

Theory of spin-dependent tunnelling in magnetic junctions

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Abstract

Rigorous theory of the tunnelling magnetoresistance (TMR) based on the real-space Kubo formula and fully realistic tight-binding bands fitted to an *ab initio* band structure is described. It is first applied to calculate the TMR of two Co electrodes separated by a vacuum gap. The calculated TMR ratio reaches $\approx 65\%$ in the tunnelling regime but can be as high as 280% in the metallic regime when the vacuum gap is of the order of the Co interatomic distance (abrupt domain wall). It is also shown that the spin polarization P of the tunnelling current is negative in the metallic regime but becomes positive $P \approx 35\%$ in the tunnelling regime. Calculation of the TMR of an epitaxial Fe/MgO/Fe(001) junction is also described. The calculated optimistic TMR ratio is in excess of 1000% for an MgO barrier of ≈ 20 atomic planes and the spin polarization of the tunnelling current is positive for all MgO thicknesses. It is also found that spin-dependent tunnelling in an Fe/MgO/Fe(001) junction is not entirely determined by states at the Γ point ($k_{\parallel} = 0$) even for MgO thicknesses as large as ≈ 20 atomic planes. Finally, it is demonstrated that the TMR ratio calculated from the Kubo formula remains non-zero when one of the Co electrodes is covered with a copper layer. It is shown that non-zero TMR is due to quantum well states in the Cu layer which do not participate in transport. Since these only occur in the down-spin channel, their loss from transport creates a spin asymmetry of electrons tunnelling from a Cu interlayer, i.e. non-zero TMR. Numerical modelling is used to show that diffuse scattering from a random distribution of impurities in the barrier may cause quantum well states to evolve into propagating states, in which case the spin asymmetry of the non-magnetic layer is lost and with it the TMR.

The conductance $\Gamma(H_s)$ of a tunnel junction with two ferromagnetic electrodes whose magnetic moments are aligned parallel in an applied saturating field H_s is much higher than its conductance $\Gamma(0)$ in zero field when the moments are antiparallel [1–3]. The effect is called tunnelling magnetoresistance (TMR) and the relative change in the resistance of the junction, i.e. the so-called ‘optimistic’ magnetoresistance (MR) ratio

$$R_{\text{TMR}} = \frac{\Gamma(0)^{-1} - \Gamma(H_s)^{-1}}{\Gamma(H_s)^{-1}} \quad (1)$$

can be as high as 60%. The traditional explanation of the TMR effect is based on the assumption that electrons tunnelling from a ferromagnet are spin-polarized and their polarization P is given in terms of the spin-dependent density

of states (DOS) D^σ of the ferromagnet by $P = [D^\uparrow(E_F) - D^\downarrow(E_F)]/[D^\uparrow(E_F) + D^\downarrow(E_F)]$. Since the classical theory of tunnelling [4] states that the junction conductance is proportional to the product of the DOS of the left and right electrodes, it is easy to show that the TMR ratio (1) can be written in terms of the spin polarizations P_L, P_R of the left and right electrodes

$$R_{\text{TMR}} = \frac{2P_L P_R}{1 - P_L P_R}. \quad (2)$$

This is the well-known Julliere’s formula [5]. Although the Julliere’s formula is quite successful in predicting the TMR ratios from the observed values [4] of the spin polarization of electrons tunnelling from Fe, Ni and Co into a superconductor, it suffers from several fundamental defects. First of all, it has been known for long time that the polarization of the tunnelling

current predicted from the total DOS of the ferromagnetic electrodes has the wrong sign. One would expect from the DOS that the tunnelling current from Fe, Co, and Ni should be dominated by down-spin (minority) electrons since their DOS at E_F is high. In fact, the observed P has just the opposite sign.

The second problem is that the properties of the insulating barrier are not included in the Julliere's formula, i.e. the polarization of the tunnelling current is assumed to be just the property of the ferromagnetic electrodes.

The third problem that has come to light only recently is that the Julliere's formula when applied to a tunnelling junction with a thin non-magnetic metallic interlayer, such as Cu, inserted between one of the ferromagnetic electrodes and the insulating barrier fails to explain the observed [6] non-zero TMR ratio. In fact, since the DOS of the Cu layer adjacent to the barrier is spin independent, $P_{Cu} = 0$ and, therefore, it follows from equation (2) that $R_{TMR} = 0$, which contradicts the experiment [6].

The three problems we have identified, call into question the validity of the whole classical theory of tunnelling based on the DOS of the ferromagnetic electrodes. We shall examine the reasons for the failure of the DOS approach using the rigorous real-space Kubo formula [7, 8]. The Kubo formula is exact in the linear response regime (low bias limit).

To obtain clear-cut answers, we consider tunnelling between cobalt electrodes across a vacuum gap and coherent tunnelling in an epitaxial Fe/MgO/Fe junction. In the case of a junction with a metallic non-magnetic interlayer, tunnelling takes place from a cobalt electrode covered with an overlayer of N atomic planes of copper across a vacuum gap into another cobalt electrode. The geometry of the junctions we consider is shown in figure 1.

Initially, we assume that the electrodes are perfect so that the electron wave vector parallel to the layers \mathbf{k}_{\parallel} is conserved in tunnelling. This restriction will be relaxed later. In contrast to the junction with an amorphous Al_2O_3 barrier used in most experiments, tunnelling across a vacuum gap or in an epitaxial Fe/MgO/Fe junction has the great advantage that the real-space Kubo formula [7, 8] can be evaluated without any approximations for a fully realistic band structure of all the components of the junction. The results we obtain are, therefore, exact.

We use a tight-binding parametrization of an *ab initio* band structure of Co, Cu for vacuum tunnelling and of Fe and MgO for the epitaxial Fe/MgO/Fe junction (for details, see [8, 9]). The total conductance of the junction Γ^{σ} in a spin channel σ is expressed [10] in terms of the one-electron Green's functions $G_L^{\sigma}(E_F, \mathbf{k}_{\parallel})$, $G_R^{\sigma}(E_F, \mathbf{k}_{\parallel})$ at the left and right surfaces of the junction cut by a cleavage plane into two independent

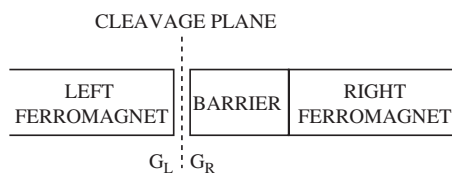


Figure 1. Schematic picture of a magnetic tunnelling junction.

parts (figure 1):

$$\Gamma^{\sigma} = \frac{4e^2}{h} \sum_{\mathbf{k}_{\parallel}} Tr [T_{\sigma} \text{Im} G_R^{\sigma}(E_F, \mathbf{k}_{\parallel})] [T_{\sigma}^{\dagger} \text{Im} G_L^{\sigma}(E_F, \mathbf{k}_{\parallel})]. \quad (3)$$

The summation in equation (3) is over the two-dimensional Brillouin zone and the trace is over the orbital indices corresponding to s, p, d orbitals. Since we use a multi-orbital band structure, G^{σ} and T_{σ} are matrices whose size depends on the number of orbitals. The matrix T_{σ} is given by

$$T_{\sigma} = t(\mathbf{k}_{\parallel}) [I - G_R^{\sigma}(E_F, \mathbf{k}_{\parallel}) t^{\dagger}(\mathbf{k}_{\parallel}) G_L^{\sigma}(E_F, \mathbf{k}_{\parallel}) t(\mathbf{k}_{\parallel})]^{-1}, \quad (4)$$

where I is a unit matrix in the orbital space and $t(\mathbf{k}_{\parallel})$ is the matrix of tight-binding hopping integrals connecting atomic orbitals in the right surface of the cut junction to atomic orbitals in the left surface.

The Kubo formula (3) has a simple physical interpretation. First of all, since we operate in the linear-response regime, the current is proportional to the conductance and, therefore, equation (3) gives effectively the tunnelling current. The quantities $\text{Im} G_{L(R)}^{\sigma}(E_F, \mathbf{k}_{\parallel})$ are (up to a factor $1/\pi$) one-dimensional DOS of the left (right) surfaces of the cut junction in the channel $(\mathbf{k}_{\parallel}, \sigma)$ and the matrix T_{σ} can be regarded as an effective tunnelling matrix. Since we assume coherent tunnelling (perfect electrodes), the current flows in independent $(\mathbf{k}_{\parallel}, \sigma)$ channels, which means that all the channels contribute additively to the total current and, hence, the sum over \mathbf{k}_{\parallel} in equation (3).

With this interpretation, the Kubo formula (3) resembles superficially the Julliere's formula. However, in contrast to the classical theory of tunnelling, the Kubo formula (3) does not assume separation of the tunnelling junction into two independent left and right parts. Although the Green's functions G_L^{σ} and G_R^{σ} are for disconnected left and right surfaces, the mutual interaction of the two surfaces is described exactly through the matrix T_{σ} defined by equation (4). It will be seen that this interaction is essential for correct treatment of the tunnelling junction with a non-magnetic metallic interlayer [12].

Since the full interaction between the left and right surfaces is contained in equation (3), it applies not only to tunnelling but also to metallic conduction. In the case of a metallic sample, one has to make sure that the resistance of the electrodes is much lower than the resistance of the sample (which is automatically satisfied for a tunnelling junction). This can be achieved experimentally in the 'pillar' geometry shown schematically in figure 2. The 'sample' in figure 2 is

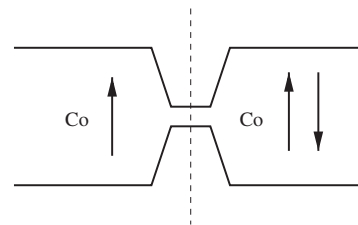


Figure 2. Schematic picture of a metallic contact in the pillar geometry. Arrows indicate the orientation of the magnetization.

the narrow region (pillar). It should be noted that calculations made in the slab geometry (figure 1) apply directly to the pillar geometry (figure 2) provided the constriction is adiabatic, i.e. the cross section of the structure decreases gradually rather than abruptly.

We first apply the Kubo formula (3) to calculate the MR ratio for two cobalt electrodes in direct (metallic) contact (figure 2). We then break the contact by introducing a vacuum gap between the left and right electrodes. This allows us to investigate how the MR evolves from the ballistic current-perpendicular-to-plane magnetoresistance (CPP GMR) for a metallic system to TMR. In calculating the MR, we assume that the magnetization in the left electrode points up and that in the right electrode can point either up or down. The MR in the unbroken (metallic) contact is, therefore, due to a completely abrupt domain wall (the orientation of the magnetization changes by 180° from one atomic plane to the next). Following Harrison [10, 11], we model tunnelling across vacuum gap (broken contact) by turning off gradually the hopping matrix $t(k_{\parallel})$ across the gap. As discussed in [10], hopping between s, p, d orbitals scales differently with the distance between the electrodes (width of the vacuum gap). This has the consequence that tunnelling between d orbitals is rapidly suppressed owing to their weak overlap across the gap. In the case of an MgO barrier discussed later, suppression of d-type tunnelling is due to the fact that there are no d orbitals present in the barrier. Given that only the s-s interaction survives in the tunnelling regime, it is appropriate to use it as a measure of the width of the vacuum gap between the Co electrodes. It is, therefore, convenient to introduce a dimensionless reduced s-s hopping parameter $0 \leq t \leq 1$ by $t = t_{ss}^{\text{gap}}/t_{ss}^{\text{bulk}}$, where t_{ss}^{bulk} is the bulk s-s hopping in Co and t_{ss}^{gap} is the hopping across the vacuum gap.

The dependence of the TMR ratio, determined from equation (3), on the reciprocal of the reduced hopping parameter $1/t$ is shown in figure 3. It can be seen that the TMR ratio drops very rapidly from its metallic value of 280% for the abrupt domain wall ($t = 1$) to about 40% and then remains almost constant in the tunnelling regime reaching about 65% for $t = 0.1$. The rapid initial decrease of the MR ratio occurs because, in the metallic limit $t \approx 1$, a significant proportion of the current in Co is carried by d electrons that are highly spin-polarized. This explains a large MR ratio in the metallic

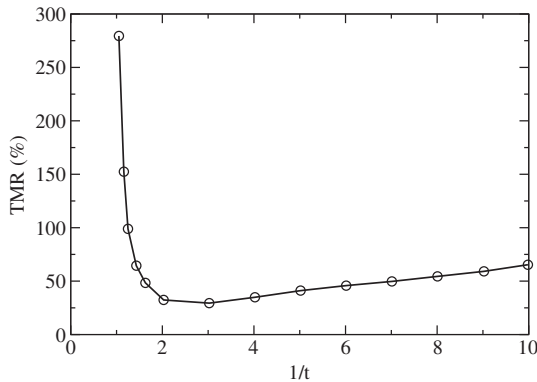


Figure 3. Dependence of the TMR of a Co(001) junction on the reciprocal of the reduced s-s hopping between the Co electrodes.

regime (abrupt domain wall). In the tunnelling regime, the current is carried only by s-p electrons which are weakly spin-polarized and, hence, the TMR ratio is much smaller. The switching from d-type to s-p type conduction, which occurs as one moves from the metallic to the tunnelling regime, has also important implications for the sign of the polarization of the tunnelling current which can be easily determined from equation (3). It should be noted that the correct definition of the spin polarization is not in terms of the DOS, as assumed in the Julliere's formula, but in terms of the partial tunnelling currents carried by \uparrow and \downarrow spin electrons. In the linear-response regime, $P_{L(R)}$ is, therefore, given by

$$P_{L(R)} = \frac{\Gamma_{L(R)}^{\uparrow} - \Gamma_{L(R)}^{\downarrow}}{\Gamma_{L(R)}^{\uparrow} + \Gamma_{L(R)}^{\downarrow}}, \quad (5)$$

where $\Gamma_{L(R)}^{\sigma}$ gives the current of electrons of spin σ tunnelling from the left (right) ferromagnet through a barrier (vacuum gap) into a suitable detector of spin-polarized current. In practice, the detector is usually a superconducting aluminum electrode. We stress that the spin polarization of the tunnelling current is not just a property of the ferromagnetic electrode (as is assumed in the Julliere's formula), but is instead the joint property of the electrode and the barrier.

The dependence of the spin polarization P of the Co junction on the width of the vacuum gap (reciprocal hopping $1/t$) obtained from equations (3) and (5) is shown in figure 4. For a small vacuum gap of the order of the lattice constant ($1/t \approx 1$), the conductance is dominated by d electrons and P has the 'wrong' sign $P < 0$ consistent with the total DOS argument of the classical theory of tunnelling [4]. However, there is a rapid crossover to $P > 0$ as the width of the gap increases. It can be seen from figure 4 that the calculated P for Co not only has the correct sign in the tunnelling regime $1/t \gg 1$, but its magnitude 30–40% is in excellent agreement with the observed [4] $P \approx 35\%$. The crossover from negative to positive P occurs because the overlap of d-orbitals decreases with increasing gap much faster than that of s-orbitals and it is, therefore, s electrons that determine the conductance in the tunnelling regime.

We now turn to spin-dependent tunnelling between two Fe(001) electrodes separated by an MgO barrier [9]. It is

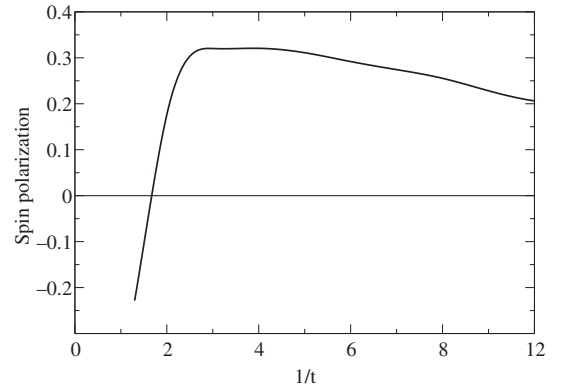


Figure 4. Dependence of the spin polarization of electrons tunnelling between two Co electrodes across a vacuum gap on the reciprocal of the reduced s-s electron hopping between the Co electrodes (width of the gap).

known experimentally [13] that thin epitaxial bcc Fe(001) films grow pseudomorphically on rocksalt MgO(001) substrate so that the Fe atoms sit above the O ions. The Fe lattice is, therefore, rotated by 45° relative to the MgO lattice. There is only a small lattice mismatch of about 3.5% between the Fe–Fe and O–O in-plane distances. Based on these results, we neglect the small lattice mismatch between Fe and MgO and assume that the whole Fe/MgO/Fe(001) junction grows epitaxially. We describe the band structure of the electrodes by tight-binding bands fitted to the *ab initio* band structure of bcc Fe [14] and that of the barrier by tight-binding bands fitted to the band structure of bulk MgO [15]. Since the whole structure is epitaxial, momentum parallel to the junction is conserved and, therefore, our Kubo formula (3) is directly applicable to the Fe/MgO/Fe(001) junction. The TMR is calculated by the same method as for the vacuum cobalt junction. For convenience, we shall use the (pessimistic) TMR ratio $R_{\text{TMR}} = (\Gamma(H_s) - \Gamma(0))/\Gamma(H_s)$ to characterize the TMR of the Fe/MgO/Fe junction. The dependence of R_{TMR} on the thickness of the MgO barrier is shown in figure 5(a). The majority-spin $\Gamma_{\text{FM}}^\uparrow$ and minority-spin $\Gamma_{\text{FM}}^\downarrow$ conductances in the ferromagnetic configuration of the junction and the conductance Γ_{AF} of electrons of either spin in the antiferromagnetic configuration

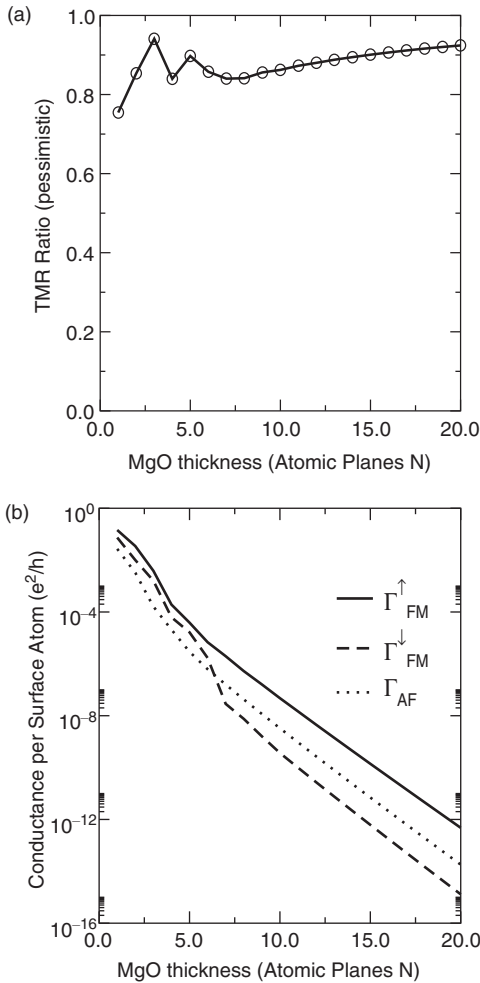


Figure 5. (a) Dependence of the pessimistic TMR ratio R_{TMR} of an Fe/MgO/Fe(001) junction on MgO thickness. (b) Dependences of the total conductances $\Gamma_{\text{FM}}^\uparrow$, $\Gamma_{\text{FM}}^\downarrow$, and Γ_{AF} on MgO thickness.

are plotted against the MgO thickness on a logarithmic scale in figure 5(b).

The TMR ratio oscillates initially with MgO thickness, but after about 7 atomic planes of MgO, stabilizes and increases only slowly reaching a very high value of 0.92 for 20 atomic planes of MgO. This corresponds to the optimistic ratio of some 1200%. The behaviour of the individual conductances is more informative. Firstly, it is clear from figure 5(b) that the majority-spin conductance is always higher than the minority-spin conductance. It follows that the calculated spin polarization of the tunnelling current is positive, as found experimentally for junctions based on Al_2O_3 barrier. It is also clear that after some 10 atomic planes of MgO, the junction reaches an asymptotic regime with all the conductances decreasing exponentially with MgO thickness. However, the slope of $\Gamma_{\text{FM}}^\uparrow$ is somewhat smaller than that of $\Gamma_{\text{FM}}^\downarrow$ and Γ_{AF} , which explains why the TMR ratio increases with MgO thickness. Different rates of decrease of the conductances $\Gamma_{\text{FM}}^\uparrow$, $\Gamma_{\text{FM}}^\downarrow$ and Γ_{AF} are due to the fact that tunnelling through MgO is not dominated by $k_{\parallel} = 0$ (the Γ point) even for thicknesses of MgO as large as 20 atomic planes.

Our calculated dependence of the TMR ratio on the MgO thickness demonstrates clearly a failure of the classical Julliere’s formula (2) which predicts a constant TMR ratio independent of the barrier thickness. The reason why the TMR depends on the barrier thickness is that the conductances in the individual spin channels $\Gamma_{\text{FM}}^\uparrow$, $\Gamma_{\text{FM}}^\downarrow$, and Γ_{AF} are determined by details of the matching of the wave functions of tunnelling electrons across the whole junction. The wave function matching is treated exactly by the Kubo formula but ignored completely in the classical theory of tunnelling [4, 5].

Finally, we shall use the Kubo formula (3) to demonstrate that TMR of a Co junction with a vacuum gap remains non-zero when one of the Co electrodes is covered with a Cu layer. This is yet another example of a system for which the exact matching of the wave functions of tunnelling electrons across the whole junction is vital.

The dependence of the TMR ratio obtained by numerical evaluation of the Kubo formula (3) on the thickness of the Cu overlayer is shown in figure 6. The calculation is for (111) orientation of the layers and vacuum gap characterized by reduced hopping $t = 0.1$. In contrast to the Julliere’s formula (2), the TMR determined from the Kubo formula (3)

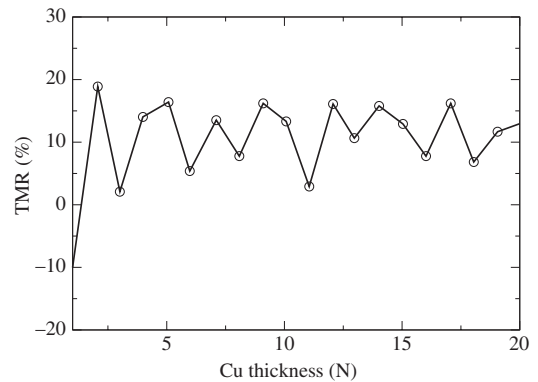


Figure 6. Dependence of the TMR of a vacuum cobalt junction with a copper interlayer on the thickness of the copper layer.

is non-zero and oscillates as a function of Cu thickness due to quantum interference of electrons on the Cu interlayer. It is interesting that for a small Cu thickness (two monolayers), the TMR ratio becomes negative. A negative TMR with a very thin gold interlayer has been observed by Moodera [17].

The physical explanation of a non-zero TMR is that the Cu layer acts as a spin filter. Since the Fermi surfaces of Cu and of the majority-spin electrons in Co are very similar (the Co majority d band lies below E_F), majority-spin electrons cross easily the Co/Cu interface and participate in tunneling as if there were no intervening Cu layer. On the other hand, there is a poor match between the Cu bands and the minority-spin bands in Co, which results in the formation of down-spin quantum well states in the Cu overlayer [18, 19]. Since the quantum well states are localized in the Cu layer they do not contribute to transport of charge in the down-spin channel, which gives rise to a spin asymmetry (non-zero polarization P) of the tunnelling current and, hence, non-zero TMR.

The apparent paradox that the Julliere's formula predicts zero TMR but the Kubo formula gives a non-zero TMR can now be easily resolved. Since the down-spin quantum well states in the Cu layer contribute to the ordinary DOS they are, incorrectly, counted in the Julliere's formula (2) as contributing to the tunnelling current. The total DOS of down-spin electrons, which is made up of propagating and quantum well states, is equal to the DOS of up-spin states which are all propagating. There is, therefore, no spin asymmetry in the DOS of the Cu overlayer and, hence, the Julliere's formula gives zero TMR. On the other hand, the Kubo formula excludes automatically all the quantum well states. Since these only occur in the down-spin channel, their loss from transport creates a spin asymmetry of electrons tunnelling from a Cu overlayer, i.e. non-zero TMR.

It is clear that for a non-zero TMR effect to occur, one needs a strong scattering at the ferromagnet/non-magnet interface in one of the spin channels and weak scattering in the other spin channel. These are the same conditions as those required for a large GMR in the corresponding ferromagnet/non-magnet multilayer. It is, therefore, clear that Co/Cu is a particularly good combination but, for example, an Al interlayer should not lead to any sizable TMR since GMR for an Al spacer is very small. This is in agreement with the observation (e.g. see [16]) that an Al interlayer kills the TMR very effectively.

To observe a non-zero TMR, quantum-well states in one of the spin channels need to be well defined. This is the case when the effect of impurities is negligible (ballistic transport across the whole junction) and scattering at the ferromagnet/non-magnet interface is specular. Scattering from impurities or/and diffuse scattering at the ferromagnet/non-magnet interface may allow quantum well states to evolve into propagating states, in which case the spin asymmetry of electrons tunnelling from the non-magnetic interlayer may be lost (and with it the TMR effect). The fact that the calculated TMR shown in figure 6 is non-decaying as a function of Cu thickness is due to our neglect of impurity/interface scattering.

To investigate qualitatively the effect of diffuse scattering on the quantum well states, we have used a one-band model of the tunnelling junction with a random distribution of impurities

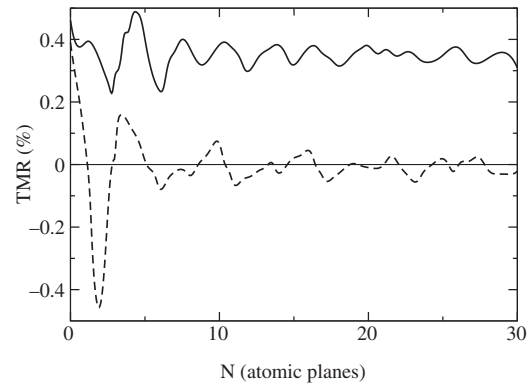


Figure 7. Dependence of the TMR ratio for a junction with a non-magnetic metallic interlayer on the interlayer thickness N . Solid curve is for a perfect junction; broken curve for a junction with random impurities in the barrier.

in the barrier. Since k_{\parallel} is no longer conserved we had to use a large in-plane supercell geometry to simulate disorder in the barrier. This method allows us to evaluate the Kubo formula without any approximations for a realistic model of disorder. The price to pay is a simplified band structure—one band model—and a junction with a relatively small cross section (supercell size). Figure 7 shows the TMR ratio as a function of the thickness of a non-magnetic metallic interlayer for a junction with (broken curve) and without disorder (solid curve). It can be seen that disorder suppresses the average TMR but, rather surprisingly, large oscillations about zero of the TMR as a function of the non-magnetic spacer thickness are not washed out by the disorder. The results shown in figure 7 are for a barrier of five atomic planes, i.e. the amount of disorder is relatively large. For a thinner barrier with disorder (1–3 atomic planes), the average TMR remains non-zero.

Acknowledgment

It is a pleasure to dedicate this paper to Professor Shinjo on the occasion of his retirement.

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