Assisted Tunneling in Ferromagnetic Junctions and Half-Metallic Oxides

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Different mechanisms of spin-dependent tunneling are analyzed with respect to their role in tunnel magnetoresistance (TMR). Microscopic calculation within a realistic model shows that direct tunneling in iron group systems leads to about a 30% change in resistance, which is close but lower than experimentally observed values. The larger observed values of the tunnel magnetoresistance (TMR) might be a result of tunneling involving surface polarized states. It is found that tunneling via resonant defect states in the barrier radically decreases the TMR by order of magnitude. One-magnon emission is shown to reduce the TMR, whereas phonons increase the effect. The inclusion of both magnons and phonons reasonably explains an unusual bias dependence of the TMR. The model presented here is applied qualitatively to half-metals with 100% spin polarization, where one-magnon processes are suppressed and the change in resistance in the absence of spin-mixing on impurities may be arbitrarily large. Even in the case of imperfect magnetic configurations, the resistance change can be a few 1000 percent. Examples of half-metallic systems are CrO$_2$/TiO$_2$ and CrO$_2$/RuO$_2$.

73.40.Gk, 73.40.Rw, 75.70.Pa, 85.70.Kh

Tunnel magnetoresistance (TMR) in ferromagnetic junctions, first observed more than a decade ago, is of fundamental interest and potentially applicable to magnetic sensors and memory devices. This became particularly relevant after it was found that the TMR for 3$d$ magnetic electrodes reached large values at room temperatures, and junctions demonstrated a non-volatile memory effect.

A simple model for spin tunneling, that has been formulated by Julliere and further developed in, is expected to work rather well for iron, cobalt, and nickel based metals. The microscopic model is in good agreement with experimental results on bulk polarizations and measured and calculated Fermi surfaces of 3$d$ metals. However, it disregards important points such as impurity-assisted and inelastic scattering, tunneling into surface states, and a reduced effective mass of carriers inside the barrier. These effects are important for proper understanding of the behavior of actual devices, like peculiarities in their $I-V$ curves and will be analyzed below. A couple of half-metallic systems, which could in principle achieve an ultimate magnetoresistance at room temperatures and low fields, will be discussed.

The model that we will consider below includes a Hamiltonian $H_0$ for non-interacting conducting spin-split electrons separated by a barrier, electron-phonon interaction $H_{ap}$, and exchange interaction of carriers with localized $d_i$ electrons $H_x$, the latter giving rise to an electron-magnon interaction. Impurity term $H_i$ will correspond to a short-range confining potential producing defect states in the barrier,

$$H = H_0 + H_{ap} + H_x + H_i. \quad (1)$$

Tunneling current is then evaluated within a general linear response formalism. Magnetoresistance (MR) is a relative change in a junction conductance with respect to the change of mutual orientation of spins from parallel ($G^P$) to antiparallel ($G^{AP}$). The MR depends only on effective polarization $P_{FB}$ of tunneling electrons.

$$MR = \frac{G^P - G^{AP}}{G^{AP}} = \frac{2P_{FB}^P}{1 - P_{FB}^P}. \quad (2)$$

The most striking feature of Eq. (2) is that the MR tends to infinity when both electrodes are made of a 100% spin-polarized material ($P = P' = 1$), because of a gap in the density of states (DOS) for minority carriers. Such half-metallic behavior is rare, but some materials possess this amazing property, most interestingly the oxides CrO$_2$ and Fe$_3$O$_4$. These oxides have potential for future applications in combination with lattice-matching materials, as illustrated below.

The full calculation of a TMR within microscopic model due to direct tunneling can be performed numerically and it gives a value of about 30% at low biases. Note that electric field present in a biased barrier skews its shape, thus making it more transparent for ‘hot’ electrons tunneling at energies where the difference between the DOS of majority and minority carriers is reduced. As a result the TMR in the direct tunneling decreases with the increased bias.

In a half-metallic case we obtain zero conductance $G^{AP}$ in the AP configuration at biases within the half-metallic band gap. Even at 20° deviation from the AP configuration, the value of MR exceeds 3,000% within the half-metallic gap, and this is indeed a very large value.

Presence of impurity/defect states in the barrier would result in a resonant tunneling of electrons. Comparing the direct and the impurity-assisted contributions to conductance, it is easy to see that the latter dominates when...
the density of impurity states exceeds \( \sim 10^{17} \text{cm}^{-3} \text{eV}^{-1} \). When impurities take over, the magnetoresistance is again given by the Julliere’s formula where the effective polarization \( \Pi \) is that of impurity ‘channels’.

\[
\text{MR}_1 = 2\Pi \Pi (1 - \Pi \Pi),
\]

and it gives a value of about 4\% for \( \text{MR}_1 \) in the case of Fe with non-magnetic impurities. The value of MR is reduced since generally \( \Pi < P_{FB}^{\uparrow} \) because of mixing of the tunneling electron wave function with non-polarized defect states. In the case of magnetic impurities (spin-flip centers) the TMR will be even smaller. At the same time, conductance may be substantially increased. These predictions have been confirmed experimentally.

Resonant diode type of structure gives similar results.

Direct tunneling, as we have seen, gives TMR of about 30\%, whereas in recent experiments TMR is well above this value, approaching 40\%. It would become clear below that this enhancement is unlikely to come from the inelastic processes.

Up to now we have disregarded the possibility of localized states at metal-oxide interfaces. Keeping in mind that the usual barrier AIO\(_x\) is amorphous, the density of such states may be high. We have to take into account tunneling into/from those states. The corresponding tunneling MR is found to be

\[
\frac{G_{\text{ins}}(\theta)}{A} = \frac{\pi^2}{\hbar} B \Pi (1 + P_{FB} P_s \cos(\theta)),
\]

\[
P_s = \frac{D_{s\uparrow} - D_{s\downarrow}}{D_{s\uparrow} + D_{s\downarrow}},
\]

\[
\Pi = \frac{1}{2}(D_{s\uparrow} + D_{s\downarrow}),
\]

where \( P_s \) is the polarization and \( \Pi \) is the average density of surface states, and \( \theta \) is the mutual angle between moments on electrodes. The parameter \( B \sim [2\pi^2 \hbar^2 m_k/(m_2^2 \nu)] \exp(-2\nu w) \), where \( w \) is the barrier width, \( \kappa \) is the absolute value of electron momentum under the barrier, \( m \) and \( m_2 \) are the free electron mass and the effective mass in the barrier, respectively. The corresponding magnetoresistance would be \( \text{MR}_{\text{ins}} = 2P_{FB} P_s / (1 - P_{FB} P_s) \).

It is easy to show that the bulk-to-surface conductance exceeds the bulk-to-bulk one at densities of surface states \( D_{s\uparrow} > D_{sc} \sim 10^{13} \text{cm}^{-2} \text{eV}^{-1} \) per spin, comparable to those found at some metal-semiconductor interfaces.

If on both sides of the barrier the density of surface states is above the critical value \( D_{sc} \), the magnetoresistance would be due to surface-to-surface tunneling with a value given by \( \text{MR}_{\text{s-s}} = 2P_{s\uparrow} P_{s\downarrow} / (1 - P_{s\uparrow} P_{s\downarrow}) \). If the polarization of surface states is larger than that of the bulk, as is often the case even for imperfect surfaces, then it would result in enhanced TMR.

Inelastic processes with excitation of magnon or phonon modes introduce new energy scales into the problem (30-100 meV) which correspond to a region where unusual \( I - V \) tunnel characteristics are seen (Fig. 1). We obtain for magnon-assisted inelastic tunneling current at \( T = 0 \):

\[
I^\uparrow = \frac{2\pi e}{h} \sum^\uparrow \frac{X^\alpha g_L^\alpha g_R^\alpha}{\int d\omega \rho_{\alpha}^{\text{mag}}(\omega)(eV - \omega)\theta(eV - \omega)},
\]

\[
I^\uparrow_{\text{AP}} = \frac{2\pi e}{h} \left[X^R g_L^R g_R^R \int d\omega \rho_{\alpha}^{\text{mag}}(\omega)(eV - \omega)\theta(eV - \omega) + X^L g_L^R g_R^R \int d\omega \rho_{\alpha}^{\text{mag}}(\omega)(eV - \omega)\theta(eV - \omega)\right],
\]

where \( X \) is the incoherent tunnel exchange vertex, \( \rho_{\alpha}^{\text{mag}}(\omega) \) is the magnon density of states that has a general form \( \rho_{\alpha}^{\text{mag}}(\omega) = (\nu + 1)\omega/\omega_{0}^{\nu+1} \), the exponent \( \nu \) depends on a type of spectrum, \( \omega_0 \) is the maximum magnon frequency, \( g_{\alpha}(\omega) \) marks the corresponding electron density of states on left (right) electrode, \( \theta(x) \) is the step function, \( \alpha = L, R \).

For phonon-assisted current at \( T = 0 \) we obtain

\[
I^\uparrow_{\alpha} = \frac{2\pi e}{h} \sum^\alpha \frac{\pi e}{\bar{\omega}} \int d\omega \rho_{\alpha}^{\text{ph}}(\omega) P_\alpha(eV - \omega)\theta(eV - \omega),
\]

\[
I^\uparrow_{\text{AP}} = \frac{2\pi e}{h} \sum^\alpha \frac{\pi e}{\bar{\omega}} \int d\omega \rho_{\alpha}^{\text{ph}}(\omega) P_\alpha(eV - \omega)\theta(eV - \omega),
\]

where \( P_\alpha \) is the phonon vertex, \( P_\alpha / X = \gamma_\omega / \omega_D \), where \( \gamma_\omega \) is the constant and \( \omega_D \) is the Debye frequency, \( a \) is the spin index, \( \rho_{\alpha}^{\text{ph}} \) is the phonon density of states, \( \alpha \) marks electrodes and the barrier.

The elastic and inelastic contributions together define the total junction conductance \( G = G(V,T) \) as a function of the bias \( V \) and temperature \( T \). We find that the inelastic contributions from magnons and phonons \( G_{\alpha}(V,0) \), respectively, grow as \( G^\alpha(V,0) \propto (eV/\omega_0)^{\nu+1} \) and \( G_{\text{ph}}(V,0) \propto (eV/\omega_D)^{\nu} \) at low biases. These contributions saturate at higher biases: \( G^\alpha(V,0) \propto 1 - \frac{1}{2} \frac{\omega_0}{\nu eV} \) for \( eV > \omega_0 \) and \( G_{\text{ph}}(V,0) \propto 1 - \frac{1}{4} \frac{\omega_0}{\nu eV} \) at \( |eV| < \omega_0 \). This behavior would lead to sharp features in the \( I - V \) curves on a scale of 30-100 mV (Fig. 1).

It is important to highlight the opposite effects of phonons and magnons on the TMR. If we take the case of the same electrode materials and denote \( D = g_\uparrow \) and \( d = g_\downarrow \) then we see that \( G_{\text{ph}}^\downarrow(V,0) - G_{\text{ph}}^\uparrow(V,0) \propto -(D - d)^2(eV/\omega_0)^{\nu+1} < 0 \), whereas \( G_{\text{ph}}^\uparrow(V,0) - G_{\text{ph}}^\uparrow(V,0) \propto (D - d)^2(eV/\omega_D)^{\nu} > 0 \), i.e. spin-mixing due to magnons decreases the TMR, whereas the phonons tend to reduce the negative effect of magnon emission. Different bias and temperature dependence can make possible a separation of these two contributions, which are of opposite sign.

At finite temperatures we obtain the contributions of the same respective sign as above. For magnons: \( G_{\text{ph}}^\uparrow(0,T) - G_{\text{ph}}^\uparrow(0,T) \propto -(D - d)^2(TdM/dT) < 0 \), whereas \( M = M(T) \) is the magnetic moment of the electrode at a given temperature \( T \). The phonon contribution is given by a standard Debye integral with the following results: \( G_{\text{ph}}^\downarrow(0,T) - G_{\text{ph}}^\uparrow(0,T) \propto \).
\[(D - d)^2(T/\omega_D)^4 > 0 \text{ at } T \ll \omega_D, \text{ and } G_{ph}^0(0,T) = G_{ph}^0(0,T) \propto +(D - d)^2(T/\omega_D) \text{ at } T \gtrsim \omega_D.\] It is worth mentioning that the magnon excitations are usually cut off by, e.g., the anisotropy energy \(K_{an}\) at some \(\omega_r\). Therefore, at low temperatures the conductance at small biases will be almost constant.

It is very important that in the case of half-metallics \(P_{FB} = 1\), and even with an imperfect barrier the magnetoresistance can, at least in principle, reach any value limited by only spin-flip processes in the barrier/interface. We should note that the one-magnon excitations in half-metallics are suppressed by the half-metallic gap in electron spectrum. Spin-mixing can only occur on magnetic impurities in the barrier or interface, the allowed two-magnon excitations in the electrodes do not result in spin-mixing.

The examples of systems with half-metallic behavior are \(\text{CrO}_2/\text{TiO}_2\) and \(\text{CrO}_2/\text{RuO}_2\) (Fig. 1). They are based on half-metallic \(\text{CrO}_2\), and all materials have the rutile structure with almost perfect lattice matching. This should yield a good interface and help in keeping the system at desired stoichiometry. \(\text{TiO}_2\) (\(\text{RuO}_2\)) are used as the barrier (spacer) oxides. The half-metallic behavior of the corresponding multilayer systems is demonstrated by the band structures calculated within the linear muffin-tin orbitals method (LMTO) in a supercell geometry with \([001]\) growth direction (Fig. 2). The calculations show that \(\text{CrO}_2/\text{TiO}_2\) is a perfect half-metallic, whereas \((\text{CrO}_2)_2/\text{RuO}_2\) is a weak half-metallic, since there is some small DOS around \(E_F\).

The main concerns for achieving a very large value of magnetoresistance with half-metallics will be spin-flip centers and imperfect alignment of moments, provided that other, e.g., many-body, effects are small. As for conventional tunnel junctions, the present results show that the defect states in the barrier, or a resonant state like in a resonant tunnel diode type of structure, reduces their magnetoresistance by several times but may dramatically increase the current through the structure. The inelastic processes are responsible for the unusual shape of the \(I-V\) curves at low biases, and their temperature behavior, which is also affected by impurity-assisted tunneling. The surface states assisted tunneling may lead to enhanced TMR, if their polarization is higher than that of the bulk. This could open up ways to improving performance of ferromagnetic tunnel junctions.

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