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LETTER TO THE EDITOR

Modelling of spin-polarized electron tunnelling from 3d ferromagnets

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Abstract. Spin-polarized electron tunnelling from ferromagnetic Fe and Co films is modelled within a quantum-mechanical treatment of the electronic transport and a tight-binding approximation accounting for an accurate band structure of the 3d metals. Calculations have been performed assuming that the band gap of the insulator is much larger than the hopping integrals between the metal and the insulator, the electronic structure of the latter being approximated by two non-coupled s-type tight-binding bands separated by a gap. It is found that within the *ballistic* regime of conductance the spin polarization of the tunnelling current depends strongly on the type of covalent bonding between the ferromagnet and the insulator. In the case of ssσ bonding the tunnelling current is carried only by the s electrons of the ferromagnet and the spin polarization is positive. This is due to the strong s–d hybridization within the ferromagnet which reverses the sign of the spin polarization of the s-electron partial density of states at the Fermi level with respect to the total surface density of states. The absolute values of the spin polarization of the tunnelling current in this case of ssσ bonding across the metal–insulator interface are in very good agreement with experimental data on tunnelling between 3d ferromagnets and aluminium through an alumina spacer. Including the sdσ bonding at the interface, however, results in a large contribution of the d-electron tunnelling current, which reduces the spin polarization and leads to a change in its sign for the case of Co.

Recent advances in tunnelling magnetoresistance (TMR) [1–3] have demonstrated that thin-film tunnel junctions, in which ferromagnetic layers are separated by a thin insulating layer, are very promising from the point of view of applications as magnetic sensors and as magnetic random-access memory elements. The actual magnitude of TMR which is important for better performance of future devices is determined by the spin polarization of the tunnelling current from the ferromagnetic metal through the insulator film to a non-magnetic metal [4]. The spin-polarization values are known experimentally for numerous ferromagnetic metals with alumina spacers [5]. It was found that for ferromagnetic 3d metals these values are positive in all tunnelling measurements.

Various theoretical explanations were proposed in order to explain these positive values of the spin polarization (see, e.g., [5] and references therein). For example, Hertz and Aoi [6] argued that s electrons are favoured over d electrons in tunnelling, despite the much greater density of d states in ferromagnetic 3d metals. Sterns [7] related the spin polarization of the tunnelling current to the relative difference in the momentum of the itinerant d electrons determined from the dispersions of the majority and minority spin electrons in the bulk ferromagnet. Although reasonable agreement with experimental observations was obtained in both studies, no accurate calculations of the tunnelling current were performed and results were based mainly on semi-quantitative estimates.

In this letter we model the tunnelling current from ferromagnetic Co and Fe within the quantum-mechanical formulation of electronic transport taking into account realistic

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band structures of the ferromagnets. We do not take into account scattering by defects or impurities within the tunnel junction and in this sense consider only the *ballistic* regime of conductance. We analyse the contributions of the different types of electron (s, p or d) to the tunnelling current by changing the type of covalent bonding at the metal–insulator interface. We find that depending on the type of bonding at the interface (ssσ, spσ or sdσ) the spin polarization changes significantly and can even change sign. In the case of ssσ bonding, i.e. when only the s electrons of the ferromagnet contribute to the tunnelling current, we obtain positive values of the spin polarization, which are in very good agreement with experimental data on tunnelling between 3d ferromagnets and aluminium through an alumina spacer [8]. Including sdσ bonding at the interface results in a large d-electron contribution to the conductance which reduces the spin polarization of the tunnelling current and leads to a change in its sign for the case of Co.

The tunnelling current is calculated by considering two perfect semi-infinite metallic electrodes separated by a thin layer of the perfect insulator. Assuming two-dimensional periodicity in the plane perpendicular to the current, zero temperature and a small applied bias, the phonon-free conductance per spin can be written as follows [9]:

$$G = \frac{2\pi e^2}{\hbar} \sum_{\boldsymbol{k}_{||}} \mathrm{Tr}\left[\rho_1(\boldsymbol{k}_{||}) T^{+}(\boldsymbol{k}_{||}) \rho_2(\boldsymbol{k}_{||}) T(\boldsymbol{k}_{||})\right]. \tag{1}$$

Here $\rho_1(\boldsymbol{k}_{||})$ and $\rho_2(\boldsymbol{k}_{||})$ are the density-of-states operators of the isolated electrodes for a given value of the parallel momentum $\boldsymbol{k}_{||}$: $\rho_n(\boldsymbol{k}_{||}) = \left(G_n^{-}(\boldsymbol{k}_{||}, E_F) - G_n^{+}(\boldsymbol{k}_{||}, E_F)\right)/2\pi\mathrm{i}$, where $G_n^{-}(\boldsymbol{k}_{||}, E_F)$ and $G_n^{+}(\boldsymbol{k}_{||}, E_F)$ are the advanced and retarded Green functions of the uncoupled electrodes ($n = 1, 2$) evaluated at the Fermi energy E_F. $T(\boldsymbol{k}_{||})$ is the T-matrix which is defined by

$$T(\boldsymbol{k}_{||}) = V(\boldsymbol{k}_{||}) + V(\boldsymbol{k}_{||}) G^{+}(\boldsymbol{k}_{||}, E_F) V(\boldsymbol{k}_{||}) \tag{2}$$

where $G^{+}(\boldsymbol{k}_{||}, E_F)$ is the retarded Green function of the whole coupled system and $V(\boldsymbol{k}_{||}) = V_1(\boldsymbol{k}_{||}) + V_2(\boldsymbol{k}_{||})$ is the sum of the coupling between the left electrode and the insulator, $V_1(\boldsymbol{k}_{||})$, and between the right electrode and the insulator, $V_2(\boldsymbol{k}_{||})$. Summation over $\boldsymbol{k}_{||}$ in (1) should be carried out within the two-dimensional Brillouin zone. Equation (1) for the conductance is equivalent to the real space representation of the Kubo formula [10–12].

In this letter we assume that the electronic band gap E_g within the insulator is much larger than the magnitude of the hopping integrals between the metals and the insulator. This is a reasonable approximation for insulators like Al_2O_3 where the band gap is about 9 eV which is much larger than a typical value of the tight-binding hopping integrals for 3d metals which are of the order of 1 eV or less. In this case we can consider $V(\boldsymbol{k}_{||})$ in (2) as a perturbation. Retaining only terms up to $\mathrm{O}(V^2)$ in (2) which give a non-zero contribution to the tunnelling current we arrive at the following expression for the conductance:

$$G = \frac{2\pi e^2}{\hbar} \sum_{\boldsymbol{k}_{||}} \mathrm{Tr}\left[\rho_1(\boldsymbol{k}_{||}) V_1(\boldsymbol{k}_{||}) G_{ins}^{-}(\boldsymbol{k}_{||}, E_F) V_2(\boldsymbol{k}_{||}) \rho_2(\boldsymbol{k}_{||}) V_2(\boldsymbol{k}_{||}) G_{ins}^{+}(\boldsymbol{k}_{||}, E_F) V_1(\boldsymbol{k}_{||})\right] \tag{3}$$

where $G_{ins}^{-}(\boldsymbol{k}_{||}, E_F)$ and $G_{ins}^{+}(\boldsymbol{k}_{||}, E_F)$ are advanced and retarded Green functions of the insulator. The error resulting from applying the perturbation theory is of the order of $(V/E_g)^{2N}$, where N is a number of insulator monolayers. It is negligible when $N \gg 1$. Equation (3) is a generalization of the result obtained earlier in [13] for a single-orbital tight-binding model to the case of a general multi-orbital band structure.

The electronic structure of fcc Co(001) and bcc Fe(001) were modelled using s, p and d tight-binding bands fitted to the band structures calculated from first principles [14].

Such a parametrization was successful for the description of GMR in magnetic multilayers [15]. The second electrode was considered to be non-magnetic and was represented by an s band. The matrix elements of the Green function for the semi-infinite electrodes which enter (1) are obtained in terms of the Green functions for the bulk metals [16]. The insulator was represented by a slab consisting of a few monoatomic layers and periodic in a lateral direction. The band structure of the insulator was modelled using two uncoupled s-like tight-binding bands separated by a gap. The Fermi level was always assumed to be in the centre of the gap. The spin polarization of the conductance was defined by $P = (G^{\uparrow} - G^{\downarrow})/(G^{\uparrow} + G^{\downarrow})$, where $G^{\uparrow}$ ($G^{\downarrow}$) is the conductance for majority- (minority-) spin electrons. Within our approximation of a large band gap for the insulator, the spin polarization P is practically independent of the value of the band. It is also stable with respect to the thickness of the insulating layer and the band filling of the non-magnetic metal electrode.

In order to analyse the dependence of the spin polarization of the tunnelling current on the type of bonding between the ferromagnet and the insulator we have performed three sets of calculations. In the first set only ssσ bonding between the ferromagnet and the insulator was taken into account. The value of the ssσ hopping integral was chosen to be the same as that for the ferromagnetic metal. In the second set spσ bonding was included in addition to the ssσ hopping. In the third set all ssσ, spσ and sdσ hopping integrals, chosen to be the same as those for the ferromagnet, were considered.

Figure 1 shows the average density of states (DOS) for the first two surface Co layers and the conductance for the majority- and minority-spin electrons as a function of the electron energy. Although only the tunnelling current at the Fermi energy is relevant for small applied biases, the energy dependence of the conductance is important for understanding the influence of the band structure on the voltage dependence of TMR. This analysis will be performed elsewhere. As is evident from the dashed lines in figure 1(a) the total surface DOS is asymmetric between the majority- and minority-spin electrons. At the Fermi energy it is a factor of eight higher for the minority electrons than for the majority electrons. This behaviour is typical for ferromagnetic cobalt where the Fermi level lies above the majority d band, but lies inside the minority d band. The solid lines in figure 1(b) represent the conductance for the case when only the ssσ interfacial bonding is included. The striking feature of this plot is the change in the sign of the spin polarization of the tunnelling current at the Fermi energy with respect to the spin polarization of the total DOS. $G^{\uparrow}$, in this case, is a factor of two higher than $G^{\downarrow}$, and the spin polarization is about 34%. This value of P is in surprisingly good agreement with the experimental result on the tunnelling between cobalt and aluminium films through an alumina spacer [8]. Taking into account spσ bonding at the cobalt–insulator interface enhances both $G^{\uparrow}$ and $G^{\downarrow}$ by a factor of two for the energies in the vicinity of the Fermi level, but does not change the qualitative features represented by the solid line in figure 1(b).

The change in the sign of P at the Fermi energy with respect to the spin polarization of the total DOS can be explained as follows. In the case when spσ interfacial bonding alone is taken into account, only the s states of the ferromagnet are coupled with those of the insulator. In this case only the s electrons of the cobalt layer contribute to the tunnelling current. Therefore, the spin-polarization of G is entirely determined by the s component of the density-of-states operator ρ_1 in equation (3). In order to reveal a correlation of the tunnelling current with the density of states we have calculated the s-orbital contribution to the surface DOS of Co which is plotted in figure 1(a) by the solid line. As can be seen from this figure the s-electron partial DOS is reduced for the energies within the d band of Co. This is a consequence of the strong s–d hybridization within the cobalt layer.

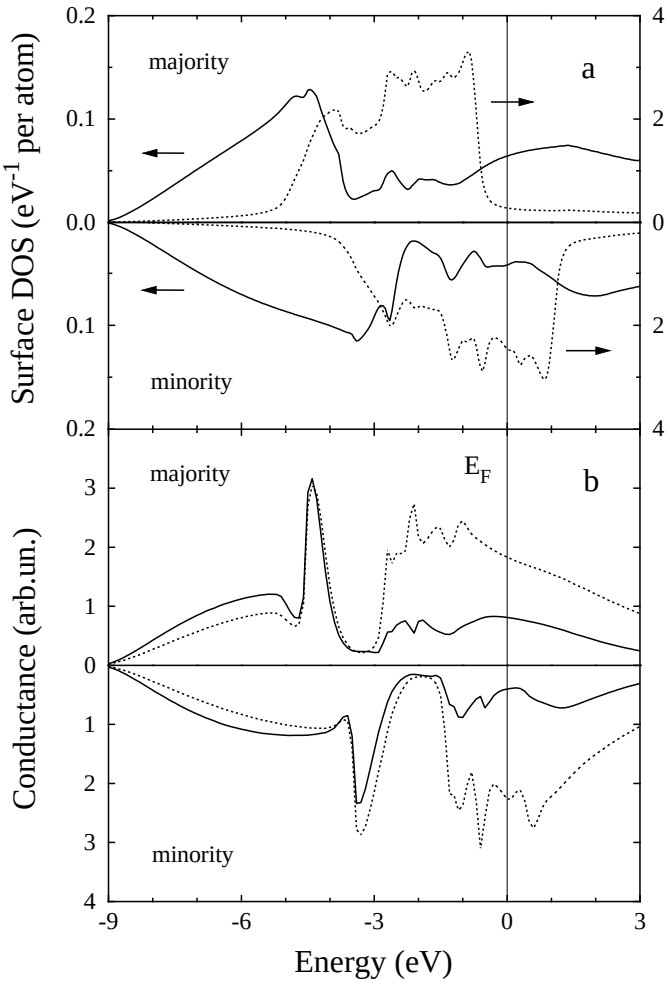

Figure 1. (a) Total density of states (dashed line) and s-electron partial density of states (solid line) averaged over two surface layers for fcc Co surface (001). Note a different scale for the total and partial DOS. (b) Spin-polarized tunnelling conductance from ferromagnetic fcc Co film to a non-magnetic metal as a function of energy. Solid line is the result of calculation when only the ssσ bonding between the ferromagnet and the insulator is taken into account. Dashed line represents the conductance in the case when all the ssσ, spσ and sdσ hopping integrals, set to be the same as in the ferromagnet, are included.

The s component of the DOS qualitatively reproduces the behaviour of the conductance versus energy (compare with the solid line in figure 1(b)). Similar to the conductance, it is higher at the Fermi energy for the majority-spin electrons than for the minority-spin electrons. Therefore, in the case of the ssσ interfacial bonding the change in the sign of P with respect to the spin polarization of the total DOS at the Fermi energy reflects the fact that the spin polarization of the partial DOS for the s electrons of Co is opposite to that for the d electrons.

The dashed line in figure 1(b) shows the conductance in the presence of the ssσ, spσ and sdσ hoppings which we set to be the same as those for cobalt. In comparison to the case

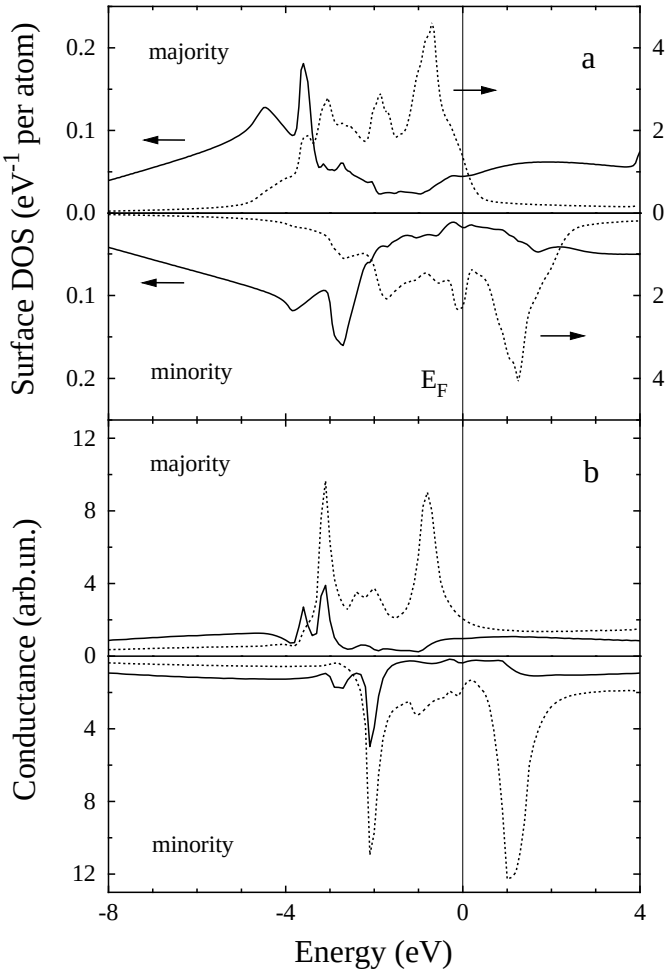

Figure 2. The same as in figure 1 for ferromagnetic bcc Fe(001) film.

of the ssσ bonding alone, the main change in the conductance appears at the energies lying within the d band for both the majority and minority spins. The significant increase in the conductance at these energies demonstrates that in the case when the electron hybridization at the interface is similar to that in the bulk ferromagnet, the d electrons give a large relative contribution to the tunnelling current. This is opposite to the predictions reported in [6]. Because of the position of the Fermi level inside the d band for the minority electrons, switching on the sdσ bonding between cobalt and the insulator results in a larger increase of $G^{\downarrow}$ with respect to $G^{\uparrow}$. This leads to a change in sign of the spin polarization, which is found to be -11%.

Figure 2 shows the surface density of states and the conductance for the majority- and minority-spin electrons in Fe. In contrast to Co the Fermi level in Fe lies inside the d band for both spins and there is no pronounced difference in the total DOS at this energy (figure 2(a)). The s-electron partial DOS has, however, a strong spin asymmetry at the Fermi level (the solid line in figure 2(a)). This asymmetry is reflected in the conductance for the case when only ssσ bonding at the interface is included (the solid line in figure 2(b)).

The value of the spin polarization at the Fermi energy is found to be 45%. Again we obtain very good agreement with the experimental data [8] on tunnelling from iron through an alumina spacer layer. Including spσ bonding at the iron–insulator interface slightly reduces the conductance but does not change qualitative features in comparison to the case when only ssσ bonding at the interface is taken into account. The dashed line in figure 2(b) shows the conductance in the presence of the ssσ, spσ and sdσ hoppings which are set to be the same as those in iron. The conductance at the Fermi energy increases by a factor of two for the majority spins and by a factor of six for the minority spins. It is clear that in this case the contribution of the d electrons to the conductance is dominant and, therefore, cannot be neglected. Switching on the sdσ bonding between iron and the insulator leads to the strong decrease in the spin polarization of tunnelling down to about 8%.

We have, therefore, found excellent agreement with experimental data of the spin polarization of the tunnelling current, provided that the tunnelling current is carried only by the s electrons in the ferromagnetic metal. This indicates that there must exist reasons (different from those based on the band structure of ferromagnets) which make the contribution of the d electrons negligible. One possible reason could be a very weak sd bonding between 3d metals and alumina. In general, the bonding between the electronic orbitals of the metal atoms and the electronic states of the oxide ions depends strongly on the band structure of the metal and the insulator and the geometry of the metal–ceramic interface [17]. Therefore, the character of the interfacial bonding could be quite different depending on the type of ferromagnet and insulator. One would expect different values of TMR depending on the type of the insulator. This was predicted by Slonczewski [18], who showed that even within the free electron approximation the value of TMR is determined not only by the ferromagnet alone but also by an additional interfacial factor which depends on the Fermi wave vectors of the electrons within the ferromagnet and insulator.

Another important reason for the negligible d-electron contribution to the tunnelling current lies beyond the ballistic formulation of the electronic transport. The influence of disorder within the ferromagnetic layer or at the ferromagnetic–insulator interface may result in a stronger scattering of the d electrons in comparison to the s electrons, because of the higher density of states of the former. This could suppress the relative contribution of the d electrons to the tunnelling current and, therefore, result in a dominant s-electron contribution. Analysis of both realistic interfacial coupling and defect scattering will be the subject of future investigations.

In conclusion, we have shown that within a ballistic treatment of conductance the spin polarization of the tunnelling current from 3d ferromagnets depends strongly on the mechanism of the covalent bonding at the interface between the ferromagnetic metal and the insulator layer. For Co and Fe we obtain very good agreement with experiments on tunnelling through an alumina spacer provided that the sdσ bonding at the metal–insulator interface is negligible so that the tunnelling current is carried solely by the s electrons of the ferromagnet. However, including the sdσ bonding at the interface causes an increase of the d-electron contribution to the conductance that results in a decrease of the spin polarization of the tunnelling current and a change in its sign for the case of Co. This indicates the need for the proper treatment of both interfacial bonding and defect scattering.

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