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We postulate localized-electron states with unpaired spin located within or at the interfaces of an otherwise nonmagnetic metallic spacer layer. Loose exchange coupling of these spins to two ferromagnets mediates a non-Heisenberg exchange coupling between them which includes a biquadratic term. A particular version of the model assumes that each interfacial layer of magnetic atoms is weakly exchange coupled to the remainder of the ferromagnets. This model permits interpretation of the biquadratic-coupling data of Gutierrez *et al.* for Fe/Al/Fe trilayers and of Fuss *et al.* for Fe/Au/Fe. According to this interpretation, the observed biquadratic coupling is intrinsic to the ideal multilayer structure rather than due to impurities or structural defects.

I. INTRODUCTION

Until recently, measurements of exchange coupling in single-crystal magnetic trilayers were interpreted exclusively with the conventional "bilinear" Heisenberg-like energy expression $E(\theta) = -J_1 \cos\theta$, where θ is the angle between the magnetic vectors of the two ferromagnets. But just last year there appeared experimental reports, based on fmr[1] and domain and hysteresis[2] studies, of an additional, non-Heisenberg term in E which we write in the "biquadratic"[2] form $J_2 \cos^2\theta$. (Our $J_2 = -2B_{12}$ of previous notation.[2]) When biquadratic exchange is dominant with $J_2 > 0$ and positive cubic anisotropy is present, it has the peculiar effect of aligning magnetic moments orthogonally instead of colinearly.[2] The need for this term or its equivalent was reported in epitaxial trilayers, grown on nonmagnetic substrates, which have the compositions Co/Cu/Co,[1] Fe/Cr/Fe,[2-4] Fe/(Au,Al)/Fe,[5] Fe/Al/Fe,[6] and Fe/Cu/Fe.[7] Evidence of orthogonal domain arrangements is also present in micrographs of Fe-whisker-based Fe/Cr/Fe trilayers.[8] Only one sign ($J_2 \geqslant 0$) has been reported in all experiments. These discoveries have opened wide the theoretical question of the nature of exchange coupling through a nonmagnetic metal, especially since J_2 is often large.

At least three types of mechanism may, in principle, give rise to biquadratic or more generally non-Heisenberg form of exchange. The first is one associated with spatial fluctuations of bilinear coupling due to terraced variations of spacer thickness in nonideal specimens.[9] The resulting torque fluctuations induce static spin-wave fluctuations whose sum of coupling and exchange-stiffness energies is minimized when the mean moments are orthogonal. The simplest version of the theory gives the relation

$$J_2 = \frac{4L(\Delta J)^2}{\pi^3 A} \coth\left(\frac{\pi D}{L}\right), \tag{1}$$

where A is the exchange stiffness within the ferromagnetic layers of thickness D. The spacer has monolayer-high terraces of width L at whose edges the local exchange J_1 has step changes of amount $\pm 2\Delta J$. Paradoxically, J_2 *increases* with L, i.e., as the specimen becomes more nearly perfect, up to the point $J_2 \simeq \Delta J$ where the theory breaks down. The fluctuation mechanism was found consistent in order of magnitude with published data for GaAs-based Fe/Cr/Fe trilayers.[2] The variation of 90° bands with phase of short-period 180° bonding in Fe-whisker based, wedge shaped, Fe/Cr/Fe is qualitatively consistent with the fluctuation mechanism.[8] The fluctuation mechanism has been confirmed by experiments and detailed analysis for trilayers of composition Fe/Cu/Fe.[7] Other theoretical work treats the same mechanism numerically to embrace large-amplitude effects.[10]

The second mechanism is the intrinsic one arising from the electron structure of an ideal trilayer. There are already six calculations of the angular dependence of intrinsic exchange energy,[11-16] all of which predict a biquadratic term. Three of these calculations[11,12,16] utilize the spin-current technique first introduced in connection with exchange through an insulating spacer.[17] In all of the calculations, J_2 is found to oscillate as a function of spacer thickness w with half the period of J_1 in the asymptotic limit of large w. Asymtotically, half of the nodes of J_2 coincide with those of J_1. However, according to one *preasymptotic* computation particularly appropriate to Fe/Cr/Fe, the nodes of J_2 are shifted with respect to those of J_1 in such a direction that the sign of J_2 is positive at each node of J_1.[11] This result thus offers a qualitative explanation of the orthogonal domains, implying $J_2 > 0$, found at the nodes of J_1 in whisker-based trilayers Fe/Cr/Fe.[8] In addition, the interpretation of data for Fe/Cu/Fe (Ref. 7) admits the possibility of an appreciable intrinsic J_2 contribution (along with that due to the spacer fluctuations) as estimated from the hole-gas model.[12]

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Experiments with epitaxial Fe/Al/Fe trilayers[6] revealed a biquadratic coupling which depends on temperature too strongly to be explained easily by the fluctuation mechanism. Moreover, the temperature dependence of the intrinsic mechanism is also too slow to explain these results.[12] Very recent measurements for Fe/Au/Fe,[18] GaAs-based Fe/Cr/Fe,[19] and Fe/Al/Fe (Ref. 20) also give remarkably consistent results exhibiting steep temperature dependences of J_2 for all spacer thicknesses. Since J_1, and therefore ΔJ, is found to vary little with temperature, Eq. (1) rules out the fluctuation mechanism. Always $J_2>0$ is found, thus ruling out the intrinsic mechanisms mentioned above because they predict oscillations about 0. The inconsistency of this new body of data with existing theory has stimulated the present proposal and investigation of an additional mechanism.

Our new *loose-spin* mechanism postulates localized atomic-electron states which contribute a non-Heisenberg energy to the exchange coupling between the pair of ferromagnets. The underlying mechanism is an indirect exchange, such as that of Ruderman, Kittel, Kasuya, and Yosida (RKKY), coupling each localized spin $\mathbf{S}_i$ to other spins through the polarizability of the conduction band.[21,22] By assumption, these atoms do not lie within one of the ferromagnets, so their exchange fields may be weak and we term them "loose" spins. Any such loose spins present within the spacer or adjacent to its interfaces contribute an effective exchange coupling between the ferromagnetic films. The angular dependence of this coupling energy departs from the conventional bilinear form because of the non-linearity connected with saturability of the loose-spin polarization. One finds $J_2>0$ always. A similar effect should arise from a very thin additional ferromagnetic layer deposited artificially within the spacer.

Generally, the predicted positive sign of J_2 implies that the effect of loose spins is always additive, unlike the above intrinsic J_1 and J_2 which have RKKY oscillations. Therefore, experiments with deliberately introduced impurities could test the loose-spin theory and reveal the presence of the spacer polarization induced by the ferromagnets even if it is made so incoherent by interface roughness as to cause intrinsic exchange to be greatly weakened by cancellations.

We attempt to fit predictions of this loose-spin model to existing experimental data for biquadratic exchange in three (100)-epitaxial trilayers of composition Fe/Al/Fe,[6] and one of Fe/Au/Fe.[18] One type of fit assumes that the active loose spins comprise the two ferromagnetic atomic layers adjoining each side of the spacer. If this particular mechanism, which we call that of *loose interfacial spins*, is correct, it should also be considered intrinsic, strictly speaking, because only the ideal atomic structure, without impurities or defects, is involved. This attribution of a special role to the interfacial layer resembles previous suggestions.[13,23]

II. COUPLING THROUGH LOOSE SPINS

Referring to Fig. 1, let $F1$ and $F2$ be two identical saturated semi-infinite ferromagnets having unit magnetization vectors $\mathbf{m}_1$ and $\mathbf{m}_2$. They are composed of atomic

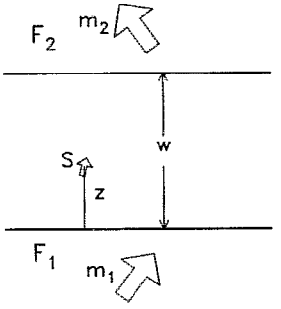

FIG. 1. Cross section showing two semi-infinite ferromagnets $F1$ and $F2$, with unit moments $\mathbf{m}_1$ and $\mathbf{m}_2$, joined by a nonmagnetic spacer of thickness w. The small arrow represents a loose spin $\mathbf{S}$ situated at a distance z from $F1$.

spins $\hbar S_f$. In a continuous representation, their edges lie at $z=0$ and $z=w$, respectively. Consider also the additional single atomic electron state with local spin quantum number S and momentum operator $\hbar\mathbf{S}$, at position z $(0<z<w)$. It is subject to local, conveniently dimensioned exchange-coupling fields $\mathbf{U}_1=U_1(z)\mathbf{m}_1$ and $\mathbf{U}_2=U_2(z)\mathbf{m}_2$, which are due to the respective conduction-electron polarization fields induced by the two ferromagnets. As $\mathbf{m}_1$ and $\mathbf{m}_2$ are mutually independent, the vector sum of these fields gives rise to the effective local-spin Hamiltonian $H=(\mathbf{U}_1+\mathbf{U}_2)\cdot\mathbf{S}/S$.

An expression for the coupling $U_1(z)$ between a semi-infinite ferromagnet and *an atom* is inherent in the RKKY-type theory for coupling between a semi-infinite ferromagnet and an atomic monolayer.[24] Thus we may write, in our own notation,

$$U_1(z)=\frac{-J_f J_l m^* S_f S a^2 d}{32\pi^2\hbar^2 k_F}\,\frac{\cos(2k_F z+\psi)}{z^2}, \tag{2}$$

which is valid asymtotically for $k_F z\to\infty$. Here, m^* is the effective mass of the relevant Bloch state having extremal Fermi vector k_F lying orthogonal to the spacer plane. Also, J_f and J_l (J/Ω in the previous notation[24]) have the dimension of energy and measure the strength of the intraatomic exchange between a local electron spin and those of the itinerant electrons; ψ is a phase constant dependent on hybridization.[25] Because the ferromagnetic layers are assumed identical, we have $U_2(z)=U_1(w-z)$.

The energy levels of the defect are $\epsilon_m=-Um/S$ with $m=-S,-S+1,\ldots S$. Here

$$U(\theta)=|\mathbf{U}_1+\mathbf{U}_2|=(U_1^2+U_2^2+2U_1U_2\cos\theta)^{1/2}, \tag{3}$$

where θ is the angle between $\mathbf{m}_1$ and $\mathbf{m}_2$. Since U is likely to be relatively weak, it makes sense to consider thermal excitation of the loose spins. We neglect thermal excitation of the strongly coupled spins located within the ferromagnets. From conventional statistics, the free energy per loose spin is

$$f(T,\theta)=-k_BT\ln\left(\frac{\sinh\{[1+(2S)^{-1}]U(\theta)/k_BT\}}{\sinh[U(\theta)/2Sk_BT]}\right). \tag{4}$$

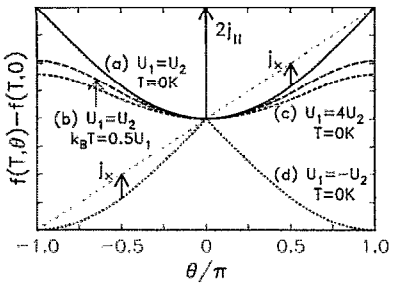

FIG. 2. The free-energy difference $f(T,\theta)-f(T,0)$ per loose spin located inside or at an interface of the spacer, as a function of angle θ between the magnetization vectors of the two ferromagnets. Units of f are arbitrary. Local exchange-field parameters U_1, U_2, and temperature T are varied as indicated. The loose-spin quantum number is $S=1$. Geometric constructions of colinear ($j_\parallel$) and orthogonal ($j_\times$) exchange-coupling coefficients are indicated for cases (a) and (d).

This represents an additive contribution to the θ-dependent energy coupling the two ferromagnetic layers. The possibilities of the local moment being destroyed at low temperatures by delocalization or Kondo effect, or of taking part with other spins in a spin-glass transition, are neglected throughout.

Figure 2 illustrates $f(T,\theta)-f(T,0)$ vs θ for various cases, exhibiting these characteristics: For $U_1U_2>$ or <0, we have $f(T,\pi)>$ or $<f(T,0)$, and the coupling is called "ferromagnetic" or "antiferromagnetic," respectively. In the limit $T=0$ K, $f(0,\theta)=\epsilon_S=-U(\theta)$. In the special case $T=0$ K *and* $U_1=\pm U_2$ this reduces to the nonanalytic form

$$f(0,\theta)=-2\left|U_1 \begin{matrix}\cos\\ \sin\end{matrix}\left(\frac{\theta}{2}\right)\right|,$$

which has the slope discontinuities seen in curves (a) and (d) of Fig. 2. As illustrated by curves (b) and (c), $f(T,\theta)$ assumes analytic forms for $T\neq 0$ or $U_1\neq U_2$.

The macroscopic free energy F per unit spacer area is phenomenologically expandable in the form

$$F(\theta)=J_0-J_1\cos\theta+J_2\cos^2\theta+J_3\cos^3\theta+\cdots. \quad (5)$$

In connection with the nonanalytic loose-spin energies shown in Figs. 2(a) and 2(d), this series does not converge rapidly for small T if $|U_1|$ is close to $|U_2|$. Therefore, it is convenient to define two new exchange-coupling coefficients[14]: One is the *colinear* coupling $J_\parallel=(1/2)[F(\pi)-F(0)]$. The other is the *orthogonal* coupling $J_\times$ equal to the difference between the mean energy of the two colinear alignments $\theta=(0,\pi)$ and the energy of orthogonal alignment $\theta=\pi/2$:

$$J_\times=(1/2)F(0)+(1/2)F(\pi)-F(\pi/2). \quad (6)$$

Whenever F is well approximated by the first three terms of Eq. (5), we have $J_\parallel=J_1$ and $J_\times=J_2$. These definitions of $J_\parallel$ and $J_\times$ are particularly appropriate when cubic magnetocrystalline anisotropy orients the moments of the ferromagnets strongly along (100) axes, as in the often-used case of iron, because then each equilibrium value of θ in M-H studies of (100)-epitaxial multilayers is nearly equal

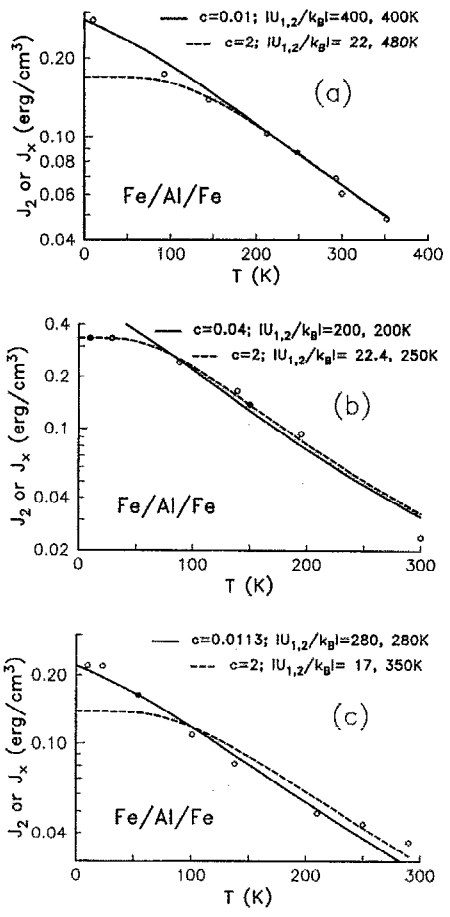

FIG. 3. Comparison of loose-spin theory for $J_\times$ with experimental J_2 vs temperature T for three (100) trilayer specimens (a, b, and c) of composition Fe/Al/Fe, reported by Gutierrez *et al.* (see Ref. 6). In each case, one curve assumes $|U_1|=|U_2|$ while $|U_1|$ and the fractional atomic concentration c (based on one atomic plane) are adjusted. The other curve in each plot assumes $c=2$ and $|U_1|$ and $|U_2|$ are adjusted individually.

to 0, $\pi/2$, or π. Figure 2 indicates geometric constructions of the corresponding atomic coefficients $j_\parallel=J_\parallel\rho^{-1}$ and $j_x=J_x\rho^{-1}$, where $\rho=ca^{-2}$ is the areal density of the loose spins. Here a is the nearest-neighbor distance (2.87 Å in Fe, 2.86 Å in Al) between atoms in one (100) plane, and c is the fractional concentration of loose spins.

Our Figs. 3 and 4 show examples of theoretical $J_\times$ in the loose-spin model, using semilog plots and assuming

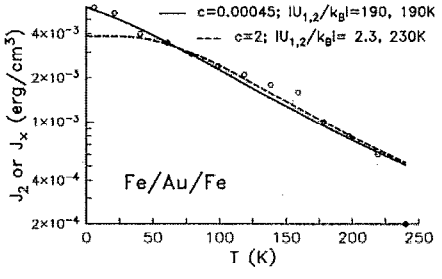

FIG. 4. Theoretical fits of $J_\times$ to J_2 data for a (100) trilayer of composition Fe/Au/Fe, taken by Fuss, Wolf, and Grünberg (see Ref. 18), as in Fig. 3.

$S=1$. Other values of S do not give markedly different predictions. In the special case $U_1=\pm U_2$, $J_\times(T)$ is nearly exponential, as illustrated by the solid curves. For $|U_1|$ significantly different from $|U_2|$, the function $J_\times=J_2(T)$ has a plateau at small T, followed by the nearly exponential decay evident in the dashed curves. For $|U_1|\ll|U_2|$, the bilinear coupling reduces unsurprisingly to $j_1=U_1B(U_2/k_BT)$, where $B(x)$ is the well-known Brillouin function

$$B(x)=\left(\frac{2S+1}{2S}\right)\coth\left(\frac{2S+1}{2S}x\right)-\frac{1}{2S}\coth\left(\frac{x}{2S}\right). \quad (7)$$

The oscillatory behavior of U_1 according to Eq. (2) implies that J_1 due to loose spins oscillates. For $U_1\ll U_2$ and $T=0$, we have $j_1=U_1$ sgn U_2, $j_2=U_1^2/2|U_2|$. The latter relation is equivalent to one due to Barnas' and Grünberg.[13] The sign $j_2\geqslant 0$ implies that J_2 may dominate experimentally, even though it is higher order than J_1, because the oscillation amplitude of the latter may be attenuated by fluctuations of spacer thickness.

III. LOOSE INTERFACIAL SPINS

In recent experiments, three (100) epitaxial trilayer specimens of composition Fe/Al/Fe, which we designate (a), (b), and (c) had Al thicknesses and preparation conditions such that J_1 was too small to be measured.[6] They differed notably in the growth temperature of the second Fe film: (a) 150 °C, (b) −10 °C, (c) 180 °C. The data for J_2, obtained from *M-H* loops, are plotted in Fig. 3. Specimens (a) and (c) behave quite exponentially, favoring the loose-spin model with $|U_1|=|U_2|$ (solid curves). In each case the solid curve is obtained under the assumption $|U_1|=|U_2|$, with c and $|U_1|$ adjusted for a fit. This case might be approached if the loose spins were those of impurity atoms near the midplane of the spacer. However, such large exchange fields $|U_{1,2}|/k_B$ ranging between 250 and 400 K cannot be reconciled with Eq. (2). One may, however, postulate that each loose spin comprises a *cluster* of Fe atoms, with U_1 and U_2 proportional to the number of atoms in the cluster. The fitted concentrations, $c=1\%$–2%, correspond to a volume concentration of 2–4 clusters per 1000 Al atoms in the circa 13-Å-thick spacers.

On the other hand, specimen (b) gives data, replotted in Fig. 3(b), which is not simply exponential but deviates from exponential behavior much in the way the theory does for $|U_1|\ll|U_2|$. Each dashed curve assumes $c=2$ (see below), and U_1 and U_2 ($U_1<U_2$) are adjusted to fit the data, giving greater weight to large T. The question of which curve types, solid or dashed, in Fig. 3 make a better fit is made less significant by a reevaluation of one specimen, presented at this Conference, which raises J_2 at small T to values definitely higher than either type of curve.[6] This indicates the presence of additional loose spins, e.g., Fe impurities, *within* the spacer.

Experimental data for J_2 in (100) epitaxial trilayers Fe/Au/Fe were obtained by Fuss *et al.*[18] The data, together with theoretical fits of $J_\times$, like those in Fig. 3, are shown in Fig. 4. Noteworthy in this case is the value of $U_1/k_B=2.3$ K fitted under the hypothesis $c=2$. Its smallness compared to those ($U_1/k_B=17$–22 K) in Al may be attributed, according to Eq. (2), to the small effective masses $m^*/m=0.18$ and 0.27 in Au bands tabulated previously.[26] Noteworthy is the rapid decrease of experimental J_2 with temperature, falling for $T\geqslant 260$ K to values less than 1/30 of its value at $T=0$ K. The data for J_1 fall much more slowly; at $T=300$ K, J_1 still has about 80% of its value at $T=0$, ruling out interpretation in terms of the fluctuation-model Eq. (1).

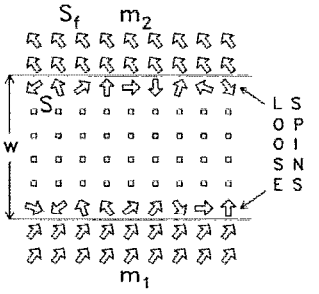

FIG. 5. Illustration of trilayer with loose interfacial spins. For correspondence to Fig. 1, and application of Eq. (2), the spacer width w is shown to include the two interfacial layers belonging to the ferromagnets.

Under the assumption $c=2$, theoretical fits of U_2/k_B in Figs. 3 and 4 range between 250 and 480 K for the Al spacer, and $U_2/k_B=230$ K for the Au spacer. Such large values of exchange might arise at the interfacial layers of the ferromagnets for *structurally perfect* specimens. We note that none of the nearest neighbors of an atom in the bcc structure of Fe lie in the same (100) plane. This makes the above mean-field treatment, which neglects correlations between the spins in this layer, reasonable in spite of their high concentration in a fully populated lattice plane. The existence of two such interfacial layers accounts for our assumption $c=2$.

Figure 5 illustrates our model of loose interfacial spins. Each ferromagnet less one interfacial layer constitutes one of the continuous semi-infinite saturated ferromagnets, with separation w, considered in Fig. 1. The two interfacial magnetic layers, indicated in Fig. 5, constitute atoms with loose spins (spin quantum number S) of the type considered in Fig. 1. Application of our above theory implies neglect of coupling between the two interfacial layers. This is justified as follows: (1) coupling between two magnetic monolayers should be weaker than that of a single monolayer to the many layers of a semi-infinite magnet at a comparable distance. (2) The d orbitals of the magnetic-interface layer will hybridize with the s orbitals of the adjoining normal metal weakening the atom-to-conduction-electron exchange. This implies $|J_l|<J_f$. For example, the d bands in bulk Fe are split by 2 eV whereas the s bands are split by only 0.4 eV.[27]

Our values of U_2/k_B, ranging between 230 and 480 K for Al and Au spacers, were fitted under the hypothesis of loose interfacial spins. They compare favorably with the $T_C=1043$ K for bulk iron in a qualitative sense, because

the interfacial layer has half as many nearest neighbors and hybridization leads to $|J_l| < J_f$ as explained above. Let us compare Eq. (2) with the corresponding U_1/k_B ranging between 17 and 22 K for Al spacers with $z \simeq 16$ Å. We note that the (001) lattice planes of the fcc aluminum lattice are separated by $d=a/2^{1/2}$. When using Eq. (2) we may consider the three conduction electrons per Al atom to write $k_F=(36\pi^2)^{3/2}/a=1.75\times10^8$ cm^{-1}. Equation (2) is then consistent with the foregoing numbers if we assume the parameter value $|J_fJ_l|^{1/2}=10$ eV and that ψ is such that $\cos(2k_Fz+\psi)=\pm1$.

Of the two hypotheses tried, $c=2$ gives the more reasonable fits for U_1 and U_2 in Figs. 3 and 4. The fact that $|J_fJ_l|^{1/2}=10$ eV of RKKY theory is unacceptably large (It should be closer to 2 eV) does not necessarily disprove the loose-interfacial-spin mechanism of biquadratic coupling. We have found in a fundamental calculation[16] that taking into account the difference between the itinerant-electron densities of the spacer and the ferromagnets can make J_1 for their mutual coupling much greater than predicted by RKKY theory when the conduction-electron band of the spacer is nearly free-electron-like with a large Fermi energy, as in Al. It is reasonable to believe that a similar correction is needed for the coupling $U_1(z)$ of a semi-infinite ferromagnet to a loose spin. In physical terms, a large $U_1(z)$ could be attributed to an enhancement of the ferromagnet-induced spin-density wave in the spacer by quantum-well reflections between the two interfaces. (The infinite-electron gas assumption of the RKKY theory does not provide an opportunity for amplitude-building reflections.) Bound quantum-well states figure in some exchange theories[28–30] and in inverse photoelectric measurements.[31] Qualitative support for this picture is seen in a very recent work[15] which explicitly describes exchange coupling between ferromagnets as occurring by repeated spin-differentiated reflection of electron waves from the interfaces. If looseness of interfacial spins is confirmed by further studies, it will indicate that free-electron-like metals with high electron density, such as Al, in principle transmit much stronger coupling than hitherto supposed. The relative modesty of *bilinear* couplings thus far observed in Al may be attributable to the destructive interference of the RKKY oscillations caused by spacer roughness.

An experimental test of the loose-spins model would be provided by measuring J_2 in a trilayer whose spacer contains deliberately deposited magnetic impurities. Such a test may be circumscribed by the Kondo effect or by the spin-glass "freezing" temperature T_{fr}. For example, one has $T_{fr}=9$, 14, 23, and 29 K, for bulk alloys having 1, 2, 5, and 8 atomic percent Fe in Au, respectively.[32] For $T<T_{fr}$, the freezing will tend to suppress local-spin coupling, but our theory is meaningful only for the paramagnetic state $T>T_{fr}$ or a ferromagnetic state. Another problem, not addressed here, is the diminution of the direct ferro-ferro coupling through the scattering of the RKKY spin-polarization waves by the loose spins at elevated temperatures.

However, if the local atomic concentration of loose spins exceeds the percolation value ($\simeq 17\%$ for the fcc lattice) then ferromagnetism with some transition temperature T_c can occur. Letting $T<T_c$, one has the possibility of observing the anomalous coupling energy $F=-ca^{-2}|U_1\cos(\theta/2)|$, resulting from $U_1=U_2$ [see Fig. 2(a)], by placing the extra ferromagnet at the center of the spacer. If the functions $U_1(z)$, $U_2(z)$ are oscillatory as suggested by Eq. (2), a judicious choice of spacer thickness and position z of the impurity layer could provide the condition $U_1=-U_2$, giving $F=-ca^{-2}|U_1\sin(\theta/2)|$. Since this condition gives antiferromagnetic alignment at vanishing external field, magnetization curves could have the possibility of testing for this form of nonconventional dependence of the energy [see Fig. 2(d)].

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