Bias dependence in spin-polarized tunneling

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We discuss the transport of electrons through ferromagnetic tunnel junctions. The spin-up and spin-down chemical potentials are different at the insulator-ferromagnet interfaces by different amounts between the parallel and the antiparallel configuration. As a result, the tunneling probabilities for the spin-up and spin-down channels change differently as the external voltages are increased. There is a strong bias dependence of the magnetoresistance ratio, consistent with experimental results. [S0163-1829(97)04810-8]

Several groups have recently observed magnetoresistance ratio of 18 (Ref. 1) and 15.6% (Ref. 2) in ferromagnet tunnel junctions at room temperature. These follow earlier work by Julliere[3] who observed a ratio of 14% in the conductance at 4.2 K and zero bias. The magnetoresistance ratio are comparable to corresponding values in the giant magnetoresistance in ferromagnetic multilayers. Because the resistance of the tunnel junction is much higher than that of the multilayers, the power consumed will be much less. This has generated interest in the practical application of the tunnel junctions. One of the puzzling features in the tunneling experiments is the large decrease of the magnetoresistance as the external voltage is increased. In a simple picture, one expects the conductance G_s for electrons of spins s to be proportional to the tunneling probability: $G_s \propto N_s^< N_s^> \exp(-\kappa d)$ where $N_s^>$ is the density of states to the left of the junction; d is the thickness; κ is the imaginary wave vector in the barrier. Here the superscripts $<,>$ denote the left- and the right-hand side of the junction, respectively. As the external voltage is increased, the barrier height and hence κ will decrease, thereby increasing the conductance. However, this change seems to be the same no matter what the spin s is. Thus in this simple picture the magnetoresistance ratio is determined by the product of densities of states and remains unchanged as the external voltage is increased.

Theoretical study of the magnetoresistance in tunneling has mostly not considered the effect of electron interaction. As we know from the classic study of the Schottky barrier (a metal-insulator interface), it is essential to include the screening of the charges. For the Schottky barrier, a dipole layer is developed because of the difference in work function between the insulator and the metal. In the present case, the external potential can induce a dipole layer with a magnitude proportional to the voltage in a corresponding manner. This is even more interesting for ferromagnetic junctions because the charge and spin degrees of freedom are coupled together. We have recently analyzed the effect of electron interaction on tunnel junctions using the Boltzmann-Vlasov-Landau (BVL) equation.[4,5] In this paper, we explain and explore some of the physical implications of that approach. In particular, we found that there is a splitting between the spin-up and spin-down chemical potential that is proportional to the external field throughout the ferromagnets. As a result the chemical potential difference and hence κ is spin dependent. A large bias dependence of the magnetoresistance ratio is obtained. This is illustrated in Fig. 1 where we plotted the magnetoresistance ratio normalized by the zero-field value as a function of the averaged resistance also normalized by the zero-field value for a model calculation with different system parameters. We next explain what we think is the essential physics.

One can think of the tunneling junction of total thickness L as a trilayer structure of two coupled ferromagnets on the left and on the right. We assume the z direction to be perpendicular to the interface which is located at $z=0$. There is an interfacial resistance for unit area of magnitude $r(1-\gamma s)$ for spin channel $s=\pm 1$ caused by the insulating barrier between the ferromagnets. (Thus r is the average resistance of the spin-up and the spin-down channel. $2\gamma r$ is the difference between these two channels.) This interfacial resistance can be obtained from the transmission matrix[6] and shall be approximated as proportional to the product of the density of states on the left and on the right in this paper.[9] Thus r, γ is different between the parallel and the antiparallel configurations. The interfacial resistance is of the order of kΩ for an area of 10^{-4} cm^2 and is much larger than the resistance of the ferromagnets r_F. Denote the chemical potential for spin s on the left (right) by $\mu_s^<$ ($\mu_s^>$) and the current density by the symbol J_s. Because of the high interfacial resistance $r(1-\gamma s)$, most of the voltage drop will occur at the interface. We obtain, from Ohm's law, $\Delta\mu_s = \mu_s^>(z=0) - \mu_s^<(z=-d) = r(1-\gamma s)J_s$. Now r, γ are functions of $\Delta\mu_s$. We expect, for a model with a barrier height of U and a width d, $r(1-\gamma s) = r_{0s}\exp\{d[\kappa_s(\mu) - \kappa_s(\mu=0)]\}$. Here the imaginary wave vector $\kappa_s \propto \hbar^2 \int_0^d dz \sqrt{U - \Delta\mu_s z/d}/2m_s$ (m_s is the mass of the electron of spin s) from the quantum mechanics of tunneling. Carrying out the integral, we thus get $\kappa_s = 2\kappa_{s0}U[1-(1-\Delta\mu_s/U)^{1.5}]/(3\Delta\mu_s)$, where $\kappa_{s0} = 4\pi\sqrt{2m_sU/\hbar^2}$, $r_{0s} = r_0(1-\gamma_0 s)$ is the resistance at zero bias. r_0, γ_0 are constants. If the chemical potentials are different between the spin-up and the spin-down electrons, from the spin dependence of the tunneling factor, it is natural to expect a bias dependence on the magnetoresistance ratio. We next point out why $\Delta\mu_+ \neq \Delta\mu_-$.

Associated with the change in the chemical potential, there is a change of the electron density of spin s by an amount given by $\delta\rho_s = \mu_s N_s$. This change in turn implies changes in the net charge (ρ) and magnetization (σ)

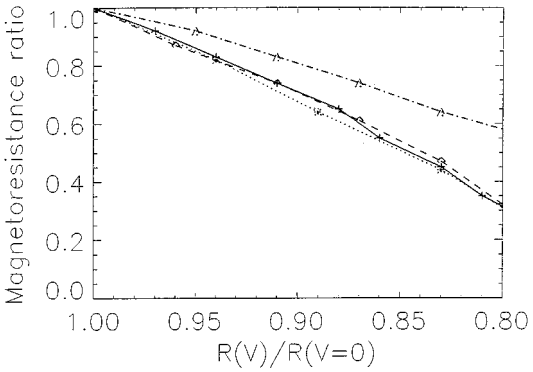

FIG. 1. Magnetoresistance ratio normalized by the zero-field value $[r_P(J_S)-r_{AP}(J_S)]/[r_P(J_S=0)-r_{AP}(J_S=0)]$ as a function of the averaged parallel and antiparallel resistance also normalized by the zero-field value, $[r_P(J_S)+r_{AP}(J_S)]/[r_P(J_S=0)+r_{AP}(J_S=0)]$. Here $r_P(J_S)$, $r_{AP}(J_S)$ are the resistance in the parallel and antiparallel configuration for total current J_S. The results are for the following choices of parameters: $\gamma=0.6$, $l_{sf}=50$, $\lambda=2$, and (solid line) $N_d/N_u=3$, $\kappa_0 d=2$, $\beta=0.8$, $b=0.8$ (dashed line), $N_d/N_u=3$, $\kappa_0 k=2$, $\beta=0.4$, $b=0.4$ (dotted line), $N_d/N_u=3$, $\kappa_0=3$, $\beta=0.8$, $b=0.8$ (dashed-dotted line), $N_d/N_u=4$, $\kappa_0 d=2$, $\beta=0.8$, $b=0.8$.

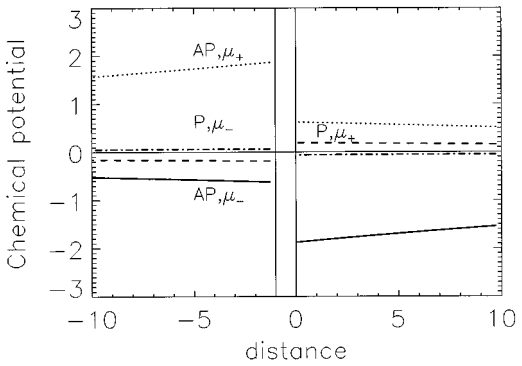

FIG. 2. The schematic behavior of the chemical potentials of the spin-up and the spin-down bands (solid lines) in a spin-polarized tunnel junction in an external field $V=0.25$. The chemical potential is expressed in units of the total voltage drop J_S/r where J_S is the total current. Quantities that change qualitatively when the magnetizations are changed from parallel to antiparallel are labeled by P and AP for the two configurations. The insulator is assumed to be from $x=-1$ to $x=0$ with its thickness $d=1$ in the present unit of distance.

densities given by $\delta\rho^{<}=\delta\rho_+^{<}+\delta\rho_-^{<}=N_+^{<}\mu_+^{<}+N_-^{<}\mu_-^{<}$, $\delta\sigma^{<}=\delta\rho_+^{<}-\delta\rho_-^{<}=N_+^{<}\mu_+^{<}-N_-^{<}\mu_-^{<}$ with similar expressions for the right-hand side of the junction where the superscript $<$ is replaced by $>$. $\delta\rho_s=(\rho+s\sigma)/2$.

We argue in the next paragraph that the physics requires that the net charge induced becomes very small, of the order of r_F/r times $N_s\mu_s$. This implies that the shifts in the chemical potentials are opposite in sign. More precisely, $\delta\rho=0.5\Sigma_s\delta\rho_s=0.5\Sigma_s\mu_s N_s\approx 0$. $\mu_+\approx -N_-\mu_-/N_+$. Since the voltage drop across the barrier is nonzero, the chemical potential change is thus different between the spin-up and the spin-down bands. As we explain below one of the essential physics is that the charge- and the spin-fluctuation decay lengths are different. Spin accumulation can also cause a splitting of the chemical potential.[7] The large induced charge and spin densities described here are due to shifts in the Fermi level and not to spin accumulation. For spin accumulation, an additional magnetization $\delta\sigma'$ of the order of $I_m T_2/\Omega$ is created on the right-hand side (of volume Ω) due to the injection magnetization current I_m from the left-hand side which takes a finite time T_2 to relax. This $\delta\sigma'$ is also present in addition to the $\delta\sigma$ discussed above but is of a much smaller magnitude. We now turn our attention to the net charge induced at the interface.

For an external electric field E, the current density can be expressed as a sum of a term $\rho_{sF}^{-1}E$ due to the external driving field in the metal and another term J_{s1} due to the potential caused by the difference between the induced charge densities and that due to the self-consistent screening potential W:[4] $J_s=J_{s1}+\rho_{sF}^{-1}E/2$; $J_{s1}\propto-\Sigma_p\tau_s\partial_z[\delta\rho_s/m_s-\chi_s W]/m_s$. Here ρ_{sF}, the resistivity of the ferromagnet, is given by $\rho_{sF}^{-1}=-\Sigma_p\tau_s\chi_0/2m_s^2$, $\chi_0=[\Sigma_s\tau_s\partial_e f_{0s}(e)/m_s^2]/(\Sigma_s\tau_s/2m_s^2)$ is a density of states factor. f_{0s} is the Fermi distribution function; τ_s, m_s are the relaxation time in the ferromagnet and the effective mass. $\chi_s=\partial_e f_{0s}/m_s$. We argue below that J_{s1} cannot be conserved unless the charge at the interface is small. Recall that the charge density of spin s is a sum of the total charge and magnetization densities $\delta\rho_s=(\rho+s\sigma)/2$. Away from the interface, the magnetization decays at a rate proportional to the renormalized[5] spin diffusion length l_{sf} (of the order of 100 Å) whereas the charge density decays at a faster rate proportional to the screening length λ (of the order of a few Å). Since the charge density dies off much faster than the magnetization density, the rate of change of $\delta\rho_s$ is approximately equal to that of $0.5\rho/\lambda$. J_{s1} is thus of the order of magnitude $\delta\rho\tau_s/m_s^2\lambda\approx\delta\rho/(\chi_0\rho_{sF}\lambda)$, on using the definition of the resistivity ρ_{sF}.

There are two resistance x (unit area) that enters into consideration: the interfacial resistance x (unit area) r and that of the ferromagnets $r_F=\rho_F l$. For problems of practical interest, for an area a of 10^{-4} cm^2, ra is of the order of kΩ,[1] whereas $\rho_F l a$ is of the order of $10^{-5}\Omega$. Thus $r\gg\rho_F l\gg\rho_F\lambda$. We expect that unless there is a cancellation, $\delta\rho_s$ and hence $\delta\rho$ is of the order of $N_s\mu_s\propto N_s r J_s$. Substituting this estimate of $\delta\rho$ into the estimate of J_{s1} in the previous paragraph, we found that J_{s1} is of the order of $rJ_s/\rho_F\lambda$ and is much larger than the current due to the external field. Because of the rapid decay of the charge densities, an electric field is created that may become much larger than the external field. This cannot be self-consistently sustained and Eq. (1) cannot be satisfied unless the change in the charge densities of each spin component are such that the net charge density is much smaller than $N_s\mu_s$.

In summary, the net charge induced is smaller than expected. This implies a splitting of the chemical potential of the spin-up and the spin-down electrons right at the interface. Because of this splitting, there is a bias dependence of the spin-dependent tunneling factor and hence the magnetoresistance. We next provide for the numerical detail of this paper.

Inside the ferromagnet, the relationship between the current and the charge densities can be obtained from the BVL equation:[8]

$$(m_s^{-1} p\cdot\nabla_R) f_s(p,R) - \int dr \int dp' \exp[i(p'-p)\cdot r]/(i(2\pi)^3)[U_{\rm eff}(s,R+r/2) - [U_{\rm eff}(s,R-r/2)]] f_s(p',R) = \partial f/\partial t|_{\rm collision}.$$

Here f_s is the distribution function for electrons with spin s, momentum p and located at R. The self-consistent potential is given by $U_{\rm eff}(s,R) = U_{{\rm ext},s} + W$, $W = \int dR' V(R-R') \int dp' \Sigma_s f_s(p',R')/(2\pi)^3$. $V(R) = e^2/R$ is the Coulomb interaction. $U_{{\rm ext},s}$ is the external potential. $\partial f/\partial t|_{\rm collision}$ is the phenomenological collision term that includes the relaxation of the electrons. We assume that the potential drop occurs mostly at the interface. Inside the ferromagnet, the potential drop is not strong. The BVL equation can be solved in a linear approximation by expressing the distribution function f as a sum of f_0 and a change which is linearly proportional to the external field. The details of this were described previously[4,5] and we summarized it briefly here. The current for spin component s can be written as $J_s = 0.5(J_S + sJ_D)$ in terms of a mean $0.5J_S$ and a difference J_D. After some calculations, from Eq. (2) we found that J_D can be expressed in terms of J_S at the interface as $J_D = BJ_S$ where $B = (b^> G^< + b^< G^>)/(G^< - G^>)$. Here $G = -0.5(b+\beta)/[\lambda^2 \rho_F (1-\beta^2)]$ where we have assumed that the resistivity of the ferromagnet for spin channel s, ρ_{sF}, can be written as $\rho_F(1-s\beta)$. b is a parameter that measures the difference in transport properties between the spin-up and the spin-down electrons given by $b = (\Sigma_s \tau_s \chi_s s/m_s)/(\Sigma_s \tau_s \chi_s/m_s)$. The parameter β for the asymmetry of the resistivity can be written in a similar fashion as $\beta = -\Sigma_s s\tau_s/2m_s^2 / \Sigma_s \tau_s/2m_s^2$. From Eq. (1) we obtain $\Delta\mu_s$ in terms of J_S, r, and γ. The last two quantities in turn depends on $\Delta\mu_s$ through the imaginary wave vector κ_s. In the presence of a finite external driving force, the system of equations can be solved self-consistently by iteration. Typical calculations converges to within 1% after two iterations. We have performed calculations for different choices of parameters. The results are similar. For illustrative purposes we describe below calculations performed for parameters $N_d/N_u = 3$, $\gamma - 0.6$, $\beta = 0.8$, $l_{sf} = 50$, $b = 0.8$, $\lambda = 2$, $\kappa_0 d = 2$.

In Fig. 2, we show the chemical potentials on both sides of the interface as a function of the position in the junction at a finite external voltage $V = 0.25$ when the magnetizations on opposite sides of the junction are parallel and antiparallel to each other. The highlights of our results are: (1) There is a splitting of the chemical potential proportional to the external voltage even at the interface. This suggests that the voltage drop and hence κ_s is a function of the spin s. The change in the chemical potentials is mostly controlled by the spin diffusion length, the net charge accumulated at the interface is quite small. (3) When the magnetizations are parallel (antiparallel) the chemical potentials of the spin-up and spin-down bands are switched (of the same sign) on opposite sides of the junction. When an external magnetic field turns the magnetizations from a parallel to an antiparallel configuration, the voltage drops and κ_s are changed by different amounts. This provides a reason for the voltage dependence of the magnetoresistance ratio. Points (1) and (2) have been explained previously. That point (3) is reasonable can be appreciated from the fact that $N_s^< = N_s^<(N_s^< = N_{-s}^>)$ in the parallel (antiparallel) configuration. This point was recently emphasized by us.[4]

In Fig. 1 we show the magnetoresistance ratio normalized by the zero-field value, $[r_{\rm P}(J_S) - r_{\rm AP}(J_S)]/[r_{\rm P}(J_S=0) - r_{\rm AP}(J_S=0)]$, $[r_{\rm P}(J_S), r_{\rm AP}(J_S)$ are the resistance in the parallel and antiparallel configuration for total current $J_S]$ as a function of the resistance averaged between the parallel and the antiparallel configurations, also normalized by the zero-field value, $[r_{\rm P}(J_S) + r_{\rm AP}(J_S)]/[r_{\rm P}(J_S=0) + r_{\rm AP}(J_S=0)]$, for different values of the input parameters. There is a substantial change of the magnetoresistance ratio as the averaged resistance is decreased. The trend and the order of magnitude of our result is the same as that of the experiment, for which the magnetoresistance ratio decreases by about 50% when the resistance has decreased by 50%. Our decrease is larger than the experimental values of Moodera *et al.*[1] but is consistent with recent unpublished results of the IBM-Brown group. The experimental junctions contain additional thin layers of Al or Mg between the ferromagnet and the insulator. Because of the simplicity of our model, we do not expect to see quantitative agreement between the two. In Fig. 1, different points correspond to increments of the applied voltage by 0.05. While the dependence of the magnetoresistance ratio and the resistance on the voltage is changed as the system parameters are changed, the dependence of the magnetoresistance ratio on the total resistance ratio depends only strongly on the ratio of density of states between the spin-up and the spin-down bands. For this reason, the magnetoresistance ratio is plotted against the averaged resistance ratio. Since the density of states ratio also controls the magnetoresistance, our result suggests that the higher the unbiased magnetoresistance, the less the bias dependence.

Because γ is a function of the external voltage, the chemical voltage can be a nonlinear function of the driving potential. The ratio of the chemical potential to the external driving voltage when the magnetizations on opposite sides of the junction are parallel is illustrated in Fig. 3. As we can see, the dependence of this ratio on the external voltage is quite weak. When the magnetizations are antiparallel, $\gamma = 0$ in our model, the nonlinear behavior disappears.

In this paper we have focused on the cases when the magnetizations are parallel and antiparallel to each other. It is possible to calculate the general situation where the magnetizations are at an angle θ with respect to each other. One expects Eq. (1) to be replaced by the equation $\mu_s^< - \mu_{s'}^> = r_{ss'} J_s$ where the generalized resistance $r_{ss'}$ can be calculated from the transmission matrix.[6] From symmetry considerations we expect the dependence to be a power series in $\cos\theta$.

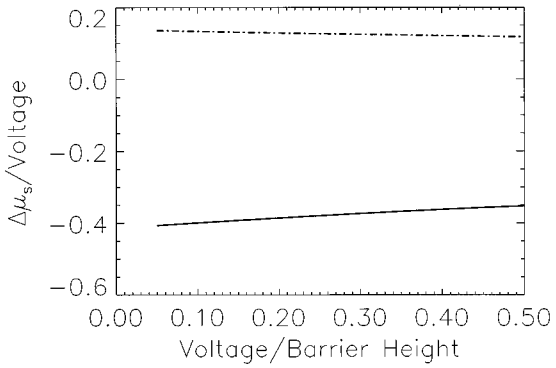

FIG. 3. Difference of chemical potential in the parallel configuration for the spin-up (solid) and spin-down (dotted-dashed line) bands in units of J_S/r as a function of the external voltage.

In summary, we found a splitting of the chemical potential between the spin-up and spin-down electrons proportional to the applied voltage at the metal-insulator interface. The splitting changes between the parallel and the antiparallel configuration. These effects affect the tunneling in a nonlinear manner and provide for a change in the magnetoresistance ratio that agrees with experimental observations in its trend and order of magnitude.

We also found that the charge induced at the insulator is much smaller than expected. Devices are often operated in the ac mode. The capacitance of the capacitor that is formed from the metal and the insulator is a quantity of interest. The small induced charge implies that the capacitance is also smaller than expected.

In real experimental systems, the barrier layer may contain impurity and surface states. If these states are nonmagnetic, they will not affect the magnetoresistance. If these states are magnetic in nature, they would seriously degrade the zero-bias magnetoresistance already. The possible effects of these impurities were not included in the present calculation for this prejudice and also because its study depends on details of the experimental sample, which is not available to the author.

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[1] J. S. Moodera, Lisa R. Kinder, Terrilyn M. Wong, and R. Meservey, Phys. Rev. Lett. **74**, 3273 (1995); J. S. Moodera and Lisa R. Kinder, J. Appl. Phys. **79**, 4724 (1996).

[2] T. Miyazaki, T. Yaoi, and S. Ishio, J. Magn. Magn. Mater. **98**, L7 (1991); T. Miyazaki and N. Tezuka, *ibid.* **139**, L231 (1995).

[3] M. Julliere, Phys. Lett. A **54**, 225 (1975).

[4] S. T. Chui, J. Appl. Phys. **80**, 1002 (1996); See also, S. T. Chui and J. Cullen, Phys. Rev. Lett. **74**, 2118 (1995); S. T. Chui, J. Magn. Magn. Mater. **151**, 374 (1995).

[5] S. T. Chui, Phys. Rev. B **52**, 3832 RC (1995).

[6] J. C. Slonczewski, Phys. Rev. B **39**, 6995 (1989). In terms of the transmission matrix T, $T_{\pm}=1/r(1-\pm\gamma)$.

[7] M. Johnson, Phys. Rev. Lett. **70**, 2142 (1993).

[8] L. Kadanoff and G. Baym, *Quantum Mechanics* (Benjamin, New York, 1962), Chap. 7.

[9] See, for example, G. D. Mahan, *Many Particle Physics* (Plenum, New York, 1981), Sec. 9.3.