnetization of the magnetic impurity that driven by spin up electrons \( (P_L = 1) \) with an external magnetic field \( \vec{B} = (0, 0, -B) \). (b) Magnetization of the magnetic impurity that driven by spin down electrons \( (P_L = -1) \) with an external magnetic field \( \vec{B} = (0, 0, B) \). The Parameters are \( \mu_L = 75 \Gamma, \mu_R = -75 \Gamma, \varepsilon_1 = 0, \varepsilon_2 = 62.5 \Gamma, \gamma \mu_B B = 135 \Gamma, j_0 = 3 \Gamma, k_B T = 12.5 \Gamma, \Gamma_L = \Gamma_R = \Gamma, P_L = 0, \hbar \Omega = 5 \Gamma, U = 10 \Gamma \).

\textbf{FIG. 3.:} (Color on line) (a) The number of Spin down electrons collected in the right side of the double QD. (b) Current with spin up and spin down as a function of time. The parameters are the same as that in Fig. 2
the inset of Fig. 2 (a). However, the initial and final magnetization appears to be regular. When the impurity spin changes to $|5/2\rangle$, it becomes parallel to the injected electron spin and there will be no spin flip anymore. Then the electron tunneling should be switched off and the spin down polarized current decreases to be zero as plotted in Fig. 3 (b). The spin up polarized current is negligible small in all the time, which is in accord with our prediction for the model. Since each electron contributes to the spin change of the magnetic atom with momentum $ℏ\mathbf{p}$, the magnetic atom initialized in the states $|ψ_M(0)⟩ = \{−5/2⟩, |−3/2⟩, |−1/2⟩, |1/2⟩, |3/2⟩, |5/2⟩$ leads to finite electrons collected in the right lead with definite numbers $⟨n_r⟩ = 5, 4, 3, 2, 1, 0$, respectively. It can be seen in Fig. 3 (a) for corresponding initial spin states. The collected electron number is calculated using the formula

$$⟨n_σ⟩ = \frac{1}{e} \int_0^∞ 1_σ(t)dt$$

where $1_σ(t)$ is current in the right side of dot 2. The current is derived from the charge fluctuating in the two dots $d⟨n_1⟩ + d⟨n_2⟩)/dt = \sum_{σ=↑,↓} (I_{2L}^σ - I_{2R}^σ)/e$, by replacing $I_{2L}^σ = I_{2R}^σ$.

In the second protocol as illustrated in Fig. 4 (c)-(h), the applied magnetic field is set to be $\vec{B} = (0, 0, B)$ and spin down polarized current is injected into the dot 1. In this case, the magnetization is changing reversely comparing with the situation discussed above. For any initial state the magnetic atom is driven to the final state $|ψ_M(0)⟩ = |−5/2⟩$ with magnetization $⟨M_z⟩ = −5ℏ/2$ as shown in Fig. 2 (b). The corresponding number of collected electrons for different initial state of the impurity is the same as that shown in Fig. 3, however, the electron spins are up polarized. For tunneling characteristic time, $1/Γ ≈ 1ns$, the orientation of Mn spin takes a few tens of ms as shown in our results. It is comparable to the time scale of optical control [13].

Now, we consider a magnetic ion with spin $M = 1/2$, such as $Cu^{2+}$. This kind of impurity has particular meaning that its maximum magnetization difference is $ℏ$, from $−ℏ/2$ to $ℏ/2$ or by inverse. Along with the change of magnetic atom from state $|−1/2⟩$ to $|1/2⟩$ only single electron is involved in the tunneling. Indeed, as shown in Fig. 4 exactly a single electron can be obtained by setting initial state of the impurity in $|−1/2⟩$. The collected electron is expected to be in spin down state and spin up electron arrive at the right lead with negligible small probability (see Fig. 4 (b)). The current variation with time is shown in the inset figure. It sharply increases and slowly decreases due to back action of the magnetic ion to the current, the back action makes electron transfer be slower [19].

In Fig. 4 (b), a small probability of spin up electron is collected, which is undesirable result in our system. Fig. 4 (a) reveals that increase of the inter-dot tunneling strength excites an electron from the dot 1 into the excited level $ε_↑$ of the dot 2 and the electron transfers to the right lead. To restrain the electron leakage through the excited level $ε_↑$, one can take a relatively small inter-dot coupling which is required to be much smaller than the decoupling $ε_↑ - ε_↓$.

The system is also sensitive to polarization of the right lead. In Fig. 5 (b), it is illustrated that when the polarization is not pure, the number of electrons collected in the right lead is larger than the expected value. Since
non-purely polarized current contains electrons with different spin orientations, thereby some electrons would be not blocked by dot 2.

The negative number of electrons in Fig. 5(c) implies that an electron in the right lead has a probability to flow into the QDs at the beginning due to the thermal excitation at high temperature, since the QDs is empty initially. After the system reaches steady state, the higher the temperature, the larger probability of the electron distributes in the QDs. The electron staying in the QDs comes from the leads. Therefore, net number of electrons collected in the right lead decreases when temperature increases.

Conclusions.— In the system, change of magnetic impurity spin is correlated to spin state and number of single electrons that tunneling through the two QDs. Based on this principle, we give the following predictions: (i) Our model works as an electron source in which number of emitted electrons can be determined beforehand by setting an initial state of the nanomagnet or using a magnetic atom with certain spin $M$. In particular, a single electron emitter is available using a magnetic atom of spin $1/2$. (ii) The number of polarized transferring electrons can be recorded in the spin state of the nanomagnets. (iii) The change in spin state of the magnetic impurity should be detected by counting the number of electrons that out from the double QD. (iv) Spin state of the magnetic impurity can be coherently controlled, in principle, by injecting suitable number of spin polarized electrons, and this controlling is reversal.

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