

Spin-flip scattering in magnetic junctions

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Processes which flip the spin of an electron tunneling in a junction made up of magnetic electrodes are studied. It is found that (i) magnetic impurities give a contribution which increases the resistance and lowers the magnetoresistance, which saturates at low temperatures. The conductance increases at high fields. (ii) Magnon assisted tunneling reduces the magnetoresistance as $T^{3/2}$ and leads to a nonohmic contribution to the resistance which goes as $V^{3/2}$. (iii) Surface antiferromagnetic magnons, which may appear if the interface has different magnetic properties from the bulk, gives rise to T^2 and V^2 contributions to the magnetoresistance and resistance, respectively. (iv) Coulomb blockade effects may enhance the magnetoresistance, when transport is dominated by cotunneling processes. [S0163-1829(98)04637-2]

I. INTRODUCTION

Magnetic junctions made up of fully saturated ferromagnets have attracted a great deal of attention, as they may lead to large magnetoresistance at low fields. This effect should be particularly enhanced in fully polarized magnets.^{1,2} The tunneling probability between two fully polarized electrodes whose magnetization are at a relative angle θ goes as $\cos^2(\theta/2)$. By averaging this quantity, we find that the conductance in the absence of an applied field, when θ can have any value, is one half of the conductance in the presence of a field, which aligns the magnetization and makes $\theta=0$. This simple analysis predicts that the maximum allowed magnetoresistance is 100%. This upper bound remains to be achieved in experiments, although enhanced effects have been reported in different experimental setups.³⁻⁶ Spin tunneling is also expected to be dominant in transport through ceramic and granular systems, where enhanced magnetoresistance at low fields has also been reported.⁷⁻¹⁰ The relevance of magnetic scattering at the interface in perovskite manganites can also be inferred by comparing with transport in related materials which exhibit colossal magnetoresistance.¹¹

In the present work, we study various effects which may limit the observed magnetoresistance. The bound discussed above assumes that the transmitted electron has a well defined spin throughout the tunneling process. If the spin can flip as the electron hops from one electrode to the other, the observed magnetoresistance will be reduced with respect to the previous value. Thus, we shall consider processes which change the spin of the electron as it tunnels.

In the following section, we analyze the role of magnetic impurities which may be present in the interface region. For simplicity, we will assume that they are magnetic ions of the same type as those which exist in the bulk of the electrodes, that is, Mn or Cr.

Section III studies spin-flip processes which involve the excitation of bulk magnons during the tunneling process. In Sec. IV, we consider the possibility that the interface has different magnetic properties than the bulk.^{6,10,12,13,14} In particular, we analyze the influence of an antiferromagnetic layer

at the interface, which may be present if it is oxidized, for instance.

Section V analyzes the main consequences of Coulomb blockade on spin polarized tunneling. Coulomb blockade requires large charging energies and small dimensions. It can be relevant to transport in granular materials. The main conclusions are presented in the last section.

II. MAGNETIC IMPURITIES

The current between the electrodes may arise from direct electron transfer or by processes in which electrons hop into impurity levels within the barrier between the electrodes. If the impurities are magnetic, these processes can change the spin of the electron, and modify the observed magnetoresistance. We will mostly assume the contribution of magnetic impurities to the total current, although at the end of the present section some comments are made on the features to be found in the opposite limit.

We consider impurities such as Mn^{3+} , Mn^{4+} , Cr^{3+} , and Cr^{4+} of the same kind as the ions present in the electrodes. These impurities have electronic levels at energies within the conduction band of the electrodes. Electrons or holes can hop from one electrode into these levels, and from there to the other electrode (see Fig. 1). We assume that the Hund's coupling between these electrodes and the core spins is much larger than any other scale in the problem. Thus, the hopping process only involves the highest spin state of the ion.

At zero temperature, an electron (or hole) which is initially polarized in the direction of the magnetization of one electrode has a finite amplitude of hopping into the impurity. In a basis aligned with the impurity spin, the initial state of the impurity is $|\Psi_0\rangle = |S, S\rangle$. After an electron hops from the left electrode, it becomes

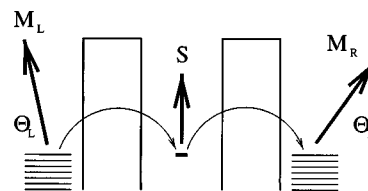


FIG. 1. Sketch of the elastic spin flip processes mediated by impurities at the interface. See text for details.

$$|\Psi\rangle = \cos\left(\frac{\theta_{LI}}{2}\right) \left|S + \frac{1}{2}, S + \frac{1}{2}\right\rangle + \frac{\sin(\theta_{LI}/2)}{\sqrt{2S+1}} \left|S + \frac{1}{2}, S - \frac{1}{2}\right\rangle + \dots, \quad (1)$$

where θ_{LI} is the relative angle between the magnetization of the left electrode and the initial spin of the impurity. Expression (1) leaves out states where the spin of the impurity is not maximum.

Let us now assume that the same electron hops coherently into the right electrode. The spin of the electron must be parallel to the magnetization of the right electrode. The direction of the magnetization forms an angle θ_{IR} with respect to the spin of the impurity, and an azimuthal angle ϕ with respect to the plane formed by the impurity spin and the magnetization of the right electrode. Then, the final state of the impurity is

$$\left[\cos\left(\frac{\theta_{LI}}{2}\right) \cos\left(\frac{\theta_{IR}}{2}\right) + \frac{\sin(\theta_{LI}/2)\sin(\theta_{IR}/2)e^{i\phi}}{2S+1} \right] |S, S\rangle + \frac{\sqrt{2S} \sin(\theta_{LI}/2)\cos(\theta_{IR}/2)}{2S+1} |S, S-1\rangle. \quad (2)$$

In the previous analysis we assumed that the impurity can accept an electron, as for Mn^{4+} . A hole current, in the opposite direction, can take place through ions such as Mn^{3+} . The corresponding calculation is straightforward, except that the angles are interchanged.

Finally, the probability that an electron hops between the two electrodes after a process such as the one described above is

$$\mathcal{T} \propto \left\langle \left[\cos\left(\frac{\theta_{LI}}{2}\right) \cos\left(\frac{\theta_{IR}}{2}\right) + \frac{\sin(\theta_{LI}/2)\sin(\theta_{IR}/2)e^{i\phi}}{2S+1} \right]^2 \right\rangle + \frac{2S}{(2S+1)^2} \left\langle \sin^2\left(\frac{\theta_{LI}}{2}\right) \cos^2\left(\frac{\theta_{IR}}{2}\right) \right\rangle, \quad (3)$$

where the brackets denote thermal averages over the values of the angles θ_{LI} , θ_{IR} , and ϕ . In the case that the direction of the impurity spin is totally random, we find that $\mathcal{T} \propto (2S^2 + 3S + 1)/2(2S + 1)^2$. For $S = \frac{3}{2}$, $\mathcal{T} \propto \frac{5}{16}$. This value increases in an applied field, as the impurities tend to be aligned by it. The corrections can be obtained by performing the averages in Eq. (3) in the presence of a field. Expanding, we find

$$\mathcal{T} = \begin{cases} \frac{2S^2 + 3S + 1}{2(2S + 1)^2} + \frac{\mu_0 SH}{k_B T} \frac{S^2 + S}{4(2S + 1)^2} + \left(\frac{\mu_0 SH}{k_B T}\right)^2 \frac{2S^2 + S + 1}{8(2S + 1)^2} + \dots, \\ \frac{\mu_0 SH}{k_B T} \ll 1, \\ 1 - \frac{k_B T}{\mu_0 SH} \frac{4S^2 + 1}{2(2S + 1)^2} + \left(\frac{k_B T}{\mu_0 SH}\right)^2 \frac{1}{(2S + 1)^2} + \dots, \\ \frac{\mu_0 SH}{k_B T} \gg 1. \end{cases} \quad (4)$$

By comparing the transmission with and without a field, we find that the contribution of impurity scattering can be included into the magnetoresistance as

$$\frac{\Delta\sigma}{\sigma} = \frac{\sigma_0(H, T) - \sigma_0(0, T) + \sigma_I(\mu_0 SH/k_B T)[(S^2 + S)/4(2S + 1)^2]}{\sigma_0(H, T) + \sigma_I\{(2S^2 + 3S + 1)/2(2S + 1)^2 + (\mu_0 SH/k_B T)[(S^2 + S)/4(2S + 1)^2]\}}. \quad (5)$$

This expression is valid to lowest order in $\mu_0 SH/k_B T$. In this expression, σ_0 stands for all contributions to the conductance other than those due to the impurities, and σ_I is the conductance through channels which involve resonant tunneling at the impurities. Exactly at resonance, each channel contributes with a term $\approx e^2/\hbar$. The magnetoresistance, when scattering by magnetic impurities is allowed, is reduced by a factor proportional to σ_I/σ_0 , and it is temperature independent at low fields. As these processes are elastic, they do not induce a dependence on applied voltage. The crossover between the low and the high field regime takes place when $\mu_0 SH/k_B T \sim 1$. If $S = \frac{3}{2}$ and $T = 300$ K, this field

is 60 T, while for $T = 4$ K, the field is 0.8 T. The corresponding figures for $S = \frac{1}{2}$ impurities are 180 T and 2.4 T.

Finally, if most of the current was due to resonant tunneling through magnetic impurities, the observed magnetoresistance will increase, instead of decreasing as in the previous case. A magnetic field will align the spin of the impurities with the magnetization of the electrodes. The impurities behave in a similar way to a third magnetic layer located between the electrodes. For instance, the current which flows from tunneling through $S = \frac{3}{2}$ impurity levels whose magnetic moments are oriented at random is $\frac{5}{16}$ that of the current when the moments are aligned by a field. The corre-

sponding figure for tunneling between magnetic electrodes is $\langle \cos^2(\theta/2) \rangle = \frac{1}{2}$. A possible situation where of the current in a magnetic junction is due to sequential (not resonant) tunneling through impurity levels is reported in Ref. 14.

III. SPIN-FLIP PROCESSES INDUCED BY BULK MAGNONS

The spin of the tunneling electron can be changed by the creation or absorption of magnons in the electrodes. These processes increase the tunneling probability between electrodes whose magnetizations are not aligned and reduce the observed magnetoresistance. We will consider that the rate of magnon induced tunneling is independent of the relative angle between the magnetization in each electrode.

If the barrier width is of length d , the electron, after a tunneling event, will be spread in a region of size d in the electrode it has hopped to. We write the electron creation operator as $\int f(r) \psi^\dagger(r) b_s^\dagger(r)$, in terms of a spinless fermion ψ and a Schwinger boson of spin s , b .¹⁵ The function $f(r)$ gives the spatial extent of the wave function of the electron after the tunneling process and $\sim d$ is the thickness of the barrier. By expanding these operators into the normal modes of the electrodes, we find that, roughly, all magnons, $b_{\vec{k}s}$ with wavelengths larger than d can be created with equal probability.

At zero temperature only magnon creation is allowed. This is possible at finite junction voltages. An electron with energy ϵ above the chemical potential of the other electrode can create any magnon with energy $\epsilon' < \epsilon$, provided that the wavelength of the magnon $a(\epsilon'/J)^{1/2}$ is less than d (a is the lattice constant). The density of states of magnons in a ferromagnetic three-dimensional system is $D(\epsilon') \propto (1/J)(\epsilon'/J)^{1/2}$. Hence, the intensity due to magnon creation is

$$I(V) \sim \frac{1}{R} \int_0^V d\epsilon \int_0^{\min[\epsilon, J(a/d)^2]} d\epsilon' \frac{1}{J} \left(\frac{\epsilon'}{J} \right)^{1/2} \sim \begin{cases} \frac{V}{R} \left(\frac{V}{J} \right)^{3/2} & V \ll J \frac{a^2}{d^2}, \\ \frac{V}{R} \left(\frac{a}{d} \right)^3 & V \gg J \frac{a^2}{d^2}. \end{cases} \quad (6)$$

This contribution is to be added to the elastic conductance $I_0(V) = V/R \langle \cos^2(\theta/2) \rangle$. We assume that magnon induced tunneling is independent of the relative angle between the magnetization in the two electrodes. Hence, the term shown in Eq. (6) reduces the magnetoresistance at finite voltages.

At finite temperatures, the electrons which tunnel can excite magnons of energy below $k_B T$. Using the previous argument, the probability that an electron excites a magnon goes as $(k_B T/J)^{3/2}$. Thus, the differential conductivity at low voltages has a contribution which goes as

$$\delta\sigma_{\text{mag}} \sim \begin{cases} \frac{1}{R} \left(\frac{k_B T}{J} \right)^{3/2} & k_B T \ll J \frac{a^2}{d^2}, \\ \frac{1}{R} \left(\frac{a}{d} \right)^3 & k_B T \gg J \frac{a^2}{d^2}. \end{cases} \quad (7)$$

This term increases the conductance of the junction, and it is independent of the relative orientation of the magnetization of the electrodes. Hence, the observed magnetoresistance decreases, as T increases, as $(k_B T/J)^{3/2}$. This contribution has the same temperature dependence as the reduction of the magnetoresistance due to the decrease in the magnetization of the electrodes. The latter effect, however, does not give rise to nonlinear I - V characteristics.

IV. SPIN FLIP PROCESSES DUE TO MAGNONS AT THE INTERFACE

It is likely that in doped manganites or in CrO_2 , the surface has a different composition than the bulk. In addition, the double exchange mechanism is weaker at a surface, as the kinetic energy of the carriers is reduced. Both effects may reduce the tendency towards ferromagnetism, leading to antiferromagnetic behavior. Note that a change in the magnetic structure of the surface leads to modifications in the height of the tunneling barrier. It is, however, unlikely that a simple dependence of the height of the barrier on the magnetic surface energy can be found.¹⁶

The contribution of spin-flip processes due to interface antiferromagnons can be estimated in the same way as in the preceding section. The only difference is the change in the density of states, due to the different dispersion relation, and to the low dimensionality. For two-dimensional antiferromagnons, this quantity is $D(\epsilon) \propto 1/J_{\text{AF}}(\epsilon/J_{\text{AF}})$. The highest-energy plasmon which can couple to the tunneling electron has energy $\sim J_{\text{AF}}(a/d)$. Hence, the intensity depends on voltage as

$$I(V) \sim \begin{cases} \frac{V}{R} \left(\frac{V}{J_{\text{AF}}} \right)^2 & V \ll J_{\text{AF}} \frac{a}{d}, \\ \frac{V}{R} \left(\frac{a}{d} \right)^2 & V \gg J_{\text{AF}} \frac{a}{d}, \end{cases} \quad (8)$$

and, at finite temperatures, we find a contribution to the conductivity such as

$$\delta\sigma_{\text{surf}} \sim \begin{cases} \frac{1}{R} \left(\frac{k_B T}{J_{\text{AF}}} \right)^2 & k_B T \ll J_{\text{AF}} \frac{a}{d}, \\ \frac{1}{R} \left(\frac{a}{d} \right)^2 & k_B T \gg J_{\text{AF}} \frac{a}{d}. \end{cases} \quad (9)$$

As in the previous case, this effect reduces the observed magnetoresistance at finite temperatures. If $a \sim d$ and $k_B T \gg J_{\text{AF}}$, the contribution of processes mediated by magnons is comparable to the purely elastic conductance. In this limit, the magnetoresistance should tend to zero.

It has been argued that in tunnel junctions based on Co, ferromagnetic magnons are localized at the interface.¹⁷ The scheme used here can be applied to this case. By inserting the appropriate density of states, we recover the results reported in Ref. 17.

V. COULOMB BLOCKADE EFFECTS

Coulomb blockade reduces the conductance of granular systems at low temperatures.^{18,19} The charging energy required to add one electron to a grain, $E_C = e^2/C$, where C is

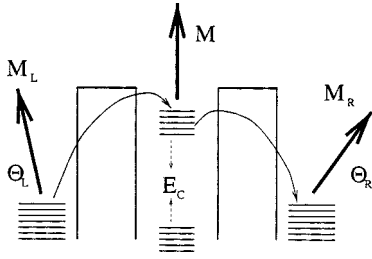


FIG. 2. Sketch of the cotunneling process through a grain with charging energy E_C .

the capacitance of the grain, is not negligible. It tends to open a gap when $k_B T \ll E_C$. The main process which suppresses this gap, at low temperatures, is inelastic cotunneling.²⁰ At finite temperatures or voltages, one electron can hop into a grain and leave it on a time scale shorter than $\hbar E_C^{-1}$, leaving an excited electron-hole pair of energy $\epsilon < k_B T, V$. A sketch of the process is depicted in Fig. 2. Cotunneling gives a conductance which goes as $(\hbar/e^2 R^2)(k_B T/E_C)^2$. This estimate is valid when a single small grain inserted between much larger grains blocks the current. If we consider N grains in series, the conductance due to cotunneling goes as $(1/R)(\hbar/e^2 R)^N (k_B T/E_C)^{N+1}$. A sketch of the process is depicted in Fig. 2.

Cotunneling requires two correlated hopping processes. In a fully polarized magnet, each hopping is reduced by a factor proportional to $\cos^2(\theta/2)$, where θ is the angle between the magnetization in the central grain and that in the right or left grains. Averaging over orientations, cotunneling is reduced by a factor $\frac{1}{2} \times \frac{1}{2} = \frac{1}{4}$, when the magnetizations are at random. The corresponding value for N grains of small capacitance is $1/2^{N+1}$. This factor becomes 1 when a magnetic field aligns the magnetization of the grains. Hence, cotunneling is more sensitive to magnetic disorder than direct tunneling. The magnetoresistance should increase when Coulomb blockade suppresses direct tunneling, and only cotunneling is allowed. Note that this effect is only important at very low temperatures, when activated processes over the Coulomb barrier are negligible. It should be much weaker in magnetic junctions where the electrodes are not fully polarized.²¹ An analysis of Coulomb blockade and cotunneling in devices where the electrodes are not fully polarized can be found in Ref. 22.

Sufficiently small grains behave similar to nonresonant (because of the Coulomb gap) magnetic impurities. Tunneling between misoriented grains is not totally suppressed, because the probability of changing the spin of the grain is not negligible, as discussed in Sec. II. This gives a limit to the maximum achievable magnetoresistance. Finite spin effects will be important for particles with a few conduction electrons, of radius $\approx 1 - 5$ nm or smaller.

At intermediate temperatures, when the contribution from cotunneling is small, the existence of a Coulomb gap leads to activated transport. In the presence of magnetic disorder, the activation energy also includes a contribution from spin flip processes, which must take place during the tunneling event.²³ This energy goes as the average magnon energy excited in the tunneling process, as discussed in Sec. III, $E_M \sim J(a^2/d^2)$. If we assume that all intergrain junctions are identical, this effect leads to a magnetoresistance which

should increase as $e^{E_M/k_B T}$. This process, however, is limited by cotunneling at low temperatures.

VI. CONCLUSIONS

The usual analysis of magnetic junctions assumes that conduction electrons tunnel elastically at the interface. The dependence of the conductance on the magnetic field arises from the differences in the density of majority and minority conduction electrons, which are modulated by the field. This approximation does not take into account processes which flip the spin of the conduction electrons as they move through the junction. Spin-flip processes reduce the magnetoresistance of junctions between fully polarized magnets. Their origin may be extrinsic, related to the different magnetic properties of the interfaces, or intrinsic, associated to the excitation of bulk magnons. In addition, they can be classified into elastic, as the scattering by magnetic impurities, or inelastic, which are mediated by magnetic excitations.

Elastic spin-flip processes could be due to magnetic impurities or other static deviations from perfect ferromagnetism, such as domain walls.²⁴ They are extrinsic, as they should not be present in perfect systems. They give rise to a temperature-independent reduction of the magnetoresistance. Assuming that the scattering by these imperfections leads to a loss of the spin orientation of the electron, the relative reduction in the magnetoresistance goes as σ_I/σ_0 , where σ_I is the contribution to the conductance from resonant tunneling via impurity states and σ_0 stands for the conductance due to other tunnel processes.

Inelastic spin-flip processes do not reduce the magnetoresistance at zero temperature and zero voltage, but give rise to nonohmic effects at finite voltages, and to changes in the conductance as function of temperature. We can distinguish between intrinsic effects, mediated by bulk magnons, and those related to magnetic excitations of the interface. Bulk magnons reduce the magnetoresistance at temperatures, or voltages, comparable to the bulk exchange coupling, which is of the order of the Curie temperature. The effect due to interface excitations shows up at the scale of the new couplings at the interface. In this work, we have considered the influence of an antiferromagnetic layer at the interface, but more complicated excitations may exist if the surface is strongly disordered.

Finally, we have analyzed the interplay of spin polarized tunneling and Coulomb blockade. We find that cotunneling processes enhance the magnetoresistance. This effect may be difficult to observe, due to the high resistance of junctions at the temperature when the Coulomb blockade is fully developed.

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