Exchange coupling between iron layers separated by bcc copper

A. T. Costa, Jr., J. d'Albuquerque e Castro, and R. B. Muniz
Instituto de Física, Universidade Federal Fluminense, Niterói, 24210-340, Brazil

M. S. Ferreira
Department of Mathematics, Imperial College, London, SW7 2BZ, United Kingdom

J. Mathon
Department of Mathematics, City University, London, EC1V 0HB, United Kingdom
(Received 11 September 1996)

The exchange coupling between Fe layers separated by bcc Cu is calculated for Fe/Cu/Fe (001) trilayers. It is shown that the coupling is basically regulated by three extrema of the bulk bcc Cu Fermi surface. The contributions from those extrema are all of the same order of magnitude, but that associated with the ''belly'' at the Γ point dominates. The calculated temperature dependence of the coupling varies considerably with spacer layer thickness. Individually, the amplitudes of these extrema contributions decrease with temperature, each according to a different rate. Such an effect may cause an actual increase of coupling with temperature for some Cu thicknesses. [S0163-1829(97)08505-6]

Although the common crystal phase of bulk Cu is fcc, it is possible to grow thin films of bcc Cu on Fe (001). The bcc stacking proceeds for up to 12 or 20 atomic planes approximately, but for larger thicknesses significant lattice modifications occur, leading to a structural transformation.[1,2]

The exchange coupling between Fe layers separated by bcc Cu has been measured by groups at Simon Fraser University (SFU) and Philips.[2,3] Both have found that the coupling oscillates with decreasing amplitude as a function of the Cu thickness, but their results disagree in several important aspects.[1] The Philips group data show well-defined short-period oscillations[2] whereas the SFU group originally observed a long-period oscillatory coupling.[3] Later, the SFU group found some indication of a short-wavelength oscillation in samples with smoother interfaces.[1] The exchange coupling in multilayers can be strongly affected by sample interface quality.[4] It is widely accepted that interface roughness tends to suppress short-wavelength oscillations and reduce the coupling amplitude. Therefore, as pointed out in Ref. 3, it is rather puzzling that the values obtained at Philips are substantially smaller than those of the SFU group.

Motivated by these apparently conflicting experimental results, we have undertaken a theoretical analysis of the exchange coupling between Fe layers across bcc Cu in Fe/Cu/Fe (001) trilayers. The coupling J is calculated for several temperatures T and spacer layer thicknesses N, using an extension of the formulation developed in Ref. 5. For sufficiently large N, we divide $J(N)$ into oscillatory components coming from extrema which are related to the spacer Fermi surface (FS). These oscillatory contributions to the coupling are calculated separately. Our results show that for perfectly smooth interfaces $J(N)$ is dominated by short-period oscillations. We find that the temperature dependence of the coupling changes significantly with spacer layer thickness. The amplitude of each oscillatory component decreases with temperature, but they do so at different rates. We show that this may cause a surprising effect which is the increase of the coupling with temperature for some Cu thicknesses.

The interlayer exchange coupling, defined as the total energy difference per surface atom between the antiferromagnetic and ferromagnetic configurations of the trilayer, is given by[6,7]

$$J=\sum_{\vec{k}_\parallel}\int d\omega f(\omega)F(\vec{k}_\parallel,\omega,N), \tag{1}$$

where

$$F=\frac{1}{\pi}\mathrm{Im}\,\mathrm{trln}[1+S^{\uparrow}(G^{c\uparrow}_{mm}-G^{c\downarrow}_{mm}) \times S^{\downarrow}(G^{c\uparrow}_{m+1,m+1}-G^{c\downarrow}_{m+1,m+1})], \tag{2}$$

$S^{\uparrow}=t^{\dagger}(1-G^{c\uparrow}_{mm}tG^{c\uparrow}_{m+1,m+1}t^{\dagger})^{-1}$ and $S^{\downarrow}=t(1-G^{c\downarrow}_{m+1,m+1}t^{\dagger}G^{c\downarrow}_{mm}t)^{-1}$. In the equations above $\vec{k}_\parallel$ are the wave vectors parallel to the layers, $f(\omega)$ is the usual Fermi-Dirac distribution function, and m is a plane index. As in Ref. 7 we consider an imaginary cleavage plane across the spacer between planes m and $m+1$, separating the trilayer into two semiinfinite systems. $G^{c\sigma}_{mm}$ and $G^{c\sigma}_{m+1,m+1}$ are matrices in orbital indices representing the surface one-electron Green's functions of the left and right cleaved systems, respectively. The trace is taken over orbital indices and t denotes the spacer hopping matrix.

This formula for the coupling is an extension of the result previously obtained in Ref. 5 and, for the one band model, reduces to the torque formula of Edwards *et al.*[8] In deriving it we have assumed that the electrons are noninteracting in the spacer and experience exchange-split one-electron potentials in the ferromagnetic layers. Most of the experimental results are for the bilinear exchange coupling term J_1 which, for perfectly smooth Fe/Cu (001) interfaces, is virtually equal to $J/2$.[5]

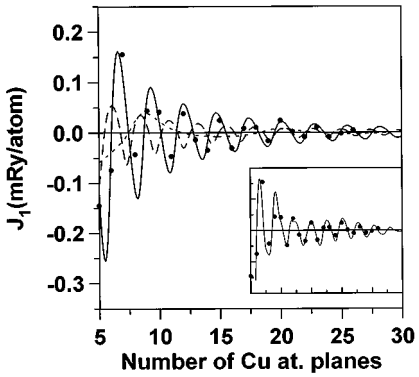

FIG. 1. Calculated bilinear exchange coupling at $T=300$ K for bcc Fe/Cu/Fe (001) trilayer vs Cu thickness (full circles). The lines are contributions from the extremal points (see text) corresponding to the Cu FS belly (full line), the neck wave vectors of set 2 (dashed line), and the neck M points (dotted line). The inset shows the total contribution from the three sets of extrema; the ticks are the same as those of the main figure.

We have calculated the required Green's functions within the tight-binding model with s,p,d orbitals and hopping to second nearest neighbors. The tight-binding parameters for all bcc Cu planes were determined from a first-principles LMTO-tight-binding electronic structure calculation of paramagnetic bulk bcc Cu. The parameters for ferromagnetic Fe were obtained from paramagnetic bulk Fe,[9] by self-consistently adjusting the on-site energies, assuming charge neutrality. The effective intraatomic electron-electron interactions were taken to be $U_{\mathrm{sp}}^{\mathrm{Fe}}=0$ and $U_{d-d}^{\mathrm{Fe}}=1$ eV.[10,11] We neglect atomic potential differences due to the magnetic configuration change, thus making the approximation known as the ''force theorem.'' The $\vec{k}_{\parallel}$ sum in Eq. (1) is performed numerically and the energy integral is evaluated in the complex plane by summing over Matsubara frequencies at finite T.

The calculated results at $T=300$ K for the bilinear exchange coupling J_1 as a function of Cu thickness are presented in Fig. 1 (full circles). Our results clearly show a short-period oscillatory exchange coupling, in excellent agreement with the Philips group data as far as the period of oscillation is concerned.

For sufficiently large spacer thickness it is possible to express the coupling as a sum of oscillatory components whose periods are determined by extrema that are related to the spacer FS.[12–16] It is essential to use a non-perturbative treatment, as in the quantum well approach, to analyze the relative importance of these contributions.[15,17] They depend upon the degree of confinement experienced by the carriers of both spin orientations in the corresponding extremum states, in the ferromagnetic and anti-ferromagnetic configurations of the magnetic layers.[7,15,17,18] The widespread practice of considering only the periods predicted by RKKY theory, and treating the amplitudes and phases of these contributions as adjustable parameters may be inadequate and rather misleading. The fitting usually involves several parameters and, in some cases, is not unique. Besides, when the spacer FS has to be regarded as consisting of more than one sheet, periods not predicted by RKKY theory may exist.[15] Moreover, at finite temperatures, the decrease of the oscillation amplitudes as N increases is different for each extremum and may deviate strongly from the $(1/N^2)$ asymptotic regime[7,17,19] usually assumed in that sort of fitting.

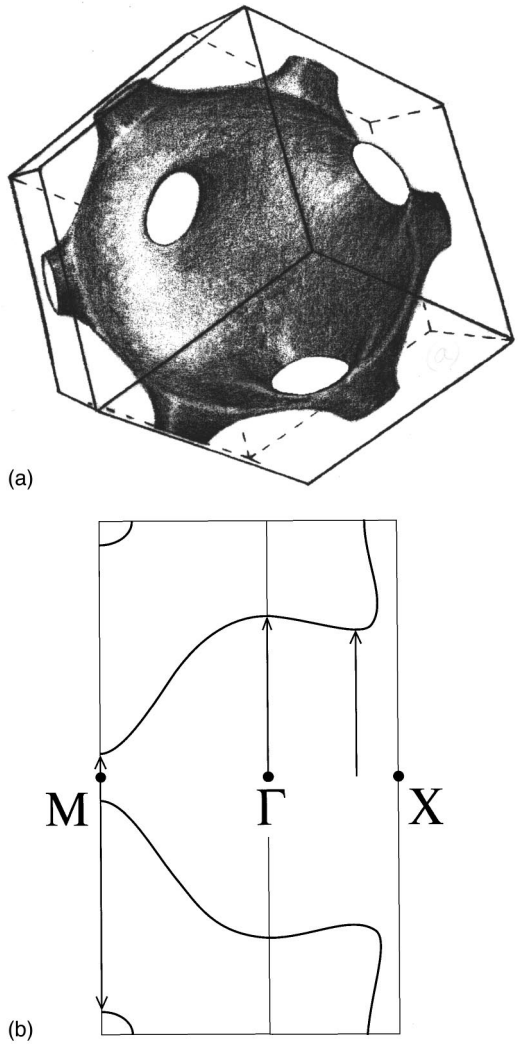

FIG. 2. Calculated bcc Cu FS (a), and its relevant cross sections for (001) (b). The arrows are the critical vectors $k^{\perp}(\vec{k}_{\parallel}^{0})$.

To identify the periods of oscillations of $J(N)$ it is useful to look at the spacer FS. In bcc Cu only one energy band $E(\vec{k}_{\parallel},k^{\perp})$ crosses the Fermi energy E_F. Its calculated FS, shown in Fig. 2(a), is basically a sphere with twelve ''necks'' developing at each face center of the bulk bcc first Brillouin zone. In the (001) direction of growth, three sets of $\vec{k}_{\parallel}^{0}$ associated with the FS extrema contribute to the coupling. The first set consists of a single wave vector $\vec{k}_{\parallel}^{0}=(0,0)$ (Γ point) related to the FS ''belly.'' The other two are associated with the ''necks.'' Set 2 consists of four vectors $\vec{k}_{\parallel}^{0}$: $(0,\pm 0.327)$ and $(\pm 0.327,0)$, and set 3 of the M points located at $(\pm 0.5,\pm 0.5)$. Here all wave vectors are given in units of $2\pi/a$ where a is the lattice constant. Due to the layered stucture of the system, it is useful to work with the layer adapted bulk Brillouin zone instead of the usual BZ. The former is defined as a prism whose base is the two-dimensional first BZ and whose height is $2\pi/d$, where d is the interplane distance perpendicular to the layers. The relevant cross sections of the spacer FS, together with the cor-

responding extremal wave vectors $\vec{k}^{\perp}(\vec{k}_{||}^{0},E_F)$, are shown in Fig. 2(b). We must distinguish sets 1 and 2 from set 3 because, for the latter, the FS can be regarded as consisting of more than one sheet. This is because more than one extrema occurs in the first prismatic bulk BZ for each wave vector $\vec{k}_{||}^{0}$ of set 3.

Considering that the integrand F in Eq. (1) is an oscillatory function of N we can expand it in a Fourier series. However, it is necessary to generalize the expansion to a multiple Fourier series,[6,15] when the equation $E(\vec{k}_{||},\vec{k}^{\perp})=\omega$ has more than one pair of solutions $\pm k_{\xi}^{\perp}(\vec{k}_{||},\omega)$. In this case, the general expansion of F is

$$F(\vec{k}_{\parallel},\omega,N)=\sum_{n_1,\ldots,n_{\xi}} c_{n_1,\ldots,n_{\xi}}(\vec{k}_{\parallel},\omega) \times\exp\left(i\sum_{\xi} n_{\xi}k_{\xi}^{\perp}(\vec{k}_{\parallel},\omega)Nd\right). \quad (3)$$

For $N\gg 1$ the exponential in Eq. (3) oscillates rapidly as a function of $\vec{k}_{||}$ and ω. Thus, the stationary phase method can be applied, and the dominant contribution to the coupling comes from $\omega=E_F$ and $\vec{k}_{||}$ in the neighborhood of points at which the argument of the exponential is stationary. In this limit both the sum in $\vec{k}_{||}$ and the energy integral in Eq. (1) can be evaluated analytically. The stationary points $\vec{k}_{||}^{0}$ are the solutions of

$$\sum_{\xi} n_{\xi}\nabla k_{\xi}^{\perp}(\vec{k}_{||},E_F)=0, \quad (4)$$

where ∇ is the two-dimensional gradient in $k_{||}$ space.

For $\vec{k}_{||}^{0}$ belonging to sets 1 and 2 only one FS sheet occurs in the first prismatic bulk BZ; the analysis then proceeds exactly as in Ref. 7. The corresponding periods are $p^b=2.69$ atomic planes and $p^n=2.36$ atomic planes, respectively. However, for the M points the FS can be regarded as consisting of two sheets. The two values of $k^{\perp}$ ($k_1^{\perp}$ and $k_2^{\perp}$) associated with $\vec{k}_{||}^{0M}$, shown by arrows in Fig. 2(b), correspond to equivalent periods $p_1^n=2\pi/2k_1^{\perp}=10.97$ atomic planes and $p_2^n=2\pi/2k_2^{\perp}=1.1$ atomic planes which cannot be distinguished just by looking at discrete values of N. However, $\nabla k_1^{\perp}$ and $\nabla k_2^{\perp}$ vanish simultaneously when calculated at $(\vec{k}_{||}^{0M},E_F)$. Thus, Eq. (4) is satisfied for any values of n_1 and n_2, yielding other periods besides p_1^n and p_2^n. The relative contributions of these extrema depend on the comparative values of the corresponding coefficients c_{n_1,n_2}. The situation is very similar to that discussed in Ref. 15. Nevertheless, our calculations have shown that the fundamental period p_1^n (which is equivalent to p_2^n) and its harmonics dominate. This is because the coefficients associated with them are far larger than those corresponding to alternative combinations of n_1 and n_2.

The calculated contributions to the coupling at $T=300$ K coming separately from each set of $\vec{k}_{||}^{0}$ are shown in Fig. 1. We note that all three contributions are comparable, but the belly (full line) clearly dominates. This contrasts with fcc Co/Cu (001) trilayers where the belly contribution is negligible in comparison with those given by the necks.[17] The main reason for such difference is that minority carriers from the vicinity of the FS belly are fully confined in bcc Cu by the Fe layers in the ferromagnetic configuration. The physical origin of such confinement is that the sp-like bcc Cu band which intersects the FS has no counterpart at the minority-spin Fe FS. A similar situation happens for the Cu FS states in the vicinity of $\vec{k}_{||}^{0}$ belonging to set 2, as shown in Fig. 3. On the other hand, the Cu FS M states of either spin can evolve into the corresponding Fe FS states because they have sp character, due to the existence of a small but finite sp-d hybridization in this k-space region. The degree of confinement experienced by the carriers in this case depends on the relative sp-d hybridization strengths. The agreement between the stationary phase approximation and numerical results verifies that the exchange coupling at room temperature in Fe/Cu (001) trilayers with perfect interfaces oscillates with a short period strongly influenced by the belly contribution. This is in accordance with the Philips group observations, as far as the period is concerned. However, the calculated strength is an order of magnitude larger than what they have observed. On the other hand, the amplitude of our long period component is about three times smaller than the short period contribution. We believe that the discrepancy between experimental and theoretical results is due to interface rough-

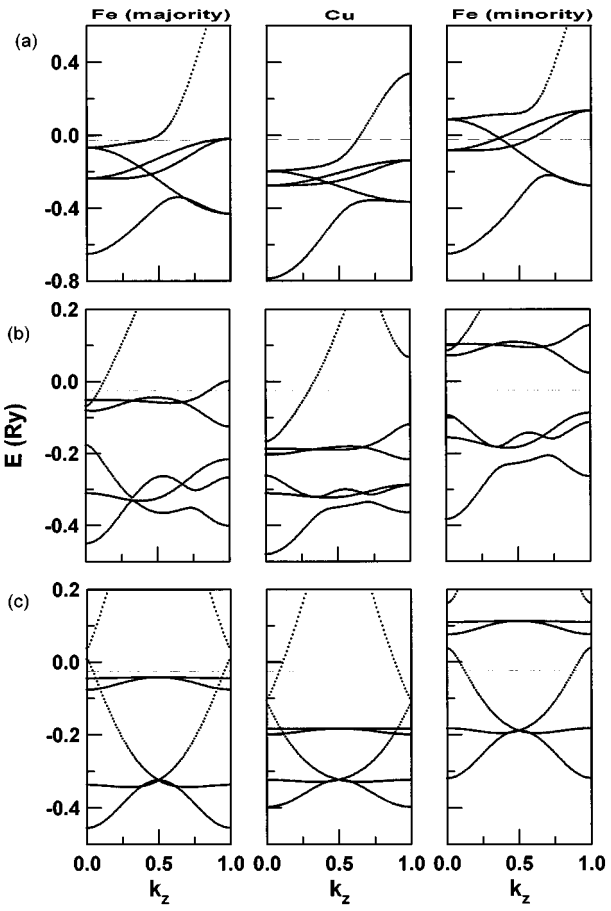

FIG. 3. Band structures of bulk bcc Cu and Fe in the relevant [001] direction for the wave vectors $\vec{k}_{||}^{0}=(0,0)$ (a), $\vec{k}_{||}^{0}=(0,0.324)$ (b) and $\vec{k}_{||}^{0}=(0.5,0.5)$ (c).

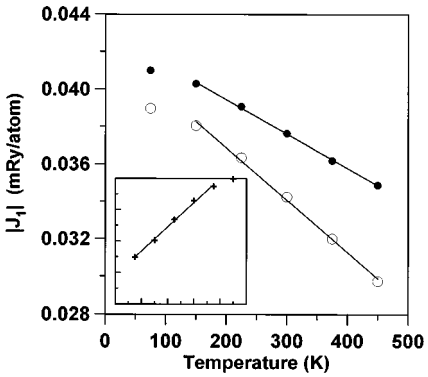

FIG. 4. Calculated temperature dependence of the bilinear exchange coupling for Fe/12Cu/Fe (full circles) and Fe/14Cu/Fe (open circles). The inset is for Fe/13Cu/Fe; the tick labels are the same as those of the main figure. The lines are simply linear fits.

ness which affects the amplitude and overall phase of the coupling.[20] The reason why the Philips results are much smaller than those of the SFU group remains unexplained.

Motivated by recent measurements of the SFU group,[21] we have calculated the temperature variation of the coupling for different Cu thicknesses. Our results, shown in Fig. 4, are for perfect interfaces where the bilinear exchange coupling J_1 is much larger than the intrinsic biquadratic term. The rate of variation of $|J_1(T)|$ with temperature clearly changes with spacer thickness. The calculated slope for Fe/12Cu/Fe agrees very well with the measured value for Fe/10Cu/Fe. The most striking result however, reproduced in the inset of Fig. 4, is the increase of the coupling with increasing temperature for some Cu thicknesses. The temperature dependence of J_1 is governed not only by the spacer FS but also by the confining strength of the ferromagnetic layers,[19] which differs for the three sets of extrema. As pointed out in Ref. 19, the energy dependence of the phases ψ of the ''Fourier'' coefficients in Eq. (3) varies according to the confinement strength and is very important in determining the temperature dependence of the coupling. It turns out that the values of $\partial\psi/\partial\omega$ calculated at the second set of extrema are about four and a half times larger than at the belly and the M points. The temperature dependence of the former contribution is then stronger than the others. Hence, the coupling is approximately given by the sum of three oscillatory functions of N with comparable amplitudes which decay differently with temperature. Therefore, at some values of N, as in Fe/13Cu/Fe, the balance is such that an overall increase in the coupling is obtained even though the amplitude of each contribution separately decreases with temperature. Such increase was not detected by the SFU group. One possible explanation is that they have observed basically just a long period component. Another reason could be the influence of spin fluctuations in the ferromagnetic layers, which is neglected in our calculations. Nevertheless, this is an interesting temperature effect which may be observed under suitable conditions.

ACKNOWLEDGMENTS

We thank S. F. Pessoa for kindly providing us with the tight-binding parameters of bcc Cu, D. M. Edwards for helpful discussions, and T. J. P. Penna for helping with the figures. This work has been financially supported by CNPq and FINEP of Brazil, and SERC of U.K.

[1] B. Heinrich and J.F. Cochram, Adv. Phys. **42**, 523 (1993).
[2] M.T. Johnson, R. Coehoorn, J.J. de Vries, N.W.E. McGee, J. aan de Stegge, and W. Hoving, Phys. Rev. Lett. **68**, 2688 (1992).
[3] Z. Celinski and B. Heinrich, J. Magn. Magn. Mater. **99**, L25 (1991).
[4] J. Unguris, R.J. Celotta, and D.T. Pierce, Phys. Rev. Lett. **67**, 140 (1991).
[5] J. d'Albuquerque e Castro, M.S. Ferreira, and R.B. Muniz, Phys. Rev. B **49**, 16 062 (1994).
[6] J. d'Albuquerque e Castro, J. Mathon, M. Villeret, and D.M. Edwards, Phys. Rev. B **51**, 12 876 (1995).
[7] J. Mathon, M. Villeret, A. Umerski, R.B. Muniz, J. d'Albuquerque e Castro, and D.M. Edwards (unpublished).
[8] D.M. Edwards, A.M. Robinson, and J. Mathon, J. Magn. Magn. Mater. **140-144**, 517 (1995).
[9] R.H. Victora, in *Magnetic and Electronic Properties of Low-Dimensional Systems*, edited by L.M. Falicov and J.L. Moran-Lopez (Springer, Berlin, 1986), p. 25.
[10] J.F. Cooke, J.W. Lynn, and H.L. Davis, Phys. Rev. B **21**, 4118 (1980).
[11] F.J. Himpsel, Phys. Rev. Lett. **67**, 2363 (1991).
[12] P. Bruno and C. Chapert, Phys. Rev. Lett. **67**, 1602 (1991); **67**, 2592(E) (1991).
[13] D.M. Edwards, J. Mathon, R.B. Muniz, and M.S. Phan, Phys. Rev. Lett. **67**, 493 (1991); **67**, 1476(E) (1991); J. Phys. Condens. Matter **3**, 3941 (1991).
[14] D.M. Edwards, J. Mathon, and R.B. Muniz, Phys. Rev. B **50**, 16 066 (1994).
[15] M.S. Ferreira, J. d'Albuquerque e Castro, D.M. Edwards, and J. Mathon, J. Magn. Magn. Mater. **154**, L1 (1996).
[16] M.D. Stiles, Phys. Rev. B **48**, 7238 (1993).
[17] J. Mathon, M. Villeret, R.B. Muniz, J. d'Albuquerque e Castro, and D.M. Edwards, Phys. Rev. Lett. **74**, 3696 (1995).
[18] J. Mathon, M. Villeret, and D.M. Edwards, J. Phys.: Condens. Matter **4**, 9873 (1992); J. Magn. Magn. Mater. **127**, L261.
[19] J. d'Albuquerque e Castro, J. Mathon, M. Villeret, and A. Umerski, Phys. Rev. B **53**, 13 306R (1996).
[20] J. Inoue, Phys. Rev. B **50**, 13 541 (1994).
[21] Z. Celinski and B. Heinrich, and J.F. Cochram, J. Magn. Magn. Mater. **145**, L1 (1995).