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Generalized Ruderman-Kittel-Kasuya-Yosida theory of oscillatory exchange coupling in magnetic multilayers

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In Ruderman-Kittel-Kasuya-Yosida (RKKY) perturbation theories of magnetic multilayers, realistic energy bands are used to obtain the nesting required for oscillatory behavior, but the Bloch functions are generally approximated by plane waves. To account for the observed long-period oscillations of the exchange coupling, we present a generalized RKKY theory, which includes both the Bloch character of the wave functions and the boundary scattering at the film edges. In contrast to existing theories, these two effects lead to long-period oscillations, that are robust with regard to roughness of the spin distribution and to oscillation amplitudes that sharply increase with the localized nature of the Bloch functions, in agreement with experiment.

Long-range oscillatory exchange coupling $J(t)$ has been observed between ferromagnetic layers that are separated by paramagnetic spacer layers of thickness t for a large number of transition- and noble-metal multilayers, such as Fe/Cr and Co/Cu.[1,2] $J(t)$ exhibits damped oscillations as t is increased, with a period varying between 9 and 18 Å, depending on the spacer metal.[2] In addition, short-period oscillations are observed in certain systems, e.g., Fe/Cr, Fe/Al, and Fe/Au multilayers, which have atomically smooth interfaces.[3] Moreover, such multilayers also exhibit giant magnetoresistance,[4,5] with potential in magnetic recording technology.[6]

The origin of such long-wavelength oscillatory exchange interactions is generally believed to be associated with the Ruderman-Kittel-Kasuya-Yosida[7,8] (RKKY) interaction $J_0(r) \sim \cos 2k_F r/r^3$ between two localized spins separated by distance r in a bulk metal, where k_F is the Fermi wave vector. When summed over spins on the interfaces,[9,10] the coupling becomes $J_{\rm RKKY} \sim \cos 2k_F t/t^2$. Presumably, this type of coupling accounts for the observed rapid oscillations.

The current explanation of the long-wavelength oscillations is based on the "aliasing" effect.[11-13] If one samples a rapidly oscillating wave precisely at each maximum, the apparent wavelength is clearly infinite. However, if the wave is discretely sampled at a slightly longer period, a slow spatial oscillation appears. Since for smooth interfaces t is incremented by multiples of the spacer lattice constant a, if the crystal lattice is nearly commensurate with the $2k_F$ wave, a long apparent period will be observed.

Recent first-principles, self-consistent energy-band calculations[14] indicate that the exchange coupling between adjacent Co layers in idealized fcc Co/Cu multilayers exhibits both long- and short-period oscillations. The coupling also depends on the crystallographic orientation of the interface. One can account for these results in terms of the prominent nesting vectors of bulk Cu, as shown in Fig. 1, where interzonal transitions $\mathbf{q}'=2\mathbf{k}_F-\mathbf{g}$ are required to account qualitatively for the numerical results. The aliasing effect provides a natural explanation for the $\mathbf{q}'=2\mathbf{k}_F-\mathbf{g}$ oscillations.

In this paper we point out that in addition to the aliasing effect there exists another source of long-period oscillations, which we term the Bloch modulation effect. This arises when one goes beyond the plane-wave approximation for the basis states in calculating the RKKY potential and uses the proper one-electron states in the presence of the lattice potential.

The RKKY exchange coupling between isolated spins or atomic moments embedded in a paramagnetic crystal-

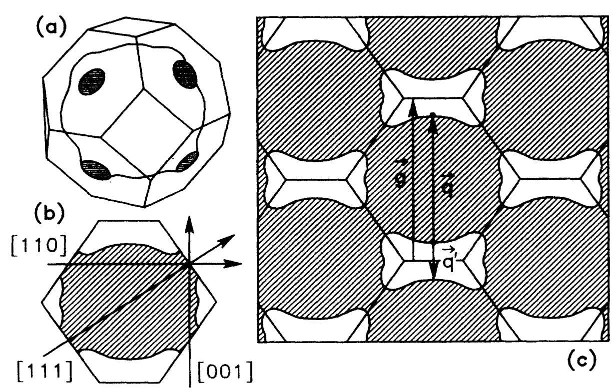

FIG. 1. Model Fermi surface for fcc Co/Cu multilayers. (a) Fermi surface and reduced zone of bulk fcc Cu. (b) Cross-sectional view of Fermi surface and reduced zone. (c) Fermi surface of Cu in extended k space showing formation of the dog-bone orbits. The longest period for [001] oscillatory coupling arises from the transition vector $\mathbf{q}'=\mathbf{q}-\mathbf{g}$, where $\mathbf{q}=\mathbf{k}'-\mathbf{k}$ is a nesting vector and $\mathbf{g}$ a reciprocal lattice vector.

line solid at positions $\mathbf{R}$ and $\mathbf{R}'$ (not necessarily lattice sites) arises from the spin polarization they induce in the solid. Initially, we neglect structural and chemical perturbations arising from the presence of the spin-bearing atoms and consider only the magnetic perturbations induced by the atomic moments $\mathbf{m}(\mathbf{r})$. Accordingly, we represent the unperturbed electronic structure of the solid by Bloch functions $\psi_\alpha(\mathbf{k},\mathbf{r})=u_\alpha(\mathbf{k},\mathbf{r})\exp[i\mathbf{k}\cdot\mathbf{r}]$ and energy eigenvalues $\varepsilon_\alpha(\mathbf{k})$ in the reduced zone scheme of the spacer material, where α is a band index. According to second-order perturbation theory, the RRKY interaction is proportional to

$$\Omega(\mathbf{R},\mathbf{R}')=\sum_{\alpha\alpha'\mathbf{k}\mathbf{k}'}\frac{\mathbf{M}_{\alpha\alpha'}(\mathbf{k},\mathbf{k}',\mathbf{R})\cdot\mathbf{M}_{\alpha'\alpha}(\mathbf{k}',\mathbf{k},\mathbf{R}')}{\varepsilon_\alpha(\mathbf{k})-\varepsilon_{\alpha'}(\mathbf{k}')}\ , \quad (1)$$

where the matrix elements are given by

$$\mathbf{M}_{\alpha,\alpha'}(\mathbf{k},\mathbf{k}',\mathbf{R})=\int\int\psi^*_\alpha(\mathbf{k},\mathbf{r})\mathbf{m}(\mathbf{r}-\mathbf{R})\psi_{\alpha'}(\mathbf{k}',\mathbf{r})d\mathbf{r}\ . \quad (2)$$

The sums on $\mathbf{k}$ and $\mathbf{k}'$ are limited to the reduced zone and are taken over occupied and unoccupied states, respectively; $\mathbf{m}(\mathbf{r})$ is the net spin density on a magnetic atom. From the Bloch periodicity condition $\psi_\alpha(\mathbf{k},\mathbf{r}+\mathbf{d})=\exp[i\mathbf{k}\cdot\mathbf{d}]\psi_\alpha(\mathbf{k},\mathbf{r})$, where $\mathbf{d}$ is a direct lattice vector, it follows that $\mathbf{M}$ can be written in the form

$$\mathbf{M}_{\alpha\alpha'}(\mathbf{k},\mathbf{k}',\mathbf{R})\equiv\exp[-i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{R}_0]\overline{\mathbf{M}}_{\alpha\alpha'}(\mathbf{k},\mathbf{k}',\delta\mathbf{R})\ , \quad (3)$$

where $\mathbf{R}_0$ is the position of the lattice site in the cell containing $\mathbf{R}$, and $\mathbf{R}=\mathbf{R}_0+\delta\mathbf{R}$. Thus, the exchange coupling becomes

$$\Omega(\mathbf{R},\mathbf{R}')=\sum_{\alpha\alpha'\mathbf{k}\mathbf{k}'}\frac{\overline{\mathbf{M}}_{\alpha\alpha'}(\mathbf{k},\mathbf{k}',\delta\mathbf{R})\cdot\overline{\mathbf{M}}_{\alpha'\alpha}(\mathbf{k}',\mathbf{k},\delta\mathbf{R}')}{\varepsilon_\alpha(\mathbf{k})-\varepsilon_{\alpha'}(\mathbf{k}')}\exp[-i(\mathbf{k}-\mathbf{k}')\cdot(\mathbf{R}_0-\mathbf{R}_0')]\ . \quad (4)$$

This formalism is readily extended to multilayers by summing the pair interactions between all magnetic atoms in successive magnetic slabs.[9,10]

If the one-electron basis functions are represented by plane waves, the numerator in Eq. (4) reduces to $|\hat{\mathbf{M}}(\mathbf{k}-\mathbf{k}')|^2\exp[-i(\mathbf{k}-\mathbf{k}')\cdot(\delta\mathbf{R}-\delta\mathbf{R}')]$, where $\hat{\mathbf{M}}(\mathbf{q})=\int\mathbf{m}(\mathbf{r})\exp[-i\mathbf{q}\cdot\mathbf{r}]d\mathbf{r}$. If the positions $\delta\mathbf{R}$, $\delta\mathbf{R}'$ in the unit cell are held fixed, and if we allow $|\mathbf{R}_0-\mathbf{R}_0'|$ to change by considering a series of multilayers having progressively thicker spacers, the period of the oscillation appears to be determined by $\mathbf{k}-\mathbf{k}'$, the intrazonal nesting vector. However, if $\mathbf{g}$ is a reciprocal lattice vector, we are free to include a factor of $\exp[-i\mathbf{g}\cdot(\mathbf{R}_0-\mathbf{R}_0')]=1$ inside the sum in Eq. (4) without changing $\Omega(\mathbf{R},\mathbf{R}')$. One is also free to choose $\mathbf{g}$ to minimize $|\mathbf{k}-\mathbf{k}'-\mathbf{g}|$ and maximize the period. Thus, a short-wavelength oscillation, when sampled periodically at nearly the same wavelength, appears to have a long wavelength. This is the aliasing effect proposed by several authors[11–13] to account for the observed long-period oscillations.

In addition to the aliasing effect, there exists another source of long-period oscillations, the Bloch modulation effect, which arises when one goes beyond the plane-wave approximation for the basis states and uses the proper one-electron Bloch states in the presence of the lattice potential. To see this, we expand the periodic part of the Bloch function in reciprocal space: $u_\alpha(\mathbf{k},\mathbf{r})=\sum(\mathbf{g})A_{\mathbf{g}\alpha}(\mathbf{k})\exp[i\mathbf{g}\cdot\mathbf{r}]$. The matrix elements $\overline{\mathbf{M}}$ then take the form

$$\begin{aligned}\overline{\mathbf{M}}_{\alpha\alpha'}(\mathbf{k},\mathbf{k}',\delta\mathbf{R})=&\sum_{\mathbf{g}=\mathbf{g}'}A^*_{\mathbf{g}\alpha}(\mathbf{k})A_{\mathbf{g}\alpha'}(\mathbf{k}')\hat{\mathbf{M}}(\mathbf{k}-\mathbf{k}')\exp[i(\mathbf{k}-\mathbf{k}')\cdot\delta\mathbf{R}]\\&+\sum_{\mathbf{g}\neq\mathbf{g}'}A^*_{\mathbf{g}\alpha}(\mathbf{k})A_{\mathbf{g}'\alpha'}(\mathbf{k}')\hat{\mathbf{M}}(\mathbf{k}-\mathbf{k}'+\mathbf{g}-\mathbf{g}')\exp[-i(\mathbf{k}-\mathbf{k}'+\mathbf{g}-\mathbf{g}')\cdot\delta\mathbf{R}]\ . \quad (5)\end{aligned}$$

From the vantage point of Eqs. (4) and (5), the aliasing effect arises from the product of the diagonal ($\mathbf{g}=\mathbf{g}'$) terms in $\overline{\mathbf{M}}_{\alpha\alpha'}(\mathbf{k},\mathbf{k}',\delta\mathbf{R})$ and $\overline{\mathbf{M}}_{\alpha'\alpha}(\mathbf{k}',\mathbf{k},\delta\mathbf{R}')$. As already noted, $\delta\mathbf{R}$ and $\delta\mathbf{R}'$, must be held fixed, and the factor of unity, $\exp[-i\mathbf{g}\cdot(\mathbf{R}_0-\mathbf{R}_0')]=1$, must be introduced in the numerator of Eq. (4). The Bloch modulation effect arises from all remaining terms, since all of these automatically include reciprocal lattice vector through the nondiagonal terms in $\mathbf{g}$. In essence, the "magnetic" $\mathbf{k}-\mathbf{k}'$ oscillation can be spatially modulated by the periodic lattice potential via $\mathbf{g}$ to produce a beat oscillation of small wave vector, $\mathbf{k}-\mathbf{k}'-\mathbf{g}$, much as the heterodyne effect acts in the time domain.[15]

Bearing in mind that the aliasing and Bloch modulation effects can give rise to the same set of long-period oscillations with small wave vectors $\mathbf{k}-\mathbf{k}'-\mathbf{g}$, can we distinguish these two effects from one another apart from the fact that they arise from different terms in the complete expression for the exchange coupling? If one restricts one's attention to periodically sampling the complete expression based on Bloch functions [cf. Eqs. (4) and (5)] at discrete lattice points, the A's only influence the amplitude of the oscillation.[16] The distinction between these two effects manifests itself when one considers spins that do not lie on lattice sites, and when we average over random arrangements of such spins.

In this context, let us consider multilayers whose interfaces are not perfectly lattice matched and atomically abrupt but are characterized by structural and chemical disorder. Such interfacial roughness is ill defined experimentally and may take many forms, depending on such factors as lattice mismatch, interfacial orientation, spacer polycrystallinity, and method of preparation. For purposes of discussion, we distinguish two types of interfacial disorder.

In type (i) disorder, the atoms occupy random sites on one or more well-defined lattice planes in the interfacial region. In magnetic rare-earth multilayers,[17] for example, where the atomic radii of the magnetic and nonmagnetic atoms, M and N, are nearly identical, the interface might consist of a single disordered *MN* plane, with nearly perfect *M* and *N* planes on either side. Broader interfaces could be produced by interdiffusion or by growth conditions that favor the formation of steps, terraces, and islands.

In type (ii) disorder, the interfacial atoms are no longer preferentially located on well-defined lattice planes. Instead, these atoms are displaced from lattice sites by misfit dislocations, lattice mismatch, or other sources of severe local strain, e.g., interdiffusion of different size atoms such as Co and Ru. A possible analogy is a semicoherent grain boundary separating two differently oriented crystallites. If the out-of-plane displacements are very small, we revert to type (i) disorder and if sufficiently large, to type (ii) disorder. In the latter case, we assume continuous atomic site distributions in three dimensions within the interfacial region, which is of order of a few interatomic distances wide.

According to the aliasing effect, interfacial roughness of type (i) has little influence on the coupling strength of long-period oscillations, as can be demonstrated by discrete spacer thickness averaging.[18] The essential point is that in type (i) disorder all spins on successive magnetic slabs are separated from one another by direct lattice vectors, even if they are distributed at random on the same or adjacent lattice planes. The aliasing effect occurs because the factor $\exp[-i\mathbf{g}\cdot(\mathbf{R}_0-\mathbf{R}_0')]$ is equal to unity for all pairs of spins.

On the other hand, interfacial roughness of type (ii) suppresses the aliasing effect, wiping out the long-period oscillations arising from this mechanism. This can be seen by explicit calculation by averaging the positions of the spins over distances of order the size of the short (nesting) wavelength: Since $|\mathbf{k}-\mathbf{k}'|$ is generally large for nesting conditions, averaging the $\mathbf{g}=\mathbf{g}'$ terms in Eq. (5) over a distribution of $\delta\mathbf{R}$ values larger than the short period $2\pi/|\mathbf{k}-\mathbf{k}'|$ leads to a small result. Clearly, the aliasing mechanism is no longer effective if the spin positions are not located on well-defined lattice planes or, more generally, at lattice sites. The exponential factor is no longer unity, since it includes a distribution of $\delta\mathbf{R}$ and $\delta\mathbf{R}'$ values.

Turning to the nondiagonal ($\mathbf{g}\neq\mathbf{g}'$) terms in Eq. (5), we note that $|\mathbf{k}-\mathbf{k}'+\mathbf{g}-\mathbf{g}'|$ can be small for appropriate values of $\mathbf{g}$ and $\mathbf{g}'$. This is true even if $\alpha=\alpha'$, i.e., even if one is dealing with *intraband* rather than interband transitions. If one now averages $\delta\mathbf{R}$ over a region smaller than the long period, the amplitude remains large. Thus, the Bloch modulation effect is robust with regard to this type of randomness for both intraband and interband processes. The corresponding transitions between plane-wave states do not exhibit this robustness.

The relative strengths of the leading $\mathbf{g}=\mathbf{g}'$ and $\mathbf{g}\neq\mathbf{g}'$ terms in $\overline{\mathbf{M}}$ for transition or noble-metal spacer materials can be estimated by using parametrized band structure schemes.[19] As will be shown in a subsequent paper,[20] the coupling strengths increase as one moves along a transition series toward increasing Z. This increase is related to the contraction of the outermost d orbitals, and is consistent with empirical observations.[2]

While we have treated the spacer as a bulk crystalline material, boundary scattering at the spacer-magnetic film interfaces as well as defect scattering in the bulk will alter the detailed form of $\Omega(\mathbf{R},\mathbf{R}')$. For example, if the spacer is treated as a free-standing film with rigid wall boundary conditions at $r=0$ and t,[20] the interaction Ω_{film} is described by $\Omega_{\text{film}}(\mathbf{R},\mathbf{R}')=\overline{\Omega}(\mathbf{R},\mathbf{R}')-\overline{\Omega}(\mathbf{R},-\mathbf{R}')$, where $\overline{\Omega}$ is given by Eq. (1) with $\mathbf{k}$ and $\mathbf{k}'$ restricted to values $k=n\pi/t$, $n=0,1,2,\ldots$. When $\mathbf{R}$ and $\mathbf{R}'$ are near opposite interfaces, $\overline{\Omega}(\mathbf{R}-\mathbf{R}')$ is small compared to $\overline{\Omega}(\mathbf{R},\mathbf{R}')$. If disorder is present in the spacer, $\overline{\Omega}$ is reduced by a factor of $\exp[-|\mathbf{R}-\mathbf{R}'|/l]$, where l is the mean free path.

In summary, we have shown that long-period oscillations in magnetic multilayers can be produced by the aliasing mechanism and also by the Bloch modulation mechanism. The fact that long-period oscillations are observed in multilayer systems having widely different degrees of lattice mismatch[1,2]—and by inference widely different types of interfacial disorder—can be explained by the existence of these two complementary RKKY-type mechanisms.

We believe that earlier studies involving the interaction of magnetic impurities in nonmagnetic solids[21] and spin-glasses[22] should be reexamined in the light of the generalized RKKY theory described here.

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[15]The asymptotic form of the RKKY interaction comes from states near $\mathbf{k}_F$ and $-\mathbf{k}_F$, where $2\mathbf{k}_F$ is the nesting vector. Using $|u_{-\mathbf{k}_F}(\mathbf{r})|^2=|u_{\mathbf{k}_F}(\mathbf{r})|^2$, the exchange interaction for the bulk is given by

$$\chi_{\mathrm{bulk}}(\mathbf{r},\mathbf{r}')=\chi_0(\mathbf{r}-\mathbf{r}')|u_{\mathbf{k}_F}(\mathbf{r})|^2|u_{\mathbf{k}_F}(\mathbf{r}')|^2 ,$$

where χ_0 is the interaction based on plane-wave basis functions but correct energy eigenvalues $\varepsilon_\alpha(\mathbf{k})$. The spatial variation of the $u_{\mathbf{k}_F}(\mathbf{r})$ gives our Bloch modulation effect. For a film with sharp edges, we obtain $\chi_{\mathrm{film}}(\mathbf{r},\mathbf{r}')=\tilde{\chi}_0(\mathbf{r}-\mathbf{r}')-\tilde{\chi}_0(\mathbf{r}+\mathbf{r}')$ if we use plane waves. If we use Bloch states instead, we obtain

$$\chi_{\mathrm{film}}(\mathbf{r},\mathbf{r}')=[\tilde{\chi}_0(\mathbf{r}-\mathbf{r}')-\tilde{\chi}_0(\mathbf{r}+\mathbf{r}')]|u_{\mathbf{k}_F}(\mathbf{r})|^2|u_{\mathbf{k}_F}(\mathbf{r}')|^2 .$$

For further details, see Ref. 20.

[16]If the Bloch functions are represented by orthogonalized plane waves, the A's are given in perturbation theory by $A=\Sigma(\mathbf{g})V(\mathbf{g})/[\varepsilon(\mathbf{k})-\varepsilon(\mathbf{k}'+\mathbf{g})]$. Thus, the A's are large if $\mathbf{k}$ is near a Brillouin-zone boundary. But this is the condition that one has a long-wave oscillation, i.e., $\mathbf{q}=2\mathbf{k}_F-\mathbf{g}$. This shows that the A's are large for long-wavelength oscillations.

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