Oscillatory Coupling between Ferromagnetic Layers Separated by a Nonmagnetic Metal Spacer

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We have derived the expression of the Ruderman-Kittel-Kasuya-Yosida coupling between two ferromagnetic layers separated by a nonmagnetic spacer, for arbitrary crystal structure and Fermi surface. For the first time, quantitative predictions of the interlayer coupling for Cu, Ag, and Au spacers are given. The results are in good agreement with recent experimental data for fcc Co/Cu and Fe/Cu systems.

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The interaction between ferromagnetic layers across a nonmagnetic metal spacer is currently attracting considerable attention. Very recently, oscillatory coupling (alternately ferromagnetic and antiferromagnetic) as a function of spacer thickness has been evidenced in Fe/Cr, Co/Cr, Co/Ru [1], Co/Cu [2,3], Fe/Cu [4], and numerous other systems [5]. The observed oscillation periods are quite large: $\Lambda \approx 5$–10 monolayers (ML). Further studies revealed the existence of multiperiodic oscillatory coupling in Fe/Cr systems [6].

This behavior is reminiscent of a Ruderman-Kittel-Kasuya-Yosida (RKKY) coupling mechanism. However, a naive application of the RKKY theory (i.e., assuming a uniform spin distribution within the ferromagnetic layers and allowing the spacer thickness to vary continuously) yields a period $\Lambda = \lambda_F/2 \approx 1$ ML [7,8], which is too short to explain the experimental results. Theories of the interlayer coupling based on *ab initio* [9] or tight-binding [10] total-energy calculations are restricted to low spacer thicknesses and are therefore not well suited for investigating long-period oscillatory coupling. Moreover, they lack physical transparency and the calculated coupling energies are often 1 order of magnitude too large.

In the present Letter, we extend the general theory of RKKY exchange [11] to the problem of interlayer coupling. Our theory provides a physically transparent explanation of most experimental observations (long-period oscillatory coupling, multiperiodic oscillations), in terms of the topological properties of the spacer Fermi surface. In particular, we show that large periods and multiperiodic oscillations are natural consequences of the discreteness of the spacer thickness and of the moment distribution within the ferromagnetic layers, *even within the free-electron approximation.* From realistic data for the Fermi surfaces of noble metals, we calculate for the first time the interlayer coupling for Cu, Ag, and Au spacers, in (111), (001), and (110) orientations. The results are in good agreement with state-of-the-art experimental data for Co/Cu and Fe/Cu systems. We discuss the effect of the roughness, and show that it may drastically influence the apparent period and magnitude of the oscillatory coupling.

Let $F1$ and $F2$ be two ferromagnetic monolayers embedded in a nonmagnetic metal. The distance between $F1$ and $F2$ is $z = (N+1)d$, where d is the interlayer spacing and N the number of atomic layers of the spacer. The magnetic layers are assumed to consist of spins $\mathbf{S}_i$ located on atomic positions $\mathbf{R}_i$ of the host metal. In a first approximation, the interaction of a conduction electron (spin $\mathbf{s}$, position $\mathbf{r}$) with $\mathbf{S}_i$ can be described by $\mathcal{V}_i(\mathbf{r},\mathbf{s}) = A\delta(\mathbf{r}-\mathbf{R}_i)\mathbf{s}\cdot\mathbf{S}_i$. It is known that such a rough approximation yields incorrect values for the oscillation phase [12]; one should therefore keep in mind that the phase obtained for the interlayer coupling cannot be expected to agree with the experimental one. The correct phase, as well as the coupling strength, could be obtained from a more sophisticated (but less transparent) approach treating explicitly the hybridization between the $3d$ bands of the ferromagnetic layers and the host-metal conduction band [13].

The spin polarization of the conduction electrons gives rise to an indirect exchange interaction $\mathcal{H}_{ij} = J(\mathbf{R}_{ij}) \times \mathbf{S}_i \cdot \mathbf{S}_j$ [14]. The exchange integral is

$$J(\mathbf{R}_{ij}) = -\left(\frac{A}{V_0}\right)^2 \frac{V_0}{(2\pi)^3} \int d\mathbf{q} F(\mathbf{q}) \exp(i\mathbf{q}\cdot\mathbf{R}_{ij}), \quad (1)$$

where V_0 is the atomic volume of the host metal and

$$F(\mathbf{q}) = \frac{V_0}{(2\pi)^3} \int d\mathbf{k} \frac{f(\epsilon_\mathbf{k})[1-f(\epsilon_{\mathbf{k}+\mathbf{q}})]}{\epsilon_{\mathbf{k}+\mathbf{q}} - \epsilon_\mathbf{k}}. \quad (2)$$

Here and in the following, we consider a single conduction band; the generalization to several bands is immediate [11]. The integration over $\mathbf{q}$ and $\mathbf{k}$ in Eqs. (1) and (2), respectively, is performed within the first Brillouin zone (FBZ). It is implicitly meant in Eq. (2) that an appropriate vector $\mathbf{G}$ of the reciprocal lattice is added to $\mathbf{k}+\mathbf{q}$ in order to keep it within the FBZ, or, equivalently, that we are working within a periodic zone scheme.

The interlayer coupling is obtained by summing $\mathcal{H}_{ij}$ over all the pairs ij, i and j running respectively on $F1$ and $F2$. We invert the order of the $\mathbf{q}$ and $\mathbf{R}_{ij}$ summations, and replace the $\mathbf{q}$ summation over the FBZ by a summation over $\mathbf{q}_\parallel$ (in-plane component of $\mathbf{q}$) running on the *two-dimensional* FBZ of the ferromagnetic plane and over q_z running from $-\pi/d$ to π/d. This amounts to replacing the FBZ by a (completely equivalent) prismatic auxiliary zone, which is better suited to our problem. The different terms of the summation over $\mathbf{R}_{ij}$ interfere destructively and yield a zero contribution unless $\mathbf{q}_\parallel = 0$.

The interlayer coupling is thus given by the *one-dimensional* Fourier transform of $F(q_z) \equiv F(\mathbf{q}_\parallel = 0, q_z)$ [7].

The interaction due to the smooth part of the function $F(q_z)$ is short range, because of destructive interferences. On the other hand, singularities of $F(q_z)$ produce a long-range oscillatory coupling. The coupling at large spacer thicknesses is therefore mainly due to the neighborhood of the singularities. The latter occur for wave vectors $\mathbf{q}_S$ linking two points $\mathbf{k}_1$ and $\mathbf{k}_2$ of the Fermi surface with antiparallel velocities $\mathbf{v}_1$ and $\mathbf{v}_2$ (Kohn singularities). Thus, the oscillation periods correspond to the $\mathbf{q}_S$ which are parallel to z, as shown in Fig. 1. From their hole confinement model, Edwards *et al.* [15] found that the periods are given by the z projection of vectors $\mathbf{q}_C$ linking a pair of points of the Fermi surface with velocities respectively parallel and antiparallel to z (calliper vectors), which is in disagreement with the present approach: Such a vector $\mathbf{q}_C$ may have a nonzero in-plane component, which is forbidden according to our theory; conversely, the velocities of the points $\mathbf{k}_1$ and $\mathbf{k}_2$ linked by a vector $\mathbf{q}_S$ parallel to z may have nonzero in-plane components.

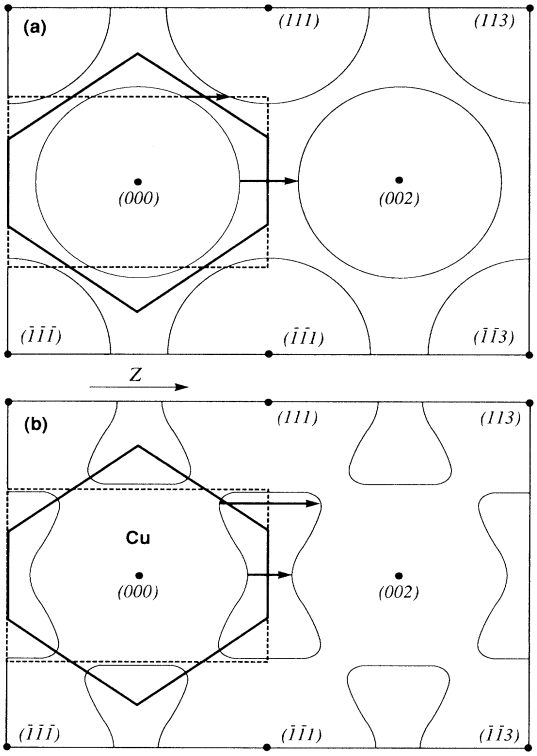

FIG. 1. Cross section of the Fermi surface, for a fcc (001) spacer. The FBZ and the auxiliary prismatic zone are represented by the bold solid and dashed contours. The wave vectors giving rise to oscillatory interlayer coupling are indicated by the horizontal bold arrows. (a) Free-electron approximation, and (b) Cu Fermi surface. The vectors giving the oscillation period for the (111) orientation are also shown (oblique arrows).

We have derived the expression of the interlayer coupling for a general Fermi surface, in an approach similar to Ref. [11]. The coupling energy per unit area can be written $E_{1,2}(z) = I_{1,2}(z)\cos(\theta_{1,2})$, where $\theta_{1,2}$ is the angle between the magnetizations of $F1$ and $F2$. The interlayer coupling constant is

$$I_{1,2}(z) = -I_0 \sum_\alpha \frac{d^2}{z^2} \frac{m_\alpha^*}{m} \sin(q_S^\alpha z + \phi_\alpha) \frac{z/L_\alpha(T)}{\sinh[z/L_\alpha(T)]}, \tag{3}$$

with

$$I_0 = (A/V_0)^2 S^2 m / 16\pi^2 \hbar^2. \tag{4}$$

α labels the Kohn singularities q_S^α of $F(q_z)$ ($0 \le q_S^\alpha \le \pi/d$), and will be omitted in the following. In Eq. (3), m^* is a characteristic mass expressing the strength of the interaction and related to the curvature tensors of the Fermi surface at $\mathbf{k}_1$ and $\mathbf{k}_2$, the phase shift ϕ is related to the topology of the Fermi surface at $\mathbf{k}_1$ and $\mathbf{k}_2$ (maxima, minima, or saddle points), and $L(T)$ is an attenuation length inversely proportional to the temperature T and related to the Fermi velocities at $\mathbf{k}_1$ and $\mathbf{k}_2$. A more detailed description of the calculations will be presented in a forthcoming publication.

For a simple illustration of this theory, we first consider a fcc spacer with one conduction electron per site, within the free-electron approximation [i.e., $k_F = (12\pi^2)^{1/3}/a$]. The Fermi surface thus consists of spheres centered on the reciprocal-lattice points [Fig. 1(a)]. We see that there may be several periods (Table I); this is an important difference with respect to earlier RKKY calculations assuming a continuous spin distribution on $F1$ and $F2$ [7,8,16]. Multiple periods arise as a consequence of the discrete spin distribution within the ferromagnetic layers, when the interatomic distances are not small as compared to the Fermi wavelength: In such cases the approximation of a uniform spin distribution breaks down, and interferences produce additional periods. This was also confirmed by direct-space numerical summations of $J(\mathbf{R}_{ij})$. The period $\Lambda = 2\pi/q_S$ can be much larger than the one expected from a naive RKKY argument ($\Lambda = \pi/k_F$) [7]. This is a direct consequence of the discrete-

TABLE I. Results of the calculations for a fcc lattice, in the free-electron approximation. For each oscillation period $\Lambda = 2\pi/q_S$, n is the number of equivalent pairs $(\mathbf{k}_1, \mathbf{k}_2)$, so that the strength of the oscillatory interaction is given by $n \times m^*$.

	d/a	Λ/d	n	m^*/m	ϕ	$L(300\text{ K})/d$
(111)	$1/\sqrt{3}$	10.26	1	1.00	π	30.67
(001)	1/2	4.58	1	1.00	π	35.43
		5.99	4	0.43	π	15.10
(110)	$1/2\sqrt{2}$	3.78	2	0.43	π	21.35
		13.30	2	0.77	π	38.52
		2.24	1	1.00	π	50.11

TABLE II. Same as Table I, for a noble-metal spacer.

	d (Å)	Λ/d	n	m^*/m	ϕ	L(300 K)/d
			Cu (a=3.603 Å)			
(111)	2.08	4.50	3	0.24	$\pi/2$	8.01
(001)	1.80	5.88	1	0.43	π	23.54
		2.56	4	0.40	$\pi/2$	18.04
(110)	1.27	3.29	2	0.58	π	8.15
		2.11	1	6.83	$\pi/2$	35.13
		9.60	1	0.29	$\pi/2$	20.52
		2.53	1	0.83	0	20.52
			Ag (a=4.069 Å)			
(111)	2.35	5.94	3	0.17	$\pi/2$	5.54
(001)	2.03	5.58	1	0.37	π	27.02
		2.38	4	0.29	$\pi/2$	12.54
(110)	1.44	3.52	2	0.34	π	11.25
		2.14	1	1.89	π	41.45
		13.36	1	0.22	$\pi/2$	13.67
		2.35	1	0.45	0	13.67
			Au (a=4.065 Å)			
(111)	2.35	4.83	3	0.16	$\pi/2$	9.46
(001)	2.03	8.60	1	0.18	π	22.41
		2.51	4	0.27	$\pi/2$	21.04
(110)	1.44	3.32	2	0.89	$\pi/2$	5.60
		2.09	1	5.17	$\pi/2$	33.33
		10.13	1	0.19	$\pi/2$	24.46
		2.49	1	0.63	0	24.46

ness of the spacer thickness, and can be interpreted, within a direct-space picture, as a vernier effect [16]; equivalently, within a reciprocal-space picture, this is due to the addition of a reciprocal-lattice vector **G** in order to keep q_z within the interval $[-\pi/d,\pi/d]$ ("umklapp" process).

The Fermi surface of noble metals differs notably from the one of free electrons [see Fig. 1(b)]. In particular, the necks around the center of the hexagonal faces of the FBZ are expected to play an important role. We have thus calculated the RKKY interlayer coupling for Cu, Ag, and Au (Table II) by using realistic Fermi-surface data obtained by Halse [17] from de Haas–van Alphen and cyclotron-resonance experiments. The results for Cu, Ag, and Au are qualitatively very similar, but present significant differences with the results of the free-electron approximation. For the (111) and (001) orientations the magnitude of the interlayer coupling is seen to decrease monotonically along the series Cu → Ag → Au. The temperature dependence of the coupling due to the smearing of the Fermi-Dirac function $f(\epsilon)$ is given by the last factor in Eq. (3). L(300 K) ranges between 5 and 30 ML for the (111) and (001) orientations, so that a sizable (but not drastic) coupling reduction is predicted at room temperature for the thickness range of experimental interest ($N \leq 30$ ML). By assuming typical values $A/V_0 \approx 1$ eV [18] and $S=1$, we obtain $I_0 \approx 13$ ergs cm^{-2}. $I_{1,2}$ is thus of the order of 0.4 erg cm^{-2} at $T=0$ for a spacer thickness of 5 ML, which is in good agreement with experimental results.

The results given in Table II, together with Eq. (3), can be used for quantitative comparison with experimental data. Figure 2(a) shows the calculated coupling for a Cu(001) spacer at $T=0$. For this system, a strong short-period oscillatory coupling and a much weaker long-period one are predicted. The apparent period thus corresponds to the short one, whereas the long-period term contributes only to a slight modulation of the oscillations. However, in the above discussion, the layers were assumed to be atomically flat, whereas real samples always present some interfacial roughness. In order to discuss the effect of the roughness, we now consider that the spacer layer of average thickness N actually consists of *large* (as compared to the spacer thickness) patches with local thickness equal to $N-1$, N, and $N+1$, with a weight equal to r, $1-2r$, and r, respectively. The coupling for a Cu(001) spacer with a roughness parameter $r=0.25$ is shown in Fig. 2(b): The coupling strength is strongly reduced, and the apparent period is increased. This is because the short period is almost suppressed by the roughness, and only the weak, long-period oscillatory coupling is seen. Thus the roughness acts as a low-pass filter for the oscillation frequency [13]. This simple example illustrates how important the influence of the

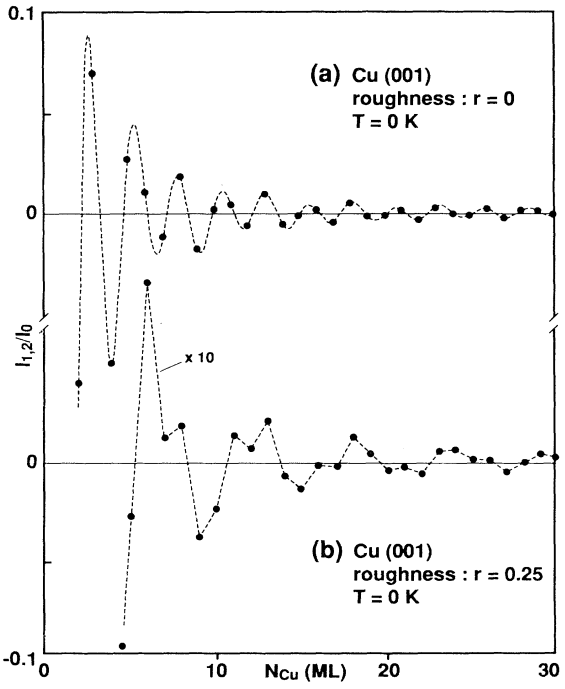

FIG. 2. Calculated interlayer coupling $I_{1,2}$ (normalized to I_0) for a Cu(001) spacer, as a function of the Cu thickness N_{Cu}, at $T=0$ K. The solid circles correspond to physically achievable thicknesses (N_{Cu} integer). (a) Zero roughness, and (b) roughness $r=0.25$.

roughness can be [6].

The systems fcc (111) Co/Cu/Co has been investigated by Mosca *et al.* and by Parkin, Bhadra, and Roche [2]. Both authors report an oscillatory interlayer coupling, with a period $\Lambda \approx 5$–6 ML; the magnitude of the coupling at the first antiferromagnetic maximum ($N_{Cu} \approx 4.5$ ML) is $I_{1,2} \approx 0.15$–0.3 erg cm^{-2}, at $T=0$. This value decreases by roughly 10% at room temperature. For the same system (assuming a roughness parameter $r=0.25$), our theory predicts $\Lambda = 4.50$ ML, $I_{1,2} \approx 0.25$ erg cm^{-2} at $N_{Cu}=4.5$ ML and $T=0$, and a 5% decrease of $I_{1,2}$ at room temperature. The phase also agrees with the experimental one, within the observed range; however, owing to the above-mentioned weakness of our model on this point, this agreement might well be fortuitous.

For the system fcc (001) Fe/Cu/Fe and fcc (001) Co/Cu/Co, Bennett, Schwarzacher, and Egelhoff [4] and Cebollada *et al.* [3] found, respectively, $\Lambda \approx 7$ ML and $\Lambda \approx 6$ ML, and no short period was observed. The observed period is rather close to the large period ($\Lambda = 5.88$ ML) calculated for a Cu(001) spacer; as discussed above, the predicted short period ($\Lambda = 2.56$ ML) is much more difficult to observe and may be hidden by the roughness.

Considering the experimental uncertainties, as well as the numerous approximations involved in the model, the agreement between our (parameter-free) theoretical predictions and the experimental observations appears fairly satisfying: The period of oscillation, the temperature dependence, and the magnitude of the interlayer coupling are well described. This is very promising. Nevertheless, further progress (both experimental and theoretical) is strongly needed for an accurate understanding of the interlayer coupling mechanism.

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