Multiscale modeling of spin transport across a diffuse interface

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We present multiscale calculations to describe the spin transport behavior of the Co/Cu bilayer stucture including the effect of the interface. The multiscale approach introduces the connection between the *ab-initio* calculation used to describe the electronic structure of the system and the generalized spin accumulation model employed to describe the spin transport behavior. We have applied our model to atomically smooth and diffuse interfaces. The results demonstrate the huge importance of the use of first principle calculations, not only due to the interfacial coordinates optimization but also the magnetic and electronic properties obtained through the electronic structure. The system including the effect of interface with and without the charge fluctuation are studied. The results indicate that changes of electronic structure at the Co/Cu interface give rise to an interfacial resistance distributed over several atomic planes, similar to the effect of interface diffusion. We argue that even atomically smooth Co/Cu interfaces have properties analogous to a diffuse interface due to the variation of electronic structure at the interface.

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The understanding of spin transport and spin torque is of increasing importance for spintronic device applications since the discovery of giant magnetoresistance (GMR)[1,2] and tunnelling magnetoresistance (TMR).[3,4] These phenomena have opened a new path for spintronic device design such as magnetic tunnelling junction (MTJ) sensors[5] and magnetoresistive random access memory (MRAM)[6] leading to the development of new generations of computer architecture. In addition, read sensors for conventional magnetic recording rely on transport properties to achieve the desired functionality. Both spin transport and spin torque are phenomena strongly affected by the interface structure and properties which will therefore play a crucial role in determining resistance arising from spin–dependent scattering at the interface.[7–11] From the theoretical point of view, the simulation of a general interface between two different materials is of great complexity. The usual and easiest way to proceed is to have both alloys in contact locating the atoms of one of the materials @(*top*,*hollow*,*bridge*) of the other. A more general situation would be when the atoms of both materials are allowed to move across to the interface leading to interdiffusion within the interfacial region. This diffusion leads in a different degrees of roughness depending on how much the alloys have mixed. Roughness at interfaces as well as the interfacial and intralayer scattering is of huge importance in relation to the objective of achieving high magnetoresistance (MR)[9,11,12].

The calculation of resistance and spin transport behavior across the diffuse interface can be investigated by injecting spin current into the magnetic system which subsequently gives rise to the spin accumulation (SA) close to the interface region. Various theoretical models have been proposed to describe the effect of interfacial roughness on the magnetoresistance[13–15], indicating that the nature of interfaces is an increasingly significant factor in the spin torque phenomenon. Theoretical approaches to spin torque are often based on the SA model of Zhang, Levy and Fert[16] which has recently been generalized to allow the investigation of diffuse interfaces.[17] The magnetic ion concentration at any given position of the system, determined via Fick's law, gives rise to a spatial variation of the transport parameters within the interface. The model described in Ref. 17 is based on an approach which allows treatment of systems with spatially varying magnetization structures by calculating the SA in a rotated coordinate system based on the direction of the local magnetization[18]. A feature of the model given in Ref. 18 is the use of a modified equation of motion for the SA, **m**, as follows

$$\frac{d\mathbf{m}}{dt} + (J/\hbar)\mathbf{m}\times\mathbf{M} = -\frac{\mathbf{m}-\mathbf{m}_\infty}{\tau_{sf}} \qquad (1)$$

where **M** is a unit vector along the local magnetization direction, J is the s−d exchange integral and τ_{sf} is the spin–flip scattering time.

The model derives stationary solutions for SA under the assumption that changes in the magnetization are much slower than the variation of the SA. A feature of Eq. (1) is the introduction of the modified definition of SA as the local value of $(n^{\uparrow}-n^{\downarrow})$ where the $n^{\uparrow(\downarrow)}$ represents the density of states (DOS) at the Fermi level, E_F. A corresponding feature of Eq. (1) is the introduction of the factor $m_\infty = (n^{\uparrow}_{eq} - n^{\downarrow}_{eq})$, where the n_{eq} is the equilibrium bulk value which can be obtained via *ab initio* calculation. This simplifies the calculation of changes in SA for a current flowing between materials with different m_∞.[17]

In this work, we focus on interface properties and their effect on the spin accumulation. First, we consider an atomically flat interface between two different materials. By means of Density Functional Theory (DFT) calculations we investigate the interface electronic structure and its effect on the spin accumulation. The interface is constructed as a periodic *bcc* structure (See Fig.1–A1 and B) without any roughness.

This multiscale approach will be applied to Co/Cu interface to investigate the spin transport behavior as well as evaluate the interfacial resistance. Secondly, we investigate the properties of a diffuse interface created by modelling interdiffusion between the layers. Interestingly it is demonstrated that the interface resistance is spread over several atomic planes in both cases, showing that modification of the interface electronic structure has a similar effect to that of a diffuse interface. The paper is structured as follows. We first describe the spin accumulation model including the calculation of m_∞. We then proceed to investigate the spin accumulation an atomically smooth interface, firstly under the simple assumption of an abrupt change of material properties at the interface. This contrasts strongly with the accumulation calculated for the realistic case taking into account the spatial dependence of m_∞ from the DFT calculations. Finally we present calculations of the spin accumulation for a diffuse interface, which shows a delocalization of the interface resistance similar to that arising from the spatial variation of m_∞.

Spin accumulation model – The full understanding of the mechanism behind GMR and TMR becomes important for the development of spin electronic technologies. The interface resistance can be calculated from the spin accumulation and subsequently gives rise to GMR. Consequently, the calculation of SA is required in order to gain insight into the spin transport behavior. Here, the SA is defined as the the difference of spin–up and spin–down electron populations available from *ab initio* calculations. This is essential to deal with multiple layers with different equilibrium value of SA. The general solution of spin accumulation is solved from Eq. (1) consisting of longitudinal ($\mathbf{m}_\parallel$) and transverse components ($\mathbf{m}_{\perp,2}$ and $\mathbf{m}_{\perp,3}$)[17] (see Supplemental Material).

Ab-initio calculation of interface electronic properties – In principle, a model that describes the most general geometry of Co/Cu interface would be composed of a diffuse interface, i.e., a geometry where the atoms belonging to both alloys are "transferred" –after performing a molecular dynamics simulation, for example– from one alloy to the other, having semi–infinite materials on both sides. Unfortunately, to model interfaces in this fashion is of extreme difficulty using pure *ab initio* calculations due to the huge number of atoms that would be involved. In the present work, we study interface effects by the simulation of three different model systems. In case 1, the interface is taken as atomically smooth and the material properties change abruptly at the interface. In case 2 the interface is again taken as atomically smooth but the SA will be calculated using atomic layer resolved values of m_∞ determined by DFT calculations on systems with relaxed atomic positions. The layers will be patterned by means of the contact of Co and Cu alloys with the same 2D periodicity and repeated periodically out–of–plane (001) as shown in figure 1–B2. Finally, in case 3 we will create a simple model of a diffuse interface by the replacement of one Co atom within the interface plane by one Cu (see figure 1–A2). Again, atomic layer-resolved values of m_∞, calcuated by DFT methods after relaxation, are used for the calculation of the SA. In this case, there is a computational price to pay, in which we require large sizes of the simulation supercells, having more atoms in the simulation.

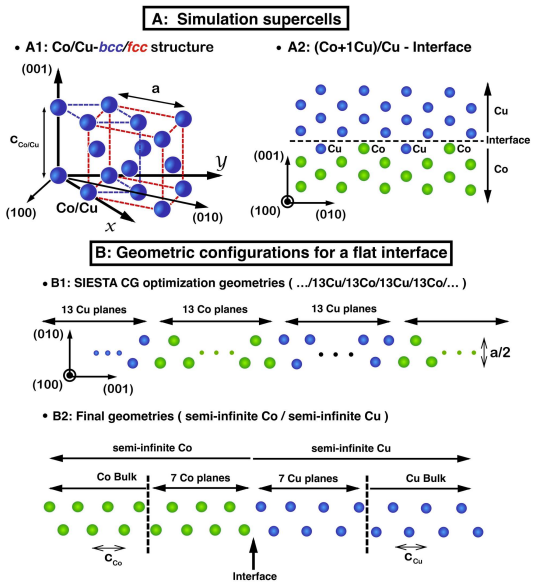

FIG. 1. (Color online) (A1) Schematic representation of the Co and Cu *bcc*/*fcc* bulk unit cells; (A2) Side view of the modeled diffusion interface employed in the present work; (B1) Periodic $\cdots$/13Co/13Cu/$\cdots$ geometric configuration used in the conjugate gradient relaxation method; (B2) Final interface geometry between two semi–infinite Co and Cu bulk alloys after extract 7Co+7Cu slice from B1 relaxed coordinates.

However, we can minimize the computational tasks by choosing conveniently the unit cell. To this end, we observe that in our system the atomic stacking is along the (001) direction and if we inspect the figure 1–A1, we can select either *bcc* or *fcc* to describe the physical systems since both unit cells depict the same structure. The choice of structure will be relevant because of the number of atoms per supercell will be different. Consequently we chose the *bcc* structure due to its reduced number of atoms and because the diffusive interface model will be straightforward to simulate. Subsequently, the spatial magnetic ion concentration is considered from the spatial variation of magnetic moment achieved from the *ab initio* calculations and it is then used to model the spatial diffusive transport parameters. (The detail of interface construction and spatial transport parameters can be seen in Supplemental Material)

To describe the physical mechanism of spin transport including the diffuse interface, we study the structure of the bilayer system of Co/Cu by employing the multiscale model, which calculates the SA with $\mathbf{m}_\infty$ determined from layer resolved *ab initio* information. We consider the bilayer system of Co/Cu with three different interfaces as described earlier. Our general solution of SA is then applied to each system discretized into many thin layers to investigate the spin transport behavior and the interfacial resistance at any position in the system. In the following we consider each case in turn before giving an overall interpretation and drawing conclusions.

Case 1: ideal interface with step change in properties – We first consider the system with the ideal interface by assuming that the concentration of magnetic ion is constant throughout

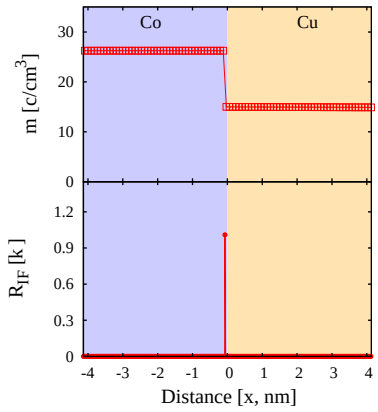

FIG. 2. (Color online) (Top) Spatial spin accumulation and (Bottom) Interfacial resistance of the Co/Cu system with an ideal interface

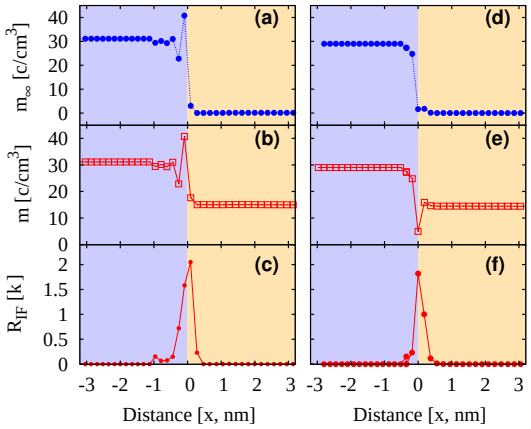

FIG. 3. (Color online) (a) (d) Spatial equilibrium value of spin accumulation (b) (e) Spin accumulation and (c) (f) Interfacial resistance of Co/Cu system with realistic case and rough interface respectively

the Co layer. This case further assumes that the transport properties are spatially invariant in the Co and Cu layers. Specifically, we assume that the equilibrium value of SA, $\mathbf{m}_{\|}(\infty)$, varies discontinuously from the bulk value of Co to that of Cu which is zero. The SA is then investigated by using our modified solution in Ref. 17. The system is discretized into many thin layers in order to calculate and observe the development of SA and spin current by applying the generalized formalism, which propagates the SA solution layer by layer throughout the system. As expected, the spin accumulation is not changed throughout Co layers with collinear magnetization due to the fact that it tends to reach the equilibrium value and its behavior depends on the concentration profile as clearly shown in Fig. 2 (top). In addition, we focus on the spin transport behavior at the interface between layers. The SA is discontinuous at the interface corresponding to the discontinuity of the spin transport parameters of Co and Cu layers at the interface. We note that **m** in the Co layer decays very slowly in the Cu consistent with experiment, due to its large spin diffusion length[19]. The spatial interfacial resistance (R_{IF}) can be calculated directly from the SA (m) and spin current (j_m) as follows,

$$R_{IF} = \frac{|\Delta m| a k_B T}{j_m e^2} \qquad (2)$$

where Δm is the difference of SA across the layers, k_BT is 10 meV or 1.6×10^{-21} J, a is the lattice constant of the material and e is the electron charge. We calculated R_{IF} between each plane using Eq. (2) as shown in Fig. 2 (bottom). In the Co the corresponding resistance is zero due to the constant value of SA. Similarly in the Cu layer the slow variation of SA leads to essentially zero resistance. The interface resistance is confined to the boundary layer due to the discontinuity in the SA.

Case 2: atomically sharp interface with ab initio *parameterization.* – In this case, we study more realistic behavior of spin transport for an atomically structured interface of Co/Cu system by means of the multiscale approach which is proceeded by relaxation of the interface and calculation of the DOS. Subsequently it leads to the calculation of the equilibrium value of SA from $DOS_{\downarrow(\uparrow)}(E_F)$[17]. The result in Fig. 3 (a) depicts the equilibrium value $\mathbf{m}_{\|}(\infty)$ from the *ab initio* calculations. Variations in the DOS close to the interface give rise firstly to a slight polarization of the Cu and secondly to oscillatory behavior in the Co layer close to the interface. The oscillations are due to the interfacial charge rearrangement between both Co and Cu species at the interface. The oscillations in the Co vanish for layers at distances greater than 1 nm, corresponding to the region where the coordinates are similar to those in the Co bulk phase. Using the *ab initio* values of $\mathbf{m}_{\|}(\infty)$ we next calculate the SA as a function of position illustrated in Fig. 3 (b). The SA closely follows the equilibrium value since the incoming spin current is fully polarized in the Co layer. Across the interface, the SA exhibits discontinuous behavior due to the different transport properties of the material of Co/Cu, gradually decreasing to zero associated with the spin diffusion length of Cu, 600 nm. Due to the oscillations in the electronic DOS in the Co, one also observes discontinuous behavior of the SA within the Co atomic layers close to the interface with the Cu.

The layer resolved R_{IF} is further calculation from SA and the spin current according to Eq. (2) as shown in Fig. 3 (c). The results contrast strongly with that of case 1: the atomically sharp interface with an assumed step change in transport properties, where the interface resistance is fully localized to the interface layer. In Fig. 3 (c) it can be seen that the interface resistance is no longer localized to the Co/Cu boundary. Instead R_{IF} is distributed over several atomic planes around the interface. A phenomenon originates as a result of changes in the electronic structure of Co/Cu close to the boundary. In case 1 we made an implicit assumption that there are no charge fluctuations around the interface region. On the contrary, in the realistic case for Co/Cu interface it is also observable that, even though without species (atoms) transferred between alloys, the properties of a real interface are quite different from those predicted by the simplified model of case 1. The electronic structure modification after the geometry optimization, such as the hybridation between different *s–d* orbitals, indicates that our multiscale model, introducing in-

terfacial effects through the *ab initio* calculations within the atomistic model plays an important role in the prediction of the of the spin transport for any system involving interfaces such as studied here.

Case 3: Simple model of a diffuse interface – In practice, sharp interfaces are not expected in real spintronic devices since the sputtering process generally builds diffuse interfaces. As previously mentioned, it is of extreme complexity to perform DFT calculations of these real systems because of the simulation supercell size and hence the large amount of atoms involved. One step forward, compared to the previously studied cases 1 and 2, is the construction of a simple rough interface, where only one atom of one material is replaced by one of the other (see figure 1–A2). In doing so, we take the first step to understand more realistic diffuse interfaces. As in case 2, the rough interface was optimized by means of CG method followed by the self-consistent calculation of the layer resoved DOS. Subsequently $\mathbf{m}_{\|}(\infty)$ is determined. In order to compare the present case with previous ones the layer resolved $\mathbf{m}_{\|}(\infty)$ values are shown in Fig. 3 (d). It reveals that the polarisation extends further into the Cu due to an enhancement of the magnetic moment, and that the rough Co/Cu interface, considerably damps out the oscillations of $\mathbf{m}_{\|}(\infty)$ present in the atomically sharp interface. The physical explanation of the reduction in the oscillatory behavior of $\mathbf{m}_{\|}(\infty)$ in the present case compared to cases 1 and 2 is lies in the smooth geometrical transition between both materials which favors the gradual charge transfer between Co and Cu at the mixed Co/Cu plane.

The transport parameters are estimated from the concentration of magnetic ions. The values of the layer resolved SA is given in Fig. 3(e). We observe that the SA exhibits discontinuous behavior close to the interface and shows damped oscillations compared to case 2. However, the spatial variation of the SA is again delocalized from the interface due to the combination of interface roughness and the spatial dependence on the electronic structure. This is reflected in the layer resolved interface resistance R_{IF} as is shown in Fig 3 (f). Similar to case 2 we note that the interface resistance is delocalized from the interface region.

In conclusion, we employed a recent developed formalism of SA that allows to describe its behavior at any position of any FM–NM interface configuration. In addition, the model makes the possibility for the treatment of systems with sharp variation of SA and also smoothly spatial magnetization concentrations. Furthermore, it is possible to use m_∞ defined as the difference of the $DOS_\downarrow$ and $DOS_\uparrow$ at Fermi level, E_F, quantities readily available from the *ab initio* calculations. Hence, we proposed a multiscale model to give rise the possibility to study the spin transport behavior at any given position of any material through the quantum mechanical calculations and atomistic simulations.

We have applied the multiscale spin accumulation (MSA) model to the Co/Cu interface for 3 cases. Firstly, we considered the ideal case of an atomically smooth interface with a step change in properties at the interface. This system exhibits discontinuous behavior of the spin accumulation and an interface resistance localized to the Co/Cu boundary. We then investigated two more realistic cases, namely, case 2 assuming an atomically smooth interface but with equilibrium polarization calculated from *ab initio* models and case 3 which introduced a first approximation to a rough interface. Both case 2 and case 3 give rise to similar delocalisation of the interface resistance, which is significant over a few lattice spacings. Interestingly, the rough interface gives rise to a smoother variation of the polarization close to the Co/Cu interface. Both case 2 and case 3 exhibit significantly increase values of interface resistance over and above the simplified model of case 1.

Clearly the presence of surface roughness; expected in practice due to the nature of the sputtering process by means of which most systems and devices are produced, has a significant effect on the transport properties as characterized here by the interface resistance. The result of a rough interface is to give rise to an increase of interface resistance which is delocalized from the Co/Cu boundary and extends over a few lattice spacings. Interestingly, a similar effect is predicted for an atomically smooth interface which might be obtained by MBE. In this case the increased interface resistance and delocalization from the Co/Cu interface results from the modification of the electronic properties of the Co and Cu due to the presence of the interface. The simple model of a bilayer as atomically smooth with a step change in properties is not sufficient, certainly to describe the properties of a Co/Cu bilayer. The properties are determined by a complex mixture of the electronic density of states which gives rise to an oscillatory polarization in the Co and the interface roughness which, although it tends to damp out the DOS oscillations, still results in an increased interface resistance delocalised from the Co/Cu boundary.

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