Oscillations of the Exchange in Magnetic Multilayers as an Analog of de Haas–van Alphen Effect

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A theory of oscillations in the exchange coupling between two transition-metal ferromagnets separated by a nonmagnetic transition-metal spacer is developed for a one-band model. A close analogy between oscillations in the exchange coupling and de Haas–van Alphen oscillations is established and exploited to show that the period, asymptotic decay, and temperature dependence of the oscillations are determined by properties of the Fermi surface in the spacer layer. The theory describes many features of the oscillations in the exchange coupling observed recently in Co/Ru, Co/Cr, and Fe/Cr superlattices.

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Antiferromagnetic coupling between the iron layers in Fe/Cr/Fe sandwiches and superlattices has been observed by several experimental techniques [1–3]. Recently, Parkin, More, and Roche [4] reported oscillations in the exchange coupling and magnetoresistance as a function of the thickness of the nonmagnetic spacer layer in Co/Ru, Co/Cr, and Fe/Cr superlattices. In this paper we describe a generalization of our previous theory [5] which shows how a one-band model can produce the following experimental features of the oscillations in the exchange coupling: (i) long oscillation period of the order of 10 interatomic distances, (ii) variable sign of the coupling for small thicknesses of the spacer layer, (iii) large overall amplitude of the coupling and an asymptotic decrease proportional to the inverse square of the spacer thickness, (iv) strong temperature dependence of the exchange coupling on a scale $\cong 100$ K.

In the previous paper [5] we proposed and investigated a specific simple model of the exchange coupling based on spatial confinement of d holes in the spacer layer. The model gave a good account of features (i)–(iii) assuming a simple cubic tight-binding band structure for the spacer layer and (100) orientation of the layers. In this paper using an analogy with the de Haas–van Alphen (dHvA) effect we develop a general theory of the exchange coupling in the asymptotic limit of a large thickness of the spacer layer. The period, asymptotic decay, and temperature dependence of the oscillations in the exchange coupling are shown to be determined by properties of the Fermi surface in the spacer layer. The theory is valid for arbitrary band and arbitrary orientation of the layers. It leads to an RKKY-like coupling in which the appearance of certain long-period oscillations is due entirely to the fact that the spacer thickness is an integral number of interlayer spacings.

To obtain the exchange coupling we calculate the difference in energy between parallel and antiparallel orientations of the magnetic moments of two infinitely thick magnetic layers separated by a nonmagnetic spacer of $N-1$ atomic planes. The ferromagnetic metal is assumed to have a full majority-spin d band and a partially occupied minority-spin d band. The nonmagnetic metal has equal numbers of holes in each spin subband. The spin subbands in the ferromagnetic and nonmagnetic spacer layers, together with the hole densities $\rho^{\uparrow},\rho^{\downarrow}$, are shown schematically in Figs. 1(a) and 1(b) for the parallel and antiparallel orientations of the layer moments. For simplicity, we assume that the number of d holes per atom of each spin in the bulk nonmagnetic metal is equal to the number of holes in the bulk ferromagnetic metal. The basic mechanism we propose for exchange coupling does not depend on this precise condition. However, it is a reasonable approximation to the actual situation in Co/Ru, Co/Cr, and Fe/Cr systems.

It is clear from Fig. 1 that deviations from bulk hole densities occur in the spacer layer near the interfaces with the ferromagnetic layers. For the parallel configuration, Fig. 1(a), both interface effects occur in the down-spin hole density and, therefore, interfere with each other. In fact, because down-spin holes are completely

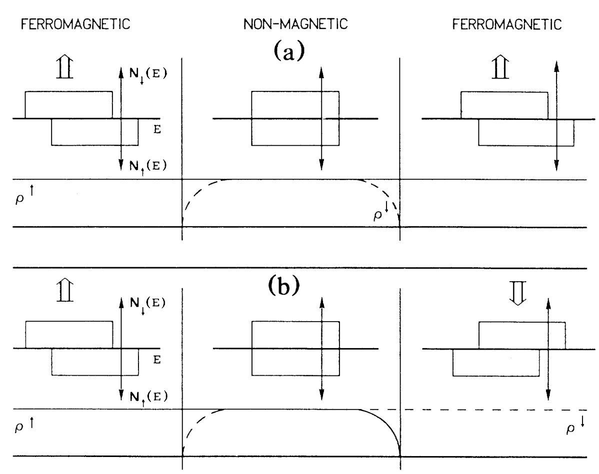

FIG. 1. Schematic plots of the densities of holes $\rho^{\uparrow}$ (solid curve) and $\rho^{\downarrow}$ (dashed curve) in a sandwich for (a) ferromagnetic and (b) antiferromagnetic alignments of the magnetic layers. Insets: The densities of states for each spin $N_{\uparrow}(E),N_{\downarrow}(E)$ in each region, the vertical axis being drawn at the Fermi level.

confined in the spacer layer by the exchange potential, the interference effect is associated with size quantization of their energy. In the antiparallel configuration, the interface effects at each end of the spacer layer occur in opposite spin densities and no interference takes place. The up-spin holes are now confined in the half space to the left of the second interface and the down-spin holes in the half space to the right of the first interface. It follows that there is a constant surface energy associated with the confinement in the antiferromagnetic configuration which is *independent* of the spacer thickness. The exchange coupling between the ferromagnetic layers, which is given by the difference in energy between the two configurations, is therefore determined entirely by the interference (size-quantization) effect in the down-spin band of the spacer layer.

It is assumed in Fig. 1 that the exchange splitting in the ferromagnetic layers is so large that holes of the wrong spin are completely excluded. We further assume that holes in the spacer layer are noninteracting. It is then clear that the energy difference $\Delta E(N-1)=E_{\uparrow\uparrow}-E_{\uparrow\downarrow}$ is given by

$$\Delta E(N-1)=E(N-1)-E(\infty), \tag{1}$$

where $E(N-1)$ is the kinetic energy of the down-spin holes trapped in the spacer layer of a sandwich with ferromagnetically aligned magnetic layers measured relative to a $(N-1)$-plane reference state with bulk density. The energy $E(\infty)$ is just the constant surface term present in the antiferromagnetic configuration.

Formally our calculation corresponds to an exact Hartree-Fock treatment of a Hubbard-like one-band tight-binding model with on-site interaction $U=0$ in the spacer layer and with $U=\infty$ in the magnetic layers.

To conserve the number of particles overall we must consider instead of the total energy of holes in the spacer layer their thermodynamic potential

$$\Omega(N-1)=E(N-1)-E_F n(N-1), \tag{2}$$

where E_F is the Fermi energy. Both $\Omega(N-1)$ and the number of holes $n(N-1)$ are again relative to a $(N-1)$-plane reference state with bulk density. The change in energy between the two configurations conserving the number of particles is, therefore, given by

$$\Delta\Omega(N-1)=\Omega(N-1)-\Omega(\infty). \tag{3}$$

Following Parkin, More, and Roche [4] we finally define an exchange coupling constant per unit area for a spacer layer of $N-1$ atomic planes by

$$J(N-1)=\Delta\Omega(N-1)/A. \tag{4}$$

At finite temperatures the total thermodynamic potential $\Omega_{\text{tot}}=\Omega+\Omega_{\text{ref}}$ is clearly given by

$$\Omega_{\text{tot}}=-T\sum_{\mathbf{k},r}\ln\left[1+\exp\left(\frac{\mu-E(\mathbf{k},r)}{T}\right)\right], \tag{5}$$

where μ is the chemical potential, $E(\mathbf{k},r)$ is the hole energy, $\mathbf{k}$ is the wave vector parallel to the sandwich, and $r=1,\ldots,N-1$ labels the discrete energy levels of holes ($k_B=1$).

To determine the coupling, we require the dependence of the thermodynamic potential Ω of size-quantized holes in a layer of $N-1$ atomic planes on the thickness of the layer $d=Na$. This is a familiar problem in the theory of the de Haas–van Alphen effect where two-dimensional quantization of the carrier energy in a magnetic field takes place in a plane perpendicular to the field. In the present problem, we have one-dimensional quantization in the direction perpendicular to the sandwich. We can, therefore, adapt the conventional theory of the de Haas–van Alphen effect [6].

To evaluate the discrete sum over r in Eq. (5), we apply the Poisson summation formula

$$\sum_{r=1}^{\infty}\Phi(r)=2\,\text{Re}\sum_{s=1}^{\infty}\int_0^{\infty}\Phi(\zeta)e^{2\pi i s\zeta}d\zeta+(\text{nonoscillatory term}), \tag{6}$$

where the discrete quantum number r is replaced by a continuous variable ζ.

It is easy to show that the oscillatory term in the Poisson formula picks from Ω_{tot} precisely the required interference contribution $\Delta\Omega$. The exchange coupling J is, therefore, given by

$$J=-2T\frac{1}{(2\pi)^2}\,\text{Re}\sum_{s=1}^{\infty}\int\int_{\text{BZ}}dk_x\,dk_y\int_0^{N-1}d\zeta\ln\left[1+\exp\left(\frac{\mu-E(k_x,k_y,\zeta)}{T}\right)\right]\exp(2\pi i s\zeta), \tag{7}$$

where the integral with respect to k_x,k_y is over the two-dimensional Brillouin zone (BZ) in the sandwich plane which is assumed to be parallel to the x-y plane.

To proceed further, we have to make some assumption about the size quantization of the carrier energy. In [5] we modeled the spacer layer by a simple cubic tight-binding band,

$$\varepsilon(k_x,k_y,k_z)=-[\cos(k_xa)+\cos(k_ya)+\cos(k_za)], \tag{8}$$

where the energy is measured in units of the hopping $2|t|$. For such a band and a sandwich parallel to the x-y plane, the quantization of k_z is given by

$$k_z=r\pi/Na,\quad r=1,2,\ldots,N-1, \tag{9}$$

where a is the distance between neighboring planes. It is easy to show that the quantization (9) holds for any single band and any orientation of the sandwich. Hence $E(k_x,k_y,\zeta)=\varepsilon(k_x,k_y,\zeta\pi/Na)$.

Assuming the quantization (9), integrating in Eq. (7) by parts with respect to ζ, and then passing from integration over ζ to integration over the energy ε, we obtain

$$J=-\frac{1}{(2\pi)^2}\mathrm{Re}\sum_{s=1}^{\infty}\frac{1}{s\pi i}\int_0^{N-1}d\varepsilon\left[1+\exp\left(\frac{\varepsilon-\mu}{T}\right)\right]^{-1}\int\int_{\mathrm{BZ}}dk_x\,dk_y\exp(2isNak_z)\,, \tag{10}$$

with $k_z\equiv k_z(\varepsilon,k_x,k_y)$. For large N, the factor $\exp(2isNak_z)$ oscillates rapidly and only the regions in the k_x-k_y plane where $k_z(\varepsilon,k_x,k_y)$ is stationary with respect to k_x,k_y contribute to the Brillouin-zone integral. We can, therefore, use the method of stationary phase to approximate the integral for large N. The remaining energy integral is evaluated as in the theory of de Haas–van Alphen effect [6], i.e., only the contribution from the vicinity of $\varepsilon=\mu$ is included. This leads to the following general asymptotic formula:

$$J(N-1)=\frac{1}{4\pi Na}\mathrm{Re}\sum_{s=1}^{\infty}\frac{\sigma}{s^2}\left|\frac{\partial k_z^2}{\partial k_x^2}\frac{\partial k_z^2}{\partial k_y^2}\right|^{-1/2}\frac{\exp[2isNak_z^0(\mu)]}{T^{-1}\sinh[2\pi sNaT\,\partial k_z/\partial\varepsilon]}\,, \tag{11}$$

$$\sigma=\begin{cases} i, & \text{both second derivatives} > 0\,,\\ -i, & \text{both derivatives} < 0\,,\\ 1, & \text{one derivative} > 0,\ \text{the other} < 0\,.\end{cases}$$

Here, $k_z^0(\mu)$ is an extremal radius of the Fermi surface in the direction perpendicular to the layers (half the caliper measurement) and all the derivatives in Eq. (11) are taken at the stationary point $k_z^0\equiv k_z^0(\mu,k_x^0(\mu),k_y^0(\mu))$. When there is more than one stationary point the contributions of all such points have to be included in Eq. (11).

The consequences of the asymptotic formula for $J(N)$ are the following:

(i) The period of oscillations in the exchange coupling is determined by the factor $\exp[2iNak_z^0(\mu)]$. Clearly, owing to the discrete thickness Na of the spacer layer, $k_z^0(\mu)$ may be replaced by $k_z^0(\mu)-\pi/a$. Therefore, long periods are obtained either when the radius $k_z^0(\mu)$ is small or when the Fermi surface approaches the zone boundary at π/a so that $k_z^0(\mu)-\pi/a$ is small.

(ii) The amplitude contains a factor determined by the curvature of the Fermi surface at its extremal points.

(iii) The temperature dependence of the oscillations is governed by the velocity of carriers at the extremal points.

(iv) The asymptotic decay at $T=0$ is proportional to $1/N^2$.

To make contact with our model calculation [5], we assume (100) orientation of the layers and compare the results of the general asymptotic formula (11) at $T=0$ for the simple cubic band (8) with the exact results calculated numerically using Eq. (3) of [5]. To evaluate J from Eq. (11), we have to determine all the extrema of $k_z(E_F,k_x,k_y)$, the Fermi energy E_F playing the role of the chemical potential at $T=0$.

For $-3<E_F<-1$, it is clear from Eq. (8) that there is a single maximum of k_z located at $k_x=k_y=0$. It leads to long-period oscillations in the exchange coupling for $E_F\lesssim -1$. These oscillations together with the relevant Fermi-surface cross sections are shown in Fig. 2 for $E_F=-1.05$ (open squares) and compared with the exact result (solid squares). The initial sign of J is antiferromagnetic and the period is $\cong 10$ interatomic distances, which is as observed in structures such as Co/Ru [4]. A long period is obtained because the Fermi surface for $E_F=-1.05$ nearly touches the zone boundary and twice the distance from the extremum to the zone boundary ($2\pi/a$ minus the caliper measurement) is small.

For $E_F>-1$, the Fermi surface develops four necks in the k_x-k_y plane (equivalent to two saddle points) which are the extrema that determine oscillations in the exchange coupling for $-1<E_F<0$. Such oscillations are shown in Fig. 2 for $E_F=-0.95$ (open triangles) together with the exact result (solid triangles). The period is again long because the diameter of the necks is small for $E_F=-0.95$. Compared with $E_F=-1.05$, there is a

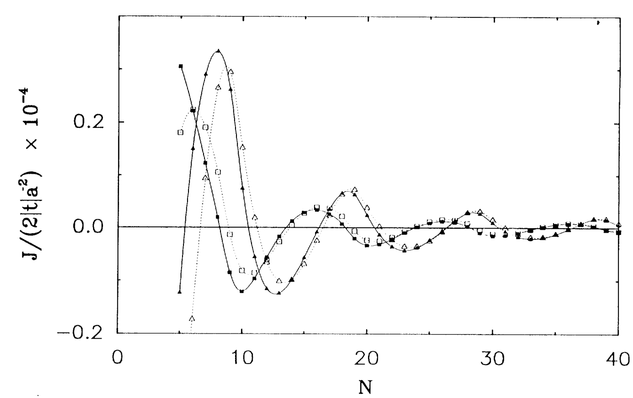

FIG. 2. Comparison of the exchange coupling J obtained from the asymptotic formula (11) (open symbols) with the exact result (solid symbols): Squares are for $E_F=-1.05$; triangles for $E_F=-0.95$. Inset: The corresponding Fermi-surface cross sections. Arrows indicate the relevant caliper measurements proportional to (period)$^{-1}$, an extended zone scheme being used in the left-hand inset to show the role of the $2\pi/a$ shift.

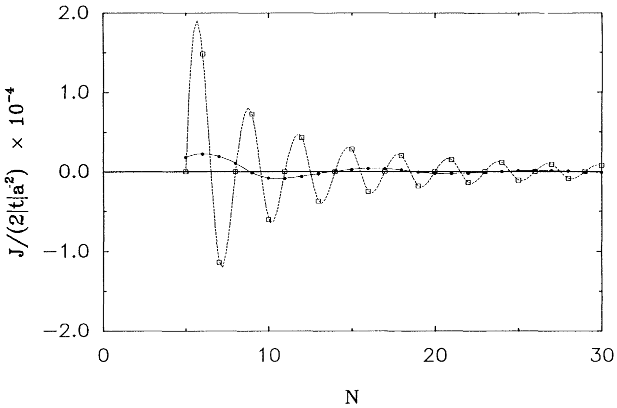

FIG. 3. Comparison of the amplitudes and periods of oscillations in the exchange coupling for two different fillings of the band: $E_F = -1.05$ (solid curve) and -2.5 (dashed curve).

phase shift in the oscillations and the sign of J for small N is now ferromagnetic. The phase shift is obtained because the factor σ in Eq. (11) takes a value $\sigma = 1$ for a saddle point. Clearly the asymptotic formula is rather accurate for $N \geq 5$.

The dependence of the oscillation amplitude on the curvature of the Fermi surface is illustrated in Fig. 3. The smaller amplitude of the oscillations for $E_F = -1.05$ (solid circles) compared with $E_F = -2.5$ (open squares) is due to the greater curvature of the Fermi surface at the extremum. In practice, roughness of the surfaces may lead to a variable effective spacer thickness which would tend to suppress short-period oscillations, such as those for $E_F = -2.5$, due to an averaging effect.

Although the amplitude of J for $E_F = -1.05$ (long-period oscillations) is relatively small, numerical evaluation of Eq. (3) at $T = 0$ K for a spacer layer consisting of two atomic planes (see [5]) gives $J \cong 1$ $\mathrm{erg\,cm^{-2}}$. This is comparable with the largest experimental value $J \cong 6$ $\mathrm{ergs\,cm^{-2}}$ obtained for Co/Ru structures [4].

Finally, we turn to the temperature dependence of the exchange coupling. It is controlled in our theory by the sinh factor in Eq. (11) which has exactly the same form as in the dHvA effect [6] provided the following correspondence is made:

$$N \frac{\partial k_z a}{\partial \varepsilon} \rightarrow \frac{1}{\hbar \omega_c}, \tag{12}$$

where ω_c is an effective "cyclotron" frequency. Substituting in Eq. (12) typical values of E_F, it is easy to see that the effective cyclotron frequency in the present problem is $\hbar\omega_c \cong 10^3$ K for small N. The dominant temperature-dependent factor in Eq. (11) is $T/\sinh(2\pi T/\hbar\omega_c)$ and hence the temperature dependence of the exchange coupling is on the scale $\cong 100$ K, as observed by Parkin, More, and Roche [4]. An effective cyclotron frequency $\hbar\omega_c \cong 10^3$ would correspond in the de Haas–van Alphen effect to an applied field of $\cong 10^3$ T which explains why the oscillations in the exchange coupling are seen at room temperature and, presumably, are not easily washed out by impurity scattering. Thus, study of layered magnetic structures with different crystal orientations of the spacer layer might be used to determine extremal cross sections of the Fermi surface and effective masses for concentrated alloys and other systems (possibly including exotic materials like heavy fermions and high-T_c superconductors) not susceptible to de Haas–van Alphen investigation. Before a detailed comparison with real systems can be made, however, it is essential to extend the calculations beyond the present one-band model and include the *sp* band. The extended theory should then be applicable to noble-metal spacer layers.

Finally, we wish to mention a connection between the oscillations of J obtained in our theory and RKKY. Our asymptotic results obtained to $O(1/N^2)$ concerning the period, rate of decay, and even the temperature dependence of J are directly comparable with the asymptotic RKKY results obtained by Roth, Zeiger, and Kaplan [7] for a general shape of the Fermi surface. It is only necessary to transpose their results to the appropriate planar geometry and introduce the discrete nature of the spacer thickness. However, the overall amplitude of the exchange coupling and its initial sign and behavior at short distances are all model-dependent features and cannot be predicted from the conventional RKKY theory. Furthermore, differences from an RKKY-type theory show up to $O(1/N^3)$ [8]. They appear because in our nonperturbative theory the interference between the disturbances in the electron density in the nonmagnetic spacer, arising from the coupling to the two ferromagnetic layers, is treated exactly.

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