Non-RKKY oscillations of exchange coupling in magnetic multilayers

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Abstract

It is shown how the exchange coupling between two ferromagnetic planes embedded in an infinite non-magnetic metal, regarded as a function of the distance between the planes, may contain important components which oscillate with periods not predicted by RKKY theory. The interesting case of a FCC(110) structure with a Cu-like Fermi surface is discussed in detail. 75.50.Fr, 75.30.Et, 75.50.Rr

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Oscillatory exchange coupling between metallic magnetic layers across a non-magnetic spacer has been intensively studied over the last five years. One of the main issues in this area has been the determination of the oscillation periods of the coupling as a function of the spacer thickness.

The physical mechanisms which have been proposed for explaining this phenomenon include the quantum well theory (QWT) of Edwards *et al.* [1–3] and an extension of the RKKY theory to the multilayer geometry due to Bruno and Chappert [4,5]. In the QWT electrons propagating across the multilayer structure experience spin-dependent potential wells whose depths depend on the exchange interaction in the ferromagnetic layers. The oscillatory behaviour of the interlayer coupling arises as a consequence of quantum interference effects inside the wells, and bears a formal analogy to de Haas-van Alphen oscillations [1]. The existence of these quantum wells has been confirmed experimentally by photoemission measurements [6].

In the RKKY theory the oscillation periods are directly related to the spacer Fermi surface (FS) and are given by the wave vectors \mathbf{q}_z perpendicular to the layers that span the FS across those parts whose group velocities are mutually antiparallel. There is a general belief that all oscillation periods are given by the RKKY theory. Indeed, it has been shown that in certain simple models [1–3] the periods predicted by the QWT coincide with the RKKY ones and are given by the extremal dimensions of the spacer FS in the direction perpendicular to the layers. In another case [7], where the lattice lacks reflection symmetry about a layer plane, the correspondence between the quantum well and the RKKY periods is more subtle but still obtains. Of course in the models mentioned above harmonics of the RKKY appear but no new fundamental periods. Furthermore, d'Albuquerque e Castro *et al.* [8] showed analytically for a very general model that RKKY theory holds in the limit of very small exchange splitting in the ferromagnetic material. However, van Schilfgaarde and Harrison found that real systems, such as Fe/Cr, are not in this limit [9], although they were convinced that the oscillation periods were derivable from the RKKY. Here we show, however, that under certain conditions, including those met in FCC(110) multilayers with

a Cu-like spacer FS, oscillation periods exist which are not predicted by RKKY. These are in addition to the usual RKKY periods.

A FCC(110) magnetic multilayer within the one-band nearest-neighbour tight-binding model is an interesting system because of the difficulty in determining analytically the energies of the resonances and size quantized states in such a structure [10]. Here, however, we approach the problem using the formalism of Ref. [8], which gives the coupling in terms of the one-electron propagators. This enables us to find the periods of oscillation analytically and to evaluate the exchange coupling numerically.

We consider a multilayered system consisting of two parallel ferromagnetic atomic planes embedded in an infinite non-magnetic material. We label these two planes 0 and n, so that the number of atomic planes in the spacer layers is equal to n-1. It has been shown recently that as far as the interlayer coupling as a function of the spacer thickness is concerned, the thickness of the magnetic layers affects the phase and amplitude of the oscillations, but not the periods [11].

Within the single band model, the expression for the exchange coupling J, defined as the difference in the thermodynamical potential between the ferromagnetic and antiferromagnetic configurations, is given by [8]

$$J = -\frac{1}{\pi} \sum_{\mathbf{q}_{\parallel}} \int d\omega f(\omega) F(\mathbf{q}_{\parallel}, \omega), \qquad (1)$$

where

$$F(\mathbf{q}_{\parallel},\omega) = Im \ln \left\{ 1 + 4V_{ex}^2 G_{n0}^{\uparrow}(\mathbf{q}_{\parallel},\omega) G_{0n}^{\downarrow}(\mathbf{q}_{\parallel},\omega) \right\},$$
(2)

and $G_{n0}^{\sigma}(\mathbf{q}_{\parallel},\omega)$ is the off-diagonal matrix element between planes 0 and n of the Green's function for an electron with spin σ in the ferromagnetic configuration of the system, $f(\omega)$ is the Fermi function, V_{ex} is the exchange interaction in the ferromagnetic layers, and the summation over \mathbf{q}_{\parallel} is restricted to the two-dimensional Brillouin zone (BZ). We assume for simplicity that the site energies in the ferromagnetic material are $\epsilon_{\uparrow,\downarrow} = \epsilon_0 \mp V_{ex}$, where ϵ_0 is the spacer on-site energy. In this situation, the off-diagonal propagators in Eq.(2) can be written for each \mathbf{q}_{\parallel} and ω as

$$G_{n0}^{\uparrow,\downarrow} = \pm \frac{\tau_{\uparrow,\downarrow} g_{n0} \left(1 - \tau_{\uparrow,\downarrow} g_{00}\right)}{V_{ex} \left(1 - \tau_{\uparrow,\downarrow} g_{n0} \tau_{\uparrow,\downarrow} g_{0n}\right)},\tag{3}$$

where $\tau_{\uparrow,\downarrow} = \pm V_{ex}(1\pm V_{ex}g_{00})^{-1}$, and g_{n0} is the matrix element of the bulk spacer Green's function. A similar expression can be obtained for $G_{0n}^{\uparrow,\downarrow}$. In general, within the one-band model, g_{0n} is equal to g_{n0} , apart from a possible \mathbf{q}_{\parallel} -dependent phase factor.

It is clear that the behaviour of the coupling as a function of the spacer thickness is related to the dependence of g_{n0} on n, which can be determined as follows. The matrix element of g between arbitrary planes l and m is given by

$$g_{lm} = \left(\frac{1}{2\pi}\right) \int_{-\frac{\pi}{d}}^{\frac{\pi}{d}} dq_{\perp} \frac{e^{-iq_{\perp}(l-m)d}}{\omega - E(\mathbf{q}_{\parallel}, q_{\perp}) + i \, 0^{+}}$$
(4)

where q_{\perp} is the wave vector perpendicular to the layers, d is the interplane distance, and $E(\mathbf{q}_{\parallel}, q_{\perp})$ describes the bulk spacer band structure. We evaluate the above expression for g_{lm} with l < m by integrating around the boundary of the semi-infinite rectangle $-\pi/d \leq Re q_{\perp} \leq \pi/d$; $Im q_{\perp} \geq 0$, in the complex q_{\perp} -plane. This procedure is completely general and can be applied, within the one-band model, to any lattice structure and layer orientation, with hoppings to arbitrary number of neighbours. For the FCC(110) case, $E(\mathbf{q}_{\parallel}, q_{\perp}) = \epsilon(\mathbf{q}_{\parallel}) + 2t_1(\mathbf{q}_{\parallel})cos(q_{\perp}d) + 2t_2(\mathbf{q}_{\parallel})cos(2q_{\perp}d)$, where $\epsilon(\mathbf{q}_{\parallel}) = -2t_0 \cos(2q_x d)$, $t_1(\mathbf{q}_{\parallel}) = -4t_0 \cos(q_x d)cos(\sqrt{2}q_y d)$, and $t_2(\mathbf{q}_{\parallel}) = -t_0$. Here q_x and q_y are the components of \mathbf{q}_{\parallel} , $-t_0$ is the hopping between first nearest neighbour atoms, and the origin of energy is chosen such that $\epsilon_0 = 0$. t_1 and t_2 are the hoppings to first and second nearest planes, respectively. Using the contour integration described above, we obtain

$$g_{n0}(\mathbf{q}_{\parallel},\omega) = A_1(\mathbf{q}_{\parallel},\omega)e^{iq_1(\mathbf{q}_{\parallel},\omega)nd} + A_2(\mathbf{q}_{\parallel},\omega)e^{iq_2(\mathbf{q}_{\parallel},\omega)nd},$$
(5)

where $\cos(q_j d) = -[\gamma + (-1)^j \sqrt{\gamma^2 + 8(\omega - \epsilon + 2t_2)/2t_2}]/4$, $A_j = [2i(\cos(q_1 d) - \cos(q_2 d))(1 - \cos^2(q_j d))^{1/2}]^{-1}$, for j = 1, 2, and $\gamma = t_1/t_2$. Here $q_{\perp} = \pm q_1$ and $q_{\perp} = \pm q_2$ are the roots of the equation $E(\mathbf{q}_{\parallel}, q_{\perp}) = \omega$. In the present case $g_{0n} = g_{n0}$ and the above expression for g_{n0} can be extended to continuous values of n. Eq.(5) shows that for values of \mathbf{q}_{\parallel} and ω , for which q_1 and q_2 are real, g_{n0} oscillates with the superposition of two periods, $2\pi d/|q_1|$ and

 $2\pi d/|q_2|$, which are in general incommensurate. In those cases g_{n0} exhibits a quasi-periodic dependence on n. As it is shown below, this fact may have a striking effect on the coupling.

The function F in Eq.(1) varies with n through g_{n0} and therefore exhibits the same quasi-periodic behaviour. In order to deal with this quasi-periodic function, we make use of a procedure analogous to the one recently proposed [11] to investigate the dependence of the coupling on the magnetic layer thickness. It consists in replacing n in expression (5) for g_{n0} by fictitious variables n_1 and n_2 , which multiply q_1 and q_2 , respectively. The real physical situation corresponds to $n_1 = n_2 = n$. The extended function $F(n_1, n_2)$ is then periodic in each variable separately, and can be Fourier analysed in the usual way. Thus we find

$$F = \sum_{m_1, m_2} C_{m_1, m_2} e^{i(m_1 q_1 + m_2 q_2)nd},$$
(6)

where $C_{m_1,m_2}(\mathbf{q}_{\parallel},\omega)$ are the Fourier coefficients, and m_1 and m_2 are integers. Since F is in fact a function of g_{n0}^2 , it follows that $C_{m_1,m_2} = 0$ unless $m_1 + m_2$ is even. These coefficients contain all the information about the electron potential in the ferromagnetic layers. In particular, they depend on the magnitude of V_{ex} and vanish for $V_{ex} = 0$. In the RKKY limit, where F is replaced by the leading second-order term of its expansion in powers of V_{ex} , the Fourier coefficients can be determined analytically. In this limit only six coefficients appear, namely $C_{\pm 2,0}$, $C_{0,\pm 2}$, and $C_{\pm 1,\pm 1}$, whose values are given by $C_{2,0} = V_{ex}^2 A_1^2/2i$, $C_{0,2} = V_{ex}^2 A_2^2/2i$, $C_{1,1} = V_{ex}^2 A_1 A_2/i$, and the property $C_{-m_1,-m_2} = C_{m_1,m_2}^*$.

By inserting Eq.(6) into Eq.(1) we find that, according to the usual stationary phase method [1–3,11], for sufficiently large values of n, the nonzero contributions to the coupling come from ω equal to the Fermi energy E_F and \mathbf{q}_{\parallel} in the neighbourhood of those points at which the argument of the exponential is stationary. They are given by the equation

$$m_1 \nabla_{\parallel} q_1(\mathbf{q}_{\parallel}, E_F) + m_2 \nabla_{\parallel} q_2(\mathbf{q}_{\parallel}, E_F) = 0, \tag{7}$$

where ∇_{\parallel} is the two-dimensional gradient in \mathbf{q}_{\parallel} space. The position of the extremal points can be determined exactly from the analytical expressions for q_1 and q_2 . The weight of the contribution to the coupling from each solution of Eq.(7) depends on the magnitude of the corresponding Fourier coefficient and partial derivatives of $\phi = m_1 q_1 + m_2 q_2$ with respect to q_x, q_y , and ω .

Clearly the surfaces $q_{\perp} = \pm q_1(\mathbf{q}_{\parallel}, E_F)$ and $q_{\perp} = \pm q_2(\mathbf{q}_{\parallel}, E_F)$ map out the FS. Fig. 1 shows a cross-section perpendicular to the atomic planes of the FS for $E_F/2t_0 = 1.64$ and $q_y = \pi \sqrt{2}/4d$. Note that $q_1 > 0$ and $q_2 < 0$. Full lines correspond to the surface $\pm q_1$ and dashed lines to $\pm q_2$. The nature of the FS depends on E_F , and three distinct energy regions are to be considered, namely $-12t_0 \leq E_F \leq -4t_0$, $-4t_0 \leq E_F \leq 0$, and $0 \leq E_F \leq 4t_0$. The two boundary values, $-4t_0$ and 0, correspond to those values of E_F at which the FS first touches the layer geometry BZ, and develops necks, respectively. As we show below, the numbers of periods we obtain in the three regions are different.

It is interesting to examine first the predictions for the periods in the RKKY limit, where only six integers pairs $m_1 m_2$ are to be considered in Eq.(7). In the first energy region we find only one period $\lambda_{2,0}^a = \pi d/|q_1(\mathbf{q}_{\parallel}^a, E_F)|$, with $\mathbf{q}_{\parallel}^a = (0,0)$. In the second region an additional period $\lambda_{1,1}^b = 2\pi d/|q_1(\mathbf{q}_{\parallel}^b, E_F) + q_2(\mathbf{q}_{\parallel}^b, E_F)|$ appears, where $\mathbf{q}_{\parallel}^b = (0, \pm \pi \sqrt{2}/4d)$. Finally, in the third region RKKY predicts four periods, namely, $\lambda_{2,0}^a$, $\lambda_{2,0}^c = \pi d/|q_1(\mathbf{q}_{\parallel}^c, E_F)|$, $\lambda_{0,2}^{c} = \pi d/|q_{2}(\mathbf{q}_{\parallel}^{c}, E_{F})|, \text{ and } \lambda_{1,1}^{c} = 2\pi d/|q_{1}(\mathbf{q}_{\parallel}^{c}, E_{F}) + q_{2}(\mathbf{q}_{\parallel}^{c}, E_{F})| = \lambda_{1,1}^{d} = 2\pi d/|q_{1}(\mathbf{q}_{\parallel}^{d}, E_{F}) + q_{2}(\mathbf{q}_{\parallel}^{c}, E_{F})| = \lambda_{1,1}^{d} = 2\pi d/|q_{1}(\mathbf{q}_{\parallel}^{d}, E_{F}) + q_{2}(\mathbf{q}_{\parallel}^{c}, E_{F})| = \lambda_{1,1}^{d} = 2\pi d/|q_{1}(\mathbf{q}_{\parallel}^{d}, E_{F})| = \lambda_{1,1}^{d} = 2\pi d/|q_{1}(\mathbf{q}_{\parallel}^{c}, E_{F})|$ $q_2(\mathbf{q}_{\parallel}^d, E_F)|$, with $\mathbf{q}_{\parallel}^c = (\pm \pi/2d, \pm \pi\sqrt{2}/4d)$ and $\mathbf{q}_{\parallel}^d = (\pm \pi/2d, 0)$. The Fourier coefficients associated with the period $\lambda_{m_1,m_2}^{\alpha}$ are $C_{\pm m_1,\pm m_2}(\mathbf{q}_{\parallel}^{\alpha}, E_F)$. These periods are shown in Fig. 2 as functions of E_F . For the present model we find that $q_1(\mathbf{q}_{\parallel}^c, E_F)$ and $q_2(\mathbf{q}_{\parallel}^c, E_F)$, which are represented in Fig. 1, satisfy the relation $q_1(\mathbf{q}_{\parallel}^c, E_F) - q_2(\mathbf{q}_{\parallel}^c, E_F) = \pi$. Thus, the oscillation periods $\lambda_{2,0}^c$ and $\lambda_{0,2}^c$ cannot be distinguished just by looking at discrete integer values of n. We recall that in FCC Cu E_F lies in the third energy region with FS necks. A quantitative description of the oscillation periods for Cu can be obtained within the present framework by going beyond nearest neighbours and using the tight-binding parameters of Halse [12]. Then $\lambda_{1,1}^c$ and $\lambda_{1,1}^d$ become distinct periods and, by taking into account the equivalence of $\lambda_{2,0}^c$ and $\lambda_{0,2}^c$ for a discrete lattice, we find exactly the four RKKY periods of Bruno and Chappert [4].

However, it follows from Eq.(7) that there are contributions to the coupling with periods

other than those which arise in the RKKY limit discussed above. In fact, it is easy to show that for E_F in the second and third energy regions we find an infinite number of solutions, corresponding to infinitely many values of m_1 and m_2 . Some of these solutions correspond merely to harmonics of the fundamental RKKY periods but some fundamentally new periods can arise. The simplest and most interesting case actually occurs in the top energy region, where $\nabla_{\parallel}q_1$ and $\nabla_{\parallel}q_2$ vanish simultaneously at \mathbf{q}^c_{\parallel} . Thus, Eq.(7) is automatically satisfied for any values of m_1 and m_2 . The corresponding periods are $\lambda_{m_1,m_2}^c = 2\pi d/|m_1q_1 + m_2q_2|$. Fig. 3 exhibits the coupling J at temperature $T = 2.0 \times 10^{-3} W/k_B$ as a function of the spacer thickness for $E_F/2t_0 = 1.64$ and $V_{ex} = 0.15W$, where $W = 16t_0$ is the spacer band-width. We chose this value of E_F so that an important new period $\lambda_{3,1}^c$, plotted as a function of E_F in Fig.2, is well separated from the RKKY periods. The full line in Fig.3 corresponds to the result obtained from Eq.(1), and the dashed line to the RKKY approximation scaled down by a factor of 8. In both cases n was treated as a continuous variable, but the physical discrete values are indicated. For this E_F the long period dominates both curves although with an amplitude differing by a large factor, but the interesting fine structure is different and reflects contributions beyond the fundamental RKKY periods. To make this point explicit, we present in the inset the absolute value of the ratio between some coefficients C_{m_1,m_2} and $C_{1,1}$, the largest coefficient of a fundamental RKKY period, as a function of V_{ex} and for $\mathbf{q}^{c}_{\parallel}$. As expected, for very small exchange splittings, the magnitudes of higher order Fourier coefficients relative to that of the fundamental RKKY one are negligible. However, they rapidly increase with V_{ex} , and the ratio $|C_{m_1,m_2}|/|C_{1,1}|$ becomes significant. From the inset in Fig.3, we see that for $V_{ex} = 0.15W$, there are additional contributions to the coupling coming not just from harmonics of the RKKY frequencies, which correspond to m_1 and m_2 even, but a very important one coming from a new period $\lambda_{3,1}^c = 4.5d$. In fact, the contribution from this new period can be calculated separately using the stationary phase method [1–3,11]. The result is shown in Fig.4, together with those corresponding to two of the fundamental RKKY periods, namely, $\lambda_{2,0}^a$ and $\lambda_{2,0}^c$. It is clear that this new period is as important as the RKKY ones, except for the dominant long period whose large amplitude is due to FS geometry which causes some second derivatives of ϕ to vanish, making a stationary phase evaluation of this contribution impossible. The amplitude of the new contribution falls off as $1/n^2$ just like the normal RKKY components and their harmonics [1–5].

The appearance of the non-RKKY periods is clearly related to the spacer off-diagonal propagators oscillating as a function of the spacer thickness with more than one period, for fixed energy and \mathbf{q}_{\parallel} . As we have shown, such a behaviour can be found even in the one-band model, for which the FS is simple and has a single sheet. It can also be found in those cases in which the spacer FS has more than one sheet. Thus we may expect the occurrence of non-RKKY periods in the coupling through a non-magnetic transition metal. Therefore, in those cases, the interpretation of the results in terms of just the RKKY theory may be misleading.

In conclusion, we have shown that the relation between the oscillation periods of the coupling and the spacer FS is more complex and subtler than has been assumed so far, making possible the appearance of non-RKKY periods. This is the central result in this communication, which settles the long standing question about whether or not the quantum well and the RKKY theories always give the same periods of oscillation.

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FIGURES

FIG. 1. Cross-section of the spacer Fermi surface perpendicular to the layers for $E_F/2t_0 = 1.64$ (see text). Full lines correspond to the surface $\pm q_1$ and dashed lines to $\pm q_2$. The two vectors $q_1(\mathbf{q}_{\parallel}^c, E_F)$ and $q_2(\mathbf{q}_{\parallel}^c, E_F)$ are indicated.

FIG. 2. RKKY periods as a function of E_F . Curves 1,2,3,4, and 5 correspond to $\lambda_{2,0}^a$, $\lambda_{1,1}^b$, $\lambda_{2,0}^c$, $\lambda_{0,2}^c$, and $\lambda_{1,1}^c = \lambda_{1,1}^d$, respectively. The dot-dashed curve is a new period $\lambda_{3,1}^c$.

FIG. 3. Exchange coupling as a function of the spacer thickness for $E_F/2t_0 = 1.64$, $V_{ex} = 0.15W$ and $k_BT = 2.0 \times 10^{-3}W$ (full line). The dashed line corresponds to the RKKY result scaled down by a factor of 8. The inset shows the ratio $|C_{3,1}|/|C_{1,1}|$ (full line), $|C_{4,2}|/|C_{1,1}|$ (dashed line), and $|C_{4,0}|/|C_{1,1}|$ (dot-dashed lined) as a function of V_{ex} , for \mathbf{q}_{\parallel}^c .

FIG. 4. Contributions to the coupling coming from the non-RKKY period $\lambda_{3,1}^c$ (full line), and two RKKY periods $\lambda_{2,0}^a$ (dashed line) and $\lambda_{2,0}^c$ (dot-dashed line) (see text).







