Geometrical Resonance in Magnetic Multilayers

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Experimentally, the magnetic coupling strength in Co/*TM* and Fe/*TM* multilayer systems, for which *TM* is a nonmagnetic transition metal, increases dramatically as a function of the electron per atom ratio, e/a, in the *TM* spacer layers. On the basis of simple model calculations we argue that this effect is a finite resonant increase of the coupling due to the variation of the band offset between the *TM* spacer and the magnetic host. We find that the specific form of the correlation is strongly influenced by the resonant coincidence of the *TM* bands with either the spin up or the spin down bands of the host, for particular values of e/a. We suggest new experiments to test the model.

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The discovery [1] of the oscillatory coupling between two adjacent ferromagnetic layers separated by paramagnetic transition metal (*TM*) spacers continues to attract both experimental [2,3] and theoretical [4–6] attention.

Most of the existing theories [4–6] interpret these oscillations as consequences of RKKY interactions between two adjacent magnetic metal (*MM*) layers. Within this picture, the magnetic coupling J, defined as the difference per unit interface area between the grand potentials of the ferromagnetic ($\uparrow\uparrow$) and antiferromagnetic ($\uparrow\downarrow$) configurations, behaves as

$$J(L) = A^{-1}[\Omega_{\uparrow\uparrow}(L) - \Omega_{\uparrow\downarrow}(L)] = C^{(m)} L^{-2} \sin(2k_F L + \phi^{(m)}) + O(L^{-3}), \quad (1)$$

where L is the thickness of the spacer layers. The L^{-2} dependence in Eq. (1), the signature of RKKY interactions between planar magnetic "defects," is clearly confirmed by the experiments [7]. The predicted spatial periodicity is $(2k_F)^{-1}$, where $2k_F$ is one of the extremal diameters in the Fermi surface of the spacer. The amplitude, $C^{(m)}$, and the phase, $\phi^{(m)}$, in Eq. (1), may be regarded, at this stage, as phenomenological parameters. Although experiments report larger periodicities, usually about 10 Å, this does not contradict the above picture because both perturbative treatments [5] and exact model calculations [8] indicate that RKKY oscillations, when sampled with the periodicity of the spacer lattice in the growth direction, d, can give rise to longer periods. This so-called "aliasing" effect, i.e., the interference between the two characteristic lengths, d and $(2k_F)^{-1}$, may generate the experimentally observed periodicities.

However, to date, much less is known about the strength of the coupling, as measured by the variations of both $C^{(m)}$ and $\phi^{(m)}$, in Eq. (1), when the material constituting the spacer or the magnetic layers is changed. For instance, the empirical correlation discovered by Parkin [3], that, for Co/*TM* and Fe/*TM* systems, this strength increases exponentially with the total number of valence electrons per atom in the spacer, has not yet received an explanation. In this Letter we shall give a qualitative description of this surprising phenomenon, using a very simple inhomogeneous jellium model [9,10]. According to our picture, the strength of the magnetic coupling is determined by the differential band offset of the spin-up and spin-down electronic structures. We find that, as the electron per atom ratio increases, this mechanism gives rise to a resonant enhancement of the amplitude of the magnetic oscillations. To put this in another way, the increase of J with e/a is due to an increase in the difference between the extent to which spin-up and -down electrons are confined in the spacer layers.

In addition to analyzing Parkin's data, we shall illustrate this mechanism by applying our model to the study of a (Fe_cV_{1-c})/Cr system, where the band structure of the *MM* layers is changed continuously. Interestingly, at those concentrations for which one of the two spin polarizations feels the multilayer as homogeneous (e.g., no offset), we shall find a resonant increase in the magnetic coupling.

We consider the geometry of a "sandwich" [Fig. 1(a)]. Two planar parallel interfaces of area A are separated by

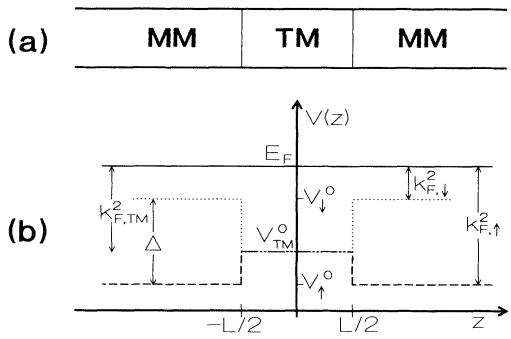

FIG. 1. (a) Schematic representation of the "sandwich" geometry for magnetic multilayers: The *MM* occupies the regions $|z| > L/2$; the *TM* layers are bounded between the interfaces. (b) Model electronic potentials for majority (dashed line) and minority (dotted line) carriers in the ($\uparrow\uparrow$) configuration. The full line corresponds to the Fermi level. The potentials in the ($\uparrow\downarrow$) configuration (not reported here) may be obtained exchanging the spin-up and -down potentials in the right *MM* region.

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a thickness L of spacer TM layers and the MM occupies the two external (semi-infinite) volumes. Conveniently and without any loss of generality, the grand potential of this system can be decomposed into the following contributions [9]:

$$\Omega(T,\mu,V,A,L)=V_{MM}\omega_{MM}(T,\mu)+V_{TM}\omega_{TM}(T,\mu)+2A\sigma(T,\mu)+A\omega_{II}(T,\mu,L)\,, \quad (2)$$

where T and μ are the temperature and the chemical potential (we assume $T=0$; therefore, $\mu=E_F$), V_{MM},V_{TM} and ω_{MM},ω_{TM} are the volumes and the grand potential densities for the two bulk metals, $V=V_{MM}+V_{TM}$, σ is the interface energy for a single TM/MM interface, and ω_{II} an interface-interface interaction (III) energy per unit area. As physical intuition would indicate $\omega_{II}(T,\mu,L)$ vanishes when $L\rightarrow\infty$ [9]. Since the first two terms in Eq. (2) correspond to bulk systems and are linear in L and the single interface energy σ does not depend at all on L, oscillations with L must arise from $\omega_{II}(E_F,L)$.

Furthermore, we model the sandwich using a simple noninteracting-electron-jellium model. Namely, the crystal potential is taken to be constant within each layer, with finite discontinuities occurring at the interfaces [Fig. 1(b)]. For the ($\uparrow\uparrow$) configuration this means

$$V_{\uparrow(\downarrow)}(z)=V^0_{\uparrow(\downarrow)}\Theta(|z|-L/2)+V^0_{TM}\Theta(L/2-|z|)\,, \quad (3.1)$$

for the $\uparrow$ and $\downarrow$ electrons, respectively. In our free-electron scheme, the constants in Eq. (3.1) are given by

$$E_F-V^0_\alpha=k^2_{F,\alpha}=E_{F,\alpha}=(6\pi^2N_\alpha/v_\alpha)^{2/3} \quad (3.2)$$

for $\alpha=\uparrow,\downarrow$, or TM,

where E_F is the Fermi energy, and N_α and v_α are the total numbers of valence (s, p, and d) electrons per atom and the Wigner-Seitz volumes. Of course $v_\uparrow=v_\downarrow$ and $N_\uparrow+N_\downarrow=N_{MM}$, the total number of valence electrons per atom in the MM [11].

In the absence of electron-electron interactions the grand potentials of the two spin subsystems are additive, $\Omega=\Omega_\uparrow+\Omega_\downarrow$, and a very useful description of the model can be given [9,12]. For the sake of simplicity, we report here only the formula holding when $V^0_{\uparrow(\downarrow)}<V^0_{TM}$ and there are no states bounded in the spacer layers:

$$\omega_{II\uparrow(\downarrow)}(L)=\omega_{exc\uparrow(\downarrow)}(L)-\omega_{exc\uparrow(\downarrow)}(\infty)\,, \quad (4.1)$$

$$\omega_{exc\uparrow(\downarrow)}(L)=A^{-1}[\Omega_{\uparrow(\downarrow)}(L)-(V_{MM}\omega_{MM}+V_{TM}\omega_{TM})]=\frac{1}{4\pi^2}\int_{V^0_{\uparrow(\downarrow)}}^{E_F}dE(E-E_F)g_{\uparrow(\downarrow)}(E)\,, \quad (4.2)$$

$$g_{\uparrow(\downarrow)}(E)=\tan^{-1}[\tfrac{1}{2}(k_{\uparrow(\downarrow)}/k_{TM}+k_{TM}/k_{\uparrow(\downarrow)})\tan(Lk_{TM})]-Lk_{TM}+m\pi\,, \quad (4.3)$$

where $k^2_\alpha=E-V^0_\alpha$. Here $g(E)$ is the sum of the one-dimensional phase shifts corresponding to the potential well (3.1), with a contribution linear in L subtracted off. In the Friedel sum given in Eq. (4.3), the integer m is incremented each time the resonance condition $Lk_{TM}/\pi-\frac{1}{2}=n$ is satisfied. The integrand in Eq. (4.2) has $2Lk_{F,TM}/\pi$ zeros in the integration domain; consequently, as L increases, more oscillations are integrated and ω_{exc} turns out to be an oscillatory function of L [9].

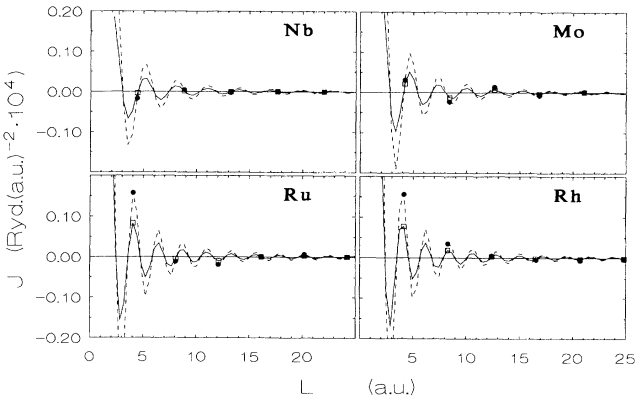

FIG. 2. The calculated magnetic coupling $J(L)$. Here the MM is Co (full lines) or Fe (dashed lines) and the spacer is a 4d TM (Nb, Mo, Ru, and Rh, with increasing e/a). Symbols mark spacer thicknesses corresponding to an integer number of layers. According to Eq. (1), antiferromagnetic coupling corresponds to a positive value of J. The growth orientations in the multilayers are the same as in Ref. [3].

In Fig. 2, we plot our results, $J(L)$, for a selection of Co and Fe/4d TM multilayers. As expected, the magnetic coupling is easily fitted by the functional form of Eq. (1). This fit, as it is clear also from the figure, shows that the amplitude $C^{(m)}$ and the phase $\phi^{(m)}$, *both* increase with the electron per atom ratio in the TM. To compare our results with the experiment, we have plotted in Fig. 3 the constant J_0, defined by Parkin [3] as the strength of the coupling at the first antiferromagnetic peak normalized for a thickness of 3 Å, for Fe and Co/TM multilayers. The agreement with Ref. [3] is quite remarkable: Our model is able to reproduce the qualitative trends, mainly the sharp increase, throughout most of the periodic table, *without any adjustable parameter* [11]. By contrast, our results for the spatial periodicity of the coupling are quite poor. Of course, such failure is not unexpected

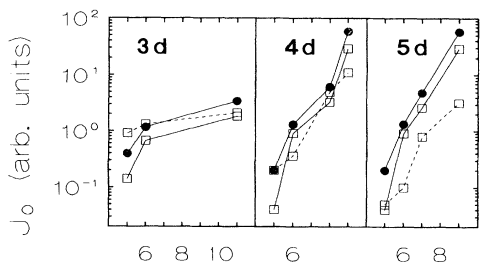

FIG. 3. The normalized magnetic coupling constant J_0 defined by Parkin [3] for Co (open squares) and Fe (black circles)/TM multilayers. The spacers are 3d, 4d, and 5d TM. Full lines: present calculations; dashed lines: experiment [3].

since, according to the aliasing argument, the periodicity is a quite delicate function of the Fermi wave vector and therefore one cannot expect a free-electron theory to reproduce the empirical trends. In fact the surprise is that the theory works so well for J_0.

To analyze the source of this success, it is convenient to concentrate on the dominant L^{-2} term in the III energy corresponding to a single polarization:

$$\omega_{\mathrm{III}\uparrow(\downarrow)}(L)\cong C_{\uparrow(\downarrow)}L^{-2}\sin(2k_{F,TM}L+\phi_{\uparrow(\downarrow)}) . \quad (5)$$

The behavior of the parameters in Eq. (6), as a function of the electronic structure of the *MM* and *TM* layers, is mainly determined by the sudden truncation of the integration domain in (4.2) at the Fermi energy and therefore by $g(E_F)$. Furthermore, it can be readily argued that

$$C_{\uparrow(\downarrow)}\cong(V^0_{\uparrow(\downarrow)}-V^0_{TM})f(x_{\uparrow(\downarrow)}) , \quad (6.1)$$

$$\phi_{\uparrow(\downarrow)}\cong\phi(x_{\uparrow(\downarrow)}) , \quad (6.2)$$

where the parameter $x_{\uparrow(\downarrow)}=k_{F,TM}/k_{F\uparrow(\downarrow)}$ and the functions $f(x)$ and $\phi(x)$ are dimensionless. As shown in Fig. 4, these scaling hypotheses are confirmed by the analysis of many calculations. Note the most striking feature at $x=1$, where the jump in the potential at the interfaces vanishes and the phase ϕ increases discontinuously by π [13]. This is the signature of a geometrical resonance: For $x>1$, $V_{TM}-V_{\uparrow(\downarrow)}<0$ and the spacer layers bound electrons in the growth direction. This is the consequence of the fact that the Schrödinger equation for the one-dimensional potential (3.1) has at least one such bound state for any *arbitrarily small* negative value of $V^0_{TM}-V^0_{\uparrow(\downarrow)}$. Moreover, since there is no restriction on the momentum in the other directions, we have an infinite number of such 2D states for any $x>1$ (and for any value of L). These have no analogs when $x<1$ because the electrons of the *MM* encounter a potential barrier at the interfaces instead of a potential well. As it turns out, these two regions are connected by a jump in the phase. In fact, the configurations with $x<1$ and $x>1$ are mutually orthogonal; therefore, at $x=1$, as in the case of the well-known Anderson infrared catastrophe [14], the theory is not analytic.

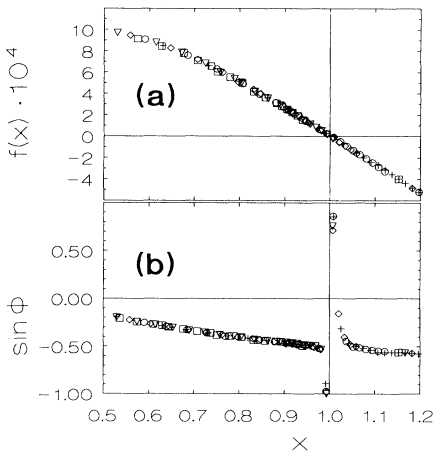

FIG. 4. The scaling functions $f(x)$ and $\phi(x)$ [see Eq. (6)] vs $x=k_{F,TM}/k_{F\uparrow(\downarrow)}$. Different symbols refer to different values of $k_{F\uparrow(\downarrow)}$, as follows: (+) 1.20, (○) 1.28, (◇) 1.36, (▽) 1.43, and (□) 1.53, in atomic units. For all the spacers considered, except Cu, $x_\downarrow$ lies in a quite narrow interval to the left of 1.

We are now able to write down an expression for the coefficients of the leading term (1) in the magnetic coupling. From Eqs. (1), (5), and (6), we find

$$C^{(m)}\cos\phi^{(m)}=\Delta[f(x_\uparrow)\cos\phi(x_\uparrow)-f(x_\downarrow)\cos\phi(x_\downarrow)] , \quad (7.1)$$

$$C^{(m)}\sin\phi^{(m)}=\Delta[f(x_\uparrow)\sin\phi(x_\uparrow)-f(x_\downarrow)\sin\phi(x_\downarrow)] , \quad (7.2)$$

where $\Delta=V^0_\downarrow-V^0_\uparrow$ is the exchange splitting in the magnetic layers.

The behavior of $C^{(m)}$ and $\phi^{(m)}$ is entirely determined by the "universal" functions $f(x)$ and $\phi(x)$, and hence depends only on the electronic densities in the two metals [15]. Thus the occurrence of the geometrical resonance is, to some extent, model independent.

Let us now discuss the behaviors of $C^{(m)}$ and $\phi^{(m)}$ in terms of Eqs. (7), with reference to Fig. 3. When the band structures in the *TM* and the *MM* are different *for both the polarizations*, $|x_{\uparrow(\downarrow)}-1|\gg 0$ and the phases of up and down charge oscillations are very similar, $C^{(m)}\propto|f(x_\uparrow)-f(x_\downarrow)|$ and $\phi^{(m)}\approx\phi(x_\uparrow)\approx\phi(x_\downarrow)$. However, if for one of the polarizations the band structures become similar in the *MM* and *TM* layers, the corresponding phase jumps by π, reflecting the geometrical resonance. The amplitude $C^{(m)}$ peaks and the phase $\phi^{(m)}$ changes discontinuously from one to the other side of $x_{\uparrow(\downarrow)}=1$. In general, increasing e/a in the spacer layers, the band structure in the nonmagnetic metal may become resonantly similar to that of minority carriers in the *MM*, as shown in Table I; in other words, $x_\downarrow$ increases from the left toward 1. This increase, because of Eq. (7), is large but finite at variance with the conclusions of Parkin [3]. We note that, with the exception of Cu, for all the spacers considered, $x_\downarrow$ is quite close to the resonant value 1. Evidently, if it were possible to increase $x_\downarrow$ beyond the resonance one should observe a decrease in the coupling.

TABLE I. Free-electron model Fermi energies [see Eq. (3.2)] for 3*d*, 4*d*, and 5*d* transition metals. *N* is the total number of *s*, *p*, and *d* electrons in the outer shell. For Fe and Co two values are reported, corresponding to different spin polarizations.

N	3*d*	E_F	4*d*	E_F	5*d*	E_F
5	V	1.36	Nb	1.15	Ta	1.15
6	Cr	1.70	Mo	1.42	W	1.41
7					Re	1.64
8	Fe	1.72,2.39	Ru	1.89		
9	Co	2.06,2.60	Rh	2.03	Ir	1.99
1	Cu	0.52				

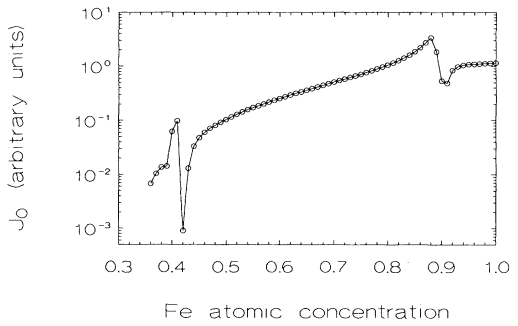

FIG. 5. J_0 vs Fe concentrations in (Fe_cV_{1-c})/Cr multilayers.

To lend support to the above suggestion we have performed calculations for Cr multilayers with the alloy Fe_cV_{1-c} as *MM*. Alloying the *MM* was modeled by varying the magnetic splitting as well as e/a. We have assumed a linear dependence on the concentration for the lattice parameter and have used the experimental fact that a magnetic moment appears at $c=0.30$ and then increases linearly with the concentration. As it turns out, we may expect the resonances to occur at $c=0.42$ and $c=0.88$, where the up and down Fermi energies in the alloy cross that in Cr. Our calculations confirm the hypothesis concerning the resonant behavior. In particular, we observe two sharp peaks in $C^{(m)}$ at $c=0.42$ and $c=0.88$, superimposed to the parabolic trend that can be obtained by a Taylor expansion in Δ of Eqs. (7). At the same concentrations a discontinuity in the phase occurs. The corresponding behavior of Parkin's J_0 is shown in Fig. 5. Here, on going from $c=1$ to $c=0.88$, the resonant condition $x_\downarrow=1$ is approached, not adding electrons to the spacer as in Fig. 3 but subtracting carriers from the magnetic layers. In the present case the resonance is somehow smoothened since the magnetic splitting Δ is proportional to the Fe concentration.

In conclusion, we have provided an interpretation of the "exponential" increase of the coupling strength observed by Parkin [3] in Co/*TM* and Fe/*TM* multilayers in terms of the geometrical resonance that occurs when the differential band offset for one of the electronic polarizations becomes small. This may happen for pure *MM* layers and high-valence *TM* spacers or for magnetic layers weakened by alloying. In the latter case this should lead to an observable nonmonotonic behavior of the coupling as a function of the concentration. Of course, our model is extremely simple and its quantitative predictions can be regarded only as a rough guide to what may happen in actual experiments. In particular, the resonance phenomena might be quite sensitive to interface roughness, which could broaden the features shown in Fig. 5. This would be consistent with the fact that our model overestimates (see Fig. 3) the increase of J_0 in the case of pure Co multilayers.

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