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Model for ferromagnetic nanograins with discrete electronic states

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We propose a simple phenomenological model for an ultrasmall ferromagnetic grain, formulated in terms of the grain's discrete energy levels. We compare the model's predictions with recent measurements of the discrete tunneling spectrum through such a grain. The model can qualitatively account for the observed features if we assume (i) that the anisotropy energy varies among different eigenstates of one grain, and (ii) that nonequilibrium spin accumulation occurs.

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What are the properties of individual quantum states in the electronic excitation spectrum of a nanometer-scale ferromagnetic particle? This is becoming an increasingly important question, since the size of memory elements in magnetic storage technologies is decreasing extremely rapidly,[1] and particles as small as 4 nm are coming under investigation.[2] In this size regime, the excitation spectrum becomes *discrete*; indeed, Guéron, Deshmukh, Myers, and Ralph (GDMR),[3] have recently succeeded in resolving individual quantum states in the spectrum of ferromagnetic Cobalt nanograins, using single-electron tunneling spectroscopy. They found *complex nonmonotonic and hysteretic energy level shifts* in an applied magnetic field and an unexpected *abundance of low-energy excitations*, which could not be fully understood within the simple models used for ferromagnetic nanograins so far.[3,4]

In this Communication, we propose a phenomenological model for ferromagnetic nanograins that is explicitly formulated in terms of the discrete states occupied by the itinerant conduction electrons and capable of qualitatively explaining the observed features. The model is similar in spirit to that advanced independently by Canali and MacDonald,[4] but our analysis includes two further ingredients beyond theirs: (i) mesoscopic fluctuations of the anisotropy energy (i.e., it may vary among different eigenstates), and (ii) nonequilibrium spin accumulation.

Experimental Results. GDMR studied Co particles 1–4 nm in diameter. Assuming a hemispherical shape, the number of atoms in such grains is in the range $N_a \approx 20$–1500, and the total spin, $s_0 \approx 0.83 N_a$,[5] thus is $s_0 \approx 17$–1250. In GDMR's devices, a grain is connected to two aluminum electrodes via aluminum oxide barriers. Its tunneling conductance consists of a series of distinct peaks (see Fig. 2 in Ref. 3), whose positions yield a set of tunneling energies of the form[6] $\Delta E_{fi}^{\pm} \equiv E_f^{N\pm1} - E_i^N$, each corresponding to the energy cost of some rate-limiting electron tunneling process $|i\rangle^N \to |f\rangle^{N\pm1}$ onto or off the grain. Here $|i\rangle^N$ denotes a discrete eigenstate, with eigenenergy E_i^N, of a grain with N electrons, etc.

As the magnetic field is swept, the resonances for Co grains undergo energy shifts and crossings (Fig. 3 in Ref. 3, and Ref. 7). The resulting tunneling spectra have several properties that differ strikingly from those of previously studied nonmagnetic Al and Au grains:[8,9]

(P1): Many *more low-energy excitations* are observed than expected: For all values of the magnetic field, the mean level spacing is $\overline{d}_{\rm obs} \lesssim 0.2$ meV. This is much *smaller* than expected from the naive single-particle estimates $\overline{d}_{\rm maj} \approx 4.6\ {\rm eV}/s_0$ or $\overline{d}_{\rm min} \approx 1.2\ {\rm eV}/s_0$ (with $s_0 \lesssim 1250$) for the majority- and minority-spin mean level spacings near the Fermi energy of Co.[10,5]

(P2): In the small-field regime ($\mu_0 H < 0.2$ T), discontinuous hysteretic switching occurs at a certain switching field $\mu_0 H_{\rm sw}$ (typically $0.1-0.2$ T), due to a sudden change in direction (henceforth called ''reversal'') of the magnetic moment. Moreover, the H dependence of tunneling resonance energies has continuous nonmonotonic variations, which differ seemingly randomly from level to level (Fig. 3 in Ref. 3, and Ref. 7).

(P3): In the large-field regime ($|H| \gg |H_{\rm sw}|$), the resonance energies depend roughly *linearly* on H, with H slopes that almost all have the *same sign* for a given grain; in particular, slopes of opposite signs due to Zeeman splitting of spin-up and spin-down levels[8,9] are not observed (Fig. 4 in Ref. 3, and Ref. 7).

Point (P2) indicates immediately that an independent-electron approach to the energy levels is not sufficient, because the energy of a given state depends on the orientation of the magnetic moment produced by all the electrons within the particle. We shall argue that also points (P1) and (P3) are related to the many-electron spin structure within the particle.

Model Hamiltonian. We propose to model a nanoscale magnet with discrete excitations by the following ''minimal'' Hamiltonian: $\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{\rm C} + \mathcal{H}_{\rm exch} + \mathcal{H}_{\rm Zee} + \mathcal{H}_{\rm anis}$, where $\mathcal{H}_{\rm C}$ is the Coulomb charging energy for a nanoparticle containing N electrons, and

$$\mathcal{H}_0 = \sum_{j\sigma} \varepsilon_j c_{j\sigma}^\dagger c_{j\sigma}, \quad \mathcal{H}_{\rm exch} = -\frac{U}{2} \vec{S}\cdot\vec{S}, \tag{1}$$

$$\mathcal{H}_{\rm Zee} = -hS^z, \quad \mathcal{H}_{\rm anis} = -\sum_{ab} \sum_{ij} S_i^a \mathcal{K}_{ij}^{ab} S_j^b, \tag{2}$$

with $h = g_{\rm eff}\mu_{\rm B}\mu_0 H$. Here $\mathcal{H}_0$ describes the kinetic energy of a single band of single-electron states $|j,\sigma\rangle$, labeled by a

discrete index j and a spin index $\sigma=(\uparrow,\downarrow)$, with the spin quantization axis chosen in the z direction. The exchange, Zeeman, and anisotropy terms, $\mathcal{H}_{\rm exch}$, $\mathcal{H}_{\rm Zee}$, and $\mathcal{H}_{\rm anis}$, are functions of the level-j spin operators $S_j^a = \frac{1}{2}\Sigma_{\sigma'\sigma} c_{j\sigma}^\dagger \sigma^a_{\sigma\sigma'} c_{j\sigma'}$ (where σ^a are Pauli matrices, with $a=x,y,z$), so that $\vec{S}=\Sigma_j \vec{S}_j$ is the total spin vector. $\mathcal{H}_{\rm exch}$ is a rotationally invariant term which models the effects of an exchange field and forces the system to adopt a nonzero total ground state spin, say s_0. On account of this term, spins aligned parallel or antiparallel to $\langle\vec{S}\rangle$ may be thought of as forming ''majority'' and ''minority'' bands, which effectively rotate rigidly together with the magnetization direction. We shall take the mean level spacings ($\overline{d}_{\rm maj}, \overline{d}_{\rm min}$) near the respective Fermi energies, and the exchange splitting of the Fermi energies, $\Delta_{\rm F} \equiv \varepsilon_{\rm F,maj} - \varepsilon_{\rm F,min}$ (≈ 2 eV for Co), as characteristic parameters of the model. The magnitude of U may then be estimated as $U \approx \Delta_{\rm F}/s_0$, since stability of the ground state spin s_0 implies[4] the relation $\Delta_{\rm F} = U(s_0+1/2) + d_0$, where $d_0 (\sim 1/s_0)$ is a small, grain-dependent energy satisfying $-(\overline{d}_{\rm maj} - U/2) < d_0 < \overline{d}_{\rm min} - U/2$. $\mathcal{H}_{\rm Zee}$ describes the spin Zeeman energy in an external magnetic field $\vec{H} = H\hat{z}$. Finally, $\mathcal{H}_{\rm anis}$ models the combined effects of crystalline, shape, and surface anisotropies, etc., in terms of a Hermitian, traceless tensor $[\Sigma_a \mathcal{K}_{ij}^{aa} = 0]$, which describes the energy cost for rotating the various spins $\vec{S}_j$. We split the tensor into an ''average'' and a ''fluctuating'' part by writing $\mathcal{K}_{ij}^{ab} = K^{ab} + k_{ij}^{ab}$. The K^{ab} part dominates since all levels contribute coherently, and, assuming $K^{ab} \propto 1/{\rm Vol}$, it makes an extensive ($\propto$Vol) contribution to the total energy. The simplest nontrivial form that this term might take is a uniaxial anisotropy

$$\mathcal{H}_{\rm uni} = -k_N (\vec{S}\cdot\hat{n})^2/s_0, \tag{3}$$

where $\hat{n}$ is the unit vector in the easy-axis direction (at, say, an angle θ from $\hat{z}$) and $k_N(>0)$ is a volume-independent constant. The fluctuating term k_{ij}^{ab} causes the total anisotropy energy to depend on which single-particle levels are occupied. It is a different ingredient relative to previous models of magnetic switching, which required only a single anisotropy energy function for the whole system, as is appropriate when only the ground state magnetic properties are pertinent.[11]

Basis states. It is convenient to use the eigenstates of $\mathcal{H}(\mathcal{K}=0)$ to construct a set of ''bare'' basis states. Since $[\mathcal{H},\vec{S}]=0$, these states can be grouped into spin multiplets that are labeled by their $\vec{S}\cdot\vec{S}$ and S_z eigenvalues, say $s(s+1)$ and m. For example, the bare ground state of $\mathcal{H}(\mathcal{K}=0)$ for given N, s, and h (>0), say

$$|s,s\rangle_0^N \equiv \prod_{j=1}^{n_\uparrow} c_{j\uparrow}^\dagger \prod_{j=1}^{n_\downarrow} c_{j\downarrow}^\dagger |{\rm vac}\rangle, \tag{4}$$

is a member of a spin multiplet of $2s+1$ states, $|s,m\rangle_0^N \propto (S_-)^{(s-m)} |s,s\rangle_0^N$. Here $n_{\uparrow/\downarrow} = N/2 \pm s$, and $S_- = S_x - iS_y$ is the spin-lowering operator. For $K^{ab} \neq 0$ (but still $k_{ij}^{ab}=0$) the true low-energy eigenstates, $|s,m\rangle^N$, are linear superpositions of the bare states in the multiplet $|s,m\rangle_0^N$ (with $|s,m\rangle^N \to |s,m\rangle_0^N$ as $|K^{ab}|/h \to 0^+$). We shall call the states $|s,m\rangle^N$ the *spin wave multiplet*, since each can be viewed as a homogeneous spin wave. By creating additional single-particle excitations, other, higher-energy multiplets can be built. However, their eigenenergies lie higher than those of the spin wave multiplet $|s,m\rangle^N$ by an amount which is at least of order the single-electron level spacing, i.e., rather large compared to $\overline{d}_{\rm obs}$ [cf. (P1)]; thus the mechanism causing the observed abundance of low-energy excitations, whatever it is, must have its origin in spin excitations, not purely in single-particle excitations.

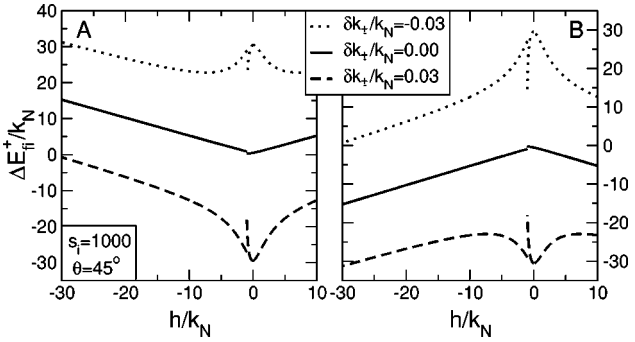

FIG. 1. Tunneling energies $\Delta E_{fi}^{\pm}$ for $\mathcal{H}_{\rm Zee} + \mathcal{H}_{\rm uni}$, plotted as functions of h/k_N sweeping positive to negative, illustrating the effects of anisotropy fluctuations ($\delta k_\pm = k_{N\pm1} - k_N$) for the transitions from $|s_i,s_i\rangle^N$ to (A) $|s_i - \frac{1}{2}, s_i - \frac{1}{2}\rangle^{N+1}$ and (B) $|s_i + \frac{1}{2}, s_i + \frac{1}{2}\rangle^{N+1}$.

Anisotropy fluctuations. Let us first turn to the behavior of the tunneling resonances for small magnetic fields [see (P2)]. The jumps at the switching field have been attributed to a sudden reversal of the nanoparticle's magnetic moment,[3] which occurs when the energy barrier between a metastable state and the true ground state is tuned to zero by the applied field. To illustrate how such jumps arise in our model, let us (for simplicity) take the anisotropy to be uniaxial [Eq. (3)] and consider the case in which the changing magnetic field only rotates the total spin moment, without changing its magnitude.[12] We have numerically diagonalized $\mathcal{H}_{\rm Zee} + \mathcal{H}_{\rm uni}$ as a function of h/k_N for $s_i = 1000$ and $s_f = s_i \pm 1/2$ to determine the ground state energies and the corresponding tunneling energies $\Delta E_{fi}^{\pm}$ (Fig. 1). The latter indeed show a jump at $h_{\rm sw}$. However, if we neglect anisotropy fluctuations by choosing $k_N = k_{N+1}$ (Fig. 1, solid lines), the $\Delta E_{fi}^{\pm}$ lines also have two unsatisfactory features: (i) An upward (downward) jump in $\Delta E_{fi}^{+}(h)$ as $|h|$ increases past $|h_{\rm sw}|$ is *always* followed by a positive (negative) large-h slope, whereas it is observed experimentally [e.g., Fig. 3(a) in Ref. 3] that *either upward or downward* jumps can occur for states having a given large-h slope; and (ii), beyond the switching field, the dependence on h is monotonic (close to linear), in disagreement with recent data (P3).[7] All attempts we made to explain such behavior by choices of K^{ab} corresponding to more complicated than uniaxial anisotropies, or by higher order terms such as $K^{abcd} S_a S_b S_c S_d$,[13] were unsuccessful.

Now, the very fact that the field dependence of each reso-

nance in Refs. 3 and 7 differs so strikingly from that of all others implies that the anisotropy energy fluctuates significantly from eigenstate to eigenstate, which we associate with $k_{ij}^{ab} \neq 0$ in our model. Although the contribution of such random fluctuations to the total energy is nonextensive, their effect on energy *differences* in which extensive contributions largely cancel, can be very significant. A detailed statistical analysis of anisotropy fluctuations is beyond the scope of this paper. Instead, we shall mimic the effects of $k_{ij}^{ab} \neq 0$ by simply using two different anisotropy constants in $\mathcal{H}_{\rm uni}$, say k_N and $k_{N\pm1} \equiv k_N + \delta k_\pm$, for N- or $(N\pm1)$-electron states. k_N can be estimated from the switching field $k_N \approx \mu_0 \mu_B H_{\rm sw}$ (cf. Fig. 1) yielding $k_N \approx 0.01$meV.[14] Now, as illustrated in Fig. 1, $\delta k_\pm / k_N$ in the range of a few percent is *sufficient to reverse the sign of the energy jumps at* $H_{\rm SW}$. Note that $\delta k_\pm \neq 0$ also causes the spectral lines to exhibit rather strong nonmonotonic ''kinks'' near $h_{\rm sw}$, whose amplitudes are of order $s_0 \delta k_\pm$. Qualitatively similar nonmonotonicities have indeed been observed recently,[7] with kink amplitudes on the scale of a few 0.1 meV, in rough agreement with $s_0 \delta k_\pm$ for $s_0 \simeq 1000$.

Anisotropy fluctuations in the range of a few percent are not unreasonable in nm-scale devices. Calculations for transition-metal clusters show that single spin flips can produce a significant change in the magnetic anisotropy energy,[15] and measurements of Gd clusters indicate that anisotropy energies can vary significantly for clusters differing only by a single atom.[16]

We now turn to the low-energy excitations observed in Refs. 3 and 7 (P1). It is natural to ask[3,4] whether these might correspond to spin wave transitions of the form $|s_i, s_i\rangle_N \rightarrow |s_f, m_f\rangle_{N\pm1}$ for different m_f values. However, this does not seem to be the case, for three reasons: (i) It can be shown that only two transitions (namely $|s_0, s_0\rangle \rightarrow |s_0 \pm 1/2, s_0 \pm 1/2\rangle$) have significant weight.[4,17] Resonances associated with final states $|s_f, m_f\rangle$ that differ only in m_f would (ii) have a spacing of order $k_N (\approx 0.01$ meV), which is significantly *smaller* than observed, and would (iii) exhibit a *systematic* increase in the magnitude of their slope ($\propto |s_i - m_f|$) for high magnetic fields that was not observed in experiment.

Nonequilibrium. Since the large density of resonances (P1) cannot be explained by equilibrium transitions (neither single particle excitations nor spin wave excitations), we must explore nonequilibrium effects: In general, N-electron states other than the ground state can be populated during the process of current flow, and this may affect the experimental tunneling spectrum.[18,6] Figure 2 illustrates the consequences as applied to a ferromagnetic grain. Even if a first tunneling event causes a ''charging'' transition from the N-electron ground state $|\mathrm{G}\rangle^N$ to the $(N\pm1)$-electron ground state $|\mathrm{G}\rangle^{N\pm1}$, it may be energetically possible for the subsequent ''discharging'' tunneling transition to return the particle to an excited N-electron state $|\alpha\rangle^N$ instead of $|\mathrm{G}\rangle^N$, provided the applied voltage is sufficiently large, $eV \geqslant E_\alpha^N - E_{\rm G}^N$.[19] Likewise, further charging and discharging transitions may allow any of a large ensemble of states to be occupied at higher and higher levels of an energy ladder, terminating only when an

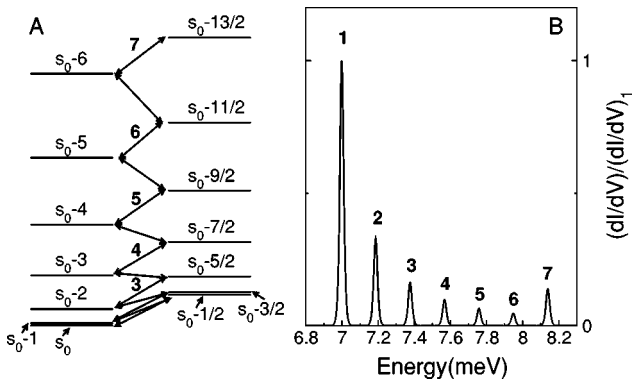

FIG. 2. Nonequilibrium spin accumulation in a ferromagnetic nanoparticle: (A) Tunneling transitions can cause an energy ladder of states with different total spins to be populated (s_0 denotes the ground state of the spin-s_0 multiplet, etc.). (B) The corresponding differential conductance as a function of energy, normalized by its first maximum and calculated by standard methods (Ref. 21). [Parameter choices: $h=0$, $\mathcal{H}_{\rm anis}=0$, $s_0=1000$, $\Delta E_{0,\rm tot}^{\alpha p}=0$, $T=80$ mK, $B_p=0.4$, a tunnel junction resistance ratio of $R_L/R_R=0.1$, and a Coulomb blockade threshold of 7 meV (see sample 3 in Fig. 2 in Ref. 3); we neglected energy and spin relaxation and assumed that charging transitions add minority electrons, so that $\delta E_{\rm res} = \overline{d}_{\rm min} - U/2$.] No significance should be attached to peak heights here, since they depend on (unknown) tunneling matrix elements, which for simplicity were all taken to be equal.

energy-increasing transition requires more energy than the applied voltage provides. As the voltage is increased, the total current (conductance) may increase stepwise (show peaks) when thresholds are crossed to allow higher-energy transitions up the nonequilibrium ladder, thereby changing the occupation probabilities of the ensemble of nonequilibrium states and opening new tunneling channels.

In a ferromagnetic particle, in addition to the nonequilibrium occupation of single-electron states discussed previously for nonmagnetic particles,[18] nonequilibrium spin excitations are possible, too, if the spin-flip rate $\Gamma_{\rm sf}$ is smaller than the tunneling rate $\Gamma_{\rm tun}$.[20] In this case, a ladder of transitions [illustrated in Fig. 2(A)] will occur between states with different total spin s, causing each to have a finite occupation probability and thus leading to *spin accumulation* on the grain.[21] Figure 2(A) illustrates this for the simplest nontrivial case, namely a ladder of spin multiplet ground states $|s,s\rangle$.[22] Figure 2(B) shows the corresponding differential conductance, calculated by solving a master equation for the population of the states of the ladder. The resonance peak spacing ($\delta E_{\rm res}$) and the number of peaks ($n_{\rm res}$) for such a ladder can readily be calculated [using Eq. (2) of Ref. 4]. $n_{\rm res}$ depends on whether the charging transition adds/removes an electron to/from the grain (to be distinguished by an index $p=\pm1$), and on whether it is a majority/minority electron (to be distinguished by an index $\alpha=\pm1$). One finds $\delta E_{\rm res} = \overline{d}_{\rm min} - U/2$ for $\alpha=-1$, and $\overline{d}_{\rm maj} - U/2$ for $\alpha=1$. Using the model parameters estimated above with $s_0 \approx 1000$, the first quantity gives a spacing of ≈ 0.2 meV, as is observed. The second quantity is larger, ≈ 3.6 meV. A detailed analysis[17] shows that $n_{\rm res} - 1$ equals the smallest integer larger or equal to

$$\frac{2E_C^{\text{thresh}} - 2B_p \Delta E_{0,\text{tot}}^{\alpha p}}{(B_p - B_{-p})(\overline{d}_{\text{maj}} + \overline{d}_{\text{min}} - U) - \alpha(\overline{d}_{\text{maj}} - \overline{d}_{\text{min}})}, \qquad (5)$$

where $B_p = [1 + C_p / C_{-p}]^{-1}$ contains the ratio of junction capacitances involved in processes p and $-p$, $\Delta E_{0,\text{tot}}^{\alpha p}$ is the energy difference between states $|s_0 + \alpha p, s_0 + \alpha p\rangle$ and $|s_0, s_0\rangle$, and E_C^{thresh} is the threshold charging energy (energy of the first peak in the differential conductance). The prediction that n_{res} increases linearly with E_C^{thresh} (Ref. 22) is in qualitative agreement with Fig. 2 of Ref. 3, and could be checked explicitly in future devices with gate electrodes, which would allow E_C^{thresh} to be tuned.

A nonequilibrium scenario can also account naturally for the fact (P3) that the vast majority of the observed transitions within a given sample shift in energy with a similar slope for large magnetic fields. This will happen when all the nonequilibrium threshold transitions correspond to tunneling events with the same change of S_z (see Fig. 2), and therefore the same Zeeman shift.

In conclusion, we have proposed a phenomenological model for nanoscale magnets that treats magnetic interactions within a many-electron picture. Its parameters were estimated from bulk properties of Co or experiment, except for the total spin s_0 and the strength of anisotropy fluctuations, which were used as free parameters. The model offers a framework for understanding recent experiments measuring the discrete excitations of magnetic nanograins, provided that we assume (i) anisotropy fluctuations of a few percent between different eigenstates within the same nanograin, and (ii) nonequilibrium spin accumulation.

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[10] We use $\overline{d}_\sigma = 1/[N_a \mathcal{N}_\sigma(\varepsilon_F)]$, where $\mathcal{N}_\sigma(\varepsilon)$ is the spin-dependent bulk density of states per atom (Ref. 5). Using bulk values is appropriate for nanoscale grains, since quantum chemical calculations (Ref. 23) on Ni clusters have shown that $\mathcal{N}_\sigma(\varepsilon)$ has converged to its bulk shape already for N_a as small as 6.

[11] See, e.g., E. Bonet *et al.*, Phys. Rev. Lett. **83**, 4188 (1999), and references therein.

[12] To change the ground state spin value by applying a magnetic field requires in general a field of $\delta h \sim (\overline{d}_{\text{maj}} + \overline{d}_{\text{min}} - U)/2$, which is generically (but not necessarily) large in nanomagnets ($\mu_0 \delta H \gtrsim 25$ T for $s_0 \lesssim 1250$).

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[14] This value is in good agreement with that found recently by Jamet *et al.* (Ref. 24) (our k_N corresponds to their $K_1 v / s_0 \approx 0.017$ meV). The estimate $k_N \approx 0.1$ meV of Ref. 3 is unreliable, since derived from spectral jump sizes. These, however, can change by a factor of ten or more as $\delta k_\pm$ is varied (cf. Fig. 1). This fact was overlooked in Ref. 3, since k_N fluctuations were not considered there.

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[19] If $|\alpha\rangle$ is the spin $(s_0 \mp 1)$-ground state, we find [using Eq. (2) of Ref. 4] that $E_\alpha^N - E_G^N = \overline{d}_{\text{min/maj}} - U/2 \mp d_0$, whose maximum value is of order $\gtrsim 1$ meV for $s_0 \lesssim 1000$.

[20] For GDMR's Co nanograins, we estimate Γ_{tun} from the conductance to be $\Gamma_{\text{tun}} \approx G \overline{d}_{\text{min}}/e^2 \approx 10^9$ s^{-1}, and the rate for spin-conserving electronic relaxation [by Eq. (8) of Ref. 18] to be $\Gamma_{\text{el}} \approx 10^9$ s^{-1}. Since in general $\Gamma_{\text{sf}} \ll \Gamma_{\text{el}}$, we expect that $\Gamma_{\text{sf}} \ll \Gamma_{\text{tun}}$ for GDMR's grains.

[21] Previous predictions of spin accumulation always involved ferromagnetic leads; see, e.g., J. Barnaś *et al.*, Phys. Rev. B **62**, 12 363 (2000), and references therein.

[22] For simplicity, we have not included the possibility of a nonequilibrium population of either single-particle excitations or spin-wave excitations (i.e., states of the form $|s,m\rangle$, with $m < s$) in our calculations or in Fig. 2. However, they certainly exist, and if taken into account, would lead to additional resonances in the tunneling spectrum. Note, though, that the threshold spacing expected for transitions involving spin-wave excitations is k_N / s_0 (or $2|\delta k_\pm|$) if $\delta k_\pm = 0$ (or $\neq 0$) which is exceedingly small in both cases.

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