Quantum Well Theory of the Exchange Coupling in Co/Cu/Co(001)

J. Mathon and Murielle Villeret
Department of Mathematics, City University, London EC1V 0HB, United Kingdom

R. B. Muniz and J. d'Albuquerque e Castro*
Departamento de Fisica, Universidade Federal Fluminense, Niteroi, Brazil

D. M. Edwards
Department of Mathematics, Imperical College of Science, Technology and Medicine, London SW7 2BZ, United Kingdom
(Received 13 June 1994; revised manuscript received 14 November 1994)

The exchange coupling between two semi-infinite Co layers separated by a Cu(001) spacer is calculated by two different methods using s, p, and d tight-binding bands fitted to *ab initio* band structures of Cu and ferromagnetic Co. The contributions to the coupling from the belly (long period) and necks (short period) of the Cu Fermi surface (FS) are determined separately. It is shown that the short-period oscillation due to the minority electrons from the vicinity of the Cu FS necks is dominant, the contribution of the long-period oscillations being negligible. This is due to the existence of quantum well bound states for the minority electrons at the Cu FS necks in the ferromagnetic configuration.

PACS numbers: 75.70.Fr, 75.30.Et, 75.50.Rr

Oscillatory exchange coupling between magnetic layers separated by a nonmagnetic spacer has been observed in a large number of metallic multilayers [1]. Ortega and Himpsel [2] showed recently that the photoemission intensity from overlayers of noble metals (Cu, Ag) on magnetic substrates (Co, Fe) also oscillates as a function of the overlayer thickness with the same period as the exchange coupling. A consistent theory of the oscillatory exchange must, therefore, also account for the oscillations in the photoemission intensity. Since detailed photoemission [2] and magnetic measurements [3] are available for Co/Cu(001), this particular system is a good testing ground for various theories of the exchange coupling.

We proposed a quantum well theory of the coupling [4,5] which assumes that the exchange potentials of the magnetic layers create quantum wells in the spacer layer. As the thickness of the spacer is varied, quantum well states crossing the Fermi surface (FS) cause periodic oscillations of the spectral density [6] and these in turn lead to oscillations of the coupling. The periodic behavior of the spectral density is exploited in the quantum well theory [5,6] to perform analytically the difficult energy and k space sums required in the evaluation of the coupling.

However, until now the quantum well theory could be applied quantitatively only to simplified models which yield coupling strengths comparable with [4,5] or even smaller [7] than the experimental values. On the other hand, *ab initio* numerical calculations [8] give coupling strengths much larger than observed. Such calculations are very difficult because of the large number of k space points required to achieve convergence [5]. Recently, Lang *et al.* [9] used a KKR Green's function formalism to calculate the coupling between two monolayers of Co in Cu at finite temperature. They argued that convergence in k space, which is not feasible in their method at $T = 0$, can be achieved at room temperature. However, the coupling strength at the first antiferromagnetic peak they obtain for Co/Cu(001) is still an order of magnitude larger than observed. The theoretical situation for Co/Cu thus remains unsatisfactory.

To resolve this problem, we report here the results of two parallel calculations of the exchange coupling in a Co/Cu(001) trilayer with thick (semi-infinite) Co layers. In the first calculation, we used the quantum well theory with fully realistic s, p, d tight-binding bands with hopping to first and second nearest neighbors fitted to the *ab initio* band structures of Cu [10] and ferromagnetic Co [11]. To check the accuracy of this approach, we have generalized our new torque formula for the coupling [12,13] to the same multiorbital band structure of Co/Cu(001) and evaluated the coupling numerically at finite temperature (details will be published elsewhere). The torque method has an exact correspondence with the method of Lang *et al.* [9] and it converges at room temperature for the same number of k space points. However, the torque formula [12] has the advantage that the coupling can be evaluated for an arbitrary thickness of Co with no extra computational effort. This is crucial since the measurements of the coupling [3] were made not for monolayers but for thick layers of Co.

Applying the quantum well theory to Co/Cu(001), we determine separately the long- (belly) and short-period (neck) contributions to the coupling. Contrary to popular belief, we find that the short-period oscillation dominates, the contribution of the long-period oscillation being negligible. This result is confirmed by the full numerical calculation and the quantum well theory provides a simple interpretation in terms of the band structure in the direction perpendicular to the layers. As expected, for thick

 0031-9007/95/74(18)/3696(4)$06.00

Cu layers the quantum well method gives the same results as the numerical calculation. Finally, we show explicitly that the spectral density in Co/Cu(001) oscillates periodically with Cu thickness. Since the spectral density controls the photoemission intensity, its oscillations explain the oscillatory behavior of both the coupling and the photoemission intensity.

We Consider N (001) planes of Cu with the bulk lattice constant sandwiched between two semi-infinite slabs of ferromagnetic fcc Co. A small lattice mismatch between Co and Cu is neglected. Following our original approach [4–6], also adopted by Lang *et al.* [9], we assume that the local potentials in the Cu and Co layers are frozen, i.e., they do not change in going from the ferromagnetic (FM) to the antiferromagnetic (AF) configuration of the trilayer. We also assume an abrupt interface between Co and Cu.

The exchange coupling, defined as the total energy difference per unit area between the ferromagnetic (FM) and antiferromagnetic (AF) configurations of the trilayer, is expressed in terms of the thermodynamic potentials Ω^s for electrons of spin s,

$$J(N) = \{[\Omega^{\uparrow}(N) + \Omega^{\downarrow}(N)]_{\mathrm{FM}} - [\Omega^{\uparrow}(N) + \Omega^{\downarrow}(N)]_{\mathrm{AF}}\}/A\,, \tag{1}$$

where A is the cross-sectional area. The thermodynamic potential for a given magnetic configuration at temperature T is given by

$$\Omega^s = -k_B T \sum_{k_\parallel} \int_{-\infty}^{+\infty} \ln\{1 + \exp[(\mu - E)/k_B T]\} \times \mathcal{D}^s(E, \mathbf{k}_\parallel, N)\, dE\,, \tag{2}$$

where μ is the chemical potential and $\mathcal{D}^s$ is the spectral density for particles of spin s in the trilayer having that configuration. Because of the in-plane translational invariance, we label all the trilayer states by the plane index i and by the wave vector $\mathbf{k}_\parallel$ parallel to the layers. The spectral density $\mathcal{D}^s$ is given by

$$\mathcal{D}^s(E, \mathbf{k}_\parallel, N) = -\frac{1}{\pi} \mathrm{Im\ Tr} \sum_i G^s_{ii}(E, \mathbf{k}_\parallel, N)\,, \tag{3}$$

where G^s_{ii} is the diagonal matrix element of the one-electron Green's function, the trace is over all atomic orbitals, and the sum over i is over all atomic planes.

The problem now reduces to the calculation of the spectral density and evaluation of the difficult k space and energy sums. Assuming that the normalized spectral density $(1/N)\Delta\mathcal{D}(\mu, \mathbf{k}_\parallel, N) = (1/N)\{[\mathcal{D}^{\uparrow} + \mathcal{D}^{\downarrow}]_{\mathrm{FM}} - [\mathcal{D}^{\uparrow} + \mathcal{D}^{\downarrow}]_{\mathrm{AF}}\}$ is a periodic function of N, we have shown elsewhere [6,14] that, for a thick spacer layer ($N \gtrsim 5$–6), both the summation over $\mathbf{k}_\parallel$ and the energy integral in Eq. (2) can be carried out analytically using the stationary phase method. This leads to the following formula for the exchange coupling $J(N)$ which is asymptotically exact for large N:

$$J = \mathrm{Re} \sum_{s=1}^{\infty} \frac{\sigma}{2s} \frac{\Delta c_s e^{2inNdk_\perp}}{2sNd\, \partial k_\perp/\partial E + \partial\psi_s/\partial E} \times \frac{k_B T d |(\partial^2 k_\perp/\partial k_x^2)\partial^2 k_\perp/\partial k_y^2|^{-1/2}}{\sinh[\pi k_B T(2sNd\, \partial k_\perp/\partial E + \partial\psi_s/\partial E)]}\,. \tag{4}$$

Here, d is the interplanar distance, $\sigma = i$ when both second derivatives in Eq. (4) are positive, $\sigma = -i$ when they are negative, and $\sigma = 1$ when the derivatives have opposite signs. The oscillation periods $\pi/k_\perp$ are determined by the extremal radii $k_\perp$ of the bulk spacer FS in the direction perpendicular to the layers. Finally, $\Delta c_s(\mu, \mathbf{k}_\parallel)$ are the Fourier components of the difference between the normalized spectral densities in the FM and AF configurations. We have included in Eq. (4) the energy dependence of the phase ψ_s of the complex Fourier coefficient Δc_s. In agreement with the torque calculation it leads to an initial decay of the coupling with Cu thickness N slower than the usual $1/N^2$ dependence. The Fourier coefficients and all the derivatives in Eq. (4) are evaluated at $E = \mu$ and at the points $\mathbf{k}_\parallel = \mathbf{k}^0_\parallel$ where $k_\perp(\mu, \mathbf{k}^0_\parallel)$ is stationary and the contributions of all the stationary points must be included.

We begin the evaluation of Eq. (4) for Co/Cu(001) with the factors that depend only on the bulk Cu FS. The oscillation periods $\pi/k^0_\perp$ were obtained from the Cu FS extremal radii in the [001] direction. There are two extremal radii $k^0_\perp$ and they occur for $\mathbf{k}^b_\parallel = (0,0)$ (belly) and $\mathbf{k}^n_\parallel a = (\pm 2.53, \pm 2.53)$ (necks) where a is the lattice constant of Cu. The corresponding periods are $p^b = 5.7$ atomic planes (~10.3 Å) and $p^n = 2.6$ atomic planes (~4.7 Å), respectively. The factor $\sigma = i$ for the belly and $\sigma = 1$ for the necks. The Cu FS curvature and the FS velocity $\partial E/\partial k_\perp$ were determined from the Cu band structure.

The last ingredient in Eq. (4) is the Fourier analysis of the spectral density. We first computed the spectral density for discrete (physical) values of the Cu thickness $L = Nd$ by the method of adlayers [15] and then continued it analytically to all real L. The calculation for discrete $L = Nd$ is based on the surface Green's function for a semi-infinite slab of ferromagnetic Co, which was determined by the decimation method [16].

The normalized spectral density $\Delta\mathcal{D}/N$ can be Fourier analyzed only if it is known for all values of the "continuous Cu thickness" L in the interval $(-\pi/2k_\perp, \pi/2k_\perp)$, where $k_\perp$ is either the belly or neck FS radius. We first generated the spectral densities for trilayers with the number of Cu atomic planes N ranging from 400 to 600. The spectral densities normalized to N were then shifted to the first period $(-\pi/2k_\perp, \pi/2k_\perp)$ by subtracting from N the appropriate integral number of periods p. As anticipated, all the shifted points condense on a continuous curve, which demonstrates explicitly that $(1/L)\Delta\mathcal{D}$ is a periodic function of the continuous Cu thickness L.

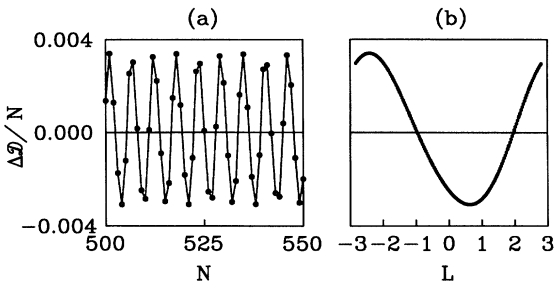

FIG. 1. Normalized spectral density $\Delta\mathcal{D}/N$ (in Ry^{-1} per atomic plane) at E_F and the extremal point $\mathbf{k}_\parallel^0 = 0$ corresponding to the belly of the Cu FS. (a) Raw computed data as a function of discrete Cu thickness N in atomic planes. (b) Data shifted to the first period of oscillations $(-\pi/2k_\perp, \pi/2k_\perp)$ as a function of the "continuous" Cu thickness L.

The contributions to the coupling from the belly and neck extrema evaluated from Eq. (4) at room temperature will now be discussed separately. We start with the long-period component which originates from the belly extremum. The raw computed spectral density $(1/N)\Delta\mathcal{D}(E_F, \mathbf{k}_\parallel = 0, N)$ is shown in Fig. 1(a) together with the data shifted to the first period [Fig. 1(b)]. The corresponding coupling $J^{\mathrm{belly}}(N)$ is shown in the inset in Fig. 2. It is far too weak to account for the total observed [3] coupling strength. This is not surprising since it is apparent from Fig. 1(b) that the peaks in the spectral density, as they move through the Fermi energy with varying spacer thickness, are broad resonances and the confinement in both spin channels is, therefore, only partial and weak.

We now turn to the short-period neck contribution which is much more interesting. Examination of all three components of the spectral density reveals that carriers

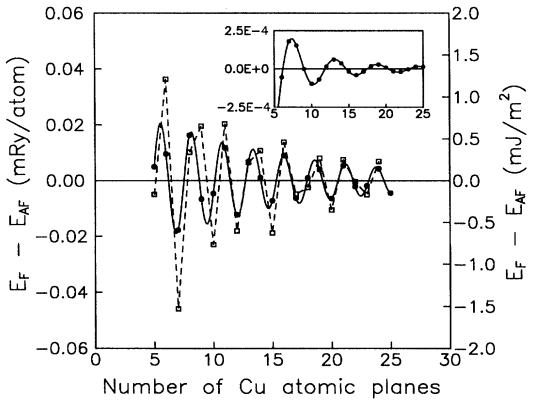

FIG. 2. Comparison between the exchange coupling versus Cu thickness for a Co/Cu/Co(001) trilayer calculated from Eq. (4) (solid circles) and from the torque formula (squares). The line passing through the solid circles is obtained from Eq. (4) for continuous Cu thickness. The inset shows the long-period component of the coupling originating from the belly of the Cu FS in mRy per atom.

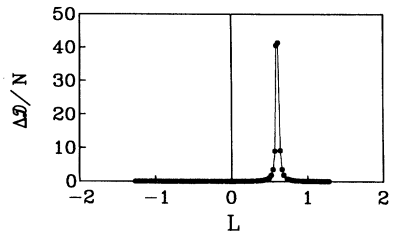

FIG. 3. Same as in Fig. 1(b) but for the extremal points $\mathbf{k}_\parallel^0$ corresponding to the necks of the Cu FS. The normalized spectral density is a set of delta function peaks corresponding to bound states localized in the Cu spacer.

of both spin orientations in the AF configuration, and also the majority spin carriers in the FM configuration, are only weakly confined (broad resonances). However, the minority spin carriers become completely confined in a quantum well in the FM configuration. Their spectral density is a set of delta functions which is shown in Fig. 3 and it dominates the total normalized spectral density.

To explain the physical origin of the complete confinement we reproduce in Fig. 4 the band structures of bulk Cu and Co in the relevant [001] direction for one of the neck wave vectors, $\mathbf{k}_\parallel^n a = (2.53, 2.53)$. The sp-like Cu band which intersects the FS, and hence determines the coupling, has no counterpart at the minority spin Co FS. The minority spin carriers must, therefore, be fully confined in Cu in the FM configuration. On the other hand, there is an sp-like band intersecting the majority Co FS into which the corresponding Cu band can evolve. The confinement of the majority-spin carriers is, therefore, only partial. This argument shows that full confinement of minority spin carriers, which dominate the coupling, takes place regardless of the details of the interfacial potential and this justifies our assumption of abrupt interfaces.

The total coupling at $T = 316$ K calculated from Eq. (4) is compared in Fig. 2 with the numerical result obtained from the torque formula. The left-hand scale in Fig. 2 gives the coupling in mRy per atom in the (100) surface. The right-hand scale gives the conversion to the

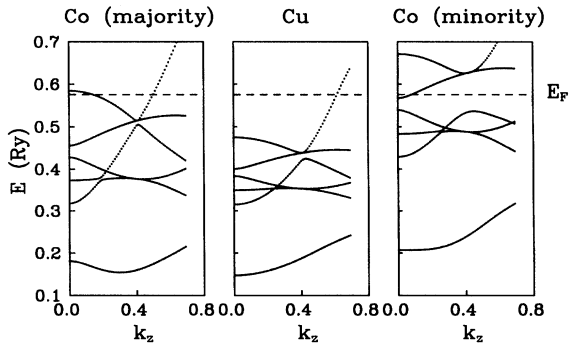

FIG. 4. Band structures of bulk Cu and ferromagnetic fcc Co in the relevant [001] direction for one of the neck wave vectors $\mathbf{k}_\parallel^0 a = (2.53, 2.53)$. k_z is given in units of $2\pi/a$.

units (mJ/m^2) commonly used by experimentalists. The observed coupling strength [3] at the first antiferromagnetic peak [about 6 monolayers (ML) of Cu] is 0.4 mJ/m^2 which is to be compared with our calculated value of 1.2 mJ/m^2 obtained from the torque formula. On the other hand, the coupling strength at the first antiferromagnetic peak obtained by Lang *et al.* [9] is about 5 mJ/m^2. This large value is probably due to the fact that Lang *et al.* calculate the coupling for monolayers rather than thick layers of Co. This is certainly the reason why, in contrast to our calculation, they find a relatively large amplitude of the long-period oscillations. It is only when the Co layers become bulklike that the band structures of Co and Cu along the relevant Γ-X line match well, which leads to weak confinement and, hence, to a very weak belly contribution to the coupling. However, it is interesting that, for large Cu thickness, all three calculations lead to broadly comparable overall strengths.

The calculations reported here have all been carried out at $T = 316$ K. If the summation over $\mathbf{k}_\parallel$ is done numerically, as in the present torque method calculations and in the work of Lang *et al.* [9], it is essential to work at finite temperature to secure convergence with only a few thousand $\mathbf{k}_\parallel$ points. This is not feasible at $T = 0$ [5]. A great advantage of evaluating the $\mathbf{k}_\parallel$ sum analytically by the stationary phase method is that the result, Eq. (4), is valid over the whole temperature range, including $T = 0$. In the present example we find that on going from $T = 316$ K to $T = 0$ the coupling increases by only 30% for a 6 ML spacer but by a factor 2 for a 25 monolayer spacer. The second advantage of the stationary phase method, fully exploited in this paper, is the explicit separation of contributions to the exchange coupling from different Fermi surface extrema. In view of these strengths of the asymptotic method, exact for large spacer thickness, the independent verification of its accuracy over a wide range of spacer thickness for a real system (Fig. 2) is very important for future work.

We are grateful to the Science and Engineering Research Council, to the Nuffield Science Foundation (U.K.), and to CNPq of Brazil for financial support.

*Present address: Department of Mathematics, City University, London, United Kingdom.

[1] *Ultrathin Magnetic Structures I, An Introduction to the Electronic, Magnetic and Structural Properties,* edited by J. A. C. Bland and B. Heinrich (Springer-Verlag, Berlin, 1994).

[2] J. E. Ortega and F. J. Himpsel, Phys. Rev. Lett. **69**, 844 (1992).

[3] M. T. Johnson, P. J. H. Bloemen, R. Coehoorn, J. J. de Vries, N. W. E. McGee, R. Jumgblut, A. Reinders, and J. aan de Stegge, Mater. Res. Soc. Symp. Proc. **313**, 93 (1993).

[4] D. M. Edwards, J. Mathon, R. B. Muniz, and M. S. Phan, Phys. Rev. Lett. **67**, 493 (1991).

[5] D. M. Edwards, J. Mathon, R. B. Muniz, and M. S. Phan, J. Phys. Condens. Matter **3**, 4941 (1991).

[6] J. Mathon, M. Villeret, and D. M. Edwards, J. Phys. Condens. Matter **4**, 9873 (1992).

[7] J. Mathon, M. Villeret, J. M. Mander, D. M. Edwards, and R. B. Muniz, Mater. Res. Soc. Symp. Proc. **313**, 171 (1993).

[8] F. Herman, J. Sticht, and M. van Schilfgaarde, J. Appl. Phys. **69**, 4783 (1991).

[9] P. Lang, L. Nordström, R. Zeller, and P. H. Dederichs, Phys. Rev. Lett. **71**, 1927 (1993).

[10] D. A. Papaconstantopoulos, *Handbook of The Band Structure of Elemental Solids* (Plenum, New York, 1986).

[11] V. L. Moruzzi, J. F. Janak, and A. R. Williams, *Calculated Electronic Properties of Metals* (Pergamon, Oxford, 1978).

[12] D. M. Edwards, A. M. Robinson, and J. Mathon (to be published).

[13] J. d'Albuquerque e Castro, M. S. Ferreira, and R. B. Muniz, Phys. Rev. B **49**, 16 062 (1994).

[14] J. Mathon, M. Villeret, and D. M. Edwards, J. Magn. Magn. Mater. **127**, L261–L266 (1993).

[15] J. Mathon, J. Phys. Condens. Matter **1**, 2505 (1989).

[16] M. P. Lopez Sancho, J. M. Lopez Sancho, and J. Rubio, J. Phys. F **15**, 851 (1985).