Quenching of Magnetoresistance by Hot Electrons in Magnetic Tunnel Junctions

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A zero bias anomaly is observed at low temperatures in the current-voltage characteristics of ferromagnetic tunnel junctions; the drop in the junction resistance with increasing bias voltage is greater for antiparallel alignment of the magnetic moments of the magnetic electrodes than for parallel alignment. The resulting decrease in the magnetoresistance of the junction is accounted for by spin excitations localized at the interfaces between the magnetic electrodes and the tunnel barrier. [S0031-9007(97)04443-8]

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While the phenomenon of magnetoresistance in ferromagnetic tunnel junctions (which we call junction magnetoresistance, JMR) was identified two decades ago [1,2], it is only recently that one has been able to grow junctions with reproducible characteristics [3–5]. Within the framework of the transfer Hamiltonian method [6,7] the direct elastic tunneling current is proportional to the sum of the products of the densities of states of itinerant electrons at the Fermi level at the left and right magnetic electrodes for each spin channel [8]. This simple picture has been successful in interpreting current-voltage (I-V) characteristics for ferromagnet-insulator-superconductor and magnetic tunnel junctions (MTJ) [9] at low bias voltage of the order of a few millivolts. Recently, Moodera *et al.* [3] and Parkin *et al.* [5] found that the JMR is strongly reduced when the applied junction bias voltage is of the order of a few *hundred* millivolts. In this Letter, we present the voltage and temperature dependence of the JMR in magnetic tunnel junctions, and we propose that a zero bias anomaly seen below 150 mV is due to hot electrons producing excitations which reduce the magnetoresistance.

We have fabricated junctions whose magnetic electrodes consist of Fe, Co, Ni, and their alloys. Details of the experimental procedure as well as characterization of the magnetic and structure properties of these junctions have been published elsewhere [5]. In Fig. 1, we show typical I-V curves for a junction composed of $Co/Al_2O_3/CoFe$. The main features in this figure are: (1) While the resistance for both parallel (P) and antiparallel (AP) alignment of the magnetization of the two electrodes drops as the applied voltage increases, the decline is more pronounced for the AP alignment; (2) the resistance decreases with temperature rather significantly. In Fig. 1 we notice a rapid decline in the resistance of the junction with a width of about 150 mV; we have called this the "zero bias anomaly." This is superimposed on the drop in resistance ordinarily seen in tunnel junctions inasmuch as it cannot be ascribed to lowering of the barrier height for such low voltages [10]. This large an energy width for this anomaly is unusual; it seems to persist to room temperature. One cannot explain this either by electron-electron interactions as in disordered media which leads to conductance dips [11] or by paramagnetic impurities at interfaces which causes conductance peaks [12]. Another mechanism is the possibility of small metallic inclusions in the barrier region [13]; however, it is unclear if this produces any spin dependence. These mechanisms are restricted to low temperatures with much smaller bias voltages on the order of several millivolts. Since the energy scale involved in this anomaly is the same order of the magnetic excitations, which is approximately the Curie temperature, we propose that itinerant tunnel electrons with excess energies above the Fermi level (due to the applied voltage), known as "hot electrons," produce collective excitations of local spins at the interface between the insulating barrier and the magnetic

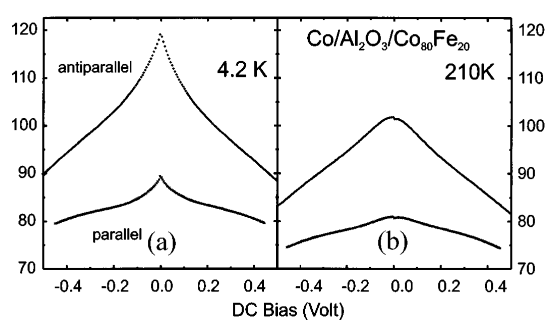

FIG. 1. Resistance as a function of voltage bias at (a) $T = 4.2$ K and (b) $T = 210$ K for parallel (P, lower curves) and antiparallel (AP, upper curves) alignments of magnetization of the two electrodes made of Co and CoFe. The drop in resistance for $V \leq 150$ mV is referred to as the zero bias anomaly.

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electrodes that are responsible for the reduction of magnetoresistance and hence the "zero bias anomaly." A similar inelastic-tunneling process in antiferromagnetic NiO barriers has been studied by Tsui *et al.* [14], and it has been recently emphasized by Moodera *et al.* [3]. By comparison with experimental data, we find our theory can reasonably account for the zero bias anomaly, i.e., for data with applied voltage less than 200 mV.

The conventional explanation for the zero bias JMR observed for ferromagnetic transition-metal electrodes is based on modeling the conduction in the magnetic electrodes by the s-d exchange Hamiltonian [15,16], and invoking the transfer Hamiltonian across the insulating barrier. The s-d exchange Hamiltonian between itinerant s and localized d electrons is

$$H_\alpha = \sum_{\mathbf{k}\sigma} \epsilon^\alpha_{\mathbf{k}\sigma} c^{\alpha +}_{\mathbf{k}\sigma} c^\alpha_{\mathbf{k}\sigma} + E_g \,, \tag{1}$$

where

$$\epsilon^\alpha_{\mathbf{k}\sigma} = \epsilon^\alpha_{\mathbf{k}} + \sigma \sum_i \int d^3\mathbf{r}\, \langle S^\alpha_i \rangle j_\alpha(\mathbf{r} - \mathbf{R}_i) \exp(i\mathbf{k} \cdot \mathbf{R}_i) \,, \tag{2}$$

α labels left (L) and right (R) electrodes, $c_{\mathbf{k}\sigma}$ is the annihilation operator of itinerant electrons, $\mathbf{S}_i$ is the spin operator for localized d electrons, $j_\alpha(\mathbf{r} - \mathbf{R}_i)$ represents the s-d exchange, and E_g is the energy of the localized spins. The energy due to the external field used to produce parallel and antiparallel alignments of the magnetic electrodes, several hundred oersted, is 2 to 3 orders of magnitude smaller than the magnon energy ω_q (see below); we will neglect it.

The barrier Hamiltonian, which leads to the transfer of electrons between left and right electrodes, and which includes magnetic excitations in the transfer, due to s-d exchange between local and itinerant electrons at the interfaces, is written as

$$\begin{aligned} H_B = &\sum_{\mathbf{k}\mathbf{k}',\sigma} T^d_{\mathbf{k}\mathbf{k}'} (c^{L+}_{\mathbf{k}\sigma} c^R_{\mathbf{k}'\sigma} + \text{H.c.}) + \frac{1}{N^{1/2}} \sum_{\mathbf{k}\mathbf{k}'\mathbf{q}} T^J_{\mathbf{k}\mathbf{k}'\mathbf{q}} [S^z_R(\mathbf{q}) + S^z_L(\mathbf{q})] (c^{L+}_{\mathbf{k}\uparrow} c^R_{\mathbf{k}'\uparrow} - c^{L+}_{\mathbf{k}\downarrow} c^R_{\mathbf{k}'\downarrow} + \text{H.c.}) \\ &+ \frac{1}{N^{1/2}} \sum_{\mathbf{k}\mathbf{k}'\mathbf{q}} T^J_{\mathbf{k}\mathbf{k}'\mathbf{q}} [(c^{L+}_{\mathbf{k}\uparrow} c^R_{\mathbf{k}'\downarrow} + c^{R+}_{\mathbf{k}'\uparrow} c^L_{\mathbf{k}\downarrow}) (\sqrt{2S_L}\, a^{L+}_{\mathbf{q}} + \sqrt{2S_R}\, a^{R+}_{\mathbf{q}}) + \text{H.c.}] \,, \end{aligned} \tag{3}$$

where $S^z_\alpha(\mathbf{q}) = S_\alpha - a^{\alpha+}_{\mathbf{q}} a^\alpha_{\mathbf{q}}$ $(\alpha = L, R)$, and $a_{\mathbf{q}} = (1/\sqrt{2SN}) \sum_{i \in I} \exp(i\mathbf{q} \cdot \mathbf{R}_i) S^{(+)}_i$ is the magnon annihilation operator. As the summation over i is restricted to sites at the interfaces between the insulator and electrodes, the magnon wave vector $\mathbf{q}$ in Eq. (3) is restricted to two dimensions parallel to the interface. We do not intend to calculate these matrix elements which represent direct (T^d) and spin-dependent (T^J) transfers; instead, we assume them to be adjustable parameters T^d and T^J, since the magnitude of the resistance is not our primary concern. We fix T_d by fitting our expressions to the JMR at low temperature and zero bias, thereby leaving one adjustable parameter, T^J, to fit the reduction of the JMR with finite applied voltage and temperatures. Also, we will consider only incoherent tunneling, where the wave vector $\mathbf{k}$ is independent of $\mathbf{k}'$ in Eq. (3); because the insulator is amorphous and the interfaces are rough this will be more appropriate than coherent tunneling where $\mathbf{k} = \mathbf{k}'$ and $\mathbf{k} = \mathbf{k}' \pm \mathbf{q}$ in Eq. (3).

With these simplifications it is reasonably straightforward to calculate tunnel currents based on our transfer Hamiltonian Eq. (3) along with the electrode Hamiltonian, Eq. (1). By retaining terms up to second order in the transfer matrix elements (these matrix elements are exponentially small) the contributions to the current from the first two terms in Eq. (3) are the same as the usual tunneling current,

$$\begin{aligned} j_{12} = \frac{4\pi e}{\hbar} \int d\omega [|T^d|^2 + (S^2_L + S^2_R) |T^J|^2] \\ \times \rho^\sigma_L(\omega) \rho^\sigma_R(\omega + eV) \\ \times [f(\omega) - f(\omega + eV)] \,, \end{aligned} \tag{4}$$

where $\rho^\sigma_\alpha(\omega)$ is the density of states of *itinerant* electrons in the α electrode for spin σ, V is the applied voltage across the junction, and $f(\omega)$ is the Fermi distribution function. The current from the third term in Eq. (3) involves emission and absorption of magnons; there are eight processes: four of them represent electron transfer from left to right electrodes, which correspond to the emission and absorption of magnons at the left and right *interfaces* of the barrier with the electrodes. For example, a term which represents the emission of a magnon at the interface of the barrier with the right electrode for a parallel alignment (P alignment) of the magnetizations of the electrodes and for an applied voltage that raises the Fermi level of the right electrode above the left one, is

$$\begin{aligned} j^{(1)}_3 = \frac{4\pi e}{\hbar N_s} \sum_{\mathbf{q}} \int d\omega\, 2|T^J|^2 S_R \rho^m_L(\omega) \\ \times \rho^M_R(\omega + eV - \omega_{\mathbf{q}}) f_L(\omega) \\ \times [1 - f_R(\omega + eV - \omega_{\mathbf{q}})] \langle a^R_{\mathbf{q}} a^{R+}_{\mathbf{q}} \rangle \,, \end{aligned} \tag{5}$$

where N_s is the number of spins at the interfaces, ρ^m_α and ρ^M_α are density of states for itinerant minority and majority electrons, $\omega_{\mathbf{q}}$ is the magnon spectrum at the interfaces, and $\langle\rangle$ denotes the thermodynamic average of the magnon spectrum

$$\langle a^\alpha_{\mathbf{q}} a^{\alpha+}_{\mathbf{q}} \rangle = 1 + \langle a^{\alpha+}_{\mathbf{q}} a^\alpha_{\mathbf{q}} \rangle = 1 + n^\alpha_{\mathbf{q}} \,, \tag{6}$$

where $n^\alpha_{\mathbf{q}} = (e^{\beta\omega^\alpha_{\mathbf{q}}} - 1)^{-1}$ represents the number of interface magnons with wave vector $\mathbf{q}$. The other seven terms $j^{(i)}_3$ $(i = 2-8)$ can be similarly written down.

In principle, we could evaluate these terms by inserting correct density of states, transfer matrix elements and magnon spectra, in order to explain the I-V characteristics in Fig. 1. However, as we have mentioned earlier, these quantities are not well known for the junctions we fabricated. Therefore, we *limit* our discussion to low bias voltages, e.g., smaller than 200 mV, where we expect the transfer matrix elements and density of states of itinerant electrons are nearly independent of the energy. In this range the resistance of the usual tunneling current, Eq. (4), is *independent* of voltage, and we can focus on the zero bias anomaly. Furthermore, we assume the temperature is much smaller than the Fermi energy so that the Fermi distribution function is taken as a step function. With these additional simplifications, the current density due to magnon emission and absorption for P alignment is

$$j_3 = \sum_{i=1}^{i=8} j_3^{(i)} = \frac{8\pi e}{\hbar N_s} |T^J|^2 \sum_{\mathbf{q},\alpha} S_\alpha \{\rho_L^m \rho_R^M (eV - \omega_{\mathbf{q}}^\alpha)(1 + n_{\mathbf{q}}^\alpha)\theta(eV - \omega_{\mathbf{q}}^\alpha) + \rho_L^M \rho_R^m [eV + \omega_{\mathbf{q}}^\alpha - (\omega_{\mathbf{q}}^\alpha - eV)\theta(\omega_{\mathbf{q}}^\alpha - eV)] n_{\mathbf{q}}^\alpha\}. \tag{7}$$

It remains to evaluate the summation over the magnon spectrum of the interfaces. In analogy with Debye's treatment of phonons, we replace the magnon dispersion relation, by a simple isotropic parabolic one, i.e., $\omega_{\mathbf{q}}^\alpha = E_m^\alpha (q/q_m)^2$, where E_m^α is related to the Curie temperature (T_c^α) by the mean field approximation, $E_m^\alpha = 3kT_c^\alpha/(S^\alpha + 1)$, and q_m is the equivalent radius of the two dimensional first Brillouin zone, $q_m = \sqrt{4\pi n}$, where n is the density of atoms at an interface. In addition, for an isotropic Heisenberg Hamiltonian in two dimensions the number of magnons excited at finite temperature is infinite. Therefore, we need to introduce a lower wavelength cutoff E_c to avoid this divergence. Physically this cutoff represents either anisotropy, which is present for spins at the interfaces between the magnetic electrodes and the insulating spacer, or a finite coherence length due to, for example, grain boundaries. Before we present a numerical calculation for the voltage dependence of conductances at arbitrary temperatures, we consider two limiting cases.

First, the voltage dependence of conductance $G(V) \equiv j/V$ at low temperature is readily obtained, because $n_{\mathbf{q}} = 0$ and only the first term in Eq. (7) survives. By combining them with Eq. (4) we find

$$G_P(V) = \frac{4\pi e^2}{\hbar} \left\{ [|T^d|^2 + (S_L^2 + S_R^2)|T^J|^2](\rho_L^M \rho_R^M + \rho_L^m \rho_R^m) + |T^J|^2 \left(S_L \frac{eV}{E_m^L} + S_R \frac{eV}{E_m^R} \right) \rho_L^m \rho_R^M \right\} \tag{8}$$

for $eV < E_m^\alpha$. For large voltage, $eV > E_m^\alpha$, one replaces the last term in brackets by $S_L(2 - E_m^L/eV) + S_R(2 - E_m^R/eV)$. Similarly, we have derived the conductivity for the AP alignment, and find for $eV < E_m^\alpha$

$$G_{AP}(V) = \frac{4\pi e^2}{\hbar} \left\{ [|T^d|^2 + (S_L^2 + S_R^2)|T^J|^2](\rho_L^M \rho_R^m + \rho_L^m \rho_R^M) + |T^J|^2 \left(S_L \frac{eV}{E_m^L} \rho_L^m \rho_R^m + S_R \frac{eV}{E_m^R} \rho_L^M \rho_R^M \right) \right\}. \tag{9}$$

Note that from the definition of E_m [after Eq. (7)] that the voltage dependence of the conductance is scaled by the Curie temperature T_c of the spins at the interfaces.

As one can see from Eqs. (8) and (9), the conductances increase linearly with voltage at small bias. This result is due to the fact that the magnon density of states in two dimensions is a constant. Such linear dependence on the voltage is quite general, as it has been pointed out by Kirtley and Scalapino [17] in their inelastic-tunneling model for the linear conductance background in the high-T_c superconductors. In our model, however, we are able to further determine the increase of conductivities for P *and* AP alignments without introducing new parameters. In fact, we can readily see from Eqs. (8) and (9) that the increase of the conductance is faster for AP, because the *increase* of conductances are proportional to $\rho_L^M \rho_R^M + \rho_L^m \rho_R^m$ for AP and to $\rho_L^M \rho_R^m + \rho_L^m \rho_R^M$ for P. Therefore, we conclude that the JMR decreases when one increases the voltage bias.

The second limiting case is the temperature dependence of the resistance at zero voltage. By taking the limit $V \to 0$ in Eq. (7) we find that the conductivity at the temperature greater than E_c is

$$G^\gamma(V,T) = G^\gamma(V,0) - \frac{8\pi e^2 S k_B T}{\hbar E_m} B^\gamma c_1(E_c), \tag{10}$$

where, for simplicity, we assumed that the two electrodes are identical (i.e., $\rho_L^{M(m)} = \rho_R^{M(m)}$) and γ stands for P and AP configurations; $B^{AP} = (\rho^m)^2 + (\rho^M)^2$ and $B^P = 2\rho^m \rho^M$, and $c_1(E_c) \equiv \ln(1 - e^{-E_c/k_B T}) \approx -\ln(k_b T/E_c)$ for $E_c < k_B T$. Therefore, the conductivities for both P and AP configurations vary as $T \ln T$ at high temperatures. Since the prefactor B^γ in Eq. (10) is larger for AP alignment, the conductance increases faster with temperature for AP alignment and the magnetoresistance is reduced at higher temperatures. We should point out that the cutoff energy E_c is not a sensitive parameter in determining the absolute value of the temperature dependence of the conductivity because it enters in the logarithm.

We now show in Fig. 2, a comparison between our theory and experimental data of the resistances of P and AP alignments for a junction $Co/Al_2O_3/CoFe$ at two

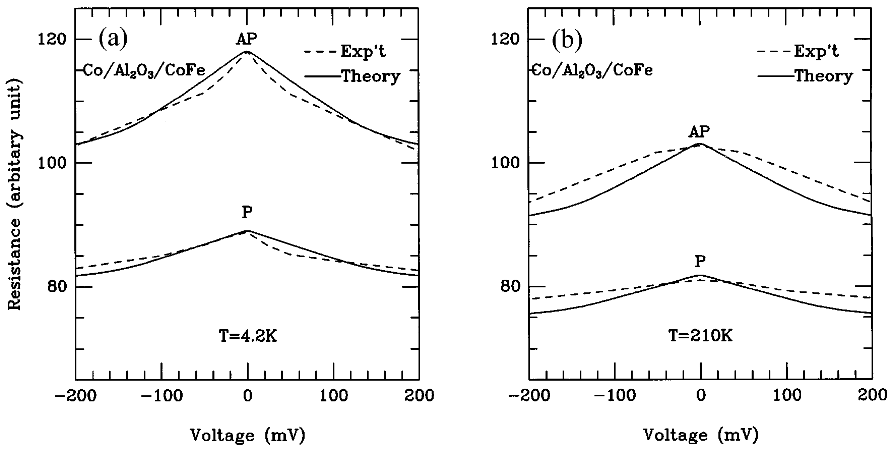

FIG. 2. Resistance as a function of voltage bias for P (lower curves) and AP (upper curves) alignments of magnetization of the two electrodes made of Co and CoFe at two temperatures, (a) 4.2 K and (b) 210 K. Dashed lines are experimental data; solid lines are theoretical results calculated from Eqs. (4) and (7). The spin polarizations of the density of the states used for Co ($\rho^M/\rho^m = 2.1$) and CoFe ($\rho^M/\rho^m = 2.2$) are derived from zero bias magnetoresistance of the junctions Co/Al_2O_3/Co and CoFe/Al_2O_3CoFe, which are consistent with Ref. [9]. We take spin $S = 3/2$, 110 meV for the Curie temperature of Co and CoFe, the magnon cutoff energy $E_c = 4$ meV, and $|T^d|^2/|T^J|^2 = 17$.

temperatures $T = 4.2$ K and $T = 210$ K. The theoretical curves (solid lines) in Fig. 2 were derived from Eqs. (4) and (7) where we determined materials parameters of the electrodes entering this expression either by using their bulk values (Curie temperature) or by using independent measurements of zero bias JMR of the junctions Co/Al_2O_3/Co and CoFe/Al_2O_3/CoFe. Therefore we have only *one* adjustable parameter, i.e., $|T^d|^2/|T^J|^2$, to fit overall voltage and temperature dependence for both P and AP alignments. For Fig. 2, we have chosen $|T^d|^2/|T^J|^2 = 17$ which is entirely reasonable since $|T^d|$ is determined by the overlap of the wave functions within the barrier while $|T^J|$ by the overlap of the wave function from one electrode at the barrier interface with the other electrode; therefore, it is expected that $|T^d|^2$ should be 1 to 2 orders of magnitude larger than $|T^J|^2$. The *one-parameter* fit to the data is reasonable for voltage smaller than 200 mV. To reproduce the disappearance as one goes from 4.2 to 210 K of the cusplike feature in the resistance, with a width of about 50 mV [compare Figs. 1(a) and 1(b)], it is necessary to introduce another adjustable parameter: a variable temperature dependent low energy cutoff in the magnon spectrum. This has been previously proposed by Kirtley and Scalapino [17].

In summary, we have proposed a new mechanism which is able to reproduce the zero bias anomaly in the conductance and magnetoresistance of ferromagnetic tunnel junctions. We are currently extending our study by including the energy dependence of transfer matrix elements and density of states at larger voltage bias, and by considering other inelastic-tunneling processes.

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