Tunable Magnetic Relaxation Mechanism in Magnetic Nanoparticles

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We investigate theoretically the magnetization dynamics of a conducting magnetic nanoparticle weakly coupled to source and drain electrodes, under the assumption that all relaxation comes from exchange of electrons with the electrodes. In the regime of sequential tunneling, the magnetization dynamics is characterized by a relaxation time t_1, which strongly depends on temperature, bias voltage, and gate voltage. While a direct measure of a nanoparticle magnetization might be difficult, we find that t_1 can be determined through a time resolved transport measurement. For a suitable choice of gate voltage and bias voltage, the magnetization performs a bias-driven Brownian motion regardless of the presence of anisotropy.

Electrical and magnetic dynamics of itinerant ferromagnets are often described using a generalization of a "Born-Oppenheimer" approximation, in which the electrons adjust instantaneously to any changes in the magnetization. This approximation, which is justified by the wide disparity of electronic and magnetic time scales, is found to hold down to the smallest size scales attainable by present-day nanofabrication techniques. The list of physical phenomena it explains includes the giant magnetoresistance effect [1], the exchange interaction between different layers in ferromagnetic multilayers [2], and the nonequilibrium spin torque [3]. The first-order effect of a changing magnetization on the electrons was considered only recently [4].

Once electronic and magnetic degrees of freedom are separated, a realistic description of magnetization dynamics requires the inclusion of a relaxation mechanism. A phenomenological description of magnetic relaxation is provided by the Gilbert damping term in the Landau-Lifschitz-Gilbert equation. The Gilbert damping term represents the magnetic relaxation caused by spin-orbit scattering, phonons, magnetic impurities, interface scattering, etc.; its microscopic origin is the subject of ongoing research [5]. Additional magnetic relaxation in thin ferromagnetic films follows from the emission of spin currents into normal metals adjacent to the ferromagnet [4]. For ferromagnetic films, the coupling to normal-metal electrodes has been found to increase the Gilbert damping coefficient by an order of magnitude, making this channel of relaxation the dominant one (see Ref. [4] and reference therein).

It is the purpose of this Letter to study time-dependent electric and magnetic properties of a magnetic nanoparticle, weakly coupled to source and drain reservoirs via tunneling contacts. Such magnetic nanoparticles have been fabricated and studied recently by Ralph and co-workers [6,7]. In the absence of strong spin-orbit coupling in the nanoparticle or for sufficiently large conductances of the tunneling barriers, all electronic and magnetic relaxation occurs via the exchange of electrons with the leads. This system is sufficiently simple that magnetic and charge degrees of freedom can be treated on equal footing and the separation of time scales for charge and magnetization dynamics can be derived from a microscopic model. Moreover, because magnetic relaxation takes place through the exchange of electrons with the leads, the magnetic dynamics crucially depends on the electric environment (bias voltage and the voltages on nearby metal gates). This dependence leads to a tunable magnetization relaxation rate. To our knowledge, this is the first system in which magnetic damping rate can be tuned by a simple gate voltage. Tunability is an important asset for potential applications of magnetic nanostructures, since optimal functioning of a nanomagnetic device requires that damping rate is matched to other relevant time scales of the system. Further, as we show below, for certain values of bias and gate voltages, the electric current creates a nonequilibrium "randomization" process that exceeds relaxation, causing a random motion of the magnetization vector, even in the presence of magnetic anisotropy.

Model.—The starting point of our discussion is a model Hamiltonian for a ferromagnetic nanoparticle, in which the ferromagnetism arises from the long-range exchange interaction [8],

$$H_{\mathrm{dot}} = \sum_{\mu,\sigma} \epsilon_\mu c_{\mu\sigma}^\dagger c_{\mu\sigma} - J\vec{S}\cdot\vec{S} - \frac{K}{S}S_z^2 + E_C(N - N_g)^2. \quad (1)$$

Here $c_{\mu\sigma}$ and $c_{\mu\sigma}^\dagger$ are annihilation and creation operators for an electron with spin σ and energy ε_μ,

$$\vec{S} = \frac{1}{2}\sum_{\mu,\sigma_1,\sigma_2} c_{\mu\sigma_1}^\dagger \vec{\sigma}_{\sigma_1\sigma_2} c_{\mu\sigma_2} \quad (2)$$

is the total spin of the nanoparticle, $\vec{\sigma}$ is the vector of Pauli matrices, J and K set the strength of the exchange interaction and anisotropy, respectively, E_C is the charging energy, and N_g is proportional to the gate voltage.

Without the anisotropy term, this model is known as the "universal Hamiltonian," which has been shown to describe nonmagnetic metal nanoparticles on energy scales below the Thouless energy [8,9]. [Note, however, that, unlike the universal Hamiltonian, Eq. (1) should be considered a model description only, since ferromagnetism implies a splitting between majority and minority electrons that exceeds the Thouless energy.]

Without exchange interaction and magnetic anisotropy, Eq. (1) is the basis for the "constant interaction model" of the Coulomb blockade [10]. For sufficiently large exchange interaction, the ground state of the Hamiltonian (1) is ferromagnetic; it has $N_s = 2S$ singly occupied single-electron levels and different Fermi levels E_M and E_m for majority and minority electrons, respectively [see Fig. 1(b)] with $E_M - E_m = 2SJ$. The excited many-electron states $|\alpha\rangle$ are characterized by occupation numbers of the single-electron levels $n_\mu = 0, 1, 2$, the total spin S, the z component of the total spin, S_z, and an additional degeneracy parameter if S is not maximal. (The z axis is assumed to be the easy axis of the magnet.) For low-lying excited states, the identity $N_s = 2S$ is preserved, since many-electron states with $N_s > 2S$ have an excitation energy of order $E_M - E_m$ [11,12]. The typical energy scale for low-lying excited states is of the order of the exchange energy J. In the nanoparticles studied in Refs. [6,7], the anisotropy K is of order J as well [11], so that the magnetic excitations $S_z \to S_z \pm 1$ have energy comparable to that of electron-hole excitations.

The time-independent low energy properties of this model have already been extensively analyzed in Refs. [11,12]. Here we study the time dependence of the
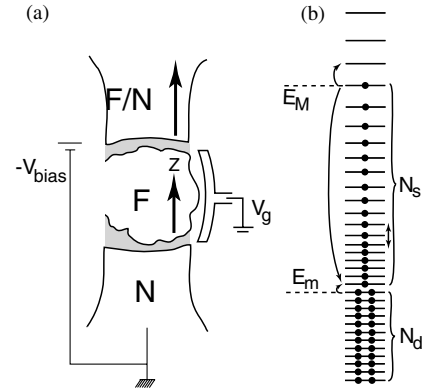

FIG. 1. (a) Schematic of the system under consideration: a ferromagnetic nanoparticle (F) is connected via tunneling junctions to two electrodes, which can be normal (N) or ferromagnetic (F) metals. A bias voltage V_{bias} is applied across the nanoparticle, while a gate voltage V_g can be applied on a gate capacitively coupled to the particle. (b) Structure of the electronic ground state of the nanoparticle. The ground state has N_s singly occupied levels and Fermi levels E_M and E_m for majority and minority electrons, respectively.

total spin $\vec{S}$ for the case when the nanoparticle is connected to source and drain reservoirs, which can be normal metals or ferromagnets [see Fig. 1(a)]. If the leads are ferromagnetic, we assume that the magnetization is collinear with the easy axis of the nanoparticle. For weak coupling (level broadening much smaller than temperature and level spacing), the electronic state of the nanoparticle can be characterized by the probability P_α to find the nanoparticle in many-electron state $|\alpha\rangle$. The probabilities are governed by a rate equation,

$$\frac{\partial P_\alpha}{\partial t} = \sum_{\alpha'} \sum_\mu \sum_{l=L,R} \sum_{\sigma=\uparrow,\downarrow} \{\Gamma_{\mu l \sigma}|\langle \alpha'|c_{\mu\sigma}|\alpha\rangle|^2 [-(1 - f_l(E_\alpha - E_{\alpha'}))P_\alpha + f_l(E_\alpha - E_{\alpha'})P_{\alpha'}]$$
$$+ \Gamma_{\mu l \sigma}|\langle \alpha'|c_{\mu\sigma}^\dagger|\alpha\rangle|^2 [-f_l(E_{\alpha'} - E_\alpha)P_\alpha + (1 - f_l(E_{\alpha'} - E_\alpha))P_{\alpha'}]\}. \quad (3)$$

Here the sum over α' extends over all many-electron states $|\alpha'\rangle$, whereas the sum over μ extends over all single-electron states μ. The distribution functions for the left and right reservoirs are denoted f_l, $l = L, R$, whereas $\Gamma_{\mu l \sigma}$ set the tunneling rate through contact l for majority ($\uparrow$) or minority ($\downarrow$) electrons to the one-body state μ. Equation (3) generalizes the rate equation used in the theory of Coulomb blockade [13]. However, one should note that, upon using an evolution equation for probabilities, information on off-diagonal terms of the density matrix is lost. This means that, while we keep track of the magnetization along the z axis, the position of the magnetization in the xy plane is not monitored. This prevents the use of this approach to the case of ferromagnetic leads with noncollinear magnetizations.

Separation of time scales.—The magnetic structure of the rate equation is encoded in the matrix elements $\langle \alpha'|c_{\mu\sigma}^\dagger|\alpha\rangle$. Only matrix elements for which the particle

number $N_{\alpha'} = N_\alpha \pm 1$ and the spin $S_{\alpha'} = S_\alpha \pm 1/2$ are nonzero. The spin dependence of the corresponding transition rate (matrix element squared) is proportional to the square of a Clebsch-Gordan coefficient [14]. Close to equilibrium ($S_z \approx S$), these two relevant Clebsch-Gordan coefficients are of order 1 and $1/\sqrt{S} \ll 1$, for up and down electrons, respectively, tunneling on a majority state, and vice versa for a minority state. As we shall see below, it is this hierarchy that leads to the separation of time scales for the charge and magnetic degrees of freedom.

We now consider Eq. (3) in the limit where both the temperature and bias voltage are much smaller than J, so that only one single-electron state μ is involved in transport, and for the case that the system is close to equilibrium, $n = S - S_z \ll S$. For definiteness, we assume that μ is a minority state. Denoting the many-electron state

with N electrons, spin S and $S_z = S - n$ by "$(n, -)$" and the many-electron state with $N + 1$ electrons, spin $S - 1/2$ and $S_z = S - 1/2 - n$ by "$(n, +)$", Eq. (3) reads

$$\partial_t P_n^+ = \sum_{l=L,R} D_{nl}^+, \qquad \partial_t P_n^- = \sum_{l=L,R} D_{nl}^-, \qquad (4)$$

where

$$D_{nl}^+ = \Gamma_{\mu l \uparrow} \frac{n+1}{2S+1}[f_{ln}^\uparrow P_{n+1}^- - (1 - f_{ln}^\uparrow)P_n^+]$$
$$+ \Gamma_{\mu l \downarrow} \frac{2S-n}{2S+1}[f_{ln}^\downarrow P_n^- - (1 - f_{ln}^\downarrow)P_n^+], \quad (5)$$

$$D_{nl}^- = \Gamma_{\mu l \uparrow} \frac{n}{2S+1}[(1 - f_{ln-1}^\uparrow)P_{n-1}^+ - f_{ln}^\uparrow P_n^-]$$
$$+ \Gamma_{\mu l \downarrow} \frac{2S-n}{2S+1}[(1 - f_{ln}^\downarrow)P_n^+ - f_{ln}^\downarrow P_n^-], \quad (6)$$

and we defined $f_{ln\downarrow} = f_l(E_n^+ - E_n^-)$ and $f_{ln\uparrow} = f_l(E_n^+ - E_{n+1}^-)$. The structure of the rate equation is similar if transport is facilitated by a majority state or if several one-electron states contribute to transport. In all cases, one retains the special hierarchy of transition rates that is present in Eqs. (5) and (6): $n = S - S_z$ stays constant in most tunneling events (transition rate of order $\Gamma_{\mu l \downarrow}$), whereas only a small fraction of tunneling events allows n to change by unity [transition rate of order $\Gamma_{\mu l \uparrow} n/(2S + 1)$].

To leading order in n/S, only the second lines in Eqs. (5) and (6), corresponding to a process where a down spin enters or exits the quantum dot, contribute. Such a process facilitates charge relaxation, the corresponding relaxation time scale being

$$t_c = (\Gamma_{\mu L \downarrow} + \Gamma_{\mu R \downarrow})^{-1}. \qquad (7)$$

This is the same time scale as for charge relaxation in a nonmagnetic Coulomb blockaded particle [13].

The magnetization dynamics appears when we consider terms of order n/S in Eqs. (5) and (6). We consider the n dependence of the Clebsch-Gordan coefficients in Eqs. (5) and (6), but neglect the (very weak) n dependence of the Fermi functions $f_{ln}^\uparrow$ and $f_{ln}^\downarrow$. Assuming that the charge degree of freedom is in equilibrium, the probability for the nanoparticle to have spin $S_z = S - n$ is $P_n = P_n^+ + P_n^- = (1 + c)P_n^-$, where c is a weighted

average of the distribution functions in the two leads,

$$c = \frac{\Gamma_{\mu L \downarrow} f_L^\downarrow + \Gamma_{\mu R \downarrow} f_R^\downarrow}{\Gamma_{\mu L \downarrow} + \Gamma_{\mu R \downarrow}}. \qquad (8)$$

(The ratio $c = P_n^+/P_n^-$ is taken n independent in view of the comments made above.) The evolution equation for P_n then reads

$$\partial_t P_n = \gamma_-(n + 1)P_{n+1} + \gamma_+ n P_{n-1}$$
$$- [\gamma_- n + \gamma_+(n + 1)]P_n, \qquad (9)$$

where we abbreviated

$$\gamma_+ = \frac{c\Gamma_{\mu L \uparrow}(1 - f_L^\uparrow) + c\Gamma_{\mu R \uparrow}(1 - f_R^\uparrow)}{2S + 1},$$
$$\gamma_- = \frac{(1 - c)\Gamma_{\mu L \uparrow} f_L^\uparrow + (1 - c)\Gamma_{\mu R \uparrow} f_R^\uparrow}{2S + 1}. \qquad (10)$$

In equilibrium one has $\gamma_- > \gamma_+$, corresponding to the stable stationary solution

$$P_n = (\gamma_+/\gamma_-)^n (1 - \gamma_+/\gamma_-). \qquad (11)$$

Equation (9) has a simple interpretation in terms of a random walker on a semi-infinite chain that has a probability $\gamma_- n$ to step towards the origin and a probability $\gamma_+(n + 1)$ to step away from the origin. The difference $\gamma_- - \gamma_+$ can be identified as the magnetic relaxation rate $1/t_1$; the sum $\gamma_+ + \gamma_-$ corresponds to a "randomization rate" for the magnetization. The identification $\gamma_- - \gamma_+ = 1/t_1$ becomes manifest when the time-dependent problem is cast in terms of an evolution equation for moments of P_n (corresponding to moments of S_z). For the first moment $\mathcal{M}(t) = \sum_n n P_n(t)$ one finds

$$\partial_t \mathcal{M}(t) = \gamma_+ - \mathcal{M}(t)/t_1, \qquad t_1^{-1} = \gamma_- - \gamma_+, \quad (12)$$

which is the equivalent of the Landau-Lifschitz-Gilbert equation for our system. Note that, while the charge relaxation rate $1/t_c$ depends on the tunneling rates only, both γ_- and γ_+ depend strongly on temperature, bias voltage, and gate voltage via the Fermi functions f_L^σ and f_R^σ.

If a bias voltage is applied on the system, the effect of the relaxation of $\mathcal{M}(t)$ toward its equilibrium value can be inferred from a small change of the measured electric current, thus allowing for an electric measurement of the magnetic relaxation time t_1,

$$I = \pm e \sum_n D_{ln}^\pm = e\Gamma_{\mu L \downarrow}(f_L^\downarrow - c) + \frac{e\mathcal{M}(t)}{2S+1}\left\{ \frac{\Gamma_{\mu R \downarrow}\Gamma_{\mu L \uparrow}}{\Gamma_{\mu L \downarrow} + \Gamma_{\mu R \downarrow}}(f_L^\uparrow - c) - \frac{\Gamma_{\mu L \downarrow}\Gamma_{\mu R \uparrow}}{\Gamma_{\mu L \downarrow} + \Gamma_{\mu R \downarrow}}(f_R^\uparrow - c) - \Gamma_{\mu L \downarrow}(f_L^\downarrow - c) \right\}. \qquad (13)$$

[In Eq. (13) constant terms of order $1/(2S + 1)$ have been dropped.] Whereas additional intrinsic relaxation mechanisms (phonon-magnon coupling or spin-orbit coupling), which have not been included in the above analysis, may alter t_1, Eq. (13) remains valid in all cases.

Nonequilibrium induced Brownian motion of the magnetization.—An interesting limit occurs when the bias voltage is bigger than $E_0^+ - E_0^-$, whereas the temperature is much smaller than $E_0^+ - E_0^-$. In that case, one may set $f_{\downarrow n}^L = f_{\uparrow n}^L = 1$ while $f_{\downarrow n}^R = f_{\uparrow n}^R = 0$. For normal-metal leads and a symmetric particle-lead coupling ($\Gamma_{L\sigma} = \Gamma_{R\sigma}$), one then obtains

that the stationary solution is given by the detailed balance solution, $P_n^+ = P_n^- = $ const, independent of the magnetization n. [This result can be verified from direct solution of the detailed balance equation associated with Eq. (3)]. This total randomization of the magnetization occurs irrespective of the presence of the anisotropy: although the (anisotropy) energy of a state increases with n, its probability P_n does not decrease. This remarkable result is closely linked to the magnetic structure of the master equation: The bias voltage serves as a driving force for magnetization randomization, whereas the magnetization relaxation is suppressed by Coulomb blockade. We note that through this mechanism, the particle's magnetic energy can increase to a value much above the ground state energy. In a real sample, this effect is limited by extra sources of relaxation that we have not included (e.g., spin-orbit scattering).

In Fig. 2 an example of the bias-voltage dependence of current and average magnetization is shown, obtained from numerical solution of the rate equation (3). Except small regions of width $\Delta V_{\text{bias}} = K/e$ near current steps, which appear when a transition $n + 1 \rightarrow n$ is allowed while the corresponding $n \rightarrow n$ transition is still blocked, one observes that the magnetization is fully randomized as soon as the bias voltage exceeds the threshold for current flow. The randomization effect is suppressed when $K \gtrsim J$. At this point, particle-hole excitations affect the hierarchy in the structure of the rate equation (3), allowing for additional relaxation mechanisms and the breakdown of detailed balance. (For example, an excited state with $N + 1$ electrons and $n = 0$, which can be accessed from the N-electron ground state with $n = 1$, can relax to the N-electron ground state with $n = 0$. However, the opposite process, a direct transition from the N-electron ground state with $n = 0$ to an excited $N + 1$-electron state with $n = 1$, is not allowed.)

Conclusion.—We have studied a simple but realistic model of a magnetic dynamics in a magnetic nanoparticle, for which relaxation occurs via the exchange of electrons with source and drain reservoirs. For this system, electric and magnetic properties can be studied on the same footing, and the separation of electric and magnetic time scales can be established explicitly. The magnetic dynamics is characterized by relaxation and randomization rates, which can both be tuned by bias voltage and gate voltage. A damping rate well matched to other time scales is crucial for magnetic dynamics that is both fast and robust. In this sense, the tunability of the damping rate discovered here may be of use in future studies of magnetic dynamics on the nanoscale. In addition, the possibility of measuring t_1 through a transport measurement removes the need for a direct magnetization measurement, which is difficult for nanoscale magnets.
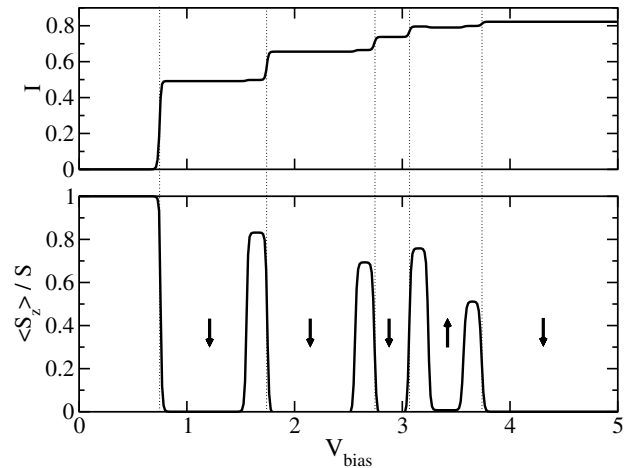

FIG. 2.   Current I (upper) and reduced magnetization $\langle S_z \rangle / S$ (lower) as a function of the bias voltage V_{bias} for a system with $J = 1$, $K = 0.1$, and $k_B T = 0.01$. The down (up) arrows correspond to minority (majority) states that become involved in the tunneling processes.

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