Spin Polarization of Epitaxial Cr on Fe(001) and Interlayer Magnetic Coupling in Fe/Cr Multilayered Structures

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Interlayer coupling in metallic multilayers is generally thought to be akin to the RKKY coupling of magnetic moments through conduction electrons. By analyzing recent experimental data on the spin polarization of chromium on iron substrates we confirm that the RKKY coupling cannot by itself explain the interlayer coupling in Fe/Cr multilayers. An additional coupling is warranted by the data, and we explain its origin.

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The layer by layer spin polarization in epitaxial Cr on Fe(001) has been observed below [1] and well above [2] the bulk Cr Néel temperature, T_N. Both of these direct observations are remarkable experimental achievements, but the existence of a spin-density-wave- (SDW-) like polarization in Cr for temperatures up to $1.8T_N$ deserves special attention. Below T_N it may be assumed that the SDW is spontaneous, with its wave vector possibly modified by epitaxial strain, and a function of temperature as it is in the bulk. The appearance of a SDW-like polarization above T_N may be explained either as an enhanced T_N due to the presence of the Fe substrate, or it may be simply a polarization wave *induced* by the Fe substrate. Since experiment suggests that the SDW has a decaying amplitude [2], we take the latter point of view here, and present the *first* calculation of the magnetization in paramagnetic Cr layers on Fe(001).

In addition we demonstrate that the interlayer coupling arising from this induced or spontaneous magnetization (the RKKY coupling) is not able by itself to account for the interlayer coupling observed in Fe/Cr multilayered structures [3,4]. While the oscillations observed in the interlayer coupling are reproduced by this RKKY mechanism, the sign and strength of the coupling are not. This provides an experimental confirmation that it is necessary to include an antiferromagnetic superexchange interaction [5, 6] to understand the interlayer coupling in Fe/Cr multilayers. As we show, this term is contained in the coupling derived by Caroli [7] between magnetic impurities in metals.

For chromium on an iron substrate it is the localized moments of the iron layer at the interface that polarizes the conduction electrons of chromium. We describe this interaction by the *s-d* mixing Hamiltonian [8]. The change in the electron density of spin σ in the paramagnetic Cr layer due to the mixing interaction is [9]

$$\delta n^{\sigma}(\mathbf{r}) = \sum_{n_1 k, n_2 k'} \left(\phi^*_{n_2 k'}(\mathbf{r}) \phi_{n_1 k}(\mathbf{r}) \int_{-\infty}^{\varepsilon_F} \frac{d\varepsilon}{2\pi i} \frac{V_{n_1 k}\, V^*_{n_2 k'}}{(\varepsilon - \varepsilon_{n_1 k})(\varepsilon - \varepsilon_{n_2 k'})[\varepsilon - \varepsilon^0_{d\sigma} - \Sigma_d(\varepsilon)]} + \text{c.c.} \right) , \tag{1}$$

where V_{nk} is the strength of the mixing interaction, ε_F is the Fermi energy, and $\Sigma_d(\varepsilon)$ is the self-energy due to the mixing interaction. After taking the contour integration in this equation, we find the Fourier transform of the spin polarization induced by a magnetic moment whose magnitude is fixed as

$$m(\mathbf{q}) = \frac{1}{2}[\delta n^{\uparrow}(\mathbf{q}) - \delta n^{\downarrow}(\mathbf{q})] = \sum_{n_1, n_2, k} \left(\frac{V_{n_1 k}\, V^*_{n_2 k'}\, M_{n_1 k, n_2 k'}(\mathbf{q})}{\varepsilon_{n_2 k'} - \varepsilon_+} \frac{\theta(\varepsilon_{n_2 k'} - \varepsilon_F) \times \theta(\varepsilon_F - \varepsilon_{n_1 k})}{\varepsilon_{n_2 k'} - \varepsilon_{n_1 k}} + \text{c.c.} \right) , \tag{2}$$

where θ is a step function, $\mathbf{k}' = \mathbf{k} + \mathbf{q} + \mathbf{G}$, $\mathbf{G}$ is a vector of the reciprocal lattice used to keep $\mathbf{k}'$ inside the first Brillouin zone, $M_{n_1 k, n_2 k'}(\mathbf{q}) \equiv \langle n_1 \mathbf{k} \mid e^{-i\mathbf{q}\cdot\mathbf{r}} \mid n_2 \mathbf{k}' \rangle$ is an atomic form factor matrix element [10], and $\varepsilon_+ \equiv \varepsilon^0_{d\downarrow} + \Sigma_d(\varepsilon)$ is the local level below ε_F; local levels above ε_F have been neglected because we are taking the limit of a large intra-atomic Coulomb interaction. We set $\mathrm{Re}[\varepsilon_+] = \varepsilon_F - E_h$ and $\mathrm{Im}[\varepsilon_+] = \Delta$, where E_h is the energy required to promote an electron from an occupied local magnetic impurity level to the Fermi level [11].

Until now we have considered one local moment in a sea of conduction electrons. For a *sheet* of local moments we must add the contributions from each moment.

Equivalently, Yafet [12] has shown that the spin polarization induced at a distance z from a sheet of spins (this simulates the interface between magnetic and paramagnetic layers) is the *one*-dimensional Fourier transform of Eq. (2). After considering the roughness of the interface and that $m(\mathbf{q})$ is complex, we obtain

$$m(z) = \frac{a}{2\pi}\int_0^\infty f_1(q_z)\, f_2(q_z)\mid m(q_z)\mid \times \cos[q_z z + \varphi_m(q_z)]\, dq_z\,, \qquad (3)$$

where

$$\mid m(q_z)\mid = \sqrt{\{\mathrm{Re}[m(q_z)]\}^2 + \{\mathrm{Im}[m(q_z)]\}^2}\,,$$

$$\varphi_m(q_z) = \tan^{-1}\{\mathrm{Im}[m(q_z)]/\mathrm{Re}[m(q_z)]\}\,,$$

a is the lattice constant, and we have introduced *two* structure form factors [6]: one to represent the roughness of the interface between the ferromagnetic substrate and the paramagnetic overlayer $f_1(q_z)$; and a second one for the roughness of the free surface of this overlayer $f_2(q_z)$. As the latter does not suffer from interdiffusion, it is reasonable to assume that the free surface is smoother than the interface and we will take $f_2(q_z) = 1$.

Away from the interface the induced spin polarization is given by the susceptibility $\chi(\mathbf{q})$, i.e., $m(\mathbf{q}) \sim \chi(\mathbf{q})$. Therefore for small $\mathbf{q}$ we make the *Ansatz* $V_{n_1k}V^*_{n_2k'} = V^2 M^*_{n_1k,n_2k'}(\mathbf{q})$. In addition, as the *s-d* mixing is local or point-contact-like, we set $V_{n_1k}V^*_{n_2k'} = \alpha V^2$ for large $\mathbf{q}$, where $\alpha << 1$ is a constant for the s and p parts of the atomic form factor. Precisely, these *Ansätze* were made in our calculation of the interlayer coupling in Fe/Cr [6].

In Fig. 1(a) we show our results on the magnetization for smooth ($p = 0$) and irregular (not very rough, $p = 1/8$) interfaces for $E_h = 0.08$ Ry [11] and $\Delta = 0$, i.e., for a narrow virtual bound state (vbs); p is an interface roughness parameter [6]. In Fig. 1(b) we consider the effects of the finite width of the vbs on the magnetization. On comparing these with the recent data [1, 2] we find a reasonable semblance. As our present calculation has not introduced the exchange enhancement in paramagnetic chromium it is not realistic to compare the magnitude of our predicted induced spin polarization with the data. Otherwise the differences between our results and the data are as follows: (1) Nodes (phase slips in the language of Ref. [2]) occur at $\frac{n}{2}l_S$ ($n = 1, 3, 5, \ldots$), where l_S is the distance between nodes; we find $l_S = 24$ ML (monolayers) whereas experimentally it is 20 ML [2]. (2) Our first node occurs in the region around 11–12 ML; this is consistent with the *apparent* phase slip at 10–11 ML observed by Walker *et al.* [1], but does not agree with the first phase slip observed at 4–5 ML by Unguris, Celotta, and Pierce [2]. It is not clear whether this is intrinsic to Cr or an artifact of how it is deposited on the iron substrate. The discrepancy in l_S may come from the strain in the chromium overlayer [13]. While the vbs *may* generate a shift in phase *if* the spectrum $m(q_z)$ has the form $m(q_z) = m(q_{\mathrm{SDW}})\,\delta(q_z - q_{\mathrm{SDW}})$, we do not find much shifting of the nodes or phase slip regions; see Fig. 1(b) [14]. Rather we find the main effects of the finite width of the vbs are (1) to *attenuate* the amplitude of the induced oscillation of the spin density, and (2) to *diminish* the ferromagnetic bias in the induced magnetization for z or t_{Cr} less than 15 ML. Therefore we conclude that for chromium the finite width of the iron vbs does not produce discernible effects such as phase shifts in the asymptotic region (large z or t_{Cr}); rather its effects are limited to the *preasymptotic* region.

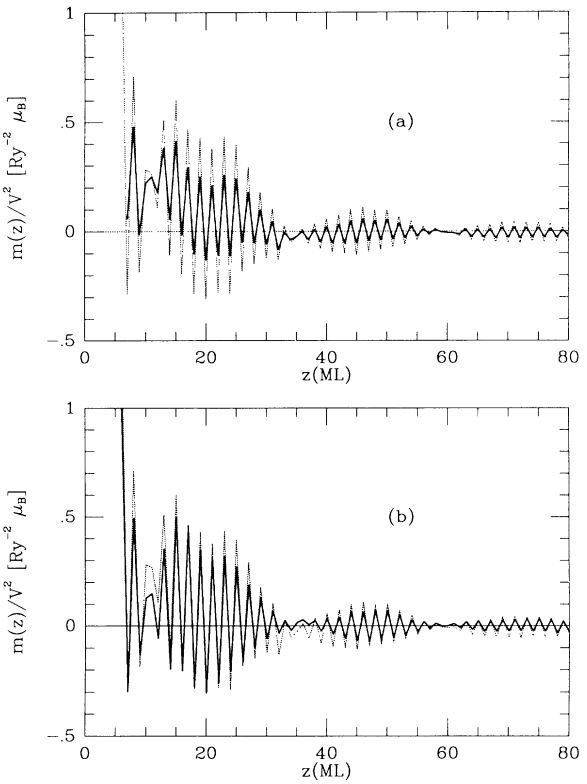

FIG. 1. (a) The spin polarization $m(z)$ in Cr on Fe(001) calculated with $E_h = 0.08$ Ry. The dotted line is for a smooth ($p = 0$) interface and the solid line for an irregular one ($p = 1/8$). V is expressed in rydbergs. (b) Comparison of spin polarization for local state with no broadening and a vbs with $\Delta = 0.04$ Ry for $E_h = 0.08$ Ry and a smooth interface ($p = 0$).

The coupling arising from the interaction of a magnetic ion with the spin polarization of the conduction electron induced by another ion, i.e., the RKKY coupling, can be expressed in reciprocal space at zero temperature as

$$j_{\mathrm{RKKY}}(\mathbf{q}) = \sum_{n_1,n_2,k}\left(\frac{\mid V_{n_1k}\mid^2 \mid V_{n_2k'}\mid^2}{(\varepsilon_{n_2k'} - \varepsilon_+)^2}\,\frac{\theta(\varepsilon_F - \varepsilon_{n_1k})\theta(\varepsilon_{n_2k'} - \varepsilon_F)}{\varepsilon_{n_2k'} - \varepsilon_{n_1k}} + \mathrm{c.c.}\right). \qquad (4)$$

According to Yafet's argument [12], the RKKY coupling $J_{\mathrm{RKKY}}(z)$ between different sheets of magnetic ions can be obtained directly from the *one*-dimensional Fourier transform of Eq. (4), as was done for the magnetization Eq. (3). However, now $f_2(q_z)$ represents the roughness of interface between the iron overlayer and the chromium. This coupling is precisely the term $J_1(z)$ we previously found in our calculation of the interlayer coupling in Fe/Cr [6] where we set $\Delta = 0$. We have calculated J_{RKKY} by using the same parameters as for the induced spin polarization and show our results in Fig. 2(a) for $p_1 = p_2 = 0$ and 1/8, i.e., we have assumed, lacking more precise information, that both interfaces are equally rough (smooth). This coupling does *not* resemble the Fe/Cr interlayer coupling observed by Demokritov *et al.* [3] or by Unguris, Celotta, and Pierce and Purcell *et al.* [4]. When we consider effects due to the finite width of the vbs it does not help the J_{RKKY} coupling look more like the coupling found from experiment [3, 4]. The best the width of the vbs can do is make J_{RKKY} oscillate about zero; a realistic finite width $\Delta = 0.04$ Ry, [see Fig. 2(b)], *reduces* the ferromagnetic bias for small z; however, it *cannot* reproduce the strong antiferromagnetic interlayer magnetic coupling seen in data for thin chromium layer thicknesses (4–10 ML) [3, 4]. Thus *we conclude that the RKKY coupling by itself cannot explain the interlayer coupling in Fe/Cr multilayers* . We have not included the exchange enhancement present in Cr [14]; while this controls some features of the magnetization and coupling, it does not alter our conclusion about the need for a coupling in addition to the one coming from the RKKY mechanism.

By using the Hartree-Fock approximation and assuming that ion-ion interactions have negligible effects on the occupation of local levels, Caroli [7] showed the magnetic interaction between two impurities based on the *s-d* mixing interaction is given by

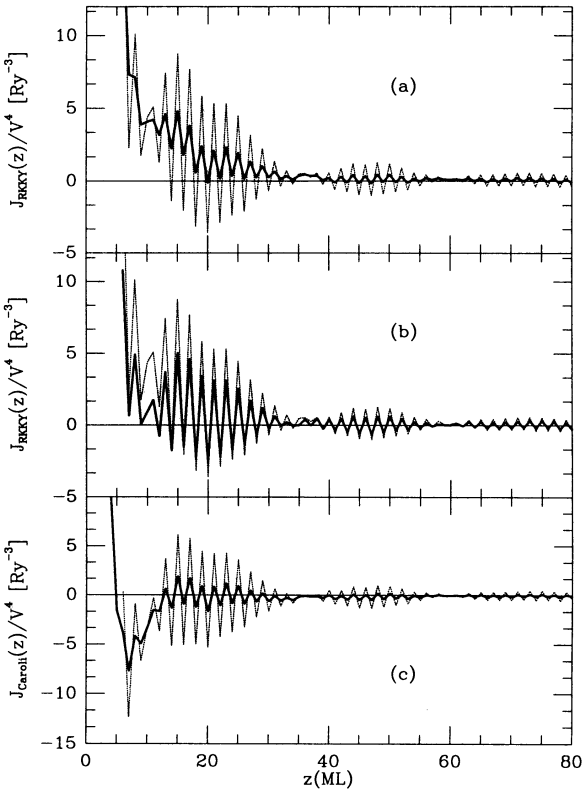

FIG. 2. (a) The RKKY-like interlayer coupling $J_{\mathrm{RKKY}}(z)$ of Fe/Cr(001) multilayers calculated with $E_h = 0.08$ Ry, and $p = 0$ (dotted line) for smooth interfaces and $p = 1/8$ (solid line) for slightly rough interfaces. The units of coupling J are rydbergs where V is expressed in rydbergs. (b) Comparison of RKKY interlayer coupling for local state with no broadening and a vbs with $\Delta = 0.04$ Ry for $E_h = 0.08$ Ry and a smooth interface ($p = 0$). (c) The total interlayer coupling $J_{\mathrm{Caroli}}(z)$ of Fe/Cr(001) multilayers calculated with $E_h = 0.08$ Ry, and $p = 0$ (dotted line) for smooth interfaces and $p = 1/8$ (solid line) for slightly rough interfaces. Here positive and negative J represent ferromagnetic and antiferromagnetic coupling, respectively.

$$J_{\mathrm{Caroli}}(\mathbf{r}) \simeq -\frac{1}{\pi}\,\mathrm{Im}\sum_{\sigma\sigma'}\sum_{n_1k,n_2k'}\sigma\sigma'\int_{-\infty}^{\varepsilon_F} d\varepsilon\,\frac{|\,V_{n_1k}\,|^2\,|\,V_{n_2k'}\,|^2\;e^{i(\mathbf{k}'-\mathbf{k})\cdot\mathbf{r}}}{(\varepsilon-\varepsilon_1^{\sigma})(\varepsilon-\varepsilon_{n_1k})(\varepsilon-\varepsilon_{n_2k'})(\varepsilon-\varepsilon_2^{\sigma'})}\,. \tag{5}$$

The contour integration that picks up poles related to the conduction band at $\varepsilon = \varepsilon_{n_1k}$ or $\varepsilon = \varepsilon_{n_2k'}$ gives the RKKY-like coupling whose form in reciprocal space is Eq. (4). The additional contribution from the pole related to *local states* at $\varepsilon = \varepsilon_1^{\sigma} = \varepsilon_2^{\sigma'} = \varepsilon_+$ gives the antiferromagnetic coupling [6] whose Fourier transform is

$$j_2(\mathbf{q}) = -\sum_{n_1,n_2,k}\left(\frac{|\,V_{n_1k}\,|^2\,|\,V_{n_2k'}\,|^2}{(\varepsilon_{n_2k'}-\varepsilon_+)^2}\,\frac{\theta(\varepsilon_{n_1k}-\varepsilon_F)\theta(\varepsilon_{n_2k'}-\varepsilon_F)}{\varepsilon_{n_1k}-\varepsilon_+} + \mathrm{c.c.}\right). \tag{6}$$

This term arises from *virtual charge excitations* and provides a second contribution to the Fe/Cr coupling, $J_2(z)$, that is in addition to the RKKY coupling [6]. This coupling comes from intermediate states which have electrons from the local iron *d* states above the Fermi sea (one from *each* iron layer). These *virtual charge excitations* of the Fermi sea [5, 15] do not enter in the description of the RKKY coupling. This latter coupling comes only from intermediate states which correspond to *spin excitations* of the Fermi sea, that is, states which correspond to electron-hole pair production in the Fermi sea with an attendant spin flip [5, 15]. The total coupling $J_{\mathrm{Caroli}}(z)$ shown in Fig. 2(c), for smooth interfaces ($p = 0$) and for slightly rough interfaces ($p = 1/8$), resembles the experimental pattern [3, 4]. The

local level $E_h = 0.08$ Ry has been chosen to produce the best fit to the data [3, 11].

To obtain the strength of the interlayer coupling in magnetic multilayers total-energy calculations are indubitably necessary. On the other hand perturbational methods of calculating this coupling provide insight as to the origin of this coupling in transition-metal multilayered structures. Here we have shown that such calculations allow us to determine what aspects of the coupling are related to the Fermi surface (RKKY-like coupling), and what parts come from states away from the Fermi surface [$J_2(z)$]. It is the denominator $(\varepsilon_{n_2k'} - \varepsilon_{n_1k})^{-1}$ in Eq. (4) at the extremal dimensions of the Fermi surface which generates the structure and peaks in $j_{\mathrm{RKKY}}(q)$, and thereby produces the *oscillations* observed in the Fe/Cr interlayer coupling. For Fe/Cr we find $J_2(z)$ provides an additional antiferromagnetic coupling which is *not* tied to the Fermi surface. This determines the sign of Fe/Cr interlayer coupling, i.e., that it is strongly antiferromagnetic in the region $4 \leq z \leq 10$ ML. When one considers sharp local states $J_2(z)$ is monatomic; the finite width of the local levels produces slight oscillations in the strongly antiferromagnetic coupling.

This additional coupling is present for systems where the dominant origin of the coupling comes from the *s-d* mixing of local and conduction electron states which yields the *virtual charge excitation* processes; it is *not* present if the local electron–conduction electron coupling is primarily due to the Coulomb exchange interaction. This latter interaction produces only *spin excitations* of the conduction electrons, and yields only RKKY coupling. For those transition-metal layered structures where the RKKY interaction *seems* to explain the interlayer coupling, one should not rush to conclude that the *s-d* mixing is inoperative; rather we have found that for systems with parabolic conduction bands at the Fermi surface, $J_2(z)$ cancels the ferromagnetic bias in the $J_{\mathrm{RKKY}}(z)$, so as to produce a coupling that oscillates around zero [16]. Therefore with parabolic bands at the Fermi surface the interlayer coupling coming from *s-d* mixing *looks like* that coming from the Coulomb exchange interaction; however, its phase and magnitude may be different [17]. For nonparabolic bands the *s-d* mixing produces the antiferromagnetic coupling observed in several multilayered systems.

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