

Interlayer Magnetic Coupling in Fe/Cr Multilayered Structures

Y. Wang and P. M. Levy

Physics Department, New York University, 4 Washington Place, New York, New York 10003

J. L. Fry

Physics Department, University of Texas at Arlington, Arlington, Texas 76019

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We have calculated the magnetic coupling of two layers of iron through an intervening layer of chromium. We use the mixing interaction between iron atoms at the interfaces with the conduction electrons of chromium and the band structure of paramagnetic chromium. When we take into account the roughness of the interface, our calculated coupling reproduces the essential features of the coupling found in Fe/Cr superlattices.

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Recently, interlayer magnetic coupling in transition-metal multilayer structures, especially iron and chromium (Fe/Cr), has attracted wide attention. For periodic Fe/Cr superlattices,¹⁻³ the Fe layers are coupled predominantly antiferromagnetically. The coupling decreases monotonically as the Cr layer thickness increases from 9 to 18 Å, and then has a slowly oscillating behavior with a period of *about* 10 ML (monolayers) out to 50 Å. Fe/Cr/Fe sandwich structures have also been studied.^{4,5} Although the details of the coupling differ, they give rise to a common question: What is the origin of this relatively long-range *antiferromagnetic* coupling in Fe/Cr multilayered structures?

We propose that the interlayer magnetic coupling between iron layers comes from the mixing or hybridization interaction between iron surface states at the interfaces with the conduction-electron states from chromium. The antiferromagnetic coupling is due to a superexchange-type interaction; the relatively long-range oscillatory coupling comes from intraband transitions, while the rapid oscillations which come from the spin-density-wave (SDW) ordering wave vector are attenuated by the roughness of the interfaces. This result is related to the specific band structures of Cr, and cannot be understood in terms of a single-band model.

To calculate interlayer coupling for a superlattice we consider a film of Cr on top of a magnetized layer of Fe. When another layer of iron atoms is deposited on top of the chromium, depending on the thickness of the intervening Cr layer, the iron magnetic moments are aligned ferromagnetically or antiferromagnetically to the first layer of iron. Subsequent planes of Fe atoms have their moments oriented by the strong magnetic coupling between iron atoms. We simplify our model by assuming the inner Fe planes are magnetically coupled only to the interfacial plane of iron atoms at the Fe/Cr interface; i.e., they are not directly coupled to the Cr conduction electrons. Rather the bulk of the iron layer is *indirectly* coupled to the Cr through its surface states. In this simplified picture the interlayer coupling is the magnetic

coupling of the surface states of one Fe layer to the magnetic disturbances set up in the intervening Cr film by the magnetized surface states of an adjacent Fe layer. Aside from differences in the density of states (DOS) for Fe this is similar to calculating the coupling between two rough planes (monolayers) of iron embedded in bulk chromium which we assume is in the paramagnetic state. This is the picture successfully used by Yafet⁶ to explain the interlayer magnetic coupling in Gd/Y superlattices. The spin-dependent scattering of Cr conduction electrons by the iron moments at the interfaces comes from two sources: the Coulomb exchange interaction and the mixing or hybridization interaction. Whereas the mixing interaction is small for the rare-earth-yttrium layered structures considered by Yafet,⁶ for 3d transition metals it is much larger. In our model we assume the mixing interaction dominates the Coulomb exchange.

The magnetic coupling $J(z)$ between two perfectly flat iron surfaces a distance z apart is given by

$$J(z) = \frac{a}{2\pi} \int_0^\infty j(q_z) \cos(q_z z) dq_z, \quad (1)$$

where a is the lattice constant of bcc Cr, $j(\mathbf{q}) = (1/N) \sum_{\mathbf{R}} e^{i\mathbf{q} \cdot \mathbf{R}} j(\mathbf{R})$, and $j(\mathbf{R})$ is the magnetic coupling between two atoms on the surfaces of the iron layers separated by \mathbf{R} . $J(z)$ is normalized to one pair of iron atoms. As we have two *planes* instead of atoms, we need only $j(q_z)$ to calculate $J(z)$; i.e., the Fourier transform is one dimensional.⁷

When one accounts for the *roughness* of interfaces, the above expression becomes

$$J(z) = \frac{a}{2\pi} \int_0^\infty f^2(q_z) j(q_z) \cos(q_z z) dq_z, \quad (2)$$

where now z should be understood as the *average* distance between two *rough* iron surfaces, and $f(q_z)$ is the structure form factor due to the roughness,

$$f(q_z) = \sum_{m=-\infty}^{\infty} p_m e^{iq_z m a/2}, \quad (3)$$

i.e., the phase factor along the z direction averaged over a rough interface. Here p_m is the probability of finding an iron atom a distance $ma/2$ away from the average position of the iron plane (for bcc lattices the spacing between planes is $a/2$), and $\sum_m p_m = 1$. For simplicity, we choose $p_{-1} = p_1 = p$ and set $p_m = 0$ for all $|m| \geq 2$; then

$$f(q_z) = (1 - 2p) + 2p \cos(q_z a/2). \quad (4)$$

If $p = 0$, $f(q_z) = 1$, there is no surface roughness and we recover Eq. (1). In deriving Eq. (2) we have assumed that there is no correlation between the roughness of two interfaces, but they have the same probability distribution p_m .

When we ignore the dispersion in energy of the surface states of iron, the $j(\mathbf{q})$ due to the mixing interaction is given as⁸

$$j(\mathbf{q}) = - \sum_{n_1, n_2} \sum_{\mathbf{k}} \frac{|V_{n_1 \mathbf{k}}|^2 |V_{n_2 \mathbf{k} + \mathbf{q}}|^2 \theta(\epsilon_{n_1 \mathbf{k}} - \epsilon_F)}{(\epsilon_F - \epsilon_{n_1 \mathbf{k}} - E_h)^2} \times \left\{ \frac{\theta(\epsilon_F - \epsilon_{n_2 \mathbf{k} + \mathbf{q}})}{\epsilon_{n_2 \mathbf{k} + \mathbf{q}} - \epsilon_{n_1 \mathbf{k}}} - \frac{\theta(\epsilon_{n_2 \mathbf{k} + \mathbf{q}} - \epsilon_F)}{\epsilon_F - \epsilon_{n_2 \mathbf{k} + \mathbf{q}} - E_h} \right\}, \quad (5)$$

where n_1, n_2 are band indices for paramagnetic Cr, and E_h is the energy required to promote an electron from an occupied local iron level to the Fermi level. For simplicity, we have assumed $E_h \ll E_e$ (the energy required to absorb an electron from the Fermi level to an unoccupied local level), so that we need only consider the emission process. The sign is chosen so that negative $j(\mathbf{R})$ signifies antiferromagnetic coupling. It is common to take the mixing parameter $V_{n\mathbf{k}}$ to be independent of \mathbf{k} . But for Cr it has been found that the matrix elements

$$M_{n_1 \mathbf{k}, n_2 \mathbf{k} + \mathbf{q}} = \langle n_1 \mathbf{k} | e^{i\mathbf{q} \cdot \mathbf{r}} | n_2 \mathbf{k} + \mathbf{q} \rangle,$$

which represent the atomic form factor, play an important role.⁹ So that our $j(\mathbf{q})$ has the form of $\chi_{Cr}(\mathbf{q})$ for large enough E_h , we take

$$|V_{n_1 \mathbf{k}}|^2 |V_{n_2 \mathbf{k} + \mathbf{q}}|^2 = V^4 |M_{n_1 \mathbf{k}, n_2 \mathbf{k} + \mathbf{q}}|^2.$$

By using Eq. (5) to calculate our coupling we are overlooking the dispersion in the density of surface states. In spite of this oversight, our simple model explains most of the features observed for the interlayer coupling in Fe/Cr superlattices.

We have calculated $j(q_z)$ by using the full band structure of bulk paramagnetic Cr; we used the same energy bands and matrix elements that have been used to calculate $\chi_{Cr}(q_z)$ and have been found to reproduce the SDW instability in Cr.⁹ We have chosen to evaluate Eq. (5) for $E_h = 0.02, 0.04$, and 0.08 Ry, because these values are probably characteristic of the surface states for iron close to the Fermi level [the t_{2g} states further from ϵ_F do not appreciably contribute to $j(q_z)$], and are appropriate for iron impurities in chromium.¹⁰ Our calculation

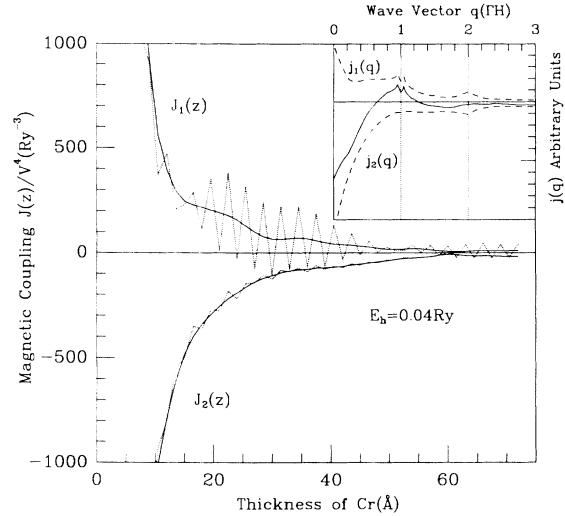


FIG. 1. The contributions to the magnetic coupling Eq. (2) from $J_1(z)$ and $J_2(z)$ for $E_h = 0.04$ Ry, $p = 0$ (dotted line) and $p = \frac{1}{4}$ (solid line). The units of J are rydbergs when V is expressed in rydbergs. Inset: $j_1(q_z)$, $j_2(q_z)$, and the total $j(q_z)$ which were used to calculate $J(z)$. The wave vector q is in units of $\Gamma H (= 2\pi/a)$.

shows that there is no appreciable difference in $J(z)$ whether we cut off $j(q_z)$ (q_z is along the ΓH direction in reciprocal space) at $3\Gamma H$, $4\Gamma H$, or $5\Gamma H$ since it becomes flat and approaches zero at large q_z ; therefore we neglect the contributions beyond the fifth Brillouin zone. In the inset in Fig. 1 we show the behavior of the two terms $j_1(q_z)$ and $j_2(q_z)$ [corresponding to the two terms in the curly brackets in Eq. (5)] as well as $j(q_z)$ for $E_h = 0.04$ Ry; other than a change of scale in magnitude, $E_h = 0.02$ and 0.08 Ry produce similar curves. We note that $j_1(q_z)$ drops off rapidly from $q = 0$ to $0.2\Gamma H$, and then remains flat over a wide range; near the zone edge, i.e., $q_{SDW} \approx 0.95\Gamma H$, the feature that gives rise to a SDW instability is noticeable.

We have calculated the coupling $J(z)$ Eq. (2) for flat ($p = 0$), irregular ($p = \frac{1}{8}$), and rough ($p = \frac{1}{4}$) interfaces. Values of $p \approx \frac{1}{8}$ have been used to explain the contribution of the interfacial roughness scattering to the resistivity of Fe/Cr superlattices.¹¹ In Fig. 1 we show the results for $E_h = 0.04$ Ry broken down into $J_1(z)$ and $J_2(z)$, and in Fig. 2 we show $J(z)$ for $E_h = 0.02, 0.04$, and 0.08 Ry. The $J_1(z)$ curve for $p = 0$ shows rapid oscillations due to the peak in $j(q_{SDW} \approx 0.95\Gamma H)$ corresponding to the SDW ordering wave vector; however, when surface roughness is introduced their amplitude is diminished. For superlattices grown by molecular-beam epitaxy (MBE) data exist primarily for $z < 18$ Å. From the curve $J_1(z)$ in Fig. 1 we note that even for $p = 0$ the amplitudes of the rapid oscillations are small in this range; this is in agreement with the existing data.² For $z > 18$ Å the rapid oscillations in $J_1(z)$ for $p = 0$ persist even for $p = \frac{1}{8}$. However, it is questionable whether they

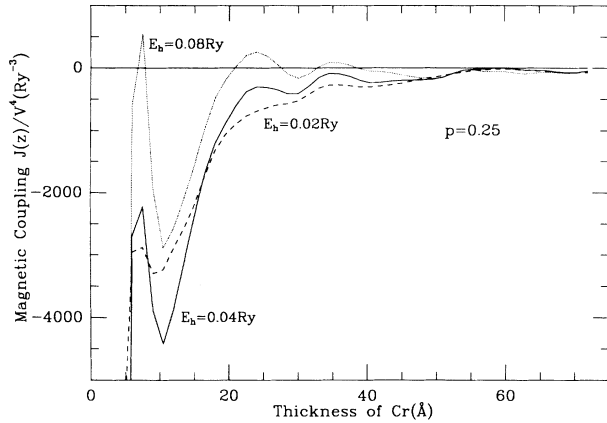


FIG. 2. The coupling $J(z)$ for $p = \frac{1}{4}$ and $E_h = 0.02$ (dashed line), 0.04 (solid line), and 0.08 Ry (dotted line). To accommodate the curves on the same figure the scale for $E_h = 0.04$ Ry has been multiplied by 10 and that for $E_h = 0.08$ Ry by 50.

would show up in MBE-grown *superlattices* because one cannot control the thickness of Cr layers precisely from one period to the next. The effect of variations of layer thickness is the same as the roughness of the interfaces; they also attenuate the rapid oscillations of the coupling. On the other hand, MBE-grown Fe/Cr/Fe *sandwich* structures do not suffer from these variations in layer thickness; the scatters found in the coupling for these structures⁵ might be interpreted as coming from the rapid oscillations present for smoother interfaces.

In the case of sputtered samples the interfaces are rough, and the curves for $p = 0.25$, see Fig. 2, have a slowly oscillating coupling which bears a remarkable resemblance to the experimental curves for the saturation field obtained by Parkin, More, and Roche.^{3,12} This slow oscillation comes from the contributions to $J_1(z)$ due to an intraband transition; it also corresponds to the turning point in $j_1(q_z)$ at $q_z \approx 0.2\Gamma H$. As $q_z = 0.2\Gamma H$ in real space corresponds to 5 lattice constants or 10 ML of Cr (about 15 Å), this is quite close to the period of the oscillations found by Parkin, More, and Roche.³ In comparing our results to data it is important to emphasize that our model calculation does not hold for small z , because the properties of ultrathin layers of Cr are strongly perturbed by the surrounding Fe layers,¹³ and cannot be treated as bulk Cr.

Generally speaking, as p increases the amplitude of the rapid oscillations decreases (see Fig. 1); the curves for the coupling become smoother and the slowly varying parts are more noticeable. Here we have used a single parameter p to describe the interface roughness; see Eq. (3). In reality, the interface roughness is not necessarily limited to the nearest-neighbor layers, $m = \pm 1$. In that case the interfacial transition region is wider, and more values of m [see Eq. (3)], i.e., more chromium layers, are sampled in calculating the interfacial magnetic coupling. This would more effectively reduce the rapid oscillations;

the results would be similar to our nearest-neighbor ($m = \pm 1$) single- p description, but with smaller values of p_m . Finally, to estimate the effect of including the dispersion in energy for the surface states of iron one can take a composite (average) of our results in Fig. 2 for the different values of E_h .

The antiferromagnetic coupling comes from j_2 . For a single free-electron-like conduction band j_1 is dominant, and by replacing the energy-dependent denominator by a constant one recovers a Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction.^{8,14} However, if the conduction-electron band is *empty*, j_1 vanishes and j_2 gives an exponentially decaying antiferromagnetic coupling; this mechanism is reminiscent of and therefore called "superexchange." For realistic band structures, e.g., for Cr, there exists a competition between the RKKY-type and superexchange-type interactions. In our calculation of $J(z)$, as E_h decreases the superexchange-type interaction decays more slowly and $J(z)$ becomes increasingly antiferromagnetic. Also the small DOS at the Fermi surface of Cr helps to reduce the RKKY-type interaction and makes the coupling go antiferromagnetic.

The adjustable parameters in our calculation are E_h , p , and V . While the first two determine the characteristics of the coupling, i.e., its strong antiferromagneticlike behavior and the slow oscillations, the mixing parameter V fixes the magnitude of the coupling. When we use $V = 0.3$ eV for $E_h = 0.04$ Ry, we find $J(z = 6 \text{ ML}) = 1.3$ meV, which is in good agreement with the data of Barthelemy *et al.*²

Our model is applicable to other systems. For example, if Co/Cr forms a bcc structure, we predict results similar to Fe/Cr, in particular the same periodicity of oscillations for the coupling; the magnitude of the coupling is different because E_h , see Eq. (5), is changed. This is in agreement with the results obtained by Parkin, More, and Roche.³ For Co/Ru the DOS of Ru at the Fermi surface is small as it is for Cr. Therefore the antiferromagnetic coupling $J_2(z)$ can prevail over the RKKY $J_1(z)$ for a range of Ru thickness as is also found experimentally by Parkin, More, and Roche.³ However, the period of the RKKY oscillations for Co/Ru will be different because the topology of the Fermi surface for hcp Ru is indubitably different from that for bcc Cr.

In summary, our model calculation has been able to reproduce the two salient features of the interlayer coupling in Fe/Cr superlattices: large but rapidly decreasing antiferromagnetism in the range 6–12 ML (9–18 Å) followed by oscillations out to at least 33 ML (50 Å). In our model the rapid oscillations, induced by the SDW ordering wave vector, are attenuated by the roughness of the interfaces.

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