RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 44, NUMBER 11 15 SEPTEMBER 1991-I

Simple theory of exchange coupling in transition-metal magnetic multilayers

D.M. Deaven and D.S. Rokhsar
Department of Physics, University of California, Berkeley, California 94720

M. Johnson
Bell Communications Research, Red Bank, New Jersey 07701
(Received 28 May 1991)

Several experiments on a variety of magnetic multilayer and sandwich structures have revealed an indirect exchange coupling between layers of ferromagnets separated by nonmagnetic spacer layers. This coupling oscillates from ferromagnetic to antiferromagnetic depending on the thickness of the spacer layers, with an anomalously long period. We study a simple model to explain the long period and compare our results with the Ruderman-Kittel-Kasuya-Yosida theory of indirect exchange coupling.

I. INTRODUCTION

The discovery of "giant magnetoresistance" in molecular-beam-epitaxy- (MBE) grown Fe/Cr multilayers[1] conjures up an abundance of technological applications and has spurred the study of other magnetic-nonmagnetic metallic multilayers. The "giant magnetoresistance" has now been observed in sputtered multilayers composed of essentially any of the transition metals,[2,3] and MBE-grown multilayers of Fe/Cu.[4,5] Magneto-optic Kerr effect measurements have demonstrated an enhanced optical rotation in Fe/Cu commensurate with the magnetoresistance anomalies.[5] This is exciting to workers seeking to improve the quality of optical storage systems based on the magneto-optic effect.

All of the systems studied thus far share a long period oscillation in the exchange coupling $\mathcal{J}(r)$ between ferromagnetic layers as a function of nonmagnetic spacer layer thickness r. Measurements of the saturation field of the magnetoresistance[2] probe $\mathcal{J}(r)$, which may also be measured via Brillouin light scattering,[4,6] ferromagnetic resonance,[4] magneto-optic rotation,[4,5,7] or spin-polarized secondary electron emission.[8] In general, these measurements show that $\mathcal{J}(r)$ oscillates around zero for r in the range $0-50$ Å. Considering the strength of the coupling ($|\mathcal{J}| \sim 5$ erg/cm^2 in Co/Ru,[2] for example), some sort of indirect exchange mechanism must be operating; magnetostatic interactions are an order of magnitude smaller. The period of the oscillation varies [$\lambda \sim 7$ Å in Fe/Cu (Ref. 5) and $\lambda \sim 18$ Å in Fe/Cr (Ref. 2)] but is substantially longer than the period predicted by the Rudermann-Kittel-Kasuya-Yosida (RKKY) theory of indirect exchange coupling.[9] In the RKKY theory the period is fixed at the reciprocal of twice the Fermi wave vector, i.e., a lattice constant.

Is this coupling simply related to the RKKY interaction, or does it represent a new type of indirect mechanism? This paper presents a calculation of $\mathcal{J}(r)$ within a simple mean-field model of itinerant electrons. The $\mathcal{J}(r)$ calculated using this model has a long period of oscillation. We also calculate the corresponding RKKY coupling, and show that our result is indeed a close cousin of the RKKY phenomenon.

II. MODEL

We are motivated to study a simple model[10] by the wide variety of systems in which qualitatively similar oscillatory couplings have been observed. This suggests that details of the electronic structure which vary among these systems will not be crucial for understanding the interlayer coupling. Accordingly, our model includes the bare minimum description of layered ferromagnetic and nonmagnetic transition metals.

Our model has a tight-binding spectrum in the z direction, perpendicular to the layer planes. Motion along the z direction is governed by the Hamiltonian

$$\mathcal{H} = -t \sum_{\langle ij\rangle,\alpha} c^\dagger_{i\alpha} c_{j\alpha} - \sum_{\langle ij\rangle} J_{ij}\mathbf{S}_i \cdot \mathbf{S}_j + \sum_{i,\alpha} \mu_i n_i, \quad (1)$$

where $c_{i\alpha}$ destroys an electron in atomic plane i with spin projection α, the spin operator $\mathbf{S}_i = \frac{1}{2}\sum_{\alpha\beta} c^\dagger_{i\alpha}\boldsymbol{\sigma}_{\alpha\beta} c_{i\beta}$ ($\boldsymbol{\sigma}_{\alpha\beta}$ are the Pauli matrices), and $n_i = c^\dagger_{i\uparrow}c_{i\uparrow} + c^\dagger_{i\downarrow}c_{i\downarrow}$. The spacially varying chemical potential μ_i is chosen to enforce uniform density, $\langle n_i\rangle = 1$.

We emphasize the difference between the one-dimensional case, Eq. (1) treated with one atom per atomic plane, and the *quasi-one-dimensional* case, in which the electrons move freely in the transverse directions. The exchange coupling is sensitive to this difference; for example, the asymptotic envelope of $\mathcal{J}(r)$ decays as $1/r$ in the one-dimensional case, but $1/r^2$ in the *quasi-one-dimensional* case.[11] The magnetic multilayers we are discussing cannot be treated simply as one-dimensional magnetic systems. We have incorporated

motion in the transverse directions by allowing the electrons in each atomic plane to occupy two-dimensional free-electron states with constant density of states $1/W$. Thus the single-particle states in our model are products of the solutions of (1) and plane waves in the x and y directions.

The tight-binding part of (1) combined with the transverse free-electron states describes a metal with a Fermi surface that is roughly spherical when $W \approx 4t$. Increasing W/t makes the Fermi surface more prolate, with a larger cutoff wave vector in the z direction. A typical transition-metal bandwidth is 5 eV, corresponding to a hopping matrix element $t \approx 1$ eV.

We treat the nearest-neighbor exchange term in (1) with the Hartree-Fock approximation, using a self-consistent mean-field magnetization $\mathbf{M}_i = \langle \mathbf{S}_i \rangle$. The second term in (1) then becomes

$$\sum_{\langle ij \rangle} J_{ij} \left(\mathbf{S}_i \cdot \mathbf{M}_j + \mathbf{M}_i \cdot \mathbf{S}_j - \mathbf{M}_i \cdot \mathbf{M}_j \right), \tag{2}$$

with the self-consistency condition $\partial \langle \mathcal{H} \rangle / \partial \mathbf{M}_i = 0$. The phenomenological exchange couplings J_{ij} are permitted three different values between neighboring atomic planes, depending on location in the multilayer. If atomic planes i and j are both magnetic, $J_{ij} \equiv J_M$. If planes i and j are both nonmagnetic, $J_{ij} \equiv J_0$. Finally, if plane i is magnetic and plane j nonmagnetic, then $J_{ij} \equiv J_I$. We use J_M simply to create a stable self-consistent magnetic layer. Inside a magnetic layer our bands are completely rigid Stoner bands, which have saturated magnetization when $J_M \gtrsim t$, and zero magnetization when $J_M \lesssim t$. Apart from this inequality, the magnitude of J_M is irrelevant and our results do not depend on the particular value of J_M. We have set $J_0 = 0$, and we will vary the remaining phenomenological exchange coupling J_I between J_0 and J_M in a crude investigation of the effects of local exchange at the magnetic-nonmagnetic interface. When $J_I = J_M$ the situation is identical to the case when $J_I = J_0$ and one atomic plane of magnetic material has been added.

In our model an indirect exchange coupling between magnetic layers occurs due to hybridization of magnetic and nonmagnetic states at the interfaces. This hybridization occurs via the hopping matrix element t connecting states in the magnetic and nonmagnetic planes and polarizes the nonmagnetic layer, leading to an indirect coupling. By calculating the coupling directly, we gain a useful reference point for evaluating the validity of perturbation theory.

By comparing the total energies of the self-consistent solutions with the magnetic layers aligned either ferromagnetically (FM) or antiferromagnetically (AFM) we obtain $\mathcal{J}(r) \equiv E(\mathrm{FM}) - E(\mathrm{AFM})$ for $r = na$. The inset in Fig. 1 illustrates our magnetic unit cell. There are two magnetic layers composed of m atomic planes each, alternated with two nonmagnetic layers composed of n atomic planes each. We have used values of m ranging from 2 (the minimum number for which a self-consistent magnetic layer is possible) to $m = 5$. Changing the number of magnetic planes does not significantly affect our results. This indicates that the interaction between the magnetic and nonmagnetic layers occurs at the interface between the two, making our coupling independent of the magnetic layer thickness, as observed experimentally.[2]

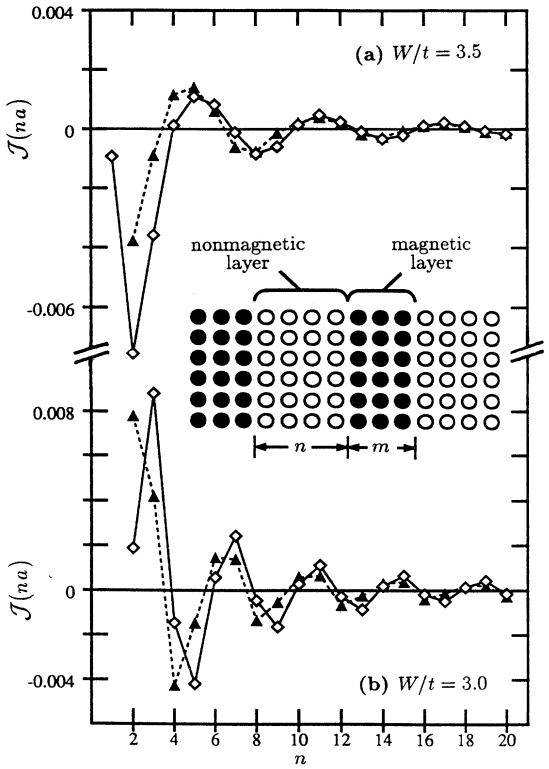

FIG. 1. Indirect exchange coupling function $\mathcal{J}(r)$ between magnetic layers in the unit cell (inset) and calculated nonperturbatively using Hamiltonian (1) in units of t. (a) $W/t = 3.5$, $J_I = 0$ (open diamonds) and $J_I = t/2$ (filled triangles). (b) Same as (a) except $W/t = 3$. Positive values of $\mathcal{J}$ correspond to antiferromagnetic coupling. There are two kinds of layers, coupled by three different near-neighbor phenomenological exchange couplings: black (magnetic) couples to black with J_M, white (nonmagnetic) to white with J_0, and black to white with J_I.

III. INDIRECT EXCHANGE COUPLING

Figure 1 shows $\mathcal{J}(r)$ calculated in our model system. We find that $\mathcal{J}(r)$ oscillates with a period λ significantly larger than π/k_F, where k_F refers specifically to the cutoff wave vector in the z direction. Increasing the ratio W/t increases k_F but leads to a longer period, *opposite* the trend predicted by the RKKY theory. However, the asymptotic decay of the envelope is the same as in the RKKY theory.[11] We find that for $r \gtrsim 10a$,

$$\mathcal{J}(r) \approx At \frac{\sin(2\pi r/\lambda)}{(2k_F r)^2}, \tag{3}$$

where A is a constant of order unity. Referring to Fig. 1, and taking a typical lattice constant to be $a \sim 3$ Å, the magnitude of the coupling at the second peak is typically

$10^{-3}t/a^2 \sim 1$ erg/cm^2, the same order of magnitude as found experimentally.[2]

The small-r limit of $\mathcal{J}(r)$ is worthy of note. We find $\mathcal{J}(a)$ (coupling through one spacer layer) to be ferromagnetic in all cases. As shown in Fig. 1, however, $\mathcal{J}(2a)$ (coupling through two spacer layers) may be either ferromagnetic or antiferromagnetic, depending on the value of W/t. Also, the phase of the oscillation depends on the phenomenological exchange coupling at the magnetic-nonmagnetic layer interface, J_I. In the experiments $\mathcal{J}(r \to 0)$ can be either ferromagnetic (as in Co/Cr), *or* antiferromagnetic (as in Co/Ru).[2] In the limit of a very thin spacer layer, the detailed electronic structure of the interface becomes more important. Slight changes in geometry resulting from interface growth and/or reconstructions affect the magnetic properties tremendously.[12] Although we cannot do justice to these important details in our simple model, the phase shift produced by varying J_I indicates a sensitivity of the coupling in the thin spacer limit to interface-specific details.

IV. COMPARISON WITH RKKY THEORY

One may well ask why our model should give a period so different from that of RKKY. Our toy system couples the ferromagnetic layers by polarizing the nonmagnetic layers; this is precisely the situation addressed by the RKKY calculation. In order to clarify the situation, we have computed the RKKY "range function" $\phi(r)$ for a homogeneous nonmagnetic host with band structure given by the tight-binding part of (1). The range function is proportional to the magnetization induced in the nonmagnetic host by a spin of infinitesimal magnitude localized at the origin. In the RKKY theory the coupling $\mathcal{J}(r)$ is proportional to $\phi(r)$. To obtain $\phi(r)$ one first calculates the zero-temperature generalized magnetic susceptibility

$$\chi(q_z) = 2\mu_B^2 \sum_{\mathbf{k}} \frac{\Theta(\epsilon_{\mathbf{k}}) - \Theta(\epsilon_{\mathbf{k}+q_z\hat{z}})}{\epsilon_{\mathbf{k}+q_z\hat{z}} - \epsilon_{\mathbf{k}}}. \tag{4}$$

Here $\mathbf{k}$ is an index representing crystal momentum. The function $\Theta(x)$ is the unit step function. The range function is then a one-dimensional Fourier transform,[11]

$$\phi(r) = \frac{a}{2\pi} \int dq_z \chi(q_z) \cos(q_z r). \tag{5}$$

This program is simply second-order perturbation theory for a contact interaction between an atomic plane of localized spins and the itinerant electrons in the metallic spacer layer conduction band, which respond via $\chi(q_z)$. High-quality band structures have been used with (4) and (5) to obtain good agreement with experimentally determined couplings in other multilayer systems such as Gd/Y.[13] This success suggests that the polarization induced via hybridization at the interface (as in our model) should be similar to that induced by a contact interaction with a localized spin. The resulting $\phi(r)$ generally does not closely resemble the result for a free-electron gas, but contains features traceable to peaks in $\chi(q_z)$ caused by Fermi-surface nesting effects.[14] However, the

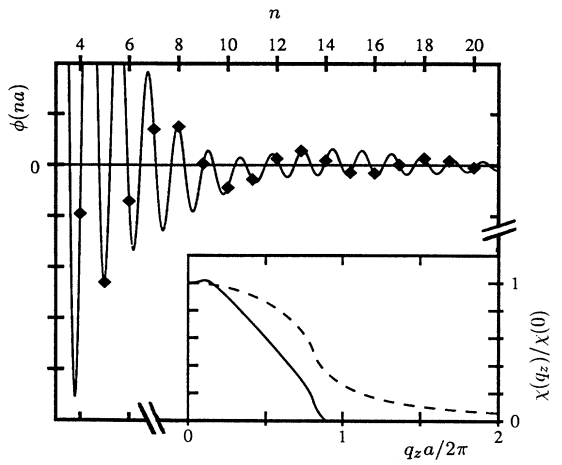

FIG. 2. RKKY "range function" $\phi(r)$ in a homogeneous system with energy given by the tight-binding part of (1), for r in the z direction (arbitrary units). When $\phi(r)$ is sampled at the discrete set of points $r = na$, n an integer, the apparent period is much larger (filled diamonds). Plotted in the inset is the generalized magnetic susceptibility $\chi(q_z)$ from which $\phi(r)$ was obtained (solid line), and the susceptibility of a free-electron gas with the same Fermi cutoff (dashed line).

Fermi surface in the tight-binding part of (1) does *not* have a spanning wave vector other than the full Fermi wave vector k_F in the z direction, or any other "nesting" features which could lead to peaks in $\chi(q_z)$. The range function therefore has a real space period $\lambda = \pi/k_F$. The inset in Fig. 2 shows the generalized susceptibility $\chi(q_z)$ calculated from the tight-binding part of (1) and for comparison, $\chi_{\mathrm{FEG}}(q)$ for a free-electron gas with cutoff wave vector k_F equal to the cutoff in the z direction of our model. The susceptibilities are clearly different, yet the *cutoff* in both occurs at $2k_F$, leading to range functions with the same period. The RKKY calculation indicates a *short-period* oscillation.

In a multilayer system, however, $\mathcal{J}(r)$ is sampled at discrete points by the multilayer structure. This leads to *aliasing*, where short-period components of $\phi(r)$ are shifted to long periods in the discretely sampled $\mathcal{J}(r)$ (see Fig. 2). The period of the aliased $\phi(r)$ matches exactly the period found in our nonperturbative calculation of $\mathcal{J}(r)$. Since the amplitude decay is also the same [Eq. (3)], the RKKY perturbation theory correctly predicts the qualitative features of $\mathcal{J}(r)$ in the strongly coupled model, when aliasing is taken into account.

To further investigate the applicability of perturbation theory, we varied the hopping matrix element between magnetic and nonmagnetic layers, t_I, while keeping the rest of the t's fixed. Empirically, the coupling strength $A \approx (t_I/t)^4$ when $t_I \ll t$, eventually saturating to $A \sim 1$ when $t_I \gtrsim t$. The period and envelope decay of the coupling are accurately described by perturbation theory even in this saturated regime.

The natural next step is to investigate the aliased range functions calculated from a realistic band structure. We believe that the long period in some of the experimen-

tal systems may be the result of aliasing. A perturbative approach is attractive since even for simplified band structures[15] the accuracy with which the total energy may be determined is smaller than $|\mathcal{J}|$. A perturbative calculation has been carried out[16] using an accurate band structure for paramagnetic Cr and yields short-period oscillations which have a period slightly longer than $2d$. They are therefore not subject to aliasing. Instead, an explanation based on interface roughness was employed.

V. CONCLUSIONS

We have presented a calculation of the indirect exchange coupling $\mathcal{J}(r)$ between ferromagnetic layers in a transition-metal multilayer using a simple model. The model incorporates tight-binding bands in both the magnetic and nonmagnetic layers, and a phenomenological exchange interaction in the magnetic layers. We find that $\mathcal{J}(r)$ exhibits a long oscillation period, $\lambda \sim 4$–6 atomic layers, similar to that found experimentally.[2–5] This result may be understood within the context of RKKY theory as a *short-period* RKKY oscillation which is sampled at a uniform set of discrete points, corresponding to the number of atomic planes in a nonmagnetic spacer layer. This aliasing effect must be considered in any theory of exchange-coupled multilayers.

When aliasing occurs, small changes of phase in the (unobserved) short-period oscillations can lead to large changes of phase in the aliased long-period oscillations. Under these circumstances, an RKKY coupling which is *always* ferromagnetic in the small-distance limit can lead to a coupling across *one* spacer layer which is either ferromagnetic or antiferromagnetic. This is consistent with the fact that both behaviors are observed experimentally.

We conclude that RKKY perturbation theory is sufficient to understand the qualitative features of our fully interacting model system. This approach is formally valid only in the limit of thin, weakly ferromagnetic layers which interact with the host conduction electrons via a contact interaction. Although these are not the conditions in our model, perturbation theory is still correct in predicting the real-space asymptotic period (given the aliasing effect) and decay envelope of the exchange coupling. This is encouraging because it suggests that a quantitative perturbation theory may apply to the real materials. We note that indirect exchange couplings in other multilayer systems such as Gd/Y (Ref. 13) have been successfully treated using RKKY perturbation theory, and consider it comforting that this is the case in our model as well.

Note added. After completing this work we received a copy of a paper (presently unpublished) describing a calculation based on aliasing.[17]

ACKNOWLEDGMENTS

We thank S.S.P. Parkin, P. Levy, M. Stiles, R. Celotta, and L.M. Falicov for useful discussions, and J. Mathon for a copy of Ref. 18. Work at Berkeley was supported by the NSF under Grant No. DMR-89-14440. D.S.R. acknowledges a grant from the Alfred P. Sloan Foundation.

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