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Magnetization reversal induced by spin accumulation in ferromagnetic transition-metal dots

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We use the orthodox theory to study current-induced magnetization reversal in ferromagnetic quantum dots. Current-induced spin accumulation causes a free energy change comparable to the charging energy. The free energy change depends both on the current direction, the direction of the equilibrium magnetization, and the characteristic features of transition-metal electronic states. Magnetization reversal occurs when the free energy change is comparable to the anisotropy energy, which is experimentally feasible.

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Controlling the magnetization direction is of fundamental interest and crucial to magnetic access memories. Current-induced dynamics of the ferromagnetic magnetization in thin films have attracted attention in the scientific community after predictions by Berger and Slonczewski that spin waves can be excited, or that torques are exerted on the magnetization.[1] Several experiments have observed spin-wave excitations and magnetization reversal by spin-polarized current in a nanoscale-sized magnetic multilayer, see, e.g., recent developments in Ref. 2 and references therein. Microscopic and phenomenological theories have been developed to clarify the mechanism of the spin transfer and spin torques exerted by the spin-polarized current.[1,3] In elemental ferromagnets like Co, Ni, and Fe spin currents transverse to the magnetization direction decay after a couple of monolayers, and the angular momentum is transferred as a spin torque on the magnetization.[1,3]

dc magnetoresistance (MR) phenomena have also been studied in much smaller systems in the mesoscopic single-electron tunneling (SET) regime.[4] Several interesting results such as the oscillation of MR as a function of bias voltage, and spin accumulation in the quantum dot have been reported. The oscillation of MR was recently measured in granular ferromagnets.[5] However, ac MR phenomena such as magnetization switching and precession in ferromagnetic dots by spin-polarized currents have, to the best of our knowledge, not been addressed. Interestingly though, spin precession in *normal metal dots* coupled to ferromagnetic leads has recently been predicted.[6] An open question is whether magnetization switching of *all-ferromagnetic SETs* is possible. Current-induced magnetization reversal in ferromagnetic single-electron systems fundamentally differs from current-induced magnetization reversal in larger metallic systems. The spin-polarized current density is much smaller in quantum dot systems than in metallic systems due to the large tunnel resistances. The current density in double-tunnel barrier quantum dot systems is smaller by an order of $10^{-5}-10^{-6}$ than in metallic systems, while the magnetization is reduced only by an order of 10^{-3}.[2,5,7] The spin transfer torques are proportional to I/M, and consequently may be too weak to cause switching or precession. On the other hand, spin accumulation in ferromagnetic dots can be large and thus affect the stability of the magnetization of the dot. The purpose of this work is thus to study the latter effect, magnetization switching induced by spin accumulation. We demonstrate that this is experimentally feasible.

To this end we compute the free energy change $\delta\Omega$ caused by spin accumulation in ferromagnetic quantum dots connected to ferromagnetic leads by treating the electronic states in transition-metal quantum dots, typically Co, within the orthodox theory of the Coulomb blockade phenomena. Magnetization reversal occurs when $\delta\Omega$ exceeds the magnetic anisotropy energy of the dot. We demonstrate that the characteristic features of the electronic state of the transition-metal dot qualitatively determine the dependence of $\delta\Omega$ on the spin accumulation.

Let us first discuss the electronic states in nanoscale quantum dots of transition-metal ferromagnets. In normal metals, without spin-orbit interaction, random matrix theory predicts the Wigner distribution $P(x)=(\pi/2)\exp(-\pi x^2/4)$ with $x=\Delta E/\delta$, where δ is a scaling parameter for the energy levels for a single band model.[8] It is not obvious, however, that this type of distribution holds for real quantum dots of transition-metals. Therefore, we perform an exact diagonalization study of the s,p,d tight-binding Hamiltonian for small clusters to study the energy level distributions. Values of the hopping integrals are taken from Ref. 9. The calculated results are averaged over 10 clusters with a truncated fcc structure with 80–100 atoms. We assume a reasonable geometric shape of the small cluster.[10] Calculated results of the distribution of the energy level spacing are shown in Fig. 1. The distribution differs strongly from the Wigner type, but may instead be fitted by *two* Wigner distribution functions with different values of the level spacings δ. The fitted curve is shown by a solid curve in Fig. 1. We interpret the sharp peak as arising from narrow d bands and the broad peak as arising from wider sp bands. The inset shows the calculated density of states (DOS) with high and narrow DOS consisting of mainly d orbitals, and low and wide DOS consisting mainly of s and p orbitals. We have thus confirmed that the DOS of ferromagnetic dots consist of d- and sp-like states and that the energy level statistics is established for each state.

We generalize our previous work[11] on ferromagnetic SETs to a model which characterizes the basic feature of the electronic states obtained above with narrow and broad bands. Since the energy levels of the former are denser than those of the latter, we model the electronic states so that the energy level spacings ΔE of lower and higher DOS regions are $\Delta E=0.3E_C$ and $0.1E_C$, respectively, where E_C is the charging (Coulomb blockade) energy. We discuss the relation between the level spacing and the charging energy below. The Fermi

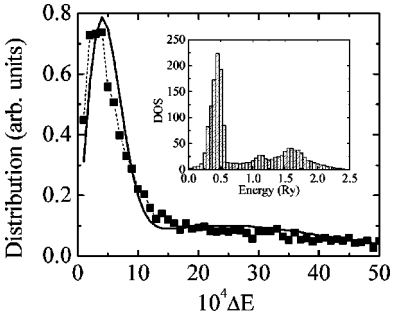

FIG. 1. Computed energy level distribution ΔE (closed squares) and a fitted curve (solid curve) with two Wigner distribution functions. Inset shows DOS summed over 10 samples.

energyε_F determines the boundary between high and low DOS states. Ferromagnetism is realized by introducing an effective field K originated from an *s-d* type exchange interaction. The values of ε_F and K determine the number of electrons $N=N_\uparrow+N_\downarrow$ and magnetic moment $M=N_\uparrow-N_\downarrow$ in the equilibrium state. Here, $N_\uparrow$ and $N_\downarrow$ are the numbers of up and down spin electrons. The magnetic energy is $-KM$, and when $K>0(<0)$, $M>0(<0)$.

In the orthodox theory[12] $N_\uparrow$ and $N_\downarrow$ determine the Gibbs free energy Ω. The equilibrium state is determined by $\partial\Omega/\partial N=0$ and $\partial\Omega/\partial M=0$. Out of equilibrium, the average number of electrons $\langle N\rangle$, the average magnetization $\langle M\rangle$, and the free energy $\langle\Omega\rangle$ are governed by a probability function $P(N_\uparrow,N_\downarrow)$ determined by detailed balance[11]

$$\langle\Omega\rangle=\sum_{N_\uparrow,N_\downarrow}\Omega(N_\uparrow,N_\downarrow)P(N_\uparrow,N_\downarrow). \tag{1}$$

The properties of the left (L) and right (R) tunnel barriers are characterized by tunnel rates, which we assume to be identical for all states, and the voltage drop at the barriers as used before,[11] that is, the voltage drop of the L and R barriers are assumed to be η eV and $(1-\eta)$ eV, respectively, with $\eta=0.1$. We consider tunnel rates $\Gamma_\uparrow^L=0.15\Gamma$, $\Gamma_\uparrow^R=0.5\Gamma$, $\Gamma_\downarrow^L=0.05\Gamma$, and $\Gamma_\downarrow^R=1.5\Gamma$. The choice $\Gamma_\uparrow^L>\Gamma_\downarrow^L$ while $\Gamma_\downarrow^R>\Gamma_\uparrow^R$ indicates that the magnetizations of the leads are antiparallel to create a large spin accumulation. We take $\varepsilon_F=77.75E_C$ and $K=0.5E_C$, which result in $N_0=74$ and $M_0=10$ in the equilibrium state. The boundary between the regions with $\Delta E=0.3E_C$ and $0.1E_C$ in the up-spin state is taken just above ε_F to model the electronic states of transition metals. A schematic figure of the electronic states and the position of the chemical potentials are shown in Fig. 2. We further assume that the spin flip lifetime is longer than the typical tunnel transport time, which is supported by a recent analysis on MR oscillation in ferromagnetic quantum dots.[5]

Numerical results of I, $\delta N=\langle N\rangle-N_0$,$\langle M\rangle$, and $\delta\Omega=\Omega(\langle N\rangle,\langle M\rangle)-\Omega(N_0,M_0)$ are shown in Figs. 3(a)–3(d), respectively, as functions of the bias voltage. Results for an initial equilibrium magnetization $M>0$ and $M<0$ as well as for both current directions are presented in each figure. We find in Figs. 3(a) and 3(b) that I and δN depend on both magnetization and current directions. When the electrons flow from L to R, the number of electrons decreases with increasing bias voltage because the tunnel rates of the right barrier, $\Gamma_\uparrow^R$ and $\Gamma_\downarrow^R$ are larger than those in the left barrier, and

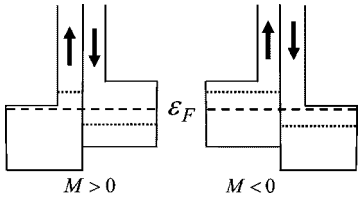

FIG. 2. Schematic figures of the density of states for up and down spin states and the position of the chemical potentials (broken and dotted lines for equilibrium and nonequilibrium states, respectively) for $M>0$ and $M<0$ with fixed current direction, from L to R.

the number of outgoing electrons through the R barrier is larger than that of incoming electrons through the L barrier. For the same reason, the number of electrons increases when electrons flow from R to L. The bias dependence of M shown in Fig. 3(c) is not symmetric with respect to the direction of the equilibrium magnetization M. When electrons flow from L to R, the decrease of electrons is caused by the decrease in the down spin electrons and therefore M increases. The increase in M for $M<0$ is slightly more efficient than for $M>0$. This is because, when $M<0$, dense energy levels below ε_F help to decrease the down (majority) spin electron and those above ε_F help to increase the up (minority) spin electrons efficiently. (See Fig. 2.) Both the distribution of the energy level spacing and the spin-dependent tunnel rates are responsible for these results. When electrons flow from R to L, more down-spin electrons flow into the dot than the up-spin electrons and therefore M decreases with increasing bias voltage. The decrease is more efficient for $M>0$ than for $M<0$. This is because the flow of down-spin electrons is hindered strongly by the wide energy level spacing in the down (majority) spin state when $M<0$.

The asymmetric feature of the calculated results with respect to the current direction and magnetization direction is due to the nonuniform energy level spacing. This interpretation is confirmed by performing similar calculations for quantum dots with equal energy level spacing. Figure 3(e) shows the numerical results of I, $|\delta N|$, and $\delta\Omega$ as functions of bias voltage for equal energy level spacing $\Delta E=0.1E_0$. We find that I, $|\delta N|$, and $\delta\Omega$ are independent of the current direction. The inset shows the spin accumulation $\langle M\rangle$ as functions of the bias voltage. We find that $\langle M\rangle$ is independent of the direction of M_0. The results may be explained phenomenologically as follows.

Let us assume that $\langle\Omega\rangle\approx\Omega(\langle N\rangle,\langle M\rangle)$ for simplicity, which is given as

$$\Omega=\frac{E_C}{2}\langle N\rangle^2+\frac{\Delta E}{2}\left(\langle N\rangle+\frac{\langle N\rangle^2+\langle M\rangle^2}{2}\right)-K\langle M\rangle-N\varepsilon_F, \tag{2}$$

in the absence of the energy level distribution, and is expanded as

$$\delta\Omega=\left(\frac{E_C}{2}+\frac{\Delta E}{4}\right)\delta N^2+\frac{\Delta E}{4}\delta M^2 \tag{3}$$

in terms of δN and $\delta M=\langle M\rangle-M_0$. No linear term appears due to the condition in the equilibrium state. Because the free energy is bilinear with respect to both δN and δM, $\delta\Omega$ in the nonequilibrium state is symmetric around M_0 irrespective of the sign of δM. In order to obtain a dependence of $\delta\Omega$ on the current direction or on the direction of δM, one should take

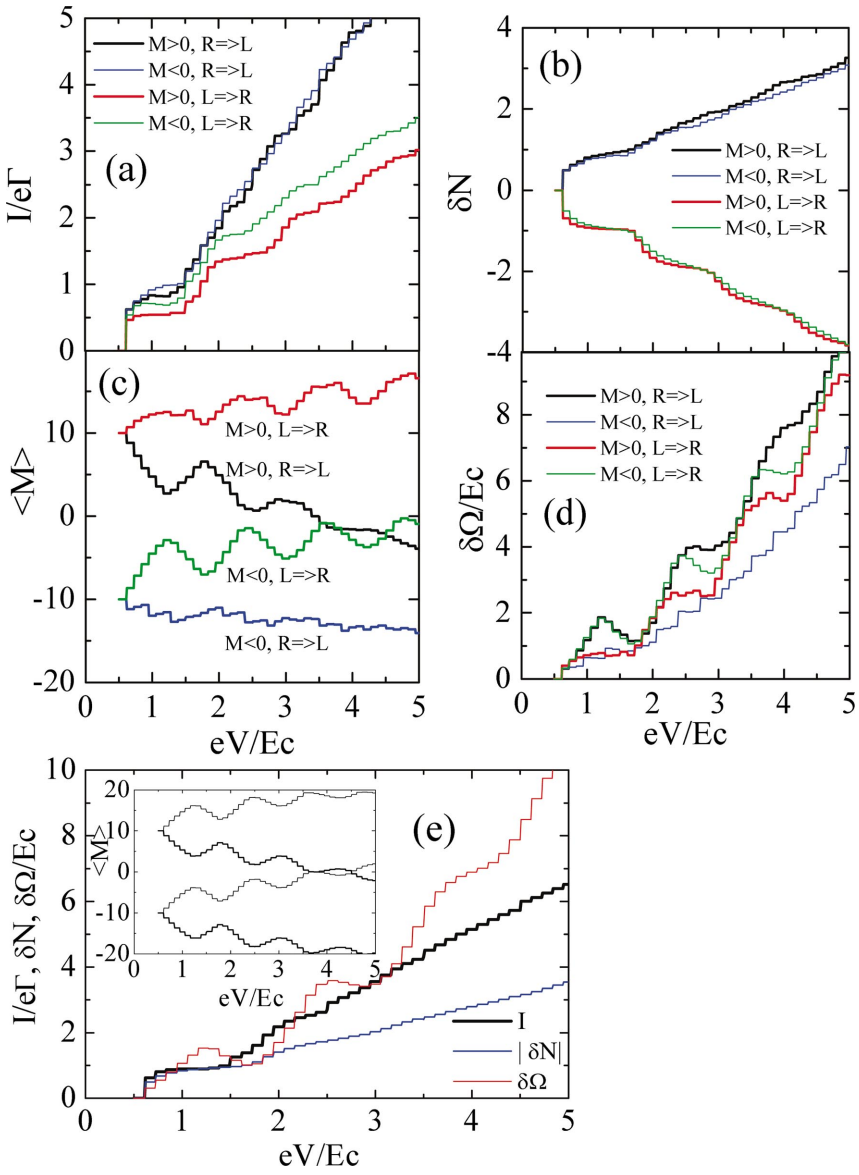

FIG. 3. (Color) (a)–(d). Calculated results of I, δN, $\langle M\rangle$, and $\delta\Omega$ as functions of applied voltage for positive and negative magnetization and for both current directions. (e). Calculated results in the simplest model where equienergy level spacing is assumed in the dot. The inset is the magnetization $\langle M\rangle$ with positive and negative initial values, for currents L to R (thin curve) and R to L (thick curve).

into account higher order terms of M as in the Landau free energy expansion, $F=(aM^2/2)+(bM^4/4)+\cdots$. It is known that a rapid change in the energy level spacings or the density of states near the Fermi level gives rise to higher order terms in the free energy in real bulk ferromagnets such as Ni, Co, and CoNi alloys.[13] Therefore, the asymmetry of the results with respect to current direction and the sign of M may be naturally realized in real ferromagnetic quantum dots. The asymmetric change in $\delta\Omega$ with respect to δM is crucial for the current-induced magnetization reversal as discussed below.

Because the spin accumulation depends on the directions of the electron flow and the direction of the magnetization M, $\delta\Omega$ also depends on the current and magnetization directions as shown in Fig. 3(d). In order to demonstrate our result more clearly, we present in Fig. 4 the energy change $\delta\Omega$ of the dot for a fixed bias voltage at 1.2 eV/E_C as functions of the direction of **M** (magnetization axis) of the dot for both current directions. Here, the magnetization axis is rotated by an angle θ with fixed magnetization directions of L and R leads. In this case, the tunnel rates of both L and R barriers induce spin mixing according to the spinor transformation. When electrons flow from R to L, $\delta\Omega$ decreases as θ changes from 0 to π. This means that the energy change due to the spin accumulation is smaller for $M<0$ than for $M>0$. On the contrary, when the electron flow is reversed from L to R, $\delta\Omega$ increases as θ changes from 0 to π, which indicates the energy change due to the spin accumulation is smaller for $M<0$ than for $M>0$. Therefore, $M<0(M>0)$ may be realized for the current from R to L (from L to R) to minimize the free energy change.

In real systems, magnetic anisotropy exists and the anisotropy energy should be added to $\delta\Omega$. The total sum determines to which angle the magnetization will rotate. If the anisotropy energy is smaller than $\delta\Omega$, magnetization switching may be induced, and the change in the direction of **M** may be measured as a change in the current with fixed bias voltage as shown in Fig. 3(a). We have shown that $\delta\Omega\sim E_C\delta M$ in our calculation, which implies $\delta\Omega\sim K\delta M$ because we have chosen $K\sim E_C$ and the free energy change is related to the magnetic energy. This indicates that E_a should be comparable to K. While E_a increases with volume, K

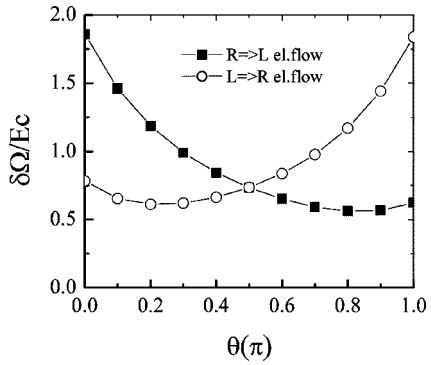

FIG. 4. Energy change $\delta\Omega$ in the dot as functions of the canting angle of the magnetization axis of the dot (squares and circles).

remains an atomic value in the range 0.1–1 eV, depending on the degree of screening. It has been reported that $E_a \sim 0.2$ eV for 4 nm FePt dots.[14] The value of the charging energy, Ec, is 10–20 meV in 4 nm dots. It therefore seems realistic that spin accumulation driven magnetization switching will occur, e.g., for FePt dots that are smaller than 4 nm where Coulomb blockade is important. In addition, the Fermi level should be close to the top of the majority d spin states in order to get a sufficiently asymmetric I-V relation at low bias voltage. The situation may be realized by choosing suitable composition of ferromagnetic alloy for dots.[15] Higher voltage bias, however, is needed when ε_F is located far from the top of the majority d spin states. Hence, choosing suitable particle size and materials, spin-accumulation-induced magnetization reversal at low temperature is feasible.

Let us finally discuss our assumptions of the level spacingvs the Coulomb charging energy. Because the number of atoms of nanoparticles with a diameter of $\sim$4 nm is of an order 10^3, which gives $\delta E \sim 1$ meV, the assumption that $\delta E = 0.1 E_C$ is reasonable. Guéron *et al.*,[16] however, observed one order of smaller energy level spacing, for which they argued this small energy level spacing might be due to spin-wave excitations. This kind of inelastic scattering might be assumed to exist to realize the magnetization switching in the nonequilibrium state. The other assumption of a longer spin-flip lifetime than transport time is supported by recent analysis of the MR oscillation of magnetic dots.[5] The existence of MR oscillations itself supports this assumption.

In summary, we have shown that the free energy change caused by the spin accumulation of ferromagnetic quantum dots depends on both current and magnetization directions, and can be comparable to the charging energy. Magnetization reversal of the quantum dot occurs when $\delta\Omega$ is comparable to or smaller than the anisotropic energy of the dot, which is experimentally feasible. The characteristic features of the electronic state of transition-metal dots qualitatively determines the asymmetric dependence of $\delta\Omega$ on the current direction, and on δM.

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[1] L. Berger, Phys. Rev. B **54**, 9353 (1996); J. C. Slonczewski, J. Magn. Magn. Mater. **159**, L1 (1996); **247**, 324 (2002).
[2] B. Özyilmaz, A. D. Kent, D. Monsma, J. Z. Sun, M. J. Rooks, and R. H. Koch, Phys. Rev. Lett. **91**, 067203 (2003); S. I. Kiselev, J. C. Sankey, I. N. Krivorotov, N. C. Emley, R. J. Schoelkopf, R. A. Buhrman, and D. C. Ralph, Nature (London) **425**, 380 (2003); S. Urazhdin, N. O. Birge, W. P. Pratt, Jr., and J. Bass, Phys. Rev. Lett. **91**, 146803 (2003).
[3] Ya. B. Bazaliy, B. A. Jones, and S.-C. Zhang, Phys. Rev. B **57**, R3213 (1998); A. Brataas, Yu. V. Nazarov, and G. E. W. Bauer, Phys. Rev. Lett. **84**, 2481 (2000); Eur. Phys. J. B **22**, 99 (2001); X. Waintal, E. B. Myers, P. W. Brouwer, and D. C. Ralph, Phys. Rev. B **62**, 12 317 (2000); K. Xia, P. J. Kelly, G. E. W. Bauer, A. Brataas, and I. Turek, Phys. Rev. B **65**, 220401(R) (2002); M. D. Stiles and A. Zangwill, Phys. Rev. B **66**, 014407 (2002).
[4] K. Ono, H. Shimada, and Y. Ootuka, J. Phys. Soc. Jpn. **66**, 1261 (1997); J. Barnas and A. Fert, Phys. Rev. Lett. **80**, 1058 (1998); K. Majumdar and S. Hershfield, Phys. Rev. B **57**, 11 521 (1998); S. Takahashi and S. Maekawa, Phys. Rev. Lett. **80**, 1758 (1998); A. Brataas, Yu. V. Nazarov, J. Inoue, and G. E. W. Bauer, Phys. Rev. B **59**, 93 (1999); X. H. Wang and A. Brataas, Phys. Rev. Lett. **83**, 5138 (1999); J. Barnas, J. Martinek, G. Michalek, B. R. Bulka, and A. Fert, Phys. Rev. B **62**, 12 363 (2000); J. Martinek, J. Barnas, S. Maekawa, H. Schoeller, and G. Schön, *ibid.* **66**, 014402 (2002).
[5] K. Yakushiji, S. Mitani, K. Takanashi, and H. Fujimori, J. Appl. Phys. **91**, 7038 (2002); F. Ernult, K. Yamane, S. Mitani, K. Yakushiji, K. Takanashi, Y. K. Takahashi, and K. Hono, Appl. Phys. Lett. **84**, 3106 (2004).
[6] J. König and J. Martinek, Phys. Rev. Lett. **90**, 166602 (2003).
[7] K. Nakajima, Y. Saito, S. Nakamura, and K. Inomata, IEEE Trans. Magn. **36**, 2806 (2000).
[8] M. L. Metha, *Random Matrics and Statistical Theory of Energy Levels* (Academic Press, New York, 1967).
[9] W. Harrison, *Electronic Structure and the Properties of Solids* (W. H. Freeman and Company, New York, 1980).
[10] M. Jamet, W. Wernsdorfer, C. Thirion, D. Mailly, V. Dupuis, P. Mélinon, and A. Pérez, Phys. Rev. Lett. **86**, 4676 (2001).
[11] A. Brataas, M. Hirano, J. Inoue, Yu. V. Nazarov, and G. E. W. Bauer, Jpn. J. Appl. Phys., Part 1 **40**, 2329 (2001).
[12] C. W. J. Beenakker, Phys. Rev. B **44**, 1646 (1991).
[13] M. Shimizu, Rep. Prog. Phys. **44**, 329 (1981).
[14] S. Sun, C. B. Murray, D. Weller, L. Folks, and A. Moser, Science **287**, 1989 (2000).
[15] S. Chikazumi, *Physics of Ferromagnetism*, 2nd ed. (Clarendon Press, Oxford, 1997).
[16] S. Guéron, M. M. Deshmukh, E. B. Myers, and D. C. Ralph, Phys. Rev. Lett. **83**, 4148 (1999).