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Perpendicular transport in layered magnetic systems: ab initio study

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Abstract

We will review the results of the ab initio study of the current-perpendicular-to-plane magnetotransport of a sample sandwiched by two ideal leads. The 'active' part of the system is either a trilayer consisting of two magnetic slabs of a finite thickness separated by a non-magnetic spacer, or a multilayer formed by alternating magnetic and non-magnetic layers. We use the Kubo-Landauer formulation of the conductance based on surface Green functions as formulated by means of the tight-binding linear muffin-tin orbital method. The formalism is extended to the case of lateral supercells with random arrangements of atoms of two types, which in turn allows to deal with specular and diffusive scattering on equal footing. Applications refer to fcc-based Co/Cu/Co(0 0 1) multilayers including the transport through layers with non-collinear alignments of their magnetizations. We consider in detail the effect of substitutional alloying in the spacer as well as interdiffusion at the interfaces between magnetic and spacer layers.

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1. Introduction

Transport in layered materials has been subject of intensive theoretical investigations, in particular in view of the discovery of the giant magnetoresistance (GMR) in metallic multilayers [1]. Most of the measurements up to date were reported for the current-in-plane (CIP) geometry [2] since the current-perpendicular-to-plane (CPP) geometry [3] is experimentally more challenging. On the other hand, from the theoretical point of view [5], the CPP current differs from the CIP current in several aspects: the high-symmetry of the CPP geometry makes its theory easier which is particularly convenient for an ab initio study, it is better suited for testing of theoretical models, and it gives larger value of the GMR as compared to the CIP geometry [6]. Last but not least, the CPP transport is also closely related to the tunneling through a nonmetallic spacer and to the ballistic transport [4].

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Ab initio calculations of the GMR are still rather rare. We mention the method of solving the Boltzmann-equation applied to multilayers [7] and a Kubo–Greenwood approach generalized to layered systems in terms of the Layer Korringa–Kohn–Rostoker (KKR) method [8] as well as in terms of the relativistic spin-polarized screened KKR method [9] but neglecting vertex corrections with respect to the configurational average of the products of two single-particle Green functions. Both approaches can, at least in principle, be used for CIP as well as for CPP.

Alternative theoretical approaches applicable to the CPP transport are based either on a non-equilibrium Green function method [10] or on a transmission matrix formalism [11,12] and were implemented within an empirical tight-binding (TB) method based on surface Green functions. In this paper we wish to present such an approach formulated within the ab initio tight-binding linear muffin-tin orbital (TB-LMTO) method [16] valid for a general stacking of random layers. The disorder is treated in terms of lateral two-dimensional supercells within each random atomic layer. The present formulation also allows to treat the non-collinear alignment of layer magnetizations which is characteristic for domain-walls [13,14] or for magnetic multilayers in external magnetic field [15].

2. Formalism

We assume that the system consists of non-random semi-infinite left ($\mathscr{L}$) and right ($\mathscr{R}$) leads sandwiching a sample consisting of a left and a right magnetic slab separated by a non-magnetic spacer of varying thickness (a trilayer geometry). Alternatively, a multilayer consists of a set of non-magnetic and magnetic layers such that a ferromagnetic and an antiferromagnetic (or, more generally, a rotated) configuration can be formed. In principle, atomic layers can be viewed in terms of $n_1 \times n_2$ supercells ($n_1 \times n_2$ two-dimensional complex lattice). In order to describe disorder (substitutional binary alloys) it is then necessary to average over different occupations of the sites within the supercell by the two constituents involved and, at the end, to check the dependence of conductances on the supercell size. Quite clearly such an approach applies to disordered spacers and/or magnetic slabs as well as to disordered interfaces. In the following we neglect possible layer and lattice relaxations in the system, i.e., all formulations are based on an infinite parent lattice.

The electronic structure of the system is described by the following TB-LMTO Hamiltonian,

$$H^{\gamma,\sigma}_{RL,R'L'} = C^{\sigma}_{RL}\ \delta_{R,R'}\delta_{L,L'} + (\Delta^{\sigma}_{RL})^{1/2} \times \{S^{\beta}(1-(\gamma^{\sigma}-\beta)S^{\beta})^{-1}\}_{RL,R'L'}(\Delta^{\sigma}_{R'L'})^{1/2}, \tag{1}$$

where R is the site index, σ is the spin index, the potential parameters C^{σ}_{RL}, Δ^{σ}_{RL}, and γ^{σ}_{RL} are diagonal matrices with respect to the angular momentum index $L = (\ell m)$. The non-random screened structure constants matrix $S^{\beta}_{RL,R'L'}$ and the screening matrix $\beta_{R,LL'} = \beta_L \delta_{L,L'}$ are spin-independent. Assuming one and the same two-dimensional translational symmetry in each layer p, $\mathbf{k}_{\parallel}$-projections can be defined, where $\mathbf{k}_{\parallel}$ is a vector from the corresponding surface Brillouin zone (SBZ). In a principal-layer formalism [17], the screened structure constants $S^{\beta}_{p,q}$ are of block tridiagonal form. Neglecting layer relaxations they are given by

$$S^{\beta}_{p,p}(\mathbf{k}_{\parallel}) = S^{\beta}_{0,0}(\mathbf{k}_{\parallel}),$$
$$S^{\beta}_{p,q}(\mathbf{k}_{\parallel}) = S^{\beta}_{0,1}(\mathbf{k}_{\parallel})\delta_{p+1,q} + S^{\beta}_{1,0}(\mathbf{k}_{\parallel})\delta_{p-1,q}. \tag{2}$$

The properties of the individual atoms are characterized by potential function matrices,

$$P^{\beta,\sigma}_{R}(z) = \frac{z - C^{\sigma}_{R}}{\Delta^{\sigma}_{R} + (\gamma^{\sigma}_{R} - \beta)(z - C^{\sigma}_{R})} \tag{3}$$

which are diagonal with respect to L and are obtained by solving the corresponding Schrödinger equation within the density functional formalism. In the case of a binary substitutional alloy the potential functions assume two different values (we thus neglect possible local environment effects within a supercell) [18]. Finally, we define the Green function matrix $g^{\beta,\sigma}(z)$ in the TB-LMTO method as

$$(g^{\beta,\sigma}(\mathbf{k}_{\parallel}, z))^{-1}_{p,q} = P^{\beta,\sigma}_{p}(z)\delta_{p,q} - S^{\beta}_{p,q}(\mathbf{k}_{\parallel}). \tag{4}$$

We refer the reader to a recent book [17] for further details concerning the TB-LMTO method

for layered systems, in particular how the problem originally infinite with respect to the layer indices p, q can be reduced to the finite problem by introducing the concept of the surface Green function.

A detailed derivation of the magnetoconductance in the present context can be found in a recent paper [18], here we give just a summary of results. The conductance $\mathscr{C}(\theta)$ is given by

$$\mathscr{C}(\theta) = \frac{e^2}{h}\frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel}\mathrm{Tr}\{B_{\mathscr{L}}(\mathbf{k}_\parallel, E_\mathrm{F}) g_{1,N}(\mathbf{k}_\parallel, \theta, E_\mathrm{F}^+) \times B_{\mathscr{R}}(\mathbf{k}_\parallel, E_\mathrm{F}) g_{N,1}(\mathbf{k}_\parallel, \theta, E_\mathrm{F}^-)\}, \quad (5)$$

where, in the spirit of Landauer formulation, the integrand can be considered as the corresponding transmission coefficient. In Eq. (5), $N_\parallel$ is the number of $\mathbf{k}_\parallel$-points in the SBZ, E_F is the Fermi energy of the system, $E_\mathrm{F}^\pm = E_\mathrm{F} \pm \mathrm{i}\delta$, Tr denotes the trace over the angular momentum index $L = (\ell m)$ and over the spin index, and θ is the angle between magnetizations in magnetic layers. It should be noted that due to non-collinear alignments of magnetizations the spin index is no longer a good quantum number. All quantities are therefore treated as 2×2-supermatrices in the spin subspace. It should be noted that Green functions $g_{1,N}(\mathbf{k}_\parallel, \theta)$ and $g_{N,1}(\mathbf{k}_\parallel, \theta)$ have to be transformed from the reference coordinate system (ferromagnetic alignment) to the local coordinate system characterized by the angle θ in a given layer by the unitary rotation corresponding to the spin 1/2. Only the potential functions need to be transformed as the structure constants are spin independent. Properties of semi-infinite leads are fully described by the quantities $B_{\mathscr{L}}$ and $B_{\mathscr{R}}$ which can be expressed in terms of lead surface Green functions

$$\begin{aligned} B^{\beta,\sigma}_{\mathscr{L}}(E) &= iS^\beta_{1,0}(\mathbf{k}_\parallel)[\mathscr{G}^{\beta,\sigma}_{\mathscr{L}}(\mathbf{k}_\parallel, z_+) - \mathscr{G}^{\beta,\sigma}_{\mathscr{L}}(\mathbf{k}_\parallel, z_-)]S^\beta_{0,1}(\mathbf{k}_\parallel), \\ B^{\beta,\sigma}_{\mathscr{R}}(E) &= iS^\beta_{0,1}(\mathbf{k}_\parallel)[\mathscr{G}^{\beta,\sigma}_{\mathscr{R}}(\mathbf{k}_\parallel, z_+) - \mathscr{G}^{\beta,\sigma}_{\mathscr{R}}(\mathbf{k}_\parallel, z_-)]S^\beta_{1,0}(\mathbf{k}_\parallel), \end{aligned} \quad (6)$$

while $g_{1,N}$ and $g_{N,1}$ are layer off-diagonal blocks of the TB-LMTO Green function of the whole system evaluated between the terminal sample layers $p = 1$ and $p = N$. Finally, we will define the magnetoresistance ratio as $\mathrm{MR} = \mathscr{R}(\theta)/\mathscr{R}(0) - 1 = \mathscr{C}(0)/\mathscr{C}(\theta) - 1$, where $\mathscr{R}(\theta) = 1/\mathscr{C}(\theta)$, and $\mathscr{C}(\theta)$ is given by Eq. (5).

A generalization to the case of $n \times n$ lateral supercells is straightforward: the equations remain formally identical, only the matrices are replaced by supermatrices with respect to inequivalent atoms in the supercell, and the $\mathbf{k}_\parallel$-integration is confined to the (n^2-times smaller) SBZ corresponding to the supercell.

3. Numerical results and discussion

The potentials of individual atoms of a binary substitutional alloy $A_{100-x}B_x$ as simulated by 5×5-supercells are approximated by the potentials of A and B atoms calculated within the coherent potential approximation (CPA), i.e., we neglect very small fluctuations due to a varying local environment of atoms A and B within a supercell. In addition, we used the same potentials for the reference ferromagnetic system and for the system with non-collinear alignment of magnetic moments which is justified by the magnetic force theorem [19]. The calculations were performed for a small value of $\delta = 10^{-7}Ry$, the conductance was evaluated by averaging over five random configurations because the current fluctuations were found to be quite small (of order of few percent [18]), and typically 1600 (64) $\mathbf{k}_\parallel$-points in the SBZ (supercell SBZ) were used.

3.1. Diffusive transport: bulk and interface resistivities

We first evaluate the resistance of an fcc(0 0 1) trilayer $5\mathrm{Co}|n\mathrm{Cu}_{84}\mathrm{Ni}_{16}|5\mathrm{Co}$ ($n = 1$–150 MLs) sandwiched by ideal semi-infinite Cu leads. The bulk resistivity of the alloy spacer is estimated from the slope of the linear dependence of the resistance as a function of the spacer thickness while the interface resistance is estimated from the offsets of the parallel (P) and antiparallel (AP) resistances (see Fig. 1). We have found the resistivity of fcc-$\mathrm{Cu}_{84}\mathrm{Ni}_{16}$ alloy 11.8 $\mu\Omega$ cm as compared to the experimental value of 15.3 $\mu\Omega$ cm. Recent theoretical

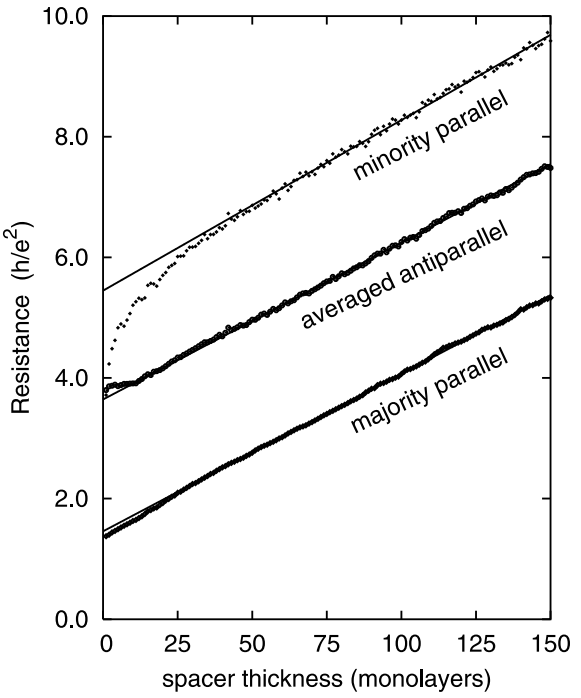

Fig. 1. Resistances of majority and minority P and AP alignments for an fcc(0 0 1) trilayer 5Co|nCu$_{84}$Ni$_{16}$|5Co sandwiched by ideal semi-infinite Cu leads. Due to a geometry, minority and majority channels for AP alignments are the same. Lines serve as a guide for eye.

calculations based on the KKR-CPA method including vertex corrections give 13.32 μΩ cm [20] for fcc-Cu$_{80}$Ni$_{20}$. It should be noted that present supercell calculations include effect of vertex corrections correctly. The interface resistivity is characterized by a dimensionless quantity $\gamma = (AR^{\downarrow} - AR^{\uparrow})/(AR^{\downarrow} + AR^{\uparrow})$ and by $AR^{*} = (AR^{\downarrow} + AR^{\uparrow})/4$, where R^{σ} ($\sigma = \uparrow, \downarrow$) denotes interface resistance and A is the area of interface. Typical experimental results for Cu/Co multilayers [6] are $\gamma = 0.71$–0.77 and $AR^{*} = 0.38$–0.51 fΩ m^2. Present results $\gamma = 0.76$ and $AR^{*} = 0.56$ fΩ m^2 compare well with the experiment. Theoretical results [21] for P and AP interface resistances for ballistic Co/Cu system are 0.35 and 1.90 fΩ m^2, respectively, 0.33 and 1.79 fΩ m^2 for Co/Cu/Co system with interface disorder [21] to be compared to 0.27/2.00 fΩ m^2 in the present Co/Cu$_{84}$Ni$_{16}$/Co system. The interface resistance thus seems to be determined primarily by differences in the electronic structure of magnetic and spacer layers at sample interfaces: the similarity between majority Co-bands and Cu-bands as contrasted to a large difference between the minority Co-bands and Cu-bands. These differences dominate also for the case of Cu-rich CuNi spacer and the effect of disorder is less important. The present approach gives a reasonable agreement with existing experimental and theoretical data for both the bulk and interface resistivities.

3.2. Diffusive transport: separation of specular and diffusive parts

The CPP transport in alloyed samples differs from that in bulk alloys and/or the CIP transport in a few aspects, one which we wish to illustrate here is the relevance of the disorder-induced vertex corrections for the CPP transport. This fact is known from model studies [5], here we wish to show it for the case of a realistic system described in detail in the previous subsection. The results are presented in Fig. 2. The parallel conductance dominates, its value is primarily due to a majority parallel channel. The most remarkable effect is the relevance of diffusive part of conductance which dominates the transport already for samples thicker than about 20 monolayers (MLs). The contribution of the ballistic or specular part of the transport becomes quickly negligible thus illustrating the relevance of vertex-corrections. Their evaluation in the framework of the CPA represents a difficult task and till now there is no such study on ab initio level. It is a great advantage of the supercell approach that vertex corrections are included from the very beginning. It should be noted that their neglect gives the result which is essentially equivalent to the specular part of the transport in Fig. 2.

3.3. Interleaved and separated geometries

The last example, motivated by a recent paper [15], will concern the ab initio study of the effect of order of magnetic and non-magnetic layers on the CPP-GMR of interleaved (IL) and separated (SP) multilayers. In particular, the emphasis is put on the effect of non-collinear alignments of layer

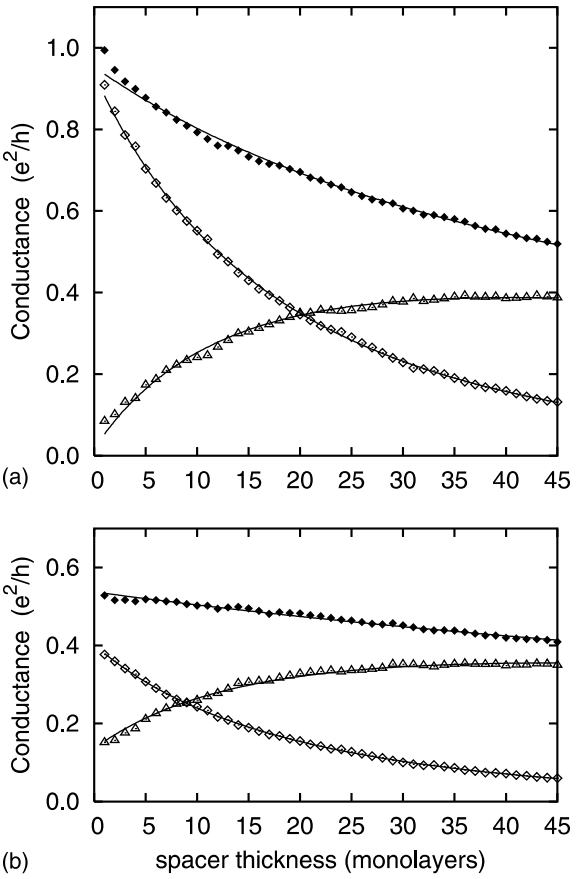

Fig. 2. Conductances of (a) P and (b) AP alignments of an fcc(0 0 1) trilayer 5Co|nCu$_{84}$Ni$_{16}$|5Co sandwiched by ideal semi-infinite Cu leads. The total (◆) conductance is separated into the ballistic or specular part (◇) and the diffusive (△) part. Lines serve as a guide for eye.

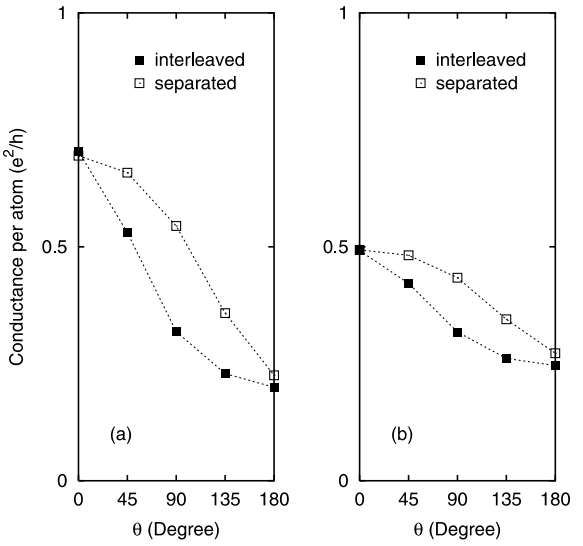

Fig. 3. The conductance of IL (full symbols) and of SP (empty symbols) multilayeres: (a) interdiffusion at sample interfaces, and (b) alloying in spacer layeres. The lines serve as a guide for eye.

magnetizations omitted in a previous study [15]. We have performed calculations for two types of fcc(0 0 1)-multilayers sandwiched by ideal Cu(0 0 1) leads which differ from each other only in the ordering of magnetic and non-magnetic layers, namely the IL and SP multilayers. The IL geometry is described by the structure formula $[M(m_1)/S(s)/M(m_2)/S(s)]_4$ while the SP geometry can be described as $[M(m_1)/S(s)]_4\ [M(m_2)/S(s)]_4$, where $M(m_1) = 10$ Co MLs and $M(m_2) = 5$ Co MLs for both IL and SP samples. We have considered two models of randomness: (i) Co/Cu multilayers with $Cu_{50}Co_{50}$ interface interdiffusion at two adjoining interface layers, and (ii) Co/$Cu_{84}Ni_{16}$ multilayers with random spacer slabs and ideal interfaces. The spacer thickness is 5 Cu MLs so that the total sample thicknesses are 95 MLs. The results for IL and SP multilayers are presented in Fig. 3 as a function of the rotation angle θ between magnetizations of magnetic slabs: $\theta = 0$ and π correspond, respectively, to P and AP alignments. The following conclusions can be drawn: (i) The differences between IL and SP geometries are quite small for collinear alignments (P and AP), for P in particular; (ii) The rotated samples show a pronounced dependence on the layer order in a qualitative agreement with the experiment [15]. There is, however, no simple relation between the angle θ and the value of the applied external field in the experiment [15]. We have also found that above differences become smaller with increasing thicknesses of disordered spacer slabs. On the contrary, the differences between the IL and SP samples increase with number of repetitions, as it can be concluded by comparing results for two and four repetitions, again in an agreement with [15]; (iii) The conductance of the SP sample is larger as compared to the IL sample in a qualitative agreement with experiment [15]: electrons

passing through the IL sample suffer additional spin scatterings at interfaces with thicker and narrower magnetic slabs with rotated magnetizations as compared to the SP sample with magnetizations rotated only at the interface which separates thicker and narrower slabs; (iv) The parallel alignment for interdiffusion case (Fig. 3a) has larger conductance as the number of disordered layers is smaller as compared to the case of random spacer sample.

4. Conclusions

The transport in magnetic multilayers is of a great scientific and technological interest (sensors, non-volatile memories, magnetic reading heads, etc.). The ab initio, i.e., a parameter-free, approach to the evaluation of the transport in magnetic multilayers is thus of a great practical importance. In the present paper we have presented such approach, particularly suitable for the CPP transport, which allows to evaluate the sample magnetoconductance under rather general conditions: both the ballistic and diffusive transport, and generally non-collinear alignments of layer magnetizations are possible. The randomness was included in terms of lateral two-dimensional supercell approach which allows to treat properly the disorder-induced vertex corrections, which are particularly important for a correct evaluation of the CPP current. We have also demonstrated that present calculations properly reproduce the bulk alloy limit as well as the interface resistivity in a good agreement with available experimental data and with other theoretical approaches. Finally, we have shown the sensitivity of the magnetocurrent to the order of magnetic and non-magnetic layers in the sample (IL and SP samples) in a qualitative agreement with a recent experiment.

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