Interlayer magnetic coupling: Effect of interface roughness

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The effect of structural interface imperfections on the interlayer magnetic coupling is studied theoretically by considering (i) a macroscopic model of roughness for possible fluctuations of spacer thickness, and (ii) a microscopic model of interdiffusion at ideal interfaces. For the Co/Cu/Co(001) system, we found a dramatic decrease of the amplitudes of the oscillations even for a small amount of interface imperfections, particularly strong for the short period oscillations, and thus indicating a possible source of discrepancy between *ab initio* calculations and experiment.

Since the discovery of the oscillatory interlayer exchange coupling (IEC) between magnetic layers separated by a nonmagnetic spacer, there has been an intense effort to understand this phenomenon theoretically and to find physical parameters which influence the periods and amplitudes of the oscillations.[1] The interest in magnetic multilayers was additionally motivated by the discovery of the giant magnetoresistance (GMR) in these materials.[2] A simple method to examine the importance of interfacial scattering proved to be the so-called planar doping,[3] namely the insertion of layers of selected elements at the interfaces between magnetic and spacer layers (experiments were performed by Guerney *et al.*[4]) Depending on the type of inserted material,[2,3] a strong influence of planar doping on both the GMR and the IEC was found. Since experimental evidence demonstrates that in reality islanding, foreign atoms (impurities), rough interfaces, or interdiffusion[5] cannot be avoided, it seems to be of quite some importance to understand in particular the scattering from nonideal interfaces with respect to the IEC and the GMR.

Up to now first-principles approaches to the IEC were applied for idealized interfaces and fall into two categories: (i) total energy calculations using mostly supercell geometries, which being numerically demanding seem to be limited to small cell sizes,[6] and (ii) methods that directly separate the small interface-interface band energy contribution to the total energy by using a Green's function particularly suited for layered structures. In connection with Lloyd formulation[7] for the integrated density of states adapted to layered structures, these methods[8–10] are also suitable for study of random systems within the well-established coherent potential approximation (CPA).[11] In the present approach[10] extensive use is made of two-dimensional translational symmetry and of the short range of the interlayer interactions in the tight-binding linear muffin-tin orbital method (TB LMTO).[12] The block tridiagonal form of the structure constant matrices with respect to layer indices allows us to calculate the coupling energies over a large number of spacer layers since the numerical effort scales linearly with the system size. A discrete Fourier transform of the coupling energies can be performed, which in turn serves to identify the periods and amplitudes of the oscillations, in particular the long period ones. In principle, according to the formulation given in Ref. 10, the potentials as well as the atomic composition can vary from layer to layer. The present approach comprises the Ruderman-Kittel-Kasuya-Yosida (RKKY) type theories and the full-confinement quantum-well approaches[1] as limiting cases of weak and strong coupling, respectively.[10]

The purpose of this paper is to investigate the role of two general cases of imperfections frequently occurring at interfaces of artificially prepared materials like multilayers, namely interfacial roughness and interfacial interdiffusion, which — as a new feature — are treated within the same, parameter-free electronic structure model. Consider the situation shown in Fig. 1 (top), namely two magnetic slabs embedded in a nonmagnetic spacer. In order to discuss the case of interfacial roughness, one can use the model proposed in Ref. 13, namely large flat terraces of monolayer heights fluctuating randomly in both directions around an ideal interface with the probability r. We assume no correlation between the formation of such terraces at neighboring interfaces. The deviations of the actual spacer thickness n from its mean value L are specified by the probabilities $w(L-n)$, where $w(0)=(1-2r)^2+2r^2$, $w(\pm 1)=2r(1-2r)$, $w(\pm 2)=r^2$, and $w(m)=0$ for $|m|>2$. In principle r is confined to $0\leqslant r\leqslant 0.5$, however, the model describes physically meaningful situations only for a smaller range, say for

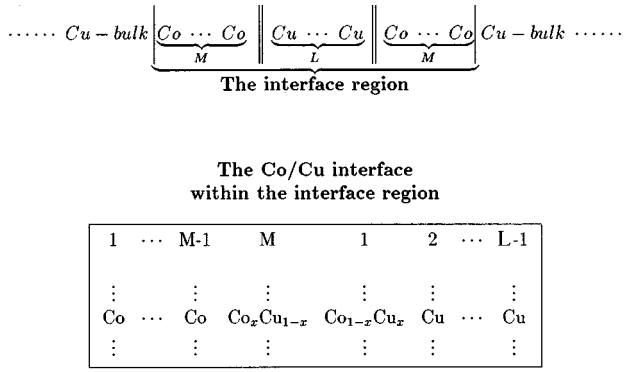

FIG. 1. Top: The interface region sandwiched between two semi-infinite fcc-Cu(001) substrates. Double and single vertical lines denote inner and outer slab interfaces, respectively. Bottom: The model of interface interdiffusion assuming intermixing of Co and Cu atoms in adjoining layers at inner slab interfaces. The same model is also used for outer interfaces.

$0 \leqslant r < \sim 0.3$. The exchange coupling energy $\mathscr{E}_x$ is defined as the energy difference between a ferromagnetic and an antiferromagnetic alignment of the magnetization in the magnetic layers [for a definition of $\mathscr{E}_x$ in the present context see Eq. (1) of Ref. 10]. Its statistical average with respect to the average spacer thickness L is given by

$$\bar{\mathscr{E}}_x(L) = \sum_n w(L-n)\mathscr{E}_x(n).$$

The convolutionlike form of the relation between $\mathscr{E}_x(n)$ and $\bar{\mathscr{E}}_x(L)$ implies that the amplitudes of oscillations with wave number k are reduced due to roughness by a factor $\tilde{w}(k) = [1 - 4r\sin^2(k/2)]^2$, namely the Fourier transform of the probabilities $w(m)$.

In the case of interfacial interdiffusion, the interdiffusion of magnetic and nonmagnetic atoms at respective interfaces into the nearest-neighboring layers of other material is assumed in terms of (random) alloying corresponding to a given concentration profile, as shown in Fig. 1 (bottom), which can be described theoretically by means of an inhomogeneous CPA.[11] We have verified numerically that the neglect of the corresponding vertex corrections[14] due to a correlated motion of two electrons in the same random field has negligible effects on the values of coupling energies.[15] The problem of statistical averaging is thus reduced to the evaluation of the coherent potentials of an isolated random slab in a spacer[11] and to the evaluation of the corresponding surface Green's function.[16] This ''frozen'' CPA (Ref. 10) is consistent with the concept of frozen potentials used throughout this paper.

The CPA, which is an effective medium theory, neglects any statistical correlations in the random system. The damping of electronic states at the Fermi energy, however, as well as concentration-dependent trends are described correctly. The present model is thus sufficiently general in the present context. It should be noted that the effect of interdiffusion in Fe/Cr multilayers was previously modeled via interfacial ordered compounds[17] using an empirical tight-binding method and completely neglecting the effect of randomness. The physical mechanism of a suppression of the amplitudes, namely the occurrence of frustrated Fe-Cr bonds at the interface and the Cr magnetic instability, differs therefore crucially from those addressed in the present paper. Quite clearly both models for interfacial imperfections can be improved, namely by considering terraces with steps higher than one monolayer and taking into account fluctuations of the thickness of the magnetic slabs, by interdiffusion comprising more interface layers, or by taking into account interface-near layers with vacancies. An extension of the present scheme to the case of periodic multilayers is also possible.

The numerical studies were performed for slabs of Co embedded in a fcc-Cu(001) spacer. The advantage of studying this system is the magnetically passive spacer and the stability of the Co moment in different configurations [as contrasted with, e.g., the Fe/Cr/Fe(001) system.[13,17]] The self-consistent potentials for the bulk Cu and for a single Co monolayer in a bulk Cu(001) are used also for the interacting slabs, since self-consistent potentials from (i) bulk fcc Cu and fcc Co calculations with aligned Fermi levels, (ii) single Co monolayer embedded in a Cu host,[9] and (iii) Co/Cu interface calculations with four layers on each side of the interface determined self-consistently in each layer yielded similar results. In order to avoid numerical difficulties with the phase of the complex logarithm needed in the Lloyd formula (see also Ref. 10), the energy integrals were performed along a contour perpendicular to the real axis, namely along $z = E_f + i\delta$, $0 \leqslant \delta \leqslant \infty$, using a Gaussian quadrature for typically 20 energy points. All calculations were performed for $T = 0$ K to subtract the net effect of disorder which might be hidden under a combined effect of the finite temperature and the spacer thickness.[18] A large number of $\mathbf{k}_\parallel$ points in the irreducible surface Brillouin zone (typically a few thousands) was used for the very first points on contour close to the real axis while for energies considerably off the real axis the number of $\mathbf{k}_\parallel$ points is significantly smaller. We have checked convergency of $L^2\mathscr{E}_x$, where L is the actual spacer thickness. A discrete Fourier transform $F(k)$ was then performed for a set of $L^2\bar{\mathscr{E}}_x$ values, where L varies typically from 10 to 50, including in some cases also values of L up to 80 and the results in both cases agreed well. The periods of oscillations Λ_i (in monolayers) were identified from the positions k_i of pronounced peaks $|F(k_i)|$ as $\Lambda_i = 2\pi/k_i$. Amplitudes of oscillations A_i were estimated from $A_i = (2/p)|F(k_i)|$, where p is the number of $L^2\bar{\mathscr{E}}_x$ values used in the Fourier analysis. It should be noted that our definition of the amplitudes of oscillations is consistent with the RKKY limit[1] and it is particularly suitable for the present study.

From the results presented in Figs. 2 and 3 and in Table I the following conclusions can be drawn:

(i) The two pronounced peaks found for Co(001) slabs of one monolayer (ML) and 5 ML thickness (Fig. 2) clearly indicate the presence of long and short period oscillations in the system. For thick magnetic slabs, namely 10 Co layers and more, the long period oscillations are suppressed and nearly vanish[10,19] in the limit of very thick slabs (Fig. 2, full line). The period of long oscillations for monolayer Co slabs[9] at $k_1 \approx 0.95$, which corresponds to 6.6 ML of the spacer, is reduced for 5 ML Co slabs to about 5.2 ML of the spacer,

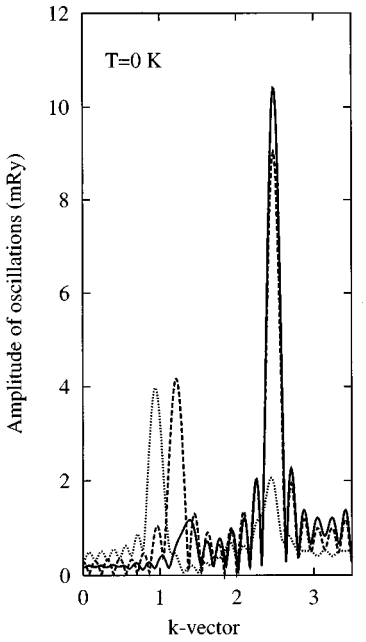

FIG. 2. Discrete Fourier transform $|F(k)|$ of $L^2\bar{\mathscr{E}}_x$ for a finite set of spacer layers ($L=10$–50 layers) and for the case of ideal interfaces: two semi-infinite Co(001) subsystems sandwiching the Cu-spacer (full line), two Co(001) slabs in fcc-Cu each 5 monolayers thick (dashed line), and two Co(001) monolayers in fcc-Cu (dotted line). The background oscillations are due to the finite data sets used for the Fourier transformation.

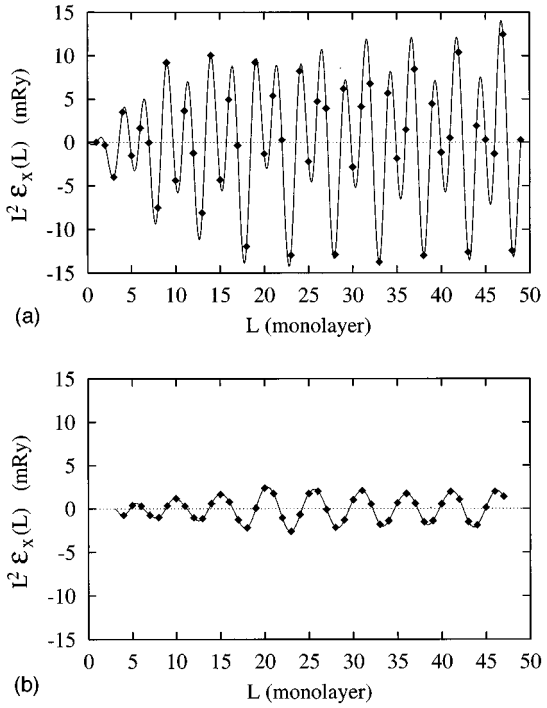

FIG. 3. The exchange coupling $L^2\bar{\mathscr{E}}_x$ as a function of the spacer thickness L for two Co(001) slabs in fcc-Cu each 5 monolayers thick and for the model of interface roughness: (a) ideal interface (roughness $r=0.0$) and (b) interface roughness $r=0.25$. Diamonds refer to the calculated values. A back Fourier transform of $F(k)$ was used to obtain a continuous interpolation (full line) between the calculated values.

while their amplitudes remain approximately the same. In contrast to the strong thickness dependence of their amplitudes, the periods of the short oscillations at $k_1 \approx 2.5$ (2.5 ML of the spacer) are insensitive to the Co slab thickness.

(ii) As can be seen from Table I, the effect of interfacial roughness on the coupling strength is quite large and is in particular strong for the short period oscillations, which are almost wiped out for $r=0.25$. The long period oscillations survive although their amplitude is reduced approximately by a factor of 2. From the analytical form of $\tilde{w}(k)$ it is obvious that mostly oscillations with a 2 ML period are suppressed, i.e., oscillations close to the short period (2.5 ML). It should be noted that the relative suppression of amplitudes of oscillations is not sensitive to the thickness of magnetic slabs and that both analytical and numerical forms give nearly identical results.

(iii) The effect of interfacial interdiffusion on the coupling strength (Table I) is even more dramatic. An appreciable suppression of the amplitudes of the short period oscillations is visible already for 2% interdiffusion and amounts to an order of magnitude for 10% interdiffusion. Clearly, the strong disorder present in the Cu-Co alloy system, in particular between Cu and down-spin Co states, is a key factor for the coupling strength.

(iv) The fact that the strong suppression of the short period coupling strength does not depend on the Co slab thickness (compare the 5 ML Co slabs and the semi-infinite Co case) confirms the decisive role of interface region. We have addressed this problem by performing calculations for slabs of 5 ML Co with 5% interdiffusion where either only the inner or only the outer slab interfaces were disordered, while the other slab interfaces were kept ideal. The relative amplitudes for the short periods A(0.05)/A(0) are 0.99 (0.36) if randomness is limited to the outer (inner) interfaces. The decisive role of the inner interfaces seems to be obvious. For the long period oscillations (the slab thickness of 5 ML is comparable to their periods) the influence of disorder at inner and outer interfaces is comparable, but weaker than in the model in which both the inner and the outer interfaces are disordered (Table I).

(v) Figure 3 shows $L^2\bar{\mathscr{E}}_x$ as a function of the spacer thickness L for an ideal and a rough interface ($r=0.25$). Two

TABLE I. Relative amplitudes A(ν)/A(0) ($\nu=r,x$) for two Co(001) slabs in fcc-Cu each 5 monolayers (ML) thick and for two semi-infinite (inf) Co(001) subsystems sandwiching the Cu spacer. The values are given for short and long periods (5 ML case) and for short periods (semi-infinite case) as a function of the interface roughness r and the interface interdiffusion concentration x (assumed to be the same at inner and outer interfaces). The quantity $A(0)$ refers to the ideal interface ($r=0.0$ or $x=0.0$).

	5 ML slab		inf		5 ML slab		inf
r	short	long	short	x	short	long	short
0.0	1.0	1.0	1.0	0.0	1.0	1.0	1.0
0.05	0.67	0.87	0.66	0.02	0.70	0.85	0.69
0.1	0.41	0.76	0.40	0.05	0.38	0.62	0.37
0.25	0.02	0.46	0.01	0.1	0.12	0.36	0.12

periods, short and long, with different amplitudes can clearly be identified for the case of the ideal interface. One can see that as compared to an ideal interface for this particular model of a rough interface only the long periods (with strongly reduced amplitudes) survive (see also Table I). Our calculations confirm an approximate L^{-2}-like behavior of $\mathscr{E}_x(L)$ in the limit of large spacer thickness L as predicted by a simple RKKY theory.[1] Note, however, that this behavior is actually only valid for a spacer thickness larger than 15 layers and it is different in the preasymptotic region. The experimental values of coupling strengths are deduced from a few first peaks of the oscillating $\mathscr{E}_x(L)$ and corresponding to a finite temperature. On the contrary, the net effect of interfacial imperfections should be deduced from the asymptotic behavior for large spacer thicknesses and for $T=0$ K. It is obvious that a comparison of theoretical and experimental values of coupling strengths is not straightforward, in particular when the quality of the interface is not precisely controlled in the experiment. New experiments more tailored to this specific problem should be performed.

In summary, we have performed *ab initio* calculations for the interlayer exchange coupling in the presence of two general kinds of interfacial imperfections, namely an interfacial roughness model with randomly distributed terraces of monolayer heights leading to fluctuations of the spacer thickness, and an interfacial interdiffusion model, in which magnetic and nonmagnetic atoms are mixed randomly at the interfaces which were treated within a unified electronic structure model. In particular a dramatic effect of the interface imperfections on the coupling strength for the short period oscillations was found in the case of interdiffusion. The present study clearly demonstrates the high sensitivity of the coupling strength with respect to various kinds of interfacial imperfections and decisive role of interface electron scattering for the exchange oscillatory coupling, and in turn indirectly also for the giant magnetoresistance in metallic multilayers. The present study also shows that care is needed when comparing theoretical and experimental values of coupling strengths.

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