Ab Initio Calculations of the Exchange Coupling of Fe and Co Monolayers in Cu

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A Korringa-Kohn-Rostoker Green's function method for planar perturbations and a frozen potential method are used to evaluate the interaction energies of Fe and Co monolayers in Cu separated by up to 23 spacer layers. For the [100] orientation a phase shift of π is obtained between the Fe and Co oscillations and the three antiferromagnetic peaks observed experimentally are well described. For [110] an interesting beat effect is obtained leading to very different results for the Fe and Co layers.

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After the discovery of antiferromagnetic coupling of Fe layers in Fe/Cr/Fe sandwiches [1] there has been a steadily increasing interest in exchange coupling in layered magnetic systems. Typically an oscillatory dependence of the coupling strength with the thickness of the spacer layer has been observed in a large variety of systems [2]. From early on this physical phenomenon has been related to RKKY interactions. Theoretical efforts have relied mostly on model calculations based on the tight binding method, as, e.g., in [3], or on the jellium model, as, e.g., in [4]. Bruno and Chappert [5] have given a detailed analysis of the asymptotic behavior of the interactions for large distances, showing that the oscillations are directly related to the detailed geometry of the Fermi surface of the spacer material and are of multiperiodical nature. While this approach successfully explains variations of the oscillations by changing the thickness and orientation of the spacer layer, it also has its limitations. First it is only valid asymptotically for large interlayer distances and second all data concerning the scattering at the magnetic layer enter as unknown parameters, making quantitative predictions impossible.

For these reasons there is a strong need for reliable *ab initio* calculations. Such calculations are, however, very difficult. In supercell calculations as in [6] the numerical work increases with the third power of the number of layers. At the same time the number of **k** points, necessary in the calculation, increases since the information about the oscillations contracts for large distances to regions around critical points on the Fermi surface. Moreover there is the problem of subtracting huge total energy values to obtain the resulting small interaction energies. In two recent reviews [7,8] it was therefore concluded that progress in total energy calculations is not to be expected in the near future.

In this paper we will show that accurate and efficient *ab initio* calculations based on Green's function methods can be performed for large systems. We perform calculations for Fe and Co monolayers with orientations [100] and [110] sandwiched in fcc Cu and separated by up to 23 spacer layers. We get good agreement with the experimentally observed antiferromagnetic peaks for the Co/Cu and Fe/Cu systems. Moreover we can explain a phase shift of π between these oscillations and can make valuable predictions for additional peaks at shorter and larger distances.

Our calculations are based on density functional theory and a newly developed Korringa-Kohn-Rostoker (KKR) Green's function method for planar defects [9]. Here the magnetic layer and the neighboring host layers are considered perturbations in an otherwise unperturbed nonmagnetic host. Because of two-dimensional (2D) periodicity of the perturbation a 2D Fourier transform of the resulting multiple scattering equations is performed. For the structural Green's function $G^{ii'}_{LL'}(\mathbf{q},E)$ of the perturbed system, depending parametrically on the 2D wave vector $\mathbf{q}$ and the energy E, the following Dyson equation has to be solved:

$$G^{ii'}_{LL'}(\mathbf{q},E)=\dot{G}^{ii'}_{LL'}(\mathbf{q},E)+\sum_{i''L''}\dot{G}^{ii''}_{LL''}(\mathbf{q},E)\Delta t^{i''}_{L''}(E)G^{i''i'}_{L''L'}(\mathbf{q},E)\,. \tag{1}$$

Here i,i' are layer and L,L' angular momentum indices. The summation includes all layers i'' and angular momenta L'' which are assumed as perturbed. $\dot{G}^{ii''}_{LL''}(\mathbf{q},E)$ denotes the structural Green's function of the host, written in the appropriate 2D representation. It is calculated from the corresponding 3D Green's function $\dot{G}_{LL'}(q+\mathbf{k}_\perp,E)$ by an integration over the wave vector $\mathbf{k}_\perp$ normal to the 2D Brillouin zone (BZ):

$$\dot{G}^{ii'}_{LL'}(\mathbf{q},E)=\frac{d}{2\pi}\int_{-\pi/d}^{\pi/d}dk_\perp e^{-i(\mathbf{q}+\mathbf{k}_\perp)\cdot(\mathbf{R}_i-\mathbf{R}_{i'})}\dot{G}_{LL'}(\mathbf{q}+\mathbf{k}_\perp,E)\,. \tag{2}$$

With the above formalism, self-consistent LDA calculations are performed for single magnetic monolayers of Fe and Co sandwiched into bulk Cu on (100) and (110) planes. The potentials are assumed to be spherically symmetric in the atomic spheres (ASA).

The resulting t matrix $\Delta t_L(E)$ for the magnetic monolayer forms, together with the host Green's function of Eq. (2),

the input for the following calculation of the exchange coupling of two magnetic monolayers separated by n spacer layers. For this we employ the frozen potential approximation [10] (this method is also referred to as the "force theorem") and use the self-consistent potential of the single monolayer for the two interacting monolayers, with the proper choice of the spin up and spin down potentials for the ferro- and antiferromagnetic configurations. The induced spin splittings of the host potentials and the resulting Δt matrices of the Cu atoms are neglected, being justified in view of the small exchange enhancement of Cu. Because of the frozen potential approximation the total energy difference between the ferromagnetic (F) and antiferromagnetic (AF) configuration is in first order given by the difference of the single particle energies

$$E_F - E_{AF} = \int_{-\infty}^{+\infty} dE (E-\mu) f_T(E) [\Delta n_F(E) - \Delta n_{AF}(E)] \tag{3a}$$

$$= -\int_{-\infty}^{+\infty} dE \frac{d}{dE} [(E-\mu) f_T(E)] [\Delta N_F(E) - \Delta N_{AF}(E)] . \tag{3b}$$

Here Δn_F and Δn_{AF} are the changes of the density of states, μ the chemical potential, and $f_T(E)$ the Fermi-Dirac distribution. By partial integration the corresponding integrated densities of states $\Delta N_{F,AF}$ are introduced in Eq. (3b), since these can be directly evaluated using modifications of Lloyd's formula [11]. The contribution $\Delta N^{12}_{F,AF}(E)$ for the interaction of two monolayers 1 and 2 coupled ferro- or antiferromagnetically is given by

$$\Delta N^{12}_{F,AF} = -\frac{1}{\pi} \mathrm{Im} \sum_\sigma \frac{1}{A_{BZ}} \int d\mathbf{q} \ln \det(\underline{1} - \dot{\underline{G}}^{12} \Delta \underline{\tau}^2_\sigma \dot{\underline{G}}^{21} \Delta \underline{\tau}^1_\sigma) , \tag{4}$$

where the determinant and the matrices refer to angular momentum space and where the $\Delta \tau^i_\sigma$

$$\Delta \underline{\tau}^i_\sigma(\mathbf{q},E) = [\underline{1} - \Delta \underline{t}^i_\sigma(E) \dot{\underline{G}}^{ii}(\mathbf{q},E)]^{-1} \Delta \underline{t}^i_\sigma(E) \tag{5}$$

represent $\mathbf{q}$-dependent t matrices for the magnetic monolayer i and spin direction σ, which include all multiple scattering effects within the monolayer.

By inserting the t matrices of the single monolayers and the host Green's function $\dot{\underline{G}}^{12}$ [Eq. (2)] into Eq. (4), the interaction energy $E_F - E_{AF}$ of Eq. (3b) can be evaluated. Since the final formula is rather lengthy we give here only the leading term, obtained by a second order expansion in powers of the dimensionless quantity $\dot{G}^{12} \Delta\tau$. According to our results this term dominates the behavior.

$$\Delta N^{12}_F - \Delta N^{12}_{AF} = \mathrm{Im} \frac{1}{A_{BZ}} \int d\mathbf{q} \, \mathrm{Tr} [\dot{\underline{G}}^{12} (\Delta \underline{\tau}_+ - \Delta \underline{\tau}_-) \dot{\underline{G}}^{21} (\Delta \underline{\tau}_+ - \Delta \underline{\tau}_-)] , \tag{6}$$

here the trace Tr refers to summation over angular momenta. Reading (6) from right to left the electrons scatter first at the "exchange potentials" $(\Delta \underline{\tau}_+ - \Delta \underline{\tau}_-)$ of monolayer 1, then propagate through the space layers to monolayer 2, scatter at these exchange potentials, and propagate back to monolayer 1.

As can be seen from (6) only matrix operations in angular momentum space have to be evaluated so that the numerical effort is independent of the spacer thickness. Therefore, as a benefit of the frozen potential approximation, calculations can be performed up to large distances, which is not possible in supercell calculations. Moreover the energy calculation is rather transparent, since the problem of subtracting large total energy contributions does not occur.

In order to evaluate the exchange interaction $E_F - E_{AF}$, the energy integration over all occupied states in (3b) and the $\mathbf{q}$ integration over the 2D Brillouin zone in (4) must be performed very accurately. For the energy integration we exploit the analytical properties of the Green's function and the Fermi-Dirac distribution and evaluate the integral by contour integration in the complex energy plane [9]. We calculate the residua at five Matsubara frequencies $z = \mu + i(2n+1)\pi kT$ explicitly. The remaining integral for energies far away from the real axis can easily be evaluated by about 16 energy points.

The $\mathbf{q}$ integration over the irreducible part of the 2D Brillouin zone is evaluated by special point methods. Especially for large distances a very high number of $\mathbf{q}$ points [as well as $k_\perp$ points in Eq. (2)] is needed [4], since all the relevant information about the interaction contracts to regions close to critical points on the Fermi surface [5]. For a temperature $T = 450$ K, corresponding to a distance 0.13 eV of the first Matsubara pole away from the real axis, our results are fully converged up to 23 spacer distances, if we choose about 500 $\mathbf{q}$ points in the irreducible part of the 2D Brillouin zone. Since the Green's functions decrease exponentially with increasing distance from the real axis, this high number of $\mathbf{q}$ points is only necessary for the one or two Matsubara energies closest to the real axis. A reduction of the temperature, to, e.g., 237 K, practically does not change the results, provided a sufficiently higher number of $\mathbf{q}$ points, e.g., 1000, is used. Angular momenta up to $l = 3$ are included in the calculation.

Figure 1 shows our results for the interlayer coupling

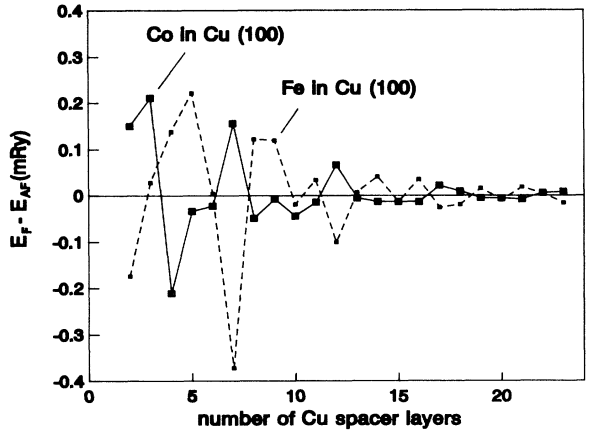

FIG. 1. Exchange coupling energies $E_F - E_{AF}$ of (100) Co and Fe monolayers sandwiched in fcc Cu versus the number of Cu spacer layers.

energies of two Co or Fe (100) monolayers sandwiched in fcc Cu and separated by a varying number of Cu layers. The existence of slowly decaying oscillations is evident from the diagram, in qualitative agreement with the RKKY picture. In the [100] direction the asymptotic expansion [5] predicts two oscillation periods arising from two different calipers of the "dogbone" structure of the Cu Fermi surface. We have therefore made a least square fit of such a RKKY expression for the oscillations to our *ab initio* data. The fit is of good quality and, in agreement with [5], yields two periods of about 2.6 and 5.9 monolayers. It also shows that for these systems the asymptotic expression is valid from about 8 spacer layers on.

By comparing the calculated values for Fe and Co in Fig. 1, we observe that the oscillations are quite similar and essentially shifted in phase by about π. For instance, at distances where the Fe layers couple ferromagnetically ($E_F - E_{AF} < 0$), the Co layers prefer antiferromagnetic coupling ($E_F - E_{AF} > 0$) and vice versa. A phase shift of π between Fe and Co has been observed in the work of Petroff *et al.* [12]. In contrast to this Johnson *et al.* [13] find a considerably smaller shift in contradiction to our results. At present the reason for the discrepancy is unclear.

If the experimentally observed peak positions for the antiferromagnetic coupling are compared with our calculated values, an overall good agreement is found. For Co layers in Cu peaks have been found for about 6, 11.5, and 17 monolayers [14], which can be identified with the three large antiferromagnetic couplings calculated for 7, 12, and 17 monolayers as shown in Fig. 1. In addition we calculate strong antiferromagnetic coupling for 2 and 3 monolayers, which experiments have problems resolving, presumably due to pinhole effects. Of course there are also additional peaks at larger distances.

For Co layers Johnson *et al.* [13] report an additional

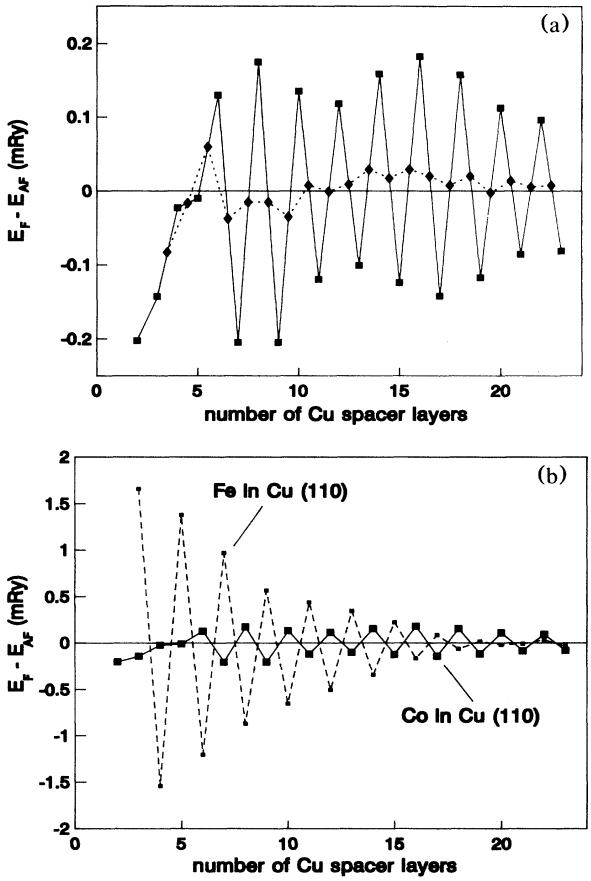

FIG. 2. (a) Exchange coupling energies for (110) Co monolayers sandwiched in fcc Cu (full line). In order to simulate roughness the dotted line gives the average $\Delta E(n+1/2)$ of neighboring values (see text). (b) Exchange coupling energies for (110) Fe monolayers (dashed) in comparison to (110) Co layers (full line).

antiferromagnetic peak at 15 monolayers, where we calculate a moderate ferromagnetic coupling. However, upon improvement of the sample quality this peak seems to decrease [15], in qualitative agreement with our results.

Not as many experiments have been reported for fcc Fe/Cu/Fe (100) layers. Peaks at around 8, 15, and 22 monolayers [16] have been found, being in qualitative agreement with our results. For instance, due to surface roughness the calculated twin peaks at 14 and 16 monolayers might merge into a single broad peak centered at 15 monolayers. Similar to the Co case, the first antiferromagnetic peak at 4–5 monolayers has apparently not been resolved in the experiment.

Compared to (100) layers, (110) layers of Fe and Co in Cu show a considerably more complicated interaction behavior. The results for (110) layers of Co in Cu are given by the full line in Fig. 2(a) and on a reduced scale in Fig. 2(b) together with the results for (110) Fe layers

(dashed line). The difference between the Co and Fe curve is striking. Whereas the interaction of the Fe layers shows in addition to the fast oscillation a slow decrease as expected from RKKY arguments, the amplitude of the Co layers does not increase at all. Nevertheless, this strange behavior can be understood from RKKY theory. For the (110) layers the asymptotic behavior is dominated by an oscillation with a period of 2.11 monolayer distances as determined by the length of the dogbone of the Cu Fermi surface [5]. The fact that this period is only slightly larger than 2 monolayer distances has two important consequences. First it explains directly the fast plus/minus changes between neighboring layers as observed for both Fe and Co. Second it leads to a long-range beating between the 2.11 and a 2 monolayer period of the ideal crystal resulting in a long-period envelope function, which enters in addition to the normal $1/R^2$ factor. The basic difference between the (110) Co and Fe layers is that at about 5 monolayers the long period beating envelope function has a node for Co and an extremum for Fe. As a result the Fe oscillations show a normal decrease whereas in the case of Co the increase of the envelope function roughly cancels the $1/R^2$ decrease leading to a more or less constant amplitude. In fact the situation is even more complicated since the Fermi surface is extremely flat at the ends of the dogbone leading to a nesting effect and a slower decay, like, e.g., $1/R^{3/2}$ or $1/R$. Moreover several additional oscillations with smaller amplitudes exist for this orientation [5].

The short oscillation obtained in our calculation has not been detected in experiment. This is plausible since the mostly unavoidable roughness of the interface acts as a low pass filter for the oscillations, as is known from the results of the Fe/Cr system. In order to model the low pass filter properties of the roughness we have plotted in Fig. 2(a) by the dotted line for the Co system the average of neighboring layers

$$\Delta E(n+\tfrac{1}{2}) := \tfrac{1}{2}[\Delta E(n)+\Delta E(n+1)] .$$

By this procedure the rapid oscillations are washed away and strong antiferromagnetic couplings are found at around 5–6 and 13–18 monolayers, which compares favorably with the experimentally observed peaks for Co/Cu at spacer thicknesses of about 7 and 16 monolayers [17].

In the present paper we have only considered the interaction of 2 monolayers. With slight modifications the above method can be extended to calculate the interaction of thin magnetic films containing more than 1 monolayer. Calculations for these systems are in progress. In line with a recent discussion by Bruno [18] and the good agreement with experiments obtained in this paper we do not expect drastic changes of the present results.

In summary, we have presented *ab initio* calculations for the magnetic exchange interactions of Fe and Co monolayers in fcc Cu. The calculations are based on a multiple scattering (KKR) Green's function method for planar perturbations. The interaction energies are calculated by the frozen potential method. The required computing time is essentially independent of the number of interlayers and calculations for large distances can be performed, thus bridging the gap between the asymptotic RKKY theory and supercell calculations possible for small distances. For the [100] orientation we obtain a phase shift of π between the Fe and Co oscillations which can explain all measured peaks and predict hitherto unresolved peaks at shorter distances. For the [110] orientation we obtain a very fast oscillation and an interesting beating effect leading to an extremely different behavior for the Fe and Co layers.

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