Oscillating exchange and spin wave stiffness in magnetic monolayers

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Spin waves in a ferromagnetic monolayer are investigated taking into account indirect exchange [Ruderman-Kittel-Kasuya-Yosida (RKKY)] interaction mediated by electrons of a nonmagnetic metal substrate. Strong anomalies are found in the spin wave stiffness when spin lattice-line distances are close to the RKKY wavelength. Conditions for such coincidence and its relevance in epitaxial magnetic layers on fcc metals are discussed taking into account realistic Fermi surface shapes, which further enhance the magnitude of the predicted effect but may change its sign. [S0163-1829(99)03402-5]

INTRODUCTION

It has been known for decades that high quality ultrathin magnetic films grown on metallic substrates retain ferromagnetic order at finite temperatures, contrary to earlier predictions of spin wave theory;[1] among factors explaining this apparent paradox are magnetic anisotropy and long-range interactions of magnetic dipolar[2] and magnetoelastic[3] origin. It has also been suspected that the proximity of a metal surface influences the exchange interaction; this aspect has recently been investigated from first principles.[4,5] We examine an intuitively simple picture of such a ''proximity effect,'' i.e., the ''indirect'' [Ruderman-Kittel-Kasuya-Yosida (RKKY)] exchange[6] mediated by the induced oscillatory polarization of the surrounding nonmagnetic metal. This is of course inspired by the success of the RKKY picture in semiquantitative interpretations of interlayer exchange effects.[7,8] Our interest was strengthened by curiosity about whether the long range of RKKY exchange can lead to substantial modification of spin wave dispersion analogous to that observed when including magnetic dipolar effects.[2]

We investigate simple models of a magnetic monolayer embedded in a metallic matrix. Wishing to look at excitations of the ordered state, we assume that the ''indirect'' oscillating exchange is superposed on stronger ''direct'' nearest-neighbor exchange, in qualitative accordance with the first-principles results.[4,5]

The model spin Hamiltonian is

$$\hat{H}=-\sum_{\mathbf{r},\mathbf{a}} J_d \mathbf{S}_{\mathbf{r}}\cdot \mathbf{S}_{\mathbf{r}+\mathbf{a}}-\sum_{\mathbf{r},\mathbf{r}'} J_{\rm osc}(\mathbf{r}-\mathbf{r}')\mathbf{S}_{\mathbf{r}}\cdot \mathbf{S}_{\mathbf{r}+\mathbf{r}'}, \qquad (1)$$

where $\mathbf{r},\mathbf{r}'$ denote the nodes of the 2D spin lattice, $\mathbf{a}$ denote the nearest-neighbor vectors, $\mathbf{S}_{\mathbf{r}}$ is the operator of spin localized at site $\mathbf{r}$, and $J_d, J_{\rm osc}$ are exchange integrals describing ''direct'' (nearest-neighbor) and oscillating (long-range) interactions.

We deal first with the classical (free-electron) RKKY model which shows some interesting analytic properties in two dimensions. Applicability of the results is then discussed in terms of the anisotropic model[9] including real Fermi surface shapes, with particular attention to the experimentally well-known cases of noble metal matrices.

THE FREE-ELECTRON MODEL

The RKKY (free-electron) approximation[6] for $J_{\rm osc}(r)$ is written as

$$J_{\rm osc}(r)=J_0F(r), \qquad (2a)$$

$$F(r)=[\sin(2k_Fr)/2k_Fr-\cos(2k_Fr)](a/r)^3, \qquad (2b)$$

$$J_0=\frac{2(A/a^3)^2}{\varepsilon_F}\left(\frac{ak_F}{4\pi}\right)^3, \qquad (2c)$$

where k_F is the Fermi wave vector, $\varepsilon_F=\hbar^2k_F^2/2m$ is the Fermi energy, $a=|\mathbf{a}|$, and A is the strength of the primary interaction between the localized and conduction electron spins, $\mathbf{s}(\mathbf{R})$: $\hat{H}_1=A\,\delta(\mathbf{R}-\mathbf{r})\mathbf{s}(\mathbf{R})\cdot\mathbf{S}_{\mathbf{r}}$.

Assuming that the localized spins are ferromagnetically ordered due to sufficiently strong direct exchange, we obtain the magnon energy $\varepsilon(k)$ from discrete Fourier transforms of the exchange integrals,[10] with two contributions $\varepsilon(k)=\varepsilon_d(k)+\varepsilon_{\rm osc}(k)$,

$$\varepsilon_d(k)=2SJ_d\sum_{\mathbf{a}}(1-\cos\mathbf{k}\cdot\mathbf{a}), \qquad (3a)$$

$$\varepsilon_{\rm osc}(k)=2SJ_0\sum_{\mathbf{r}}F(r)[1-\cos(\mathbf{k}\cdot\mathbf{r})]. \qquad (3b)$$

For long waves, $ka\ll 1$, Eq. (3a) yields the parabolic law $\varepsilon_d\cong D_dk^2$ with $D_d=(1/2)NSJ_da^2$; N is the number of nearest neighbors. The behavior of $\varepsilon_{\rm osc}(k)$ of course strongly depends on the RKKY oscillation wavelength $\Lambda=\pi/k_F$, i.e., on the product k_Fa.

Figure 1 shows the numerical results for the long-wave approximation to Eq. (3b), $D_{\rm osc}=(1/2)\partial^2\varepsilon_{\rm osc}/\partial k^2$ as a function of the product k_Fa, for a square lattice. Also shown is the RKKY contribution to the mean-field energy in the uniform state $\overline{E}_{\rm osc}=-S^2J_0\Sigma_{r\neq 0}F(r)$. An analogous figure with

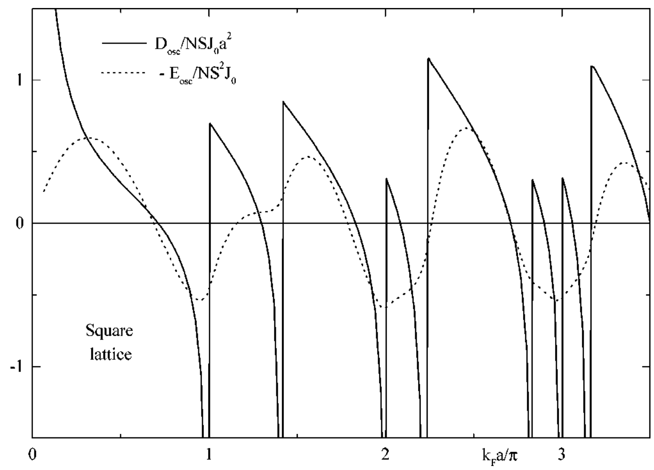

FIG. 1. RKKY contribution to spin wave stiffness $D_{\rm osc}$ and to the mean-field energy in the uniform state $\overline{E}_{\rm osc}$.

the same vertical scales is obtained for the triangular lattice; if the horizontal scale is set as $(k_F a/\pi)\sqrt{3}/2$, the peaks are again at rational positions, as explained further below.

The mean field is a smooth function of the Fermi wave vector or of the spin-lattice constant, oscillating with an amplitude about NS^2J_0. Thus for a 2D spin system with direct and oscillatory exchange, the mean field alone would predict ferromagnetic order for any $k_F a$ as long as (roughly) $J_d > J_0$. However, the RKKY contribution to the exchange stiffness $D_{\rm osc}$ shows deep negative peaks, in which the effective total $D = D_d + D_{\rm osc}$ may well be negative, indicating instability, even if the total mean field favors parallel order.

The peaks are, naturally, due to coherent interference of the RKKY wave with the spin lattice. Their positions and shapes may be analyzed using the integral 2D Fourier transform of Eq. (2b)

$$J_{\rm osc}(\mathbf{r}) = \frac{1}{4} J_0 a^3 k_F \int \Phi(\mathbf{q}/2k_F)\exp(i\mathbf{q}\cdot\mathbf{r})d^2\mathbf{q}, \tag{4a}$$

$$\Phi(x) = 1 - \frac{1}{2}x^2 \quad \text{for } x<1$$
$$= \frac{1}{\pi}\left[(2-x^2)\arcsin\frac{1}{x} + \sqrt{x^2-1}\right] \quad \text{for } x>1. \tag{4b}$$

Then Eq. (3b) may be turned into a summation over the reciprocal spin-lattice vectors $\mathbf{G}_{ij}$:

$$\varepsilon_{\rm osc}(\mathbf{k}) = -SJ_0\pi^2 k_F a\alpha^{-1}\sum_{i,j}\left[\Phi\left(\frac{\mathbf{G}_{ij}+\mathbf{k}}{2k_F}\right) + \Phi\left(\frac{\mathbf{G}_{ij}-\mathbf{k}}{2k_F}\right) - 2\Phi\left(\frac{\mathbf{G}_{ij}}{2k_F}\right)\right], \tag{5}$$

$\alpha = 1(\sqrt{3}/2)$ for a square (triangular) lattice is the area per spin in units of a^2. Expansion in small k and average over the $(\mathbf{G}_{ij},\mathbf{k})$ angle gives, with $x_{ij} = G_{ij}/2k_F$,

$$D_{\rm osc} = -SJ_0 a^2 \frac{\pi^2}{8\alpha k_F a}\sum_{i,j}\,[\Phi''(x_{ij}) + \Phi'(x_{ij})/x_{ij}]. \tag{6}$$

Since $\Phi''(x)$ is positively divergent at $x = 1+$, negative peaks in $D_{\rm osc}$ occur when $2k_F = G_{ij}$, i.e., when a spin-lattice line distance is equal to (or a multiple of) the RKKY wavelength $\Lambda = \pi/k_F$. The absolute values of G_{ij} are $(2\pi/a)(i^2+j^2)^{1/2}$ in a square lattice and $(4\pi/a\sqrt{3})(i^2+j^2-ij)^{1/2}$ in a triangular lattice. Near the first singularity we get, expanding Φ'' for $2k_F = (1-\delta)G_{10}$, $|\delta| \ll 1$,

$$D_{\rm osc} \cong -(1/2)NSJ_0 a^2(2\delta)^{-1/2} \quad \text{for } 2k_F < G_{10} \tag{7a}$$

$$\cong (\pi/4)NSJ_0a^2 \quad \text{for } 2k_F > G_{10} \tag{7b}$$

for the contributions of the $G_{\{10\}}$ set, in both lattice types, in accord with the full result (Fig. 1).

Divergence of the stiffness parameter $D_{\rm osc}$ at the singular points means that the dispersion law must be nonquadratic. Indeed, expanding Eq. (6) at $2k_F = G_{10}$, with $\Phi(1+\beta) \cong 1/2 - \beta + (11\sqrt{2}/4\pi)\beta^{3/2}$ for $\beta > 0$, we derive a ''3/2 law''

$$\varepsilon_{\rm osc}(k) = -cNSJ_0(ak)^{3/2}, \tag{8}$$

where $c \cong 1$ is actually slightly anisotropic in both lattice types.

The asymmetry of the peaks in $D_{\rm osc}$ seen in Fig. 1 and in Eqs. (7a),(7b) is a special feature of the RKKY problem in two-dimensional (2D) spin lattices. It may be alternatively analyzed in the direct summation (3b) (as shown in the Appendix), and it is also present in the case of a strictly 2D problem in which both the spin lattice and the electron gas are two-dimensional. The RKKY exchange is then described by Eq. (4a) with differently scaled J_0 and $\Phi(x)$ replaced by the 2D kernel [see Ref. 11 for the explicit $J_{\rm osc}(r)$ formula]

$$\Phi_2(x) = 1 \quad \text{for } x<1$$
$$= 1 - \sqrt{1 - \frac{1}{x^2}} \quad \text{for } x>1. \tag{9}$$

It is easy to see that Eq. (6) now gives $D_{\rm osc} < 0$ everywhere (since $\Phi_2'' + \Phi_2'/x$ is zero for $x<1$ and positive for $x>1$), and very sharp asymmetric peaks are found at the same points as above (due to one-sided divergence of $\Phi_2'' + \Phi_2'/x$).

On the other hand, the singularity of the Fourier kernel in the well-known 3D problem[6]

$$\Phi_3(x) = \frac{1}{2} + \frac{1-x^2}{4x}\ln\frac{1+x}{|1-x|} \tag{10}$$

(and also in the 1D problem[11]) is encountered on both sides of $x = 1$. We may finally note that Eq. (11) is simply connected with $\Phi(x)$ of our ''mixed'' problem

$$\Phi(x) = \frac{8}{\pi^2}\int_0^\infty \Phi_3(\sqrt{x^2+y^2})dy \tag{11}$$

(which seems to be hard to prove by direct integration).

DISCUSSION: ANISOTROPIC MODEL

The large negative values of $D_{\rm osc}/J_0$ implied by Eq. (7a) when $2k_F$ approaches G from below indicate long wave instability $(D_d + D_{\rm osc} < 0)$ if the relative distance of $2k_F$ from

G_{10} is $\delta<(J_0/J_d)^2$. In the case of exact equality, $2k_F=G$, Eq. (8) would indicate instability for long waves with $k_a<4(J_0/J_d)^2$; such an equality could only be incidental, and might result in a strong perturbation of the electron system as well. In an isolated layer (the ''truly 2D'' case), this would be a Bragg condition for electrons on the Fermi level resulting in opening of the 2D Fermi surface. For the 3D electron system, it is not the case if the $\mathbf{G}_{ij}$ of the spin lattice do not coincide with reciprocal lattice vectors $\mathbf{g}_{ijk}$ of the matrix (substrate) metal. This condition is fulfilled in important epitaxial structures using noble metals as substrates for magnetic layers.

The first monolayer in ideal epitaxy on a (001) or (111) plane of a fcc metal is a square or triangular lattice, respectively, with $a=a_m/\sqrt{2}$, if a_m is the lattice constant of the substrate. The shortest reciprocal vectors of the monolayer $\mathbf{G}_{\{10\}}$ do not belong to the substrate $\mathbf{g}_{ijk}$ set and their directions are reasonably far from the ''necks'' of the Fermi surface (FS). However, their lengths, $G_{10}=2\pi/a$ in a square lattice and $1.155\times(2\pi/a)$ in a triangular lattice, are close to the free-electron approximation for the FS ''belly'' diameter, $2k_F=(12/\pi)^{1/3}\times(2\pi/a_m)=1.105\times(2\pi/a)$, i.e., close to the coherence condition.

The free-electron model thus predicts significant reduction of exchange stiffness for monolayers grown on (111) planes [according to Eq. (7a) giving a large negative RKKY contribution], but not for layers grown on (001) planes [where Eq. (7b) would be appropriate]. This qualitative prediction is, however, quite dramatically changed when the real FS shapes of fcc metals are taken into account.

Extension of the RKKY model to real FS shapes[9] results in anisotropic $J_{\rm osc}(\mathbf{r})$ in Eq. (1). For a given direction of $\mathbf{r}$, the decisive locus or ''tip'' of the FS is that where $\mathbf{v}$, the local Fermi velocity, is parallel to $\mathbf{r}$, long-range oscillations of the asymptotic form $\cos(2k_Fr)/r^3$ as in Eqs. (2a),(2b) are found again but the value of $2k_F$ is replaced by $2k_v$, the projection of the FS diameter connecting two such ''tips'' onto $\mathbf{v}$; since there are many such diameters connecting the FS ''caliper'' points in the periodic Brillouin zone scheme (with Bragg reflections), $J_{\rm osc}(r)$ contains several wavelengths. The amplitude of each such term is enhanced (or reduced) over the free-electron value J_0 in Eq. (2c): for caliper points connected by inversion symmetry the enhancement factor may be written as

$$\epsilon_J=\frac{\hbar^3|\mathbf{v}|}{mk_F}\left(\frac{\partial^2E_m}{\partial k_x^2}\frac{\partial^2E_m}{\partial k_y^2}\right)^{-1} \qquad (12)$$

measuring the local effective mass from the electron energy dispersion $E_m(\mathbf{k})$: $k_{x,y}$ are measured perpendicular to $\mathbf{v}$, in the main normal section planes. This rule also accounts for the phase shifts[9] in $J_{\rm osc}(r)$: if the tips are saddle points, ϵ_J is negative.

We recall that the singular features in $D_{\rm osc}/J_0$ shown in Fig. 1 were first obtained by numerical summation of the direct-space series (3b). We have verified that they mainly result from long-range partial summations in Eq. (3b) in narrow cones along $\mathbf{r}$ perpendicular to $\mathbf{G}_{ij}$. Suitable approximations in such direct summations (see the Appendix) closely reproduce the analytical results (7a),(7b), which allows us to use these estimates at least for some high-symmetry caliper points of the anisotropic model.

Following Bruno and Chappert,[8] we calculated the FS shape corrections for Au, Ag, and Cu using the FS parameter sets[12] based on experimental data. In the $(\bar{1}\bar{1}1)$ plane, we find no caliper points with $\mathbf{v}$ perpendicular to $\mathbf{G}_{10}$ in Cu and Ag; in Au, such a point is incidentally close to inflexion in the $(1\bar{1}0)$ FS section where $J_{\rm osc}(r)$ according to Ref. 9 and also the direct summation estimates for $D_{\rm osc}(r)$ appear to be strongly reduced. This seems to invalidate the spherical-FS predictions for layers on (111) planes.

In the case of square lattices on (001) planes, the coincidence of $2k_v$ with G_{10} parallel to [110] is even closer than in the spherical model: we find $2k_v/G_{10}=1.050$, 1.065, and 1.041 for Cu, Ag, and Au, respectively. Near this coherence condition in the square 2D lattice, Bragg reflections in the 3D matrix have an important consequence: apart from $2k_v=(1+\delta)G_{10}$, which is on the nonsingular side corresponding to Eq. (7b), we must also include $2k_v'=g_{110}-2kv=(1-\delta)G_{10}$ since $g_{110}=(4\pi/a_m)\sqrt{2}=4\pi/a$. The [110] FS tips have high symmetry and we may use Eqs. (7a),(7b) with the appropriate enhancement (12) for the two contributions to $D_{\rm osc}$ from the two caliper pairs with $2k_v=(1\pm\delta)G_{10}$. With $\delta\cong0.05$ estimated above, the contribution to $D_{\rm osc}$ from the ''resonant'' peak of the dependence (7a) (with negative $D_{\rm osc}/J_0$) prevails over that from Eq. (7b).

The calculated absolute values of the relevant enhancement factors (12) are very large: for the [110] FS tips we find negative $\epsilon_J\cong-22$ in Au and Cu and positive $\epsilon_J\cong+6$ in Ag. The sign difference seems to be genuine, connected with the difference in the FS neck sizes. With these values, the RKKY amplitudes ϵ_JJ_0 will be negative in Cu and Au, but not in Ag. Thus the dominant first negative peak in $D_{\rm osc}/J_0$ seen in Fig. 1 would lead, in the anisotropic RKKY model, to a significant enhancement of the exchange stiffness in layers grown on (001) faces of Au or Cu, but to its weakening in layers grown on Ag.

The magnitude of the RKKY contribution to the exchange stiffness cannot be determined absolutely. With the very crude estimate of J_0 used in Ref. 8 we get estimates of $D_{\rm osc}$ in monolayers on Au or Cu of the same order of magnitude as the bulk value of D_d in Fe.

It would be tempting to extend this discussion also to the mean-field energy. In the free-electron model the RKKY mean field acts against the direct-exchange field for $2k_F\cong G_{10}$ (see Fig. 1). The enhancement and sign change of the RKKY amplitude derived above might correlate with recent experiments reporting enhanced magnetic moments[13] in thin Fe layers on fcc metals. However, the long-range approximation,[9] well applicable to the analysis of the singular behavior of the exchange stiffness, cannot be trivially extended to short-range interactions which decide about the mean energy of a dense 2D lattice (this is different from the case of interlayer exchange[7,8] across thick spacer layers).

CONCLUSIONS

The RKKY model offers an intuitively clear picture of the effect of a nonmagnetic metal matrix on the exchange stiffness (and thus on the stability of ferromagnetic order) in monolayers. The effect of weak oscillating exchange is strongly enhanced if the RKKY wavelength is close to the distance between the spin-lattice lines, which would be the case in ideal epitaxial monolayers on fcc metals. Only in special (singular) circumstances of exact coherence, the model predicts a qualitative change in spin wave dispersion, from quadratic to 3/2 power law.

The enhancement[8,9] of the RKKY interaction due to the very low curvature of relevant parts of the Fermi surface of fcc metals would lead to significant enhancement of spin-wave stiffness in layers on (001) faces of Au or Cu but to its weakening in layers on Ag. The practical relevance of such predictions is admittedly limited by the simplifications inherent to the RKKY model (dealing with spins instead of whole magnetic atoms), and also by the restriction of the model situation to an ideal magnetic planar lattice in a matrix of a different but ideal metal (disregarding all effects of structural imperfection).

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APPENDIX

For the square lattice and the spin-wave **k** along x, the long-range terms in the direct-space summation for $D_{\rm osc}$ obtained from Eq. (3b) are

$$D_{\rm osc}\cong -SJ_0a^2\sum_{m,n}(m^2/R^3)\cos(2Rk_Fa),$$

where $R=\sqrt{m^2+n^2}$ and m,n are integers. The absolute value of the oscillating summand falls off only as R^{-1} and divergent partial summations arise along the lattice directions $[i,j]$ if $k_Fa=\pi p/\sqrt{i^2+j^2}$ where p is an integer; however, such linear summations along neighboring parallel lines (offset from the origin) have alternating signs and practically cancel unless $p=i^2+j^2$. Thus divergent 2D summations appear for $k_Fa=\pi\sqrt{i^2+j^2}$, arising mainly from the close neighborhood of the $[i,j]$ lattice directions.

If J_0 does not vary much in such regions, it may be replaced by a mean value and analytical approximations for such summations may be obtained near the singular points. Setting $k_F=(1-\delta)\pi/a$ as in Eqs. (7a),(7b) near the first singularity, expanding R as $m+n^2/2m$ in a cone near the x axis, $|n|<N(m)<m$, and approximating

$$\sum_{n=1}^{N(m)}\cos\frac{\pi n^2}{m}\cong\sum_{n=1}^{N(m)}\sin(\pi n^2/m)$$
$$\cong\sqrt{m/2}\quad \text{for even } m$$
$$\cong 0\quad \text{for odd } m$$

[exact if $N(m)$ is extended to m], we get

$$D_{\rm osc}\cong -SJ_0a^2\sum_{m=1}^{\infty}[\cos(4\pi m\delta)+\sin(4\pi m\delta)]/\sqrt{m}$$

or, in terms of the generalized Riemann functions $\zeta(s,z)$,

$$\frac{D_{\rm osc}}{4SJ_0a^2}\cong\tfrac{1}{2}\,\zeta(\tfrac{1}{2},2\delta)\to -1/2\sqrt{2\delta}\quad \text{for}\quad \delta\to 0+$$
$$\cong -\tfrac{1}{2}\,\zeta(\tfrac{1}{2},1+2\delta)\to 0.73\quad \text{for}\quad \delta\to 0-,$$

in close agreement with Eqs. (7a),(7b). The anomalous dispersion (8) is also closely reproduced by an analogous but more involved analysis of partial summations in Eq. (3b).

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