P H Y S I C A L   R E V I E W   L E T T E R S

Electron-Magnon Coupling and Nonlinear Tunneling Transport in Magnetic Nanoparticles

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We present a theory of single-electron tunneling transport through a ferromagnetic nanoparticle in which particle-hole excitations are coupled to spin collective modes. The model employed to describe the interaction between quasiparticles and collective excitations captures the salient features of a recent microscopic study. Our analysis of nonlinear quantum transport in the regime of weak coupling to the external electrodes is based on a rate-equation formalism for the nonequilibrium occupation probability of the nanoparticle many-body states. For strong electron-boson coupling, we find that the tunneling conductance as a function of bias voltage is characterized by a large and dense set of resonances. Their magnetic field dependence in the large-field regime is linear, with slopes of the same sign. Both features are in agreement with recent tunneling experiments.

Metallic nanoparticles are among the best physical realizations of the concept of Fermi liquid introduced by Landau more than 50 years ago. Their discrete low-energy spectra can be put in a one-to-one correspondence with those of corresponding noninteracting electron systems. Single-electron tunneling spectroscopy [1] in normal-metal nanograins provides a vivid example of Landau's enormous simplification of interacting Fermi systems. Most of the interesting phenomena studied in these experiments can indeed be understood in terms of the quantum mechanics of confined noninteracting quasiparticles. If the grain is made of a ferromagnetic transition-metal material, however, the discrete resonant spectrum seen in tunneling experiments [2,3] is far more complex than the one predicted in an independent particle picture, and indicates that the quasiparticle states are coupled to the collective magnetic moment of the grain. Since ferromagnetic transition metals, in addition to Landau's particle-hole (p-h) excitations, support low-energy collective spin excitations, it is reasonable to assume that tunneling transport through ferromagnetic nanoparticles involves some kind of spin excitations that are the finite-system analogue of the familiar spin-waves or magnons of bulk ferromagnets. So far attempts of including spin collective modes in tunneling transport based on a simple toy model [4–6] have explained only in part the rich phenomena seen in experiment.

In this Letter we present a theoretical study of single-electron tunneling transport through a ferromagnetic metal nanoparticle based on a model that captures the salient features of its elementary excitations—p-h and spin collective—as derived from a recent *microscopic* study [7]. A few remarkable features seen in experiment emerge in a very transparent and direct way from our treatment of the electron-magnon coupling. We find that when a low-energy p-h excitation is strongly coupled to one of the spin collective modes, the tunneling differential conductance versus bias voltage displays an enhanced density of resonances with spacings smaller than the independent-electron energy mean-level spacing δ. The dependence of the tunneling resonances on external magnetic field is regulated by the behavior of the underlying quasiparticle states; it is characterized by mesoscopic fluctuations at small fields and a monotonic dependence at fields larger than the switching field. The model further predicts that in the limit of ultrasmall nanoparticles, where δ is much larger than the typical magnon energy, the conductance should display clusters of resonances separated by an energy of order δ.

The choice of our model is motivated by the microscopic analysis of Ref. [7], where the explicit derivation of the exchange-field-fluctuation propagator allows one to determine the elementary spin excitations (Stoner p-h and collective) of a magnetic grain. One finds that for a small nanoparticle there is one isolated spin collective mode below the lowest p-h excitation energy, which corresponds to the ferromagnetic resonance excitation (spatially uniform $q = 0$ spin wave), of energy $E_{res} \sim$ magnetic anisotropy energy/atom ≈ 0.1 meV in cobalt. For large nanoparticles, the ferromagnetic resonance lies in a region of p-h quasicontinuum and acquires a linewidth αE_{res}, where $\alpha < 1$ is the Gilbert damping parameter. The crossover between these two regimes occurs when one p-h excitation contributes to the resonance, namely, when $\delta = \sqrt{\alpha} E_{res}$. Although the nanoparticles investigated in Ref. [2,3] are too small to strictly satisfy this condition, interactions between p-h excitations and spin-wave modes, including the *nonuniform* ones ($q \neq 0$), will frequently occur. As long as the mechanism of the interaction of one p-h excitation with one spin-wave mode is independent on the latter being uniform or nonuniform, we can illustrate it by following Ref. [7], where the uniform case was consid-

ered. It was shown that when only one p-h excitation of energy $\epsilon_{ab} = \xi_b - \xi_a$ is close to E_{res}, the exchange-field propagator has two poles at energies

$$\omega_{\pm} = \frac{E_{res} + \epsilon_{ab}}{2} \pm \{[(E_{res} - \epsilon_{ab})/2]^2 + \gamma^2\}^{1/2}. \quad (1)$$

The avoided crossing gap γ resulting from the collective mode p-h coupling is found to be $\gamma \sim \alpha E_{res} = \frac{\Delta_{MF}}{\sqrt{2S}} |\langle b|S_x|a\rangle|$, where $2S = N_\uparrow - N_\downarrow$ is the total spin of the nanoparticle, and Δ_{MF} is the amplitude of the spin-splitting field. The presence of the matrix element $|\langle b|S_x|a\rangle|$ in the expression for γ emphasizes the fact that the coupling between spin waves or magnon and electrons is ultimately due to the exchange interaction, which conserves spin. Thus the quasiparticle states a and b should have opposite spins. In spite of the fact that most of the states lying close to the Fermi level have minority-spin character, because of spin-orbit coupling, the quasiparticle states $|I\rangle$ are in fact linear combinations of spin-up and spin-down components $|I\rangle = \alpha_I |\uparrow\rangle + \beta_I |\downarrow\rangle$, and the matrix element $|\langle b|S_x|a\rangle|$ will not vanish. Notice, however, that since spin-orbit interaction is relatively weak in transition-metal ferromagnets, the quasiparticle states can still be assumed to have in general one predominant spin character with just a small admixture of the other.

The Hamiltonian describing the isolated nanoparticle in which a magnon is coupled to one p-h excitation is

$$H_d = \sum_{i=a,b} \epsilon_i c_i^\dagger c_i + \omega \beta^\dagger \beta + \gamma(c_a^\dagger c_b \beta^\dagger + c_b^\dagger c_a \beta)$$
$$+ U\hat{n}(\hat{n} - 1), \quad (2)$$

where $c_i^\dagger$ and c_i with $i = a, b$ are Fermi operators creating and annihilating two electronic levels of energy ϵ_a and ϵ_b, respectively, with $\epsilon_a < \epsilon_b$. The Bose operators $\beta^\dagger$ and β describe a magnon of energy ω. Below we measure all energies in units of the mean-level spacing $\delta \equiv \epsilon_b - \epsilon_a$. The term $\gamma(c_a^\dagger c_b \beta^\dagger + c_b^\dagger c_a \beta)$ represents the electron-magnon coupling. It can be interpreted as a vertex describing an electron scattering from the electronic state a (respectively, b) to the state b (a), while absorbing (emitting) a magnon. We will view the coupling strength γ as a phenomenological parameter; $\gamma \sim \omega$ represents strong coupling. Recently, electron-boson interactions have been used extensively to model electron-phonon coupling in molecular single-electron transistors [8]. An interaction term more similar to ours has been used in studying magnon-assisted transport in ferromagnetic tunneling junctions [9]. The last term in Eq. (2) represents a Coulomb repulsion energy, which is nonzero when both electronic levels are occupied, $\langle \hat{n} \rangle = 2$. The model in Eq. (2), representing a double-level system coupled to one boson mode, is well known in quantum optics and cavity quantum electrodynamics under the name of the Jaynes-Cummings model [10]. The model can be solved exactly, since it conserves both the number of electrons

$n = n_a + n_b$ and the quantity $(n_b - n_a)/2 + m$, where m is the number of bosons. In the trivial cases $n = 0$ and $n = 2$ the energy spectrum is $\epsilon_m^n = \omega m + n/2(\epsilon_a + \epsilon_b + 2U)$; the corresponding eigenstates are $|0, m\rangle = (\beta^\dagger)^m|0\rangle$ and $|2, m\rangle = c_a^\dagger c_b^\dagger (\beta^\dagger)^m|0\rangle$, where $|0\rangle$ is the vacuum. The $n = 1, (n_b - n_a)/2 + m + 1/2 = k + 1$ eigenspace is spanned by the states

$$|1_a, k+1\rangle \equiv c_a^\dagger (\beta^\dagger)^{k+1}|0\rangle, \quad |1_b, k\rangle \equiv c_b^\dagger (\beta^\dagger)^k|0\rangle. \quad (3)$$

The Hamiltonian is now diagonalized within each k subspace, yielding the eigenvalues ϵ_k^+ and ϵ_k^-,

$$\epsilon_k^{\pm} = \epsilon_k^0 + \epsilon_{av} \pm \frac{1}{2}\sqrt{\epsilon_{res}^2 + 4\gamma^2(k+1)}, \quad (4)$$

where $\epsilon_{res} \equiv (\epsilon_b - \epsilon_a) - \omega$ and $\epsilon_{av} \equiv \frac{1}{2}(\epsilon_a + \epsilon_b + \omega)$. The corresponding eigenvectors are

$$|\pm, k\rangle = \delta_1^{\pm}(k)|1_a, k+1\rangle + \delta_2^{\pm}(k)|1_b, k\rangle, \quad (5)$$

where

$$\delta_1^{\pm}(k) = \frac{\gamma\sqrt{k+1}}{\sqrt{[\epsilon_k^{\pm} - \epsilon_a - \omega(k+1)]^2 + \gamma^2(k+1)}}, \quad (6)$$

$$\delta_2^{\pm}(k) = \frac{[\epsilon_k^{\pm} - \epsilon_a - \omega(k+1)]}{\sqrt{[\epsilon_k^{\pm} - \epsilon_a - \omega(k+1)]^2 + \gamma^2(k+1)}}. \quad (7)$$

On top of these states $|\pm, k\rangle$ there is also the state $|1_a, 0\rangle$ with energy ϵ_a, which forms a decoupled one-dimensional subspace in the $n = 1$ sector. We now assume that the magnetic grain is weakly coupled to metallic external electrodes and investigate single-electron tunneling transport through the grain [1]. The total Hamiltonian describing the system is $H = H_d + H_l + H_r + H_t$, where H_d is given in Eq. (2); H_l and H_r describe the left and right electrodes, assumed to be normal Fermi liquids $H_\alpha = \sum_p \xi_{p\alpha} c_{p\alpha}^\dagger c_{p\alpha}$, $\alpha = l, r$, where p is the quantum number specifying a quasiparticle of energy $\xi_{p\alpha}$ measured with respect to the chemical potential of lead α; H_t is the tunneling Hamiltonian coupling the grain to the electrodes $H_t = \sum_{p,\alpha=r,l}[t_{p\alpha} c_{p\alpha}^\dagger(c_a + c_b) + \text{H.c.}]$. In the limit of weak coupling, transport takes place via sequential tunneling, which can be described by means of a standard rate-equation formalism for the occupation probabilities of the grain many-body states [11]. We are interested in the regime where Coulomb blockade is first lifted by applying an external bias voltage, and only the two charge states $n = 0, 1$ are involved. The master equations describing the kinetics of the *nonequilibrium* occupation probabilities $P_k^n = \{P_k^0, P_0^a, P_k^-, P_k^+\}$ for the states $\{|0, k\rangle, |1_a, 0\rangle, |-, k\rangle, |+, k\rangle, k = 0, 1, \ldots\}$, are

$$\dot{P}_k^0 = -\sum_{k';\alpha}[2P_k^0(W_{0k;+k'}^\alpha + W_{0k;-k'}^\alpha + W_{0;a}^\alpha)$$
$$- P_{k'}^+ W_{+k';0k}^\alpha - P_{k'}^- W_{-k';0k}^\alpha - P_0^a W_{a;0}^\alpha], \quad (8)$$

$$\dot{P}_0^a = 2P_0^0 \sum_\alpha W_{0;a}^\alpha - P_0^a \sum_\alpha W_{a;0}^\alpha, \quad (9)$$

$$\dot{P}_k^{\pm} = -\sum_{k';\alpha}[P_k^{\pm}W_{\pm k;0k'}^{\alpha} - 2P_{k'}^0 W_{0k';\pm k}^{\alpha}]. \qquad (10)$$

The coefficients $W_{...}^{\alpha}$ appearing in Eqs. (8)–(10) are transition rates between two many-body states of the grain caused by electron tunneling from and to the leads. For instance, $W_{0k;+k'}^{\alpha}$ is the transition rate from state $|0,k\rangle$ to $|+,k'\rangle$ due to an electron tunneling from the α electrode onto the grain. The $W_{...}^{\alpha}$ are given by Fermi's golden rule

$$W_{0k;\pm k'}^{\alpha} = \Gamma_{0k;\pm k'}^{\alpha} n_F(\epsilon_{k'}^{\pm} - \epsilon_k^0 - \mu_{\alpha})[\delta_1^{\pm}(k')^2\delta_{k',k-1}$$
$$+ \delta_2^{\pm}(k')^2\delta_{k',k}], \qquad (11)$$

$$W_{0;a}^{\alpha} = \Gamma_{0;a}^{\alpha} n_F(\epsilon_a - \mu_{\alpha}), \qquad (12)$$

where μ_{α} is the electrochemical potential of lead α, which we assume to be shifted symmetrically around zero by the applied bias voltage V: $\mu_l = -\mu_r = V/2$. The transition rates $W_{\pm k';0k}^{\alpha}$ and $W_{a;0}^{\alpha}$ are obtained from $W_{0k;\pm k'}^{\alpha}$ and $W_{0;a}^{\alpha}$, respectively, by replacing the Fermi function with $[1 - n_F]$ evaluated at the same energy. The tunneling rates $\Gamma_{0k;\pm k'}^{\alpha} = \frac{2\pi}{\hbar}\sum_p |t_{p\alpha}|^2\delta[\epsilon_{p\alpha} - (\epsilon_{k'}^{\pm} - \epsilon_k)]$ will be taken for simplicity to be independent of energy and lead index, $\Gamma_{0k;\pm k'}^{\alpha} = \Gamma$. The nonequilibrium steady-state probability P_k^n is the solution of the matrix equation $\check{M}\check{P} = 0$, where the matrix $\check{M}$ includes all the transition rates, and $\check{P}$ is a vector of all the P_k^n's. The dc current through the left or right junction is then written as

$$I = (+/-)e\left\{\sum_k\left[2P_k^0\sum_{k'}(W_{0k;-k'}^{l/r} + W_{0k;+k'}^{l/r}) - P_k^-\sum_{k'}W_{-k;0k'}^{l/r}\right.\right.$$
$$\left.\left. - P_k^+\sum_{k'}W_{+k;0k'}^{l/r}\right] + 2P_0^0 W_{0;a}^{l/r} - P_0^a W_{a;0}^{l/r}\right\}. \qquad (13)$$

The transition sequence $|n=0\rangle \to |n=1\rangle \to |n=0\rangle$ allows the tunneling electron to probe the coupled p-h spin-wave excitations of the grain, which appear as resonances in the differential conductance dI/dV as a function of the bias voltage V. We discuss first the case where the p-h excitation is coupled with the uniform ($q=0$) spin-wave mode. For the nanoparticles considered in Refs. [2,3], $\delta \sim$ 1 meV, while the energy of the uniform spin wave is approximately equal to the anisotropy energy/atom $\sim$ 0.1 meV. In Fig. 1 we plot I and dI/dV vs V for the case $\omega = 0.1\delta$, which pertains to this situation. The calculations are done at temperature $T = 0.005\delta$, corresponding to the experimental $T \approx 50$ mK. When $\gamma = \omega$ [Fig. 1(a)], three sets of peaks in the conductance are visible. The first isolated peak occurs when the current starts to flow, and corresponds to the successive transitions $|0\rangle \to |1_a,0\rangle \to |0\rangle$ which are possible when $\mu_l = V/2 = \epsilon_a$ [12]. On further increasing V, the current remains constant until the next lowest charging state ϵ_0^- becomes available (at eV $= 6.8\delta$ for this case). For yet larger V higher states $|0,k\rangle$ and $|-,k\rangle$ acquire a finite nonequilibrium occupation probability, and new transport channels open up. In principle, each allowed transition $|0,k\rangle \to$

$|-,k\rangle$ gives a resonance at $\epsilon_k^- - \epsilon_k^0$, as shown in the inset of Fig. 1(a), calculated at very low temperature, $T = 0.001\delta$. But at $T = 0.005\delta$ only their envelope is visible in the form of a small bump in the conductance centered at eV $= 6.9\delta$. The third large peak, appearing at eV $= 7.0\delta$ is also the envelope of many closely spaced resonances, caused primarily by the transitions through the second group of charged states, $|+,k\rangle$, which become available at that energy. Although values of $\gamma > \omega$ are not very realistic, it is instructive to study the limit behavior of the tunneling conductance for large values of the magnon-electron coupling. In Fig. 1(b) we plot I and dI/dV vs V for $\gamma = 2\omega$. We can see that a large γ causes the sets of resonances of Fig. 1(a) to merge into one cluster, whose individual peaks now start to become visible also at $T = 0.005\delta$. Notice however, that the mean-level spacing between the peaks is $\approx 0.05\delta$, in fact much smaller than the experimentally observed resonance spacing, 0.2δ. This leads us to conclude that such a large density of resonances, caused by an unrealistically strong coupling to the uniform spin-wave mode, is *not* the one observed experimentally.

We now turn to the case where the p-h excitation is coupled to a nonuniform spin-wave mode. The exchange energy of the first nonuniform mode is $\omega \sim \Delta(a/R)^2$, where Δ is proportional to the exchange constant, a is the lattice constant, and R is the nanoparticle diameter. For a 4-nm Co nanoparticle we find $\omega \approx 1$ meV, which is approximately equal to δ [2]. In Fig. 2(a) we plot the *IV* characteristics for the resonant case, $\omega = \delta$, and two different values of γ. At small γ we have again two separate sets of resonances, which are now perfectly resolvable even at the experimental temperature. When γ is increased up to 0.8, the two sets of resonances merge into one cluster, as shown in Fig. 2(b). The number of resonances in the cluster is of the order of 15, with level spacing $\approx 0.3\delta = 0.3(\epsilon_b - \epsilon_a) = 0.3\omega$. Such a dense set of resonances with
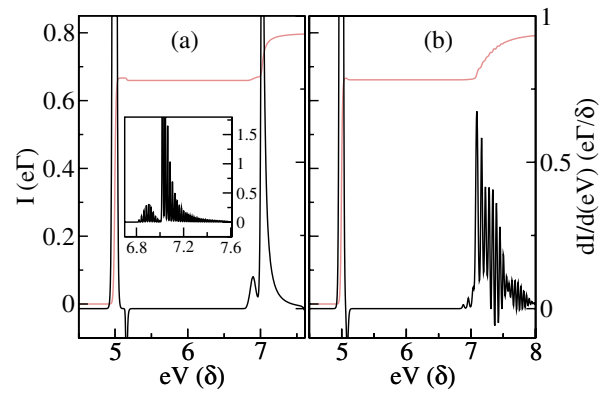

FIG. 1 (color online). Current and differential conductance versus bias voltage V for $\omega = 0.1\delta$. (a) The electron-magnon coupling strength is $\gamma = 0.1\delta$; (b) $\gamma = 0.2\delta$. The temperature T is set equal to 0.005δ, except in the inset, where it is equal to 0.001δ.
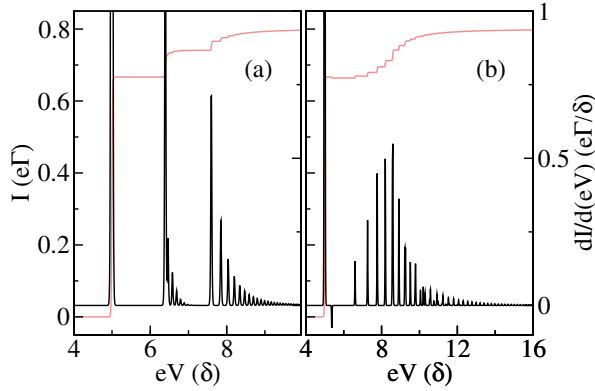
FIG. 2 (color online). Current and differential conductance versus bias voltage V, for $\omega = \delta$. (a) $\gamma = 0.3\delta$; (b) $\gamma = 0.8\delta$ The temperature T in both cases is set equal to 0.005δ.

spacing ≈ 0.2–0.5δ is one of the characteristic features observed experimentally in tunneling spectroscopy of magnetic nanograins. The results of Fig. 2 do not depend on ω being exactly equal to δ, but remain valid for $\delta \gtrsim \omega$, although the larger ω the larger is γ that takes to go from Fig. 2(a) and 2(b). For nanoparticles much smaller than the ones considered in Refs. [2,3], when $\delta \gg \omega$, our model predicts that the conductance spectrum should eventually exhibit sets of resonances separated by an energy $\approx \delta \propto 1/R^3$.

We finally discuss the magnetic field dependence of the resonance spectrum. A crucial feature of our analysis is based on the assumption that the two bare electronic states $|1_a, 0\rangle$ and $|1_b, 0\rangle$ have predominantly minority-spin character. The fact that minority electrons dominate the tunneling transitions had been originally predicted in Refs. [4,5] and was later confirmed by experiments in gated devices [3]. We consider first the regime of small external fields, where the magnetic grain is close to a reversal of the magnetic moment. The electronic states are coupled to the moment itself, and as this moves under the effect of the field, the energies of the states will be subject to random fluctuations [13–15]. Also the frequency of the ferromagnetic mode can fluctuate strongly [7]. Within our model these fluctuations will result in a quasirandom dependence of conductance resonances as a function of the field. At larger fields, after the reversal has taken place, the situation is different. The grain magnetic moment will point along the field and the energies of the minority states $|1_a, 0\rangle$ and $|1_b, 0\rangle$ will increase linearly with the field strength B, with a slope given by their effective $g_{a/b}$ factors, which are ≈ 2 since spin-orbit coupling is weak. Similarly the spin-wave energy dependence can be parameterized by $\omega(B) = \omega(0) + g_\beta \mu_B B$ [7]. We obtain $\epsilon_k^\pm - \epsilon_k^0 = \text{const} + \frac{1}{2}[g_a + g_b + g_\beta \pm \Gamma(B)]\mu_B B$ and $\epsilon_k^\pm - \epsilon_{k+1}^0 = \text{const} + \frac{1}{2}[g_a + g_b - g_\beta \pm \Gamma(B)]\mu_B B$ for the resonance excitation energies, where $\Gamma = \sqrt{(g_a - g_b - g_\beta)^2 + \text{const}/B^2}$. If we take $g_{a/b} \approx 2$ and

$g_\beta \lesssim 2$ [7], we find that the excitation energies are increasing functions of B. Thus we conclude that the conductance spectrum exhibits essentially a monotonic linear dependence on the field, and the slopes of the resonance energies have the same sign.

In conclusion, we have proposed a model that describes coupled electron-magnon excitations in a ferromagnetic metal nanoparticle. The conductance spectrum of single-electron tunneling exhibits a broad and dense set of resonances when the coupling is of the order of the magnon energy. The resonant peaks show Zeeman shifts of the same sign as a function of the external field. Both features of the model are in agreement with experiment. We expect that the resonances originate from the coupling to nonuniform spin waves; furthermore, the tunneling spectrum should break into individual clusters for ultrasmall particles.

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