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Voltage dependence of giant tunnel magnetoresistance in triple barrier magnetic systems

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Abstract. A quantum theory of the dependence on bias voltage of the tunnel magnetoresistance in a triple barrier system of the form MOMOMOM is presented. The Ms represent magnetic metallic layers and the Os are thin tunnel barriers. The two inner layers form spin-dependent quantum wells. The relative orientation of the magnetization in the successive magnetic layers can be changed from parallel to antiparallel. For a particular thickness of the inner metallic layers, a very large change of resistance occurs between the parallel and antiparallel magnetic configurations due to the spin dependence of the resonant tunnelling in these layers. It is shown that oscillations in the voltage dependence of the magnetoresistance amplitude take place associated with oscillations between resonant and antiresonant tunnelling as a function of the electrons' energy.

The recent observations of large magnetoresistance (MR) effects at room temperature in tunnel junctions of the form MOM′ (M and M′ = magnetic materials, O = oxide tunnel barrier) has stimulated a renewed interest for these systems, named magnetic valves. Besides their fundamental interest, these structures are foreseen as being candidates for very sensitive magnetic field sensors or as memory cells in magnetic random access memories. The largest MR amplitudes were observed in junctions prepared by oxygen plasma oxidation. In magnetic valves, the tunnel conductance varies as a function of the angle θ between the magnetization in the two ferromagnetic layers (magnetic valve effect). So far, the experimental results on these systems were mainly interpreted on the basis of Slonczewski's theory [1]. This author calculated the conductance of a MOM′ junction in a free electron model taking into account the exchange splitting in the d band. The calculation was made in the framework of classical quantum mechanics. No scattering of electrons in the magnetic metallic electrodes was taken into account.

Zhang *et al* [2] theoretically investigated the transport properties through double tunnel junctions of the form MOMOM. In such structure, the central magnetic layer M constitutes a spin-dependent quantum well in which resonance effects occur when the width of the well is an integer number of the electrons' wavelengths. The authors showed that at resonances, the MR ratio in these systems can be three to four times larger that in usual MOM′ magnetic valves. Furthermore, under these conditions, the conductivity through the system enhances dramatically. However, in such structures containing only one quantum well, the occurrence of the resonant tunnelling throughout the structure does not depend on the relative orientation of the magnetization in the successive magnetic layers. Consequently, the conductivity in such structures increases dramatically at resonances but not the magnetoresistance. Furthermore, the width of the resonance was assumed to be very small (proportional to $e^{-2q_0 b}$ where q_0 is the damping vector of the evanescent wave within the barrier and b is the barrier width) because the scattering in the metallic layers was neglected. However, in real systems, electron mean free paths in magnetic metallic layers are of the order of one to a few nanometres which is not much longer than the typical layer thickness. This means that electron scattering can play a significant role and must be included in the calculation of the transport in these systems. The main effect of the scattering is to decrease the sharpness of the resonances which has strong consequences for the resonant tunnelling.

Recently, we investigated, from a theoretical point of view, the transport properties in metal/oxide multilayered structures of the form $M_1OM_2OM_3OM_4$ where M represent ferromagnetic layers alternating with three insulating barriers (O = oxide) [3]. For particular thickness of the two inner magnetic layers, resonances occur in the quantum wells formed by these layers. This leads to a strong increase in the electron transmission through the insulating barriers. We showed that if the magnetization in the successive magnetic layers can be changed from parallel to anti-parallel as in spin valves, then, very large magnetoresistance effects can result due to the interplay of resonance effects in the two neighbouring quantum wells. The conductivity and magnetoconductivity were calculated within a quantum theory of linear response (Kubo formalism) taking into account the scattering in the magnetic layers. We showed that in such structures, giant tunnel magnetoconductivity can arise not only from a difference between spin up and spin down Fermi wave-vectors in the magnetic layers but also from spin-dependent mean free paths [3]. In the latter case, the effect of the scattering is to induce a spin-dependent broadening of the resonances in the quantum wells. Thus, we showed that very large magnetoresistance effects arise from the possibility to tune the position of the quantum levels in the two adjacent wells by changing the relative orientation of the magnetization in the successive magnetic layers. The idea developed in this paper may be extended to more complex multilayered structures of the form $M/(O/M)_n$ ($n > 3$) with even larger MR amplitude.

However, from a practical point of view, it is very difficult to prepare the system with a sufficient degree of control in the thickness of the various layers to satisfy the resonant conditions in the two inner magnetic layers simultaneously. One of the reasons is intrinsic: the inverse Fermi momentum and lattice constants are often non-commensurate. Furthermore, the band structure of ferromagnetic transition metals is rather complex so that the values of the electron parameters such the Fermi wave-vectors k_F are not well known. Consequently series of samples with varying thickness of the inner magnetic layers should be grown in a very controlled way to find the thickness at which the resonance occurs. An alternative procedure consists in changing the electron energy or momentum by applying an electrical field and hence satisfying the resonance condition without having to tune the thickness of the magnetic layers. Therefore, we decided to investigate the voltage dependence of the giant tunnel magnetoresistance in these multilayered $(M/O)_n$ structures. This implies calculating the non-linear response of these systems to a large applied electrical field.

We consider the transport properties across a multilayered structure of the form $M_1OM_2OM_3OM_4$ consisting of two thin magnetic layers (M_2 and M_3) inserted between three tunnel barriers with two magnetic electrodes at the edges of the structure (M_1 and M_4). We assume that, somehow, it is possible to change the relative orientation of the magnetization in the successive ferromagnetic layers from parallel to antiparallel as in giant-magnetoresistance multilayers. This can be achieved, for instance, by using ferromagnetic layers of different coercivities. The expression for the current from one metallic electrode to the other throughout the system of potential barriers has the form [4]

$$I = \frac{e}{\pi\hbar}\int_{-\infty}^{\infty}[f(E) - f(E+eV)]\,\mathrm{d}E\int |t|^2\,\mathrm{d}\vec{\kappa}$$
$$= \frac{1}{e}\int_{-\infty}^{\infty}[f(E) - f(E+eV)]\,\mathrm{d}E\int \sigma_{E,x}\,\mathrm{d}\vec{\kappa} \qquad (1)$$

where t is the transmission amplitude throughout the system, $f(E)$ and $f(E + eV)$ are the Fermi distribution for left and right electrodes, V is the applied voltage and $\sigma_{E,\kappa}(z, z')$ is the effective non-local conductivity of the system for electron with energy E and in-plane momentum $\bar{\kappa}$. The z-axis is perpendicular to the interfaces i.e. parallel to the current. The second equality in (1) comes from expression (14) in [5]. The coordinates (z, z') must be taken outside the region where the transmittance is calculated. Despite the non-local character of the conductance, it was shown in [6] and [7], that the exact expression of the non-local conductivity in the type of system under consideration does not depend on z and z'. Similarly, if one calculates the non-local conductivity approximately and takes into account vertex corrections, the obtained expression of the conductivity does not depend on z and z'. Alternatively, it was shown in [8] that it is possible to calculate the conductivity in macroscopically inhomogeneous systems without taking into account vertex corrections but by introducing effective internal electric fields into the expression of the current. These fields are defined as the gradient of electrochemical potential. Their value must be calculated in a self-consistent way to insure a uniform current in the z-direction. In the following, this latter approach was used.

We now describe our model in more detail. We consider the system M/O b/M a/O b/M a/O b/M in which we assume for simplicity that all magnetic layers are made of the same magnetic material. The two inner magnetic layers have the same thickness a. The three oxide barriers O have the same thickness b. The outer electrodes are supposed to be semi-infinite. As in [3], the electrons are assumed to form a free electron gas with exchange splitting of spin sub-bands in the ferromagnetic layers. The scattering in the metallic layers is taken into account by introducing the elastic mean free paths, which are spin dependent in the magnetic layers. l_1 and l_2 respectively represent the spin up and spin down mean free paths in M. We then calculate the field-dependent conductivity $\sigma_{E,\kappa}$ in (1) by using the Green function method which was described in our previous paper [3]. The Green functions of the system are solutions of the following equation:

$$\left[E - \frac{\hbar^2}{2m}\left(\kappa^2 - \frac{\partial^2}{\partial z^2}\right) - eV(z)\right]G(z, z') = \delta(z, z') \qquad (2)$$

where $V(z) = U - E_i z$ and E_i is the electrical field in the ith layer. To solve equation (2), we used the WKB approximation [9]. $G(z, z')$ is written in the form

$$G(z, z') = a(z')\exp\left(\int_{z_i}^{z} q(\tau)\,\mathrm{d}\tau\right) + b(z')\exp\left(-\int_{z_i}^{z} q(\tau)\,\mathrm{d}\tau\right)$$

where $a(z')$ and $b(z')$ are unknown coefficients to be determined from the condition of continuity of the Green functions and derivatives at the boundaries z_i. The function q is defined by

$$q(z) = \sqrt{\frac{2m}{\hbar^2}(U - E_F - eV(z)) + \kappa^2}$$

where U is the height of the potential barrier measured from the Fermi energy, E_F is the Fermi energy and $eV(z)$ is the z-dependent electrostatic potential.

As an example, below is the expression of the Green function for a particular interval of variables:

$$G(z_1 < z' < z < z_2) = \frac{ma_0}{\hbar^2 q(z')den}\{E^+(z_1, z')(ik_1 - q(z_1)) - E^-(z_1, z')(ik_1 + q(z_1))\}$$

$$\times \left\{ \begin{array}{l} E^+(z_2, z')[e^{ik_3 a}(ik_3 - q(z_2))F_1(k_5) - e^{ik_3 a}(ik_3 + q(z_2))F_2(k_5)] \\ -E^-(z_2, z')[e^{ik_3 a}(ik_3 + q(z_1))F_1(k_5) - e^{ik_3 a}(ik_3 - q(z_1))F_2(k_5)] \end{array} \right\} \quad (3)$$

where

$$den = e^{ik_3 a}\varphi_1^2(k_1, q(z_1); k_3, q(z_2))F_1(k_5) - e^{-ik_3 a}\tilde{\varphi}_1^2(k_1, q(z_1); k_3, q(z_2))F_2(k_5) \quad (4)$$

$$\varphi_m^n(k_i, q(z_i); k_k, q(z_l)) = E^+(z_m, z_n)(ik_i - q(z_i))(ik_k + q(z_l))$$
$$-E^-(z_m, z_n)(ik_i + q(z_i))(ik_k - q(z_l)) \quad (5)$$

$$\tilde{\varphi}_m^n(k_i, q(z_i); k_k, q(z_l)) = E^+(z_m, z_n)(ik_i + q(z_i))(ik_k - q(z_l))$$
$$-E^-(z_m, z_n)(ik_i - q(z_i))(ik_k + q(z_l)) \quad (6)$$

$$F_1(k_5) = e^{ik_5 a}\varphi_5^6(k_7, q(z_6); k_5, q(z_4))\tilde{\varphi}_3^4(k_3, q(z_2); k_5, q(z_4))$$
$$-e^{-ik_5 a}\varphi_3^4(k_5, q(z_4); k_3, q(z_2))\tilde{\varphi}_4^5(k_7, q(z_5); k_5, q(z_3)) \quad (7)$$

$$F_2(k_5) = e^{ik_5 a}\varphi_3^4(k_7, q(z_6); k_5, q(z_4))\tilde{\varphi}_3^4(k_3, q(z_2); k_5, q(z_4))$$
$$-e^{-ik_5 a}\tilde{\varphi}_3^4(k_5, q(z_4); k_3, q(z_2))\tilde{\varphi}_5^6(k_7, q(z_6); k_5, q(z_4)) \quad (8)$$

$$E^\pm(z_m, z_n) = \exp\left[\pm\int_{z_m}^{z_n} q(\tau)\,d\tau\right] \quad (9)$$

and

$$k_i = c_i + id_i = \sqrt{k_{F\sigma}^2 - \kappa^2 + eV(z_i)\frac{2m}{\hbar^2} + 2i\frac{k_{F\sigma}}{l_\sigma}}. \quad (10)$$

We neglected the drop of voltage in the metallic layers which is a very reasonable assumption considering the much higher resistance of the tunnel barriers as compared to that of the metallic layers in the direction perpendicular to the plane. $k_{F\sigma}$ and l_σ are respectively the spin-dependent electron Fermi momentum and mean free path in the magnetic layers for the spin direction σ. The coordinates z_i are defined in figure 1. They represent the positions of the various M/O interfaces.

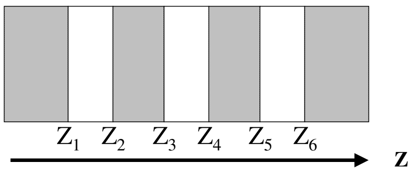

Figure 1. Schematic representation of the multilayer stacking.

To obtain the MR ratio MR $= (G^{\uparrow\uparrow} - G^{\uparrow\downarrow})/G^{\uparrow\downarrow}$ where $G^{\uparrow\uparrow}$ and $G^{\uparrow\downarrow}$ are the conductances throughout the structure respectively for parallel and antiparallel configurations of the magnetization in the successive ferromagnetic layers, the two point conductivity $\sigma(z_i, z_{i+1})$ is first calculated through every barrier using expressions (2)–(8) and the non-linear Kubo formula (1). The resulting expressions for $\sigma(z_i, z_{i+1})$ are rather complex. As an example, the expression for $\sigma(z_1, z_2)$ is written below in the limit $\exp(-q(z_i)b) \ll 1$:

$$\sigma(z_1, z_2) = \frac{e^2 q_0^2}{2\pi\hbar} \frac{c_1 c_5 q^2(z_2)(c_3^2 + q^2(z_2))}{c_1^2 + q^2(z_1)} E^+(z_1, z_2) \sinh(2d_3 a)$$
$$\times\{(\cosh(2d_3 a) - 1)(c_3^2 + q^2(z_2))^2$$
$$+2[\sin(c_3 a)(c_3^2 - q^2(z_2)) - 2c_3 q(z_1)\cos(c_3 a)]^2\}^{-1} \quad (11)$$

where $q_0 = \sqrt{(2m/\hbar^2)(U - E_F)}$.

We point out that the calculated expression of $\sigma(z, z')$ if z and z' are inside any barrier does not depend on z and z'. Furthermore the values of σ through the first and third barriers are equal. They are however different from the value of the conductivity through the second barrier. This is due to the fact that we did not take into account vertex corrections. Therefore, as discussed previously, in order to insure a uniform current along the z-direction, we calculated the voltage drop across each barrier for a fixed voltage V applied between the outer electrodes, in a self-consistent way. This calculation was carried out for both parallel and antiparallel magnetic configurations. We recall that the voltage drop in the metallic layers is negligible; the electron scattering in the outer electrodes does not influence the total resistivity at all and the scattering in the inner metallic layers defines the width of the resonance levels. The voltage drops across the first and third barriers may be quite different from the value across the second layer, especially at resonance. This may be explained by considering that electrons tunnel through the first (third) barrier from (to) a continuum of energy to (from) a resonant level. In contrast, for the second barrier, the electrons tunnel from a resonant level to another resonant level. Therefore, the second barrier is in some sense short-circuited at resonance.

To insure that the current is uniform along the z-axis, we calculated numerically, in a self-consistent way, the voltage across each barrier for a fixed voltage V applied between the outer electrodes, for both parallel and antiparallel magnetic configurations. The denominator in (11) has a resonance form. The resonances occur when the thickness a_i of the ith metallic layer satisfies the condition:

$$\tan(a_i c_i) = \frac{2q(z_i)c_i}{c_i^2 - q^2(z_i)}. \quad (12)$$

The width of the resonance is of the order of $(a_i/l_\sigma) \ll 1$. The plots of the overall conductivity and magnetoresistance (MR) versus the thickness of the metallic layer look very much the same at low and high voltages. These plots were shown in figures 1–3 of [3].

The plots of the $I(V)$ characteristics for each spin channel are shown in figures 2 and 3 for two different thicknesses a of the metallic layers. In these calculations, the following parameters were used: $l_1 = l_2 = 100$ Å, $k_{1F} = 1$ Å^{-1}, $k_{2F} = 0.4$ Å^{-1}, $q_0 = 1$ Å^{-1}. The curves exhibit several maxima, which are attributed to the occurrence of resonances at fixed thickness a when V and consequently $c_i(V)$ are varied (see (12)). The difference ΔV between two consecutive resonance values of V in the same layer is approximately equal to

$$e\Delta V \approx \frac{4\pi k_F}{a} \frac{\hbar^2}{2m}. \quad (13)$$

The behaviour of these curves is relatively complex due to the interplay of the resonances in the two inner magnetic layers for which the drop of voltage is different.

Very large MR amplitudes are obtained at the resonances. The conductance in parallel magnetic configuration can be ten times larger than in antiparallel magnetic configuration.

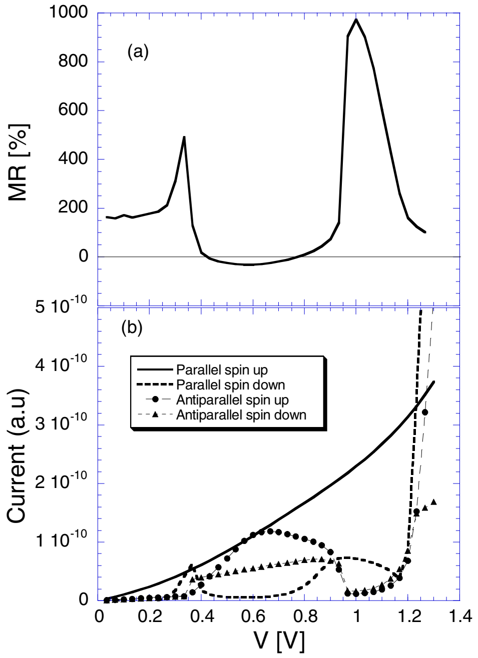

Figure 2. (a) Voltage dependence of the MR for an MOMOMOM system with $a = 11$ Å. The following values are used for the electron parameters: $l_1 = l_2 = 100$ Å, $k_{1F} = 1$ Å^{-1}, $k_{2F} = 0.4$ Å^{-1}, $q_0 = 1$ Å^{-1}. (b) Current for each spin channel in parallel and antiparallel magnetic configurations.

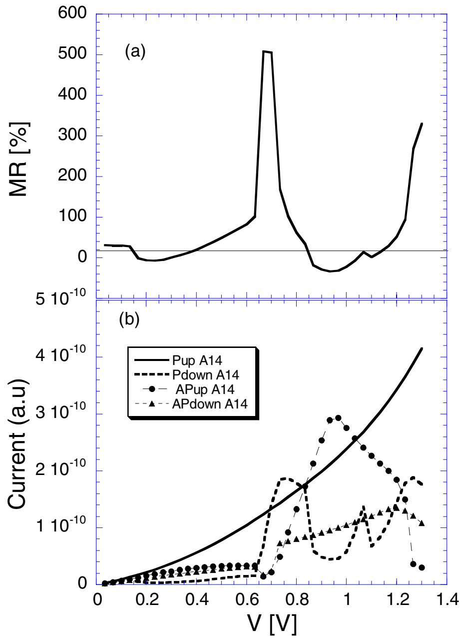

Figure 3. (a) Voltage dependence of the MR for an MOMOMOM system with $a = 14$ Å. The following values are used for the electron parameters: $l_1 = l_2 = 100$ Å, $k_{1F} = 1$ Å^{-1}, $k_{2F} = 0.4$ Å^{-1}, $q_0 = 1$ Å^{-1}. (b) Current for each spin channel in parallel and antiparallel magnetic configurations.

In figure 3, the positions of the peaks in MR are shifted as compared to figure 2 because the resonance conditions are not fulfilled for the same electron energy since the width of the quantum wells is different. It is interesting to note also that at antiresonances, the MR amplitude can even be negative. The conductance is then larger in the antiparallel magnetic configuration than in the parallel one.

Besides the oscillations in conductance and MR versus bias voltage, we also point out that at low voltage (0–0.3 V), the MR decay versus voltage is much slower in these multilayered M/O systems as compared to a single MOM tunnel junction even after normalization of the voltage by the number of junctions assuming that they are simply connected in series. To illustrate this point, we calculated the MR variation versus bias voltage using the same model and parameters as for the multilayered M/O structure but considering only one tunnel barrier. The result is plotted in figure 4. In the multilayered case, the MR is almost constant in the range 0–0.3 V whereas it drops already significantly for a single junction.

Let us now discuss the effect of roughness on the results discussed above. Three situations can be encountered:

(i) The roughness occurs at the atomic scale. The interfaces are flat but exhibit some atomic disorder because, for instance, the barrier is made of an amorphous material such as Al_2O_3. In such a case, an additional electron scattering occurs at the metal/barrier interface. This scattering comes in addition to the scattering already taken into account in the bulk of the metallic layers. It contributes to the broadening of the resonance.

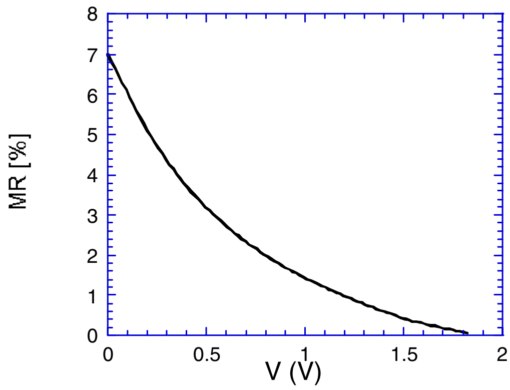

Figure 4. Voltage dependence of the MR for a single MOM junction, $k_{1F} = 1$ Å^{-1}, $k_{2F} = 0.4$ Å^{-1}, $q_0 = 1$ Å^{-1}.

(ii) The interfaces exhibit large terraces but the terraces are correlated from one interface to the next so that the thickness of each layer remains uniform. Recent studies carried out on tunnel junctions deposited on Si substrates with step bunching [10] showed that this situation can be encountered in metal/insulator systems. As in the previous case, the interfacial steps contribute to the broadening of the resonances. However, as long as the thickness of the various layers remains uniform, the overall transport properties are the same as with ideally flat interfaces.

(iii) The interfaces exhibit wide uncorrelated terraces so that the thickness of the layers varies by a large fraction of the Fermi wavelength across the sample area. This situation is the most difficult to describe theoretically. In such a case, the resonance condition is only satisfied locally. An in-plane component of the current may appear in the metallic layers in order to reach the most favourable current path. This case would certainly lead to a drastic decay of the resonance effects previously discussed.

In [11], the authors measured the MR for single and double (not triple) barrier structures and found that the MR amplitudes at low voltage are approximately the same in both structures. This is in agreement with our statement that only in triple barrier (or more) systems can an increase in MR amplitude be expected due to the interplay of spin-dependent resonant tunnelling in successive wells. Furthermore, they observed that the decrease in MR amplitude versus voltage was slower with double barrier structures than in single barrier junctions in accordance with our conclusion at low voltage.

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