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Oscillations in the exchange coupling of ferromagnetic layers separated by a non-magnetic metallic layer

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Abstract. A general theory of oscillations in the exchange coupling J between two transition metal ferromagnets separated by a non-magnetic transition metal spacer is developed. Detailed calculations of J as a function of the spacer layer thickness are made for a specific model based on a simple cubic tight-binding band structure. For a suitable choice of band filling a good account is given of the main features observed in Co/Ru, Co/Cr and Fe/Cr layered structures. An analysis of the numerical accuracy required in computing J from a total energy difference is given and this throws some light on difficulties experienced by previous workers using this approach. A general expression for J is obtained for an arbitrary band and finite temperature in the limit of large spacer thickness. A close analogy between oscillations in the exchange coupling and de Haas–van Alphen oscillations is established and the relation to RKKY theory is also discussed. It is shown that the period, asymptotic decay and temperature dependence of the oscillations in J are determined by properties of the Fermi surface of the spacer layer.

1. Introduction

Antiferromagnetic coupling between the iron layers in Fe/Cr/Fe sandwiches has been observed in light scattering experiments by Grünberg *et al* (1986) and confirmed by the SPLEED measurements of Carbone and Alvarado (1987). Similar behaviour was found in Fe/Cr superlattices, by means of magnetization measurements and neutron diffraction (Baibich *et al* 1988). Typically the antiferromagnetic coupling was observed in these experiments when the thickness of the chromium layer was less than 20–30 Å. Recently Parkin *et al* (1990) reported long-period oscillations in the exchange coupling as a function of the thickness of the non-magnetic layer in Co/Ru, Co/Cr and Fe/Cr superlattice structures. Similar well-defined oscillations with a somewhat shorter period were observed earlier in Gd/Y superlattices by Majkrzak *et al* (1986) and interpreted in terms of RKKY coupling by Yafet (1987). The oscillations observed by Parkin *et al* (1990) are also reminiscent of RKKY oscillations but the period, 15–20 Å in all cases, is unexpectedly long. The observed coupling energy is far too large to be explained by magnetostatic interactions. The measurements of Parkin *et al* (1990) are particularly important since they provide us with a large number of specific results to be explained by theory. The

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period, amplitude, initial sign, asymptotic rate of decay and temperature dependence of the coupling were all determined by Parkin *et al* (1990) for Co/Ru, Co/Cr and Fe/Cr structures.

Existing quantitative theories (Levy *et al* 1990, Hasegawa 1990, Stoeffler *et al* 1991) using realistic band structures are restricted to spacer layers of only a few atomic planes and are valid only at zero temperature. Calculations of the magnitude of the coupling by comparing the total energies of the ferromagnetic and antiferromagnetic configurations are numerically difficult (Hasegawa 1990) and do not lead to results of the correct order of magnitude (Levy *et al* 1990, Stoeffler *et al* 1991). Moreover, the physical origin of the computed effects remains obscure.

In this paper we present a theory of the exchange coupling of two transition metal ferromagnets separated by a non-magnetic transition metal spacer layer. The theory, even in its simplest form (Edwards and Mathon 1991), accounts qualitatively correctly for all the features of the exchange coupling observed by Parkin *et al* (1990). The origin of the effect is explained and it is made clear how the present theory can be used to predict specific materials and layer orientations for which long-period oscillations of the exchange coupling should occur.

The plan of the paper is as follows. In section 2 a new mechanism of the exchange coupling across a transition metal spacer is proposed and a mathematical model of the coupling is formulated. The model is then applied in section 3 to a (100) sandwich with a simple cubic tight-binding band at $T = 0$. An exact analytic formula for the exchange coupling is derived and investigated for large thicknesses of the spacer layer. A connection with RKKY is also briefly discussed. In section 4 we consider a more general band and arbitrary layer orientations at finite temperatures. A close analogy between our theory and the theory of de Haas–van Alphen effect is established and a quite general asymptotic formula for the exchange coupling is derived. It relates the period, amplitude, rate of decay and temperature dependence of oscillations in the coupling to the properties of the Fermi surface in the spacer layer. In section 5 the results of numerical calculations of the exchange coupling both at $T = 0$ and at finite temperatures using a simple cubic tight-binding band are presented and compared qualitatively with experiment. Also in section 5 the exact asymptotic results of sections 3 and 4, together with our own numerical studies, are used to examine critically the existing numerical calculations of the exchange coupling referred to earlier. Finally, the formulation of the RKKY interaction for planar structures is discussed and the RKKY range function in the planar geometry is evaluated for a simple cubic tight-binding band. The results are compared with our theory of the exchange coupling.

2. Exchange coupling mechanism

We consider a sandwich consisting of two thick layers of a ferromagnetic transition metal separated by N atomic planes of a non-magnetic transition metal. The ferromagnetic metal is assumed to have a full majority spin d band and a partially occupied spin minority d band. The nonmagnetic metal has equal numbers of holes in each spin sub-band. The spin sub-bands in the ferromagnetic and non-magnetic spacer layers, together with the hole densities $\rho_\uparrow$, $\rho_\downarrow$, are shown schematically in figures 1(a) and 1(b) for the parallel and antiparallel orientations of the ferromagnetic layer moments. For simplicity we assume that the number of d holes per atom of each spin in the bulk non-magnetic metal is equal to the number of holes in the bulk ferromagnetic metal. The basic mechanism

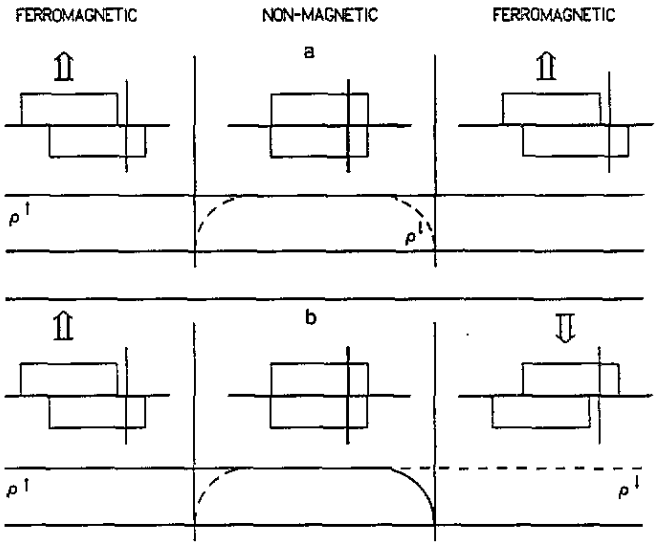

Figure 1. Schematic representation of the densities of states in the d band and of the densities of holes in a sandwich for ferromagnetic (a) and antiferromagnetic (b) alignments of the magnetic layers.

for exchange coupling described below does not depend on this precise condition. However, it is a reasonable approximation to the actual situation in Fe/Cr, Co/Ru and Co/Cr systems.

It is clear from figure 1 that deviations from bulk hole densities occur in the spacer layer near the interfaces with the ferromagnetic layers. For the parallel configuration, figure 1(*a*), both interface effects occur in the $\downarrow$-spin hole density and, therefore, interfere with each other. In the antiparallel configuration, however, the interface effects at each end of the spacer layer occur in opposite spin densities and no interference takes place. 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 effect in the down-spin band of the spacer layer.

To investigate this effect quantitatively we use a single-orbital tight-binding Hamiltonian

$$H = \sum_{i,j,\sigma} t_{ij} c_{i\sigma}^{+} c_{j\sigma} + \sum_{i} U_i n_{i\uparrow} n_{i\downarrow} \tag{2.1}$$

where $c_{i\sigma}^{+}$ creates a hole of spin σ on site i and $n_{i\sigma} = c_{i\sigma}^{+} c_{i\sigma}$. We assume the hopping parameters t_{ij} are the same in both metals and $U_i = \infty$ for sites i in the ferromagnetic layers and $U_i = 0$ in the spacer layer. The choice $U_i = \infty$ in the ferromagnetic layers means that there exist exact single-determinant eigenfunctions of the Hamiltonian corresponding to the configurations shown in figures 1(*a*) and 1(*b*). This is because the interactions U_i play no role; electrons of opposite spin completely avoid each other except in the spacer layer where $U_i = 0$. The total energies of the two configurations may, therefore, be calculated as sums of one-electron energies. To do this, we initially maintain the bulk Fermi energy E_F everywhere. It is convenient to introduce the total energy $E_{tot}(N)$ of holes of one spin confined in N atomic planes and the energy E_B per

atomic plane of holes of one spin in the bulk. If we suppose that each ferromagnetic layer in the sandwich contains M atomic planes, where M is large, then the energies of the configurations of figures 1(a) and 1(b) are, respectively, given by

$$E_{\uparrow\uparrow} = E_{\rm tot}(N) + (2M + N)E_{\rm B} \tag{2.2}$$

$$E_{\downarrow\uparrow} = E_{\rm tot}(2M + 2N). \tag{2.3}$$

The energy difference is therefore

$$\Delta E(N) = E_{\uparrow\uparrow} - E_{\uparrow\downarrow} = E(N) - E(2M + 2N) \tag{2.4}$$

where

$$E(N) = E_{\rm tot}(N) - NE_{\rm B} \tag{2.5}$$

As $M \rightarrow \infty$,

$$\Delta E(N) = E(N) - E(\infty) \tag{2.6}$$

and clearly from (2.5) $E(N)$ may be considered as the energy of holes of one spin confined to N atomic planes measured relative to an N-plane reference state with bulk density. Thus $E(N)$ describes only the end effects and the energy difference in (2.6) corresponds just to the energy associated with the interference between the two end effects.

To conserve the number of particles in calculating the energy difference we must consider the changes in the number of holes that result from keeping $E_{\rm F}$ fixed. If $\nu(N)$ is the number of holes of one spin confined in N atomic planes and $n_{\rm B}$ is the number of holes per bulk plane, we define

$$n(N) = \nu(N) - Nn_{\rm B} \tag{2.7}$$

in analogy with (2.5). We now introduce the thermodynamic potential

$$\Omega(N) = E(N) - E_{\rm F}n(N) \tag{2.8}$$

which, like $E(N)$, is measured relative to a reference state with N bulk planes. In analogy with (2.6) the energy difference between the ferromagnetic and antiferromagnetic configurations of the sandwich with particle numbers conserved is given by

$$\Delta\Omega(N) = \Omega(N) - \Omega(\infty). \tag{2.9}$$

Following Parkin *et al* (1990), we define an exchange coupling constant for a spacer layer containing N atomic planes by

$$J(N) = \Delta\Omega(N)/A \tag{2.10}$$

where A is the area of an atomic plane.

The above considerations are readily generalized to a superlattice with alternating magnetic and non-magnetic layers containing, respectively, M and N atomic planes. The energy difference per magnetic layer between the ferromagnetic and antiferromagnetic configurations is then

$$\Omega(N) - \Omega(2N + M). \tag{2.11}$$

The difference between this result and $\Delta\Omega(N)$ given by (2.9) is small for large M. Even for small M the difference is not significant for large N since it emerges in section 3 that $\Omega(N) \simeq 1/N^2$ and hence

$$|\Omega(N) - \Omega(2N)| \simeq (3/4)\,|\Omega(N) - \Omega(\infty)|. \tag{2.12}$$

In this paper we focus our attention primarily on $\Delta\Omega(N)$ given by (2.9).

In the next section we calculate $\Omega(N)$, and hence $J(N)$, for a simple cubic band at $T = 0$ and in section 4 we consider a more general situation at finite temperatures.

3. Model with a simple cubic tight-binding band at $T = 0$

We consider a simple cubic lattice with basic vectors $a(1, 0, 0)$, $a(0, 1, 0)$ and $a(0, 0, 1)$ and take the atomic planes of the sandwich to be perpendicular to the third of these vectors. We take $t_{ij} = t$ $(t < 0)$ for i, j nearest neighbours and $t_{ij} = 0$ otherwise. It is convenient to consider a non-magnetic layer with $N - 1$ atomic planes rather than N and we label them by integers $1, 2, \ldots, N - 1$. To calculate $\Omega(N - 1)$ we must consider the electrons as confined within the non-magnetic layer so that their wave functions vanish on planes 0 and N. The normalized wave functions are, therefore, of the form

$$\sum_{l=-\infty}^{\infty} \sum_{m=-\infty}^{\infty} \sum_{p=1}^{N-1} \exp[\mathrm{i}(k_x l + k_y m)a] \sin \frac{r\pi p}{N} |lmp\rangle \tag{3.1}$$

where $|lmp\rangle$ is the orbital on site $a(l, m, p)$ and the wave function is classified by the two-dimensional wave vector (k_x, k_y) and a discrete quantum number $r = 1, 2, \ldots, N - 1$. The corresponding one-electron energy is

$$\varepsilon(k_x, k_y, r) = -(\cos(r\pi/N) + \cos(k_x a) + \cos(k_y a)). \tag{3.2}$$

Throughout this section we measure all energies in units of $2|t|$. Clearly at $T = 0$

$$\begin{aligned}\Omega_{\mathrm{tot}}(N - 1) &= \Omega(N - 1) + \Omega_{\mathrm{ref}}(N - 1) \\ &= \sum_{r, k_x, k_y} (\varepsilon(k_x, k_y, r) - E_{\mathrm{F}})\, \theta(E_{\mathrm{F}} - \varepsilon(k_x, k_y, r))\end{aligned} \tag{3.3}$$

where $\theta(x)$ is the unit step function and

$$\Omega_{\mathrm{ref}}(N - 1) = (N - 1)(E_{\mathrm{B}} - n_{\mathrm{B}} E_{\mathrm{F}}) \tag{3.4}$$

corresponds to the reference state. This term just cancels the part of the right-hand side of (3.3) that is proportional to $N - 1$, and the part that tends to a constant as $N \to \infty$ cancels out in calculating $\Delta\Omega(N - 1)$ from (2.9). Hence $\Delta\Omega(N - 1)$ is given by the part of $\Omega_{\mathrm{tot}}(N - 1)$ that tends to zero as $N \to \infty$.

The summation over k_x, k_y in (3.3) can be turned into an energy integral involving the two-dimensional density of states $N_{2\mathrm{D}}(E)$

$$\Omega_{\mathrm{tot}}(N - 1) = \sum_{r=1}^{N-1} \int \mathrm{d}E\, N_{2\mathrm{D}}(E)\, (-\cos(r\pi/N) + E - E_{\mathrm{F}})\, \theta(E_{\mathrm{F}} + \cos(r\pi/N) - E). \tag{3.5}$$

This expression may be used to determine $\Delta\Omega(N - 1)$ numerically, as is discussed in section 5. However, it is instructive to obtain some analytical results by approximating the density of states $N_{2\mathrm{D}}(E)$ as follows:

$$N_{2\mathrm{D}}(E) \simeq \begin{cases} (1/2\pi) N_{\parallel} & |E| < 2 \\ 0 & |E| > 2. \end{cases} \tag{3.6}$$

Here, $N_{\parallel}$ is the number of lattice sites in one atomic plane. This is an exact representation

of $N_{2D}(E)$ at the band edges but underestimates it elsewhere. However, numerical calculations show that such an approximation reproduces all the essential features of $\Delta\Omega(N-1)$ when $-3 < E_F < -1$. Furthermore, with this approximation the asymptotic form of $\Delta\Omega(N-1)$ for large N is obtained exactly. We now proceed to evaluate Ω_{tot} for $-3 < E_F < -1$ with the approximation (3.6).

On evaluating the integral in (3.5) we find

$$\Omega_{tot}(N-1) = -\frac{N_\parallel}{4\pi}\sum_{r=1}^{\infty} f(r) \tag{3.7}$$

with

$$f(x) = (\cos^2(\pi x/N) + 2\alpha\cos(\pi x/N) + \alpha^2)\,\theta(\xi - x) \tag{3.8}$$

where $\alpha = E_F + 2$ and

$$\xi = (N/\pi)\cos^{-1}(-\alpha) \qquad 0 \leqslant \xi < N. \tag{3.9}$$

The summation in (3.7) may be carried out explicitly but the result is not very illuminating. It is more convenient to use the Poisson summation formula

$$\tfrac{1}{2}f(0) + \sum_{r=1}^{\infty} f(r) = g(0) + 2\sum_{s=1}^{\infty} g(2\pi s) \tag{3.10}$$

where

$$g(t) = \int_0^{\infty} f(\xi)\cos(\xi t)\,\mathrm{d}\xi. \tag{3.11}$$

Hence

$$g(t) = \frac{2\sin(\xi t)}{t}\left(\frac{2\alpha^2 - 1}{(Nt/\pi)^2 - 4} - \frac{\alpha^2}{(Nt/\pi)^2 - 1}\right) + \frac{6N\alpha(1-\alpha^2)^{1/2}\cos(\xi t)}{\pi[(Nt/\pi)^2 - 1][(Nt/\pi)^2 - 4]}. \tag{3.12}$$

As discussed earlier $\Delta\Omega(N-1)$ is the part of $\Omega_{tot}(N-1)$ that tends to zero as $N \to \infty$ and this corresponds to replacing the sum in (3.7) by the last term of (3.10). Hence

$$\Delta\Omega(N-1) = -\frac{N_\parallel}{2\pi}\sum_{s=1}^{\infty} g(2\pi s). \tag{3.13}$$

For a spacer layer containing more than about three atomic planes it is a good approximation to use the asymptotic form of each term in (3.12) thus neglecting terms of the order $1/N^4$. Using (3.9) and (2.10) with $A = N_\parallel a^2$, we then obtain the exchange coupling $J(N-1)$ in the form

$$J(N-1) = \frac{1}{8\pi^2 N^2 a^2}\sum_{s=1}^{\infty}\left(\pm\frac{(E_F+1)(E_F+3)}{s^3}\sin\frac{2\pi s N}{N_p}\right.$$
$$\left. - \frac{3}{2Ns^4}(E_F+2)[-(E_F+1)(E_F+3)]^{1/2}\cos\frac{2\pi s N}{N_p}\right) \tag{3.14}$$

where + holds for $E_F > -2$ and − holds for $E_F < -2$, and N_p is given by

$$N_p = \pi/\cos^{-1}|2 + E_F|. \tag{3.15}$$

As $E_F \to -1$, $J(N-1) \to 0$ and, in fact, we find that in the present approximation

$\Delta\Omega(N-1) = 0$ throughout the region $-1 < E_F < 0$. However, it will be shown in section 4 that the latter result is the consequence of the approximation (3.6) to $N_{2D}(E)$.

It is clear from (3.14) that the exchange coupling J oscillates with period N_p and with an amplitude that decays as $1/N^2$ for large N. The period becomes long when E_F is near -3 or -1. The former case corresponds to a very low hole density with a small almost spherical Fermi surface. The latter case is more interesting and corresponds to the Fermi surface nearly touching the zone boundary. In the present model the coupling strength weakens as the period lengthens because of the factor $(E_F + 1)(E_F + 3)$ in (3.14). The relation between the period and the caliper measurement of the Fermi surface, and between the amplitude and the curvature of the Fermi surface at its extremal points, is discussed for a general band in section 4. An asymptotic expression for $J(N-1)$, correct to order $1/N^2$, is derived there for a general band. It agrees with the first term of (3.14) for the present model. The close relationship between this asymptotic result and RKKY coupling is also discussed in section 5. From the RKKY point of view the overall strength of the coupling should depend on a product of the appropriate susceptibility of the spacer layer material and the square of the exchange coupling between the magnetic and non-magnetic layers. It is not clear how to calculate the latter coupling in general and an advantage of the present method is that this problem does not arise. The numerical calculations described in section 5 show that the overall strength of the coupling J obtained in our approach is of the right order of magnitude to agree with experiment.

Finally we point out that the close correspondence with RKKY is restricted to the asymptotic limit where only terms of order $1/N^2$ are important. This is clearly seen in the low-density limit when $E_F + 3$ is small. The spacer layer of thickness $d = Na$ then corresponds to a gas of holes with a spherical Fermi surface of radius k_F given by $k_F^2 a^2/2 = E_F + 3$. The leading term ($s = 1$) in the sum (3.14) is then equal to a positive constant times a factor

$$-(1/d^2)(\sin(2k_F d) + 3\cos(2k_F d)/2k_F d). \tag{3.16}$$

A simple RKKY theory of the exchange coupling between two planes of spins, distance d apart, in the same spacer medium yields the following expression for the corresponding factor:

$$-(1/d^2)(\sin(2k_F d) - \cos(2k_F d)/2k_F d). \tag{3.17}$$

4. Generalization of the model to an arbitrary band and finite temperatures

We showed in section 3 that the calculation of the exchange coupling in a sandwich reduces to the calculation of the total energy of carriers trapped in the non-magnetic spacer by the exchange potentials of the ferromagnetic layers. Since the exchange potentials are equivalent in our model to two infinitely high potential barriers, we simply require the dependence of the total energy of size-quantized carriers 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. In the case of de Haas–van Alphen oscillations, the carrier energy is quantized by a magnetic field in a plane perpendicular to the field whereas, in the present problem, one-dimensional quantization in the direction perpendicular to the sandwich is induced by the exchange potential.

It is well known that the amplitude, period and temperature dependence of de Haas–van Alphen oscillations are linked directly to the shape of the Fermi surface. It is our

aim to show that a similar general relationship exists between oscillations of the exchange coupling and the shape of the Fermi surface of carriers in the spacer layer.

Our calculation is an adaptation of the conventional theory of the de Haas–van Alphen effect (see, e.g., Abrikosov 1972). As in section 3, we consider the thermodynamic potential of carriers confined in the spacer layer

$$\Omega_{\text{tot}} = -T\sum_{k,r} \ln\left(1 + \exp\frac{\mu - E(k, r)}{T}\right) \tag{4.1}$$

where μ is the chemical potential, $E(k, r)$ is the carrier energy, k is the wavevector parallel to the sandwich, and r labels the size-quantized levels ($k_B = 1$). At $T = 0$, we have $\mu = E_F$ and (4.1) clearly reduces to Ω_{tot} defined in section 2.

To evaluate the sum in (4.1), we need to know the explicit dependence of $E(k, r)$ on the discrete quantum number r. Without loss of generality, we may assume that k lies in the (x, y) plane and quantization takes place in the z direction. We then know from section 3 that the quantization of the energy for a SC tight-binding band reduces to the quantization $k_z = r\pi/Na$ in the expression for the bulk energy $\varepsilon(k_x, k_y, k_z)$. One can easily show that the same quantization holds for any single band and any orientation of the sandwich. Hence we have quite generally

$$E(k_x, k_y, r) = \varepsilon(k_x, k_y, r\pi/Na) \tag{4.2}$$

where a is the separation between two neighbouring atomic planes in the spacer layer.

As in section 3, we can replace the sum over r in (4.1) by an integral using Poisson's summation formula and use the result already proved in section 3 that the oscillatory contribution to Ω_{tot} is in fact the required exchange energy $\Delta\Omega$. The exchange energy per unit area is, therefore, given by

$$J = -\frac{2T}{(2\pi)^2}\,\text{Re}\sum_{s=1}^{\infty}\iint_{\text{BZ}} dk_x\, dk_y \times \int_0^{N-1} d\zeta \ln\{1 + \exp[(\mu - \varepsilon(k_x, k_y, \zeta\pi/Na))/T]\}\exp(2\pi is\zeta) \tag{4.3}$$

where the integral with respect to k_x, k_y is over the two-dimensional Brillouin zone in the sandwich plane.

After integration by parts the real part of the integral over ζ becomes:

$$\text{Re}\left(\frac{1}{2\pi isT}\int_0^{N-1} d\zeta \frac{\partial\varepsilon}{\partial\zeta}\{1 + \exp[(\varepsilon(k_x, k_y, \zeta) - \mu)/T]\}^{-1}\exp(2\pi is\zeta)\right). \tag{4.4}$$

To obtain (4.4) we used the result that the Fermi function is approximately zero for ε at the top of the band. This is valid provided the distance from the Fermi level to the top of the band is much larger than T.

We can now change the integration over ζ to an integration over energy. The exchange coupling then takes the form

$$J = -\frac{1}{4\pi^3}\,\text{Re}\sum_{s=1}^{\infty}\frac{1}{si}\int_0^{N-1} d\varepsilon\,\{1 + \exp[(\varepsilon - \mu)/T]\}^{-1}\iint_{\text{BZ}} dk_x\, dk_y \exp(2isNak_z). \tag{4.5}$$

We shall next evaluate the two-dimensional integral with respect to k_x, k_y. The component k_z in this integration is a dependent variable and we can use the bulk tight binding

energy formula $\varepsilon(k_x, k_y, k_z)$ to express $k_z \equiv k_z(\varepsilon, k_x, k_y)$. It is clear that for large N the factor $\exp(2isNak_z)$ is a rapidly oscillating function and only those regions in the (k_x, k_y) plane in which k_z is stationary with respect to k_x, k_y will make significant contributions to the integral. We can, therefore, apply the method of stationary phase in which the required integral is approximated by expanding k_z in a Taylor series about its stationary points up to second-order terms.

Let us assume that $(k_x^0(\varepsilon), k_y^0(\varepsilon))$ is a stationary point. It can be always achieved by a suitable rotation in the (k_x, k_y) plane that the Taylor expansion about $(k_x^0(\varepsilon), k_y^0(\varepsilon))$ takes the simple form

$$k_z(\varepsilon, k_x, k_y) = k_z(\varepsilon, k_x^0, k_y^0) + \tfrac{1}{2}[(\partial^2 k_z/\partial k_x^2)(k_x^0 - k_x)^2 + (\partial^2 k_z/\partial k_y^2)(k_y^0 - k_y)^2] + \ldots. \tag{4.6}$$

When this result is substituted in (4.5) the two-dimensional integral in the (k_x, k_y) plane separates into two independent integrals with respect to k_x and k_y and each of them can be evaluated exactly. We obtain

$$\iint_{\mathrm{BZ}} \mathrm{d}k_x\, \mathrm{d}k_y \exp(2isNak_z) \simeq \sigma \frac{\pi}{sNa} \left| \frac{\partial^2 k_z}{\partial k_x^2} \frac{\partial^2 k_z}{\partial k_y^2} \right|^{-1/2} \exp(2isNak_z^0) \tag{4.7}$$

where k_z^0 is the stationary value of k_z and both the partial derivatives are taken at the stationary point $(k_x^0(\varepsilon), k_y^0(\varepsilon))$. The factor σ is equal to $\exp(i\pi/2)$ when both derivatives in (4.7) are positive, it is equal to $\exp(-i\pi/2)$ when they are negative and $\sigma = 1$ when the two derivatives have opposite signs.

It remains to perform the energy integral in (4.5). The integrand contains the product of the Fermi function and the factor $\exp(2isNak_z^0)$ which depends on energy via k_z^0. The region in which the Fermi function is essentially constant does not contribute to the integral because of rapid oscillations of the function $\exp(2isNk_z^0)$. The only significant contribution comes from a narrow interval around the Fermi surface where the Fermi function varies rapidly. Since k_z^0 is a slowly varying function in this interval, it can be expanded about μ:

$$k_z^0(\varepsilon) \simeq k_z^0(\mu) + (\partial k_z/\partial \varepsilon)(\varepsilon - \mu) + \ldots. \tag{4.8}$$

Substituting the expansion (4.8) and the result (4.7) in (4.5) it is straightforward to evaluate the energy integral in (4.5). This leads to the following general asymptotic formula for the exchange coupling J defined by (2.10):

$$J(N-1) = \frac{1}{4\pi Na} \mathrm{Re} \sum_{s=1}^{\infty} \frac{\sigma}{s^2} \left| \frac{\partial k_z^2}{\partial k_x^2} \frac{\partial k_z^2}{\partial k_y^2} \right|^{-1/2} \frac{\exp(2isNak_z^0(\mu))}{T^{-1} \sinh(2\pi sNaT\, \partial k_z/\partial \varepsilon)} \tag{4.9}$$

where all the derivatives are taken at the stationary point and for $\varepsilon = \mu$. Naturally, the contributions of all the stationary points of k_z need to be included in (4.9).

It is clear from (4.9) that $J(N)$ oscillates with a period N_p given by

$$N_p = \pi/ak_z^0(\mu) \tag{4.10}$$

where $k_z^0(\mu)$ is the caliper measurement of the Fermi surface in the direction perpendicular to the layers. The temperature dependence of J is determined by the velocity of carriers at the stationary points on the Fermi surface and the oscillation amplitude by the curvature of the Fermi surface at the stationary points.

As an illustration, and as a check on the general formula (4.9), we now apply it at $T = 0$ to the simple cubic band already investigated in section 3. We assume again that the z direction is normal to the layers and along a cubic axis. The Fermi surface for such a band is given by

$$E_F = -(\cos(k_x a) + \cos(k_y a) + \cos(k_z a)). \tag{4.11}$$

It is, therefore, clear that the stationary points of $k_z(E_F, k_x, k_y)$ are at $(0, 0)$, $(0, \pm\pi/a)$, $(\pm\pi/a, 0)$ and $(\pm\pi/a, \pm\pi/a)$. Since the band is symmetric about $E = 0$, there are just two cases to be considered: $-3 < E_F < -1$ and $-1 < E_F < 0$. In the former case, the only relevant stationary point is $k_x = k_y = 0$. Evaluating all the required derivatives in (4.9) at this point we find that both the second derivatives are negative, which means that $\sigma = -i$. Taking then the limit $T \to 0$, it is straightforward to show that (4.9) reduces exactly to the first term in (3.14) of section 3.

Consider now the interval $-1 < E_F < 0$. Using the approximation of a constant two-dimensional density of states (DOS), we found in section 3 no exchange coupling in this interval. This is because the constant two-dimensional DOS corresponds to a band with a parabolic dispersion in the (k_x, k_y) plane and it is clear from (4.11) that k_z for such a band has no extremum at $k_x = k_y = 0$ for $E_F > -1$. The exact treatment of the band (4.11) for $-1 < E_F < 0$ shows that additional extrema occur at $(0, \pm\pi/a)$ and $(\pm\pi/a, 0)$ and they are, in fact, the only extrema in this range of E_F. It is easy to show that the second derivatives in (4.9) have opposite signs at all these stationary points (saddle points), which means that $\sigma = 1$. Hence, in the range $-1 < E_F < 0$, the asymptotic formula for J at $T = 0$ becomes

$$J(N-1) \simeq \frac{E_F^2 - 1}{4\pi^2 N^2 a^2} \sum_{s=1}^{\infty} \frac{1}{s^3} \cos \frac{2\pi s N}{N_p} \tag{4.12}$$

where the oscillation period N_p is defined by

$$N_p = \pi / \cos^{-1}|E_F|. \tag{4.13}$$

It is clear that oscillations of the exchange coupling with a long period occur not only for $E_F < -1$ (as already shown in section 3) but also for $E_F > -1$.

We shall now return to the general formula (4.9). One can see from (4.9) that the amplitude of oscillations in the exchange coupling decreases with increasing temperature in the same way as the amplitude of de Haas–van Alphen oscillations. In fact, the sinh factor has exactly the same form as in the de Haas–van Alphen effect (Abrikosov 1972) if we make the correspondence

$$(N/\pi)\, \partial(k_z^0 a)/\partial \varepsilon \to 1/\hbar\omega_c \tag{4.14}$$

where ω_c is the cyclotron frequency. Hence the equivalent 'cyclotron' frequency in the present problem is $\hbar\omega_c \simeq W/2N$, where W is the band width. For $N \simeq 5$–10 and $W \simeq 3$ eV, we have $\hbar\omega_c \simeq 10^3$ K corresponding to an equivalent field of about 10^3 T. Taking account of the numerical factors in the sinh factor, the temperature dependence our theory predicts is on the scale $\simeq 10^2$ K, as observed by Parkin *et al* (1990).

5. Numerical studies for the simple cubic band and comparison with experiment

5.1. Exchange coupling at short distances

The general asymptotic formula (4.9) becomes poor for spacer layer thicknesses <5–10 atomic planes. To determine the initial sign and initial strength of the exchange coupling

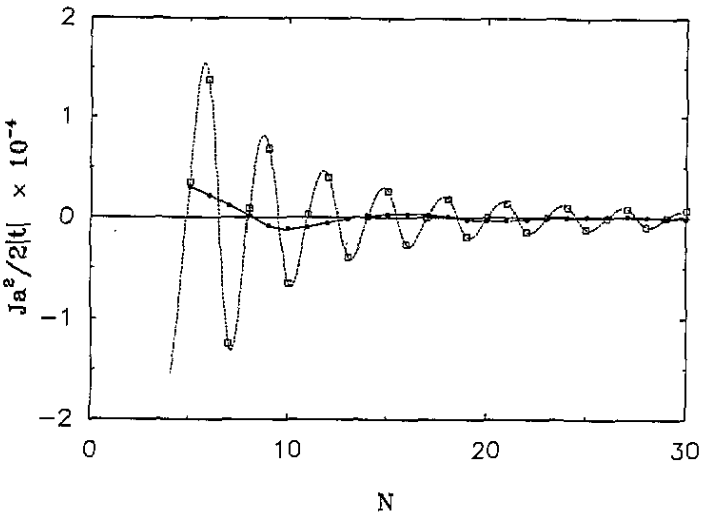

Figure 2. Comparison of the amplitudes and periods of oscillations in the exchange coupling J as a function of the number of atomic planes N in the spacer layer for two different fillings of the band: $E_F = -2.5$ *(squares) and* $E_F = -1.05$ *(circles).*

$J(N)$, we need to evaluate numerically either (3.3) or (3.5). Direct evaluation of Ω_{tot} in (3.5) using the full two-dimensional tight-binding density of states for a square lattice is straightforward and the $J(N)$ curves for two values of the Fermi energy $E_F = -2.5$ and $E_F = -1.05$ are shown in figure 2.

There are three important differences between the two curves $J(N)$ shown in figure 2. For the lower value of E_F the period is only three atomic planes but it becomes ten atomic planes for $E_F = -1.05$ when the Fermi surface nearly touches the zone boundary. Also the sign of the interaction at short distances, which is ferromagnetic for $E_F = -2.5$, changes to antiferromagnetic for $E_F = -1.05$. Finally, the amplitude of oscillations with a long period is much smaller than that of oscillations with a short period. The long period predicted by our model for $E_F = -1.05$ and the initially antiferromagnetic coupling are as observed by Parkin *et al* (1990) for Co/Ru structures.

It remains to estimate the magnitude of the effect. Parkin *et al* (1990) found $J \approx 6$ erg cm^{-2} for Co/Ru system with the thickness of the Ru layer of 3 Å corresponding to two Ru monolayers. Setting $N = 3$ in (3.5) and using the value $E_F = -1.05$ leading to long-range oscillations, we obtain

$$J \approx 1 \text{ erg cm}^{-2} \tag{5.1}$$

where we have used a typical value $W \approx 3$ eV for the band width $W = 12|t|$ of a transition metal. Given the simplicity of the single-orbital model used, we consider this value of J to be in good agreement with the observed result.

Although the long-period oscillations for $E_F = -1.05$ lead to an antiferromagnetic sign of the coupling at short distances, ferromagnetic coupling at short distances combined with a long period can also occur. This case occurs in our model when E_F is just above the value $E_F = -1$ and is illustrated in figure 3 where the exchange couplings for $E_F = -1.05$ and $E_F = -0.95$ are compared.

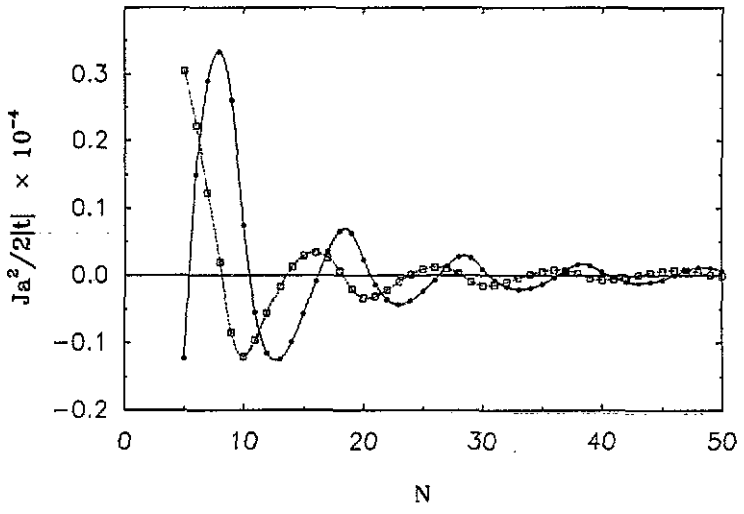

Figure 3. Comparison of the exchange coupling $J(N)$ for two different values of the Fermi energy close to the Brillouin zone boundary: $E_F = -0.95$ (circles) and $E_F = -1.05$ (squares).

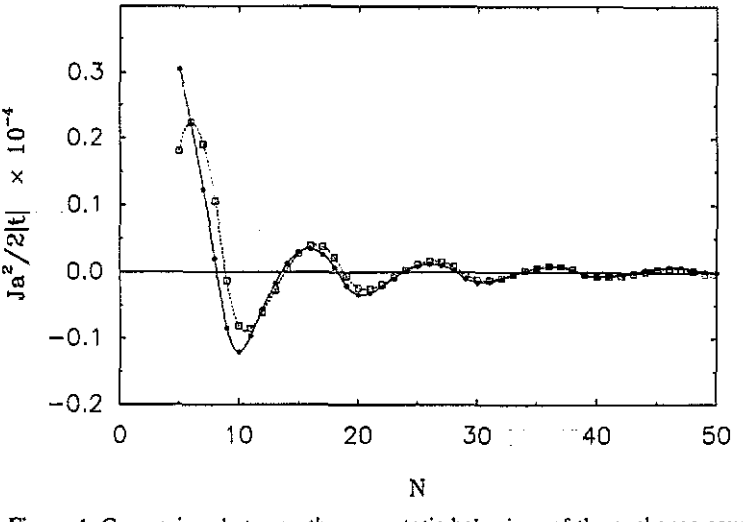

Figure 4. Comparison between the asymptotic behaviour of the exchange coupling $J(N)$ obtained from (4.9) for $E_F = -1.05$ (squares) and the exact result obtained from (3.5) (circles).

To conclude this subsection, we show in figure 4 the exchange coupling $J(N)$ for $E_F = -1.05$ evaluated from the general asymptotic formula (4.9) (broken curve) together with the exact result obtained from (3.5).

5.2. *Temperature dependence of the exchange coupling*

One of the most interesting and surprising results reported by Parkin *et al* (1990) is a strong temperature dependence of the exchange coupling on the scale $\approx 10^2$ K. As

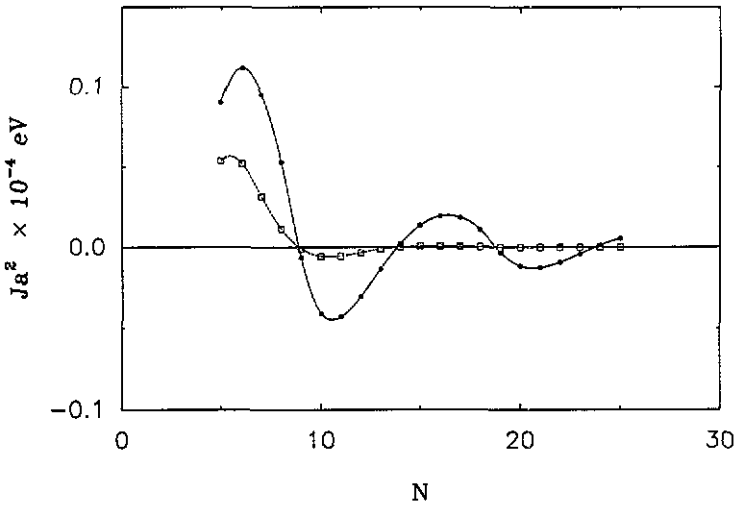

Figure 5. Temperature dependence of the exchange coupling $J(N)$ for $E_F = -1.05$: $T = 0$ (circles) and $T = 50$ K (squares).

already discussed qualitatively in section 4, a temperature dependence on the same scale is also predicted by our theory. It is interesting that a strong temperature dependence of J goes, at least for our simple one-band model, hand in hand with a long period and small amplitude of oscillations in the exchange coupling. This is because the temperature dependence of J is determined by the velocity of carriers $\partial\varepsilon/\partial k_z$ at the Fermi surface which tends, together with the amplitude of oscillations, to zero when the Fermi surface touches the zone boundary.

The exchange coupling J at $T = 50$ K determined from (4.9) is compared in figure 5 with J at $T = 0$. Both curves are for $E_F = -1.05$. It can be seen that a significant reduction of J is obtained even at such a low temperature. Our calculated temperature dependence is stronger than observed by Parkin *et al* (1990). This is not surprising since both the amplitude and $\partial\varepsilon/\partial k_z$ are proportional for a simple cubic tight-binding band to the same factor $(E_F + 1)(E_F + 3)$, and we already know from section 5.1 that such a model underestimates the amplitude of J by a factor ≈ 6. The temperature dependence of J is, therefore, overestimated by the same factor.

5.3. Direct calculation of the difference in energy between the ferromagnetic and antiferromagnetic configurations

The most direct way of calculating the exchange coupling is clearly to compute by brute force the difference in energy $\Delta\Omega$ between the ferromagnetic and antiferromagnetic configurations of a sandwich/superlattice. This has been tried for Fe/Cr structures using the local spin density functional (Levy *et al* 1990) and tight-binding (Hasegawa 1990) methods. Stoeffler *et al* (1991) and Stoeffler and Gautier (1991) also used tight-binding approximation to calculate ΔE for Fe/V, Co/Pd, Co/Ru and Fe/Cr systems. All these calculations are for spacer layers of several atomic planes only and the magnitude of the exchange coupling J obtained by Levy *et al* (1990) and by Stoeffler *et al* (1991) and Stoeffler and Gautier (1991) is much larger than observed. Hasegawa concluded that

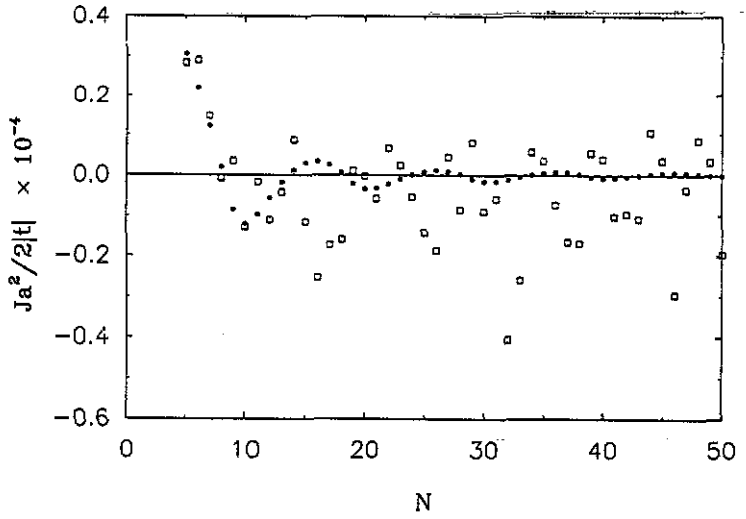

Figure 6. Exchange coupling $J(N)$ obtained by direct evaluation of (3.3) using different numbers of Cunningham special k-points in the two-dimensional Brillouin zone. Squares are for 16640 k-points and circles are for 1050624 k-points. Both results are for $E_F = -1.05$.

ΔE was too small to be determined accurately by his method. Moreover, the sign of J changes in the calculations of Stoeffler *et al* (1991) over distances of two to three atomic planes, which disagrees with the long periods observed by Parkin *et al* (1990) for Co/Ru, Co/Cr and Fe/Cr.

The exact asymptotic expansions we derived in sections 3 and 4 allow us to assess the feasibility of direct calculations of J. Every such calculation is either for a slab (Levy *et al* 1990, Hasegawa 1990) or for a cluster of atoms (Stoeffler *et al* 1991). In slab calculations, Brillouin zone summations over a two-dimensional zone have to be done numerically and it turns out that these summations are the main factor that limits the accuracy of such calculations.

In our approach, 'brute-force' calculation of J means direct evaluation of (3.3) for the total energy of carriers trapped in the spacer layer. This involves a two-dimensional BZ sum over k_x, k_y which we evaluated using the method of Cunningham special points (Cunningham 1974). Any discrete summation means that the wave vectors k_x, k_y effectively become quantized as if periodic (or other) boundary conditions were imposed over a region with transverse dimension L. It is clear that to obtain reliable results for a layered structure, the length L must be much larger than the thickness d. Taking $L/d \simeq 100$ and $N \simeq 50$, which is needed to see long-period oscillations, the number of k-points in the two-dimensional zone is $\simeq 2.5 \times 10^7$. However, even using this huge number of ordinary k-space points the computed results for J amount to more or less random noise. Even using Cunningham points we find that the correct oscillations of J only emerge when we use as many as 10^6 points. This is illustrated in figure 6. We thus conclude that direct evaluation of the exchange coupling J is possible but the size of a slab (cluster) needed to reproduce long-period oscillations with the observed amplitude is several orders of magnitude larger than those used in all existing calculations referred to above. Even with $N = 5$ the number of points required to obtain a reliable value of J far exceeds that used in any existing calculation.

5.4. *RKKY for a simple cubic tight-binding band*

We already discussed in general terms a connection between the asymptotic behaviour of the exchange coupling obtained in our theory and RKKY in section 3. However, to compare directly our numerical results discussed in section 5.1 with RKKY, we also evaluated the RKKY response of a SC lattice gas to a plane of spins.

There are several ways of calculating the RKKY interaction in the planar geometry. The most obvious method is to take the Fourier transform of the wavevector-dependent susceptibility $\chi(q)$ to obtain the RKKY range function in the direction perpendicular to interacting planes. Taking the planes to be parallel to the (x, y) plane, the appropriate Fourier transform is

$$F(z) = \frac{1}{2\pi}\int_{-\infty}^{+\infty} \chi(q_\parallel = 0, q_z)\, e^{izq_z}\, dq_z \tag{5.2}$$

where $q_\parallel$ is the wavevector parallel to the (x, y) plane. This method was applied successfully by Yafet (1987) to calculate the RKKY interaction in yttrium.

Another possibility is to calculate the exchange coupling of two (or more) planes of spins by summing up pairwise the RKKY interactions of all the individual spins occupying discrete lattice sites in the ferromagnetic planes. This method was first used by Fairbairn and Yip (1990) and more recently by Baltensberger and Helman (1990).

A third method of calculating the response to a plane of spins is by solving the corresponding scattering problem exactly; this yields the full non-linear response which reduces to the linear RKKY response for weak coupling. We have adopted this last approach. The effect of a plane of spins on electrons in a tight-binding band is modelled by a potential V_σ which is constant at each lattice site of a square lattice. The potential V_σ is attractive ($V_\uparrow = -V_0$) for up-spin carriers and repulsive ($V_\downarrow = V_0$) for down-spin carriers. It is then straightforward to solve the Dyson equation for the one-particle Green function G^σ:

$$G_0^\sigma = G_0^\sigma + G_0^\sigma V_\sigma G^\sigma \tag{5.3}$$

where G_0^σ is the Green function for the unperturbed SC tight-binding band (see Kalkstein and Soven 1971). The local occupation numbers n_i^σ for atomic planes i parallel to the perturbing plane are then determined numerically from

$$n_i^\sigma = (1/\pi)\int_{-\infty}^{E_F} \mathrm{Tr}\,\mathrm{Im}\, G_{ii}^\sigma(q_\parallel, E)\, dE \tag{5.4}$$

where the trace is over the wavevector $q_\parallel$ parallel to the perturbing plane which is taken to be the (100) plane. The spin density $S_i = n_i^\uparrow - n_i^\downarrow$ induced in the lattice gas by the perturbing plane was evaluated numerically from (5.3) and (5.4) for $V_0 = W/6$, where W is the band width. The results are shown in figure 7 for the same values of the Fermi energy as in figure 3 ($E_F = -1.05$ and $E_F = -0.95$). The most notable result is that the period of RKKY oscillations in the spin density is exactly the same as in our theory of the exchange coupling. The asymptotic decay of the oscillations is also the same as far as one can tell from the data for 20 atomic planes (the accuracy becomes too poor for $i > 20$–30 for the reasons discussed in section 5.3). However, the behaviour at short distances is clearly model dependent and is different for our theory and RKKY. This difference was already demonstrated analytically in the low-density limit in section 3.

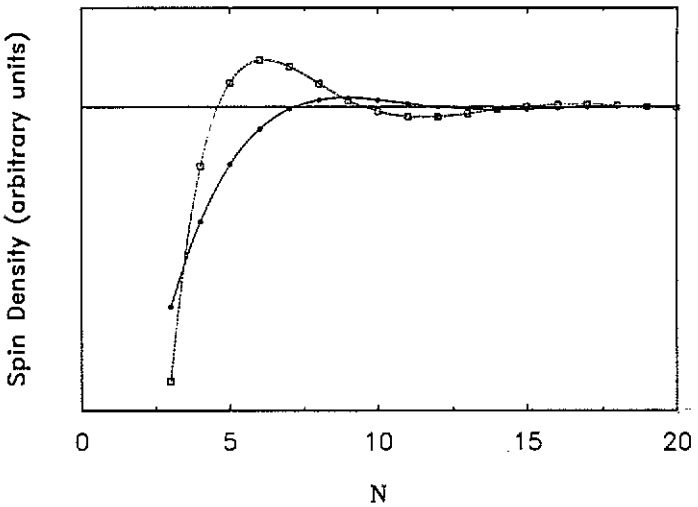

Figure 7. Spin density induced in a tight-binding lattice gas by a perturbing atomic plane at $N = 0$. Results shown are for $E_F = -0.95$ (squares) and $E_F - 1.05$ (circles).

6. Conclusions

Our results for the exchange coupling through a non-magnetic spacer layer can be broadly classified into two categories, those that are model dependent and those that are model independent.

In sections 2, 3 and 5 we proposed and investigated comprehensively a single-orbital tight-binding model for the exchange interaction of two transition metal ferromagnets separated by a non-magnetic transition metal spacer layer perpendicular to one of the cubic axes.

In section 4 we derived a general asymptotic formula for the exchange coupling, valid in the limit of a thick spacer layer. The results of section 4 apply to an arbitrary band and arbitrary orientation of the spacer layer. They are also valid at finite temperatures.

The specific model investigated in sections 2, 3 and 5 contains a single adjustable parameter: the position of the Fermi level E_F in the band. The main result obtained with this model is that the exchange coupling oscillates as a function of the thickness of the spacer layer with a period that depends on the position of E_F in the band and becomes infinite when the Fermi surface touches the zone boundary. Once the parameter E_F is fixed to reproduce the observed long period of oscillations of the exchange coupling in Co/Ru structures $\simeq 10$ atomic layers, our model yields unique predictions for the initial sign, amplitude, rate of decay and temperature dependence of the coupling. We find that the initial strength of the coupling is $J \simeq 1$ erg cm^{-2}, which should be compared with $J \simeq 6$ erg cm^{-2} obtained by Parkin *et al* (1990) for Co/Ru. The initial sign of J is antiferromagnetic and the coupling decreases with increasing thickness of the spacer layer as $1/N^2$. Both these results are as observed by Parkin *et al* (1990) for Co/Ru. Finally, the calculated temperature dependence of J is strong, on the scale $\simeq 10^2$ K, which is again in qualitative agreement with the results of Parkin *et al* (1990). For a fully quantitative theory it is clearly necessary to extend our work to more realistic band structures including both d and sp bands.

The general asymptotic formula for J derived in section 4 reveals a close analogy between oscillations of J as a function of the number of atomic planes N in the spacer layer and de Haas–van Alphen oscillations of the susceptibility as a function of $1/B$. As in the de Haas–van Alphen effect, we find that the asymptotic behaviour of the exchange coupling is completely determined by the properties of the Fermi surface in the spacer layer. In particular, the period of oscillations is determined by the caliper measurement of the Fermi surface in the direction perpendicular to the layers, the amplitude is determined by the curvature of the Fermi surface at its extremal points and the temperature dependence of the coupling is governed by the velocity of carriers at the extremal points of the Fermi surface. The first result concerning the period of oscillations is especially valuable. It can be used as a method for probing the Fermi surface complementary to the de Haas–van Alphen effect (caliper measurements of the Fermi surface are obtained instead of extremal sections). Alternatively, a simple examination of a known Fermi surface allows us to predict materials and orientations for which long-period oscillations of J should be observable. Because the quantization in the present problem is on a scale 10^2–10^3 larger than in the de Haas–van Alphen effect, oscillations of the exchange coupling are much more stable against temperature than de Haas–van Alphen oscillations. It is, therefore, likely that they are also more robust against the effects of impurities and alloying, in which case they could be used as a probe of Fermi surfaces in alloys.

We also applied our exact asymptotic formula for J in section 5.3 to assess the feasibility of direct calculations of the difference in energy between the ferromagnetic and antiferromagnetic configurations of a sandwich with a non-magnetic spacer layer. We find that such calculations are feasible for our single-orbital model. However, to obtain long-period oscillations over 50 atomic planes we had to use a slab containing more than 10^8 orbitals. This is several orders of magnitude greater than the size of clusters/slabs used in previous calculations discussed in section 5.3 and we believe that this is the main reason for all these calculations failing to reproduce the observed long-period oscillations in J.

Finally, we wish to mention a connection between the oscillations of J obtained in our theory and RKKY. Not surprisingly, our asymptotic results for large N concerning the period, rate of decay and even the temperature dependence of J are directly comparable with the asymptotic RKKY results obtained by Roth *et al* (1966) for a general shape of the Fermi surface. It is only necessary to transpose their results to the appropriate planar geometry. However, the overall amplitude of the exchange coupling, and its initial sign and behaviour at short distances are all model-dependent effects and cannot be predicted from the conventional RKKY theory. Differences between our theory and RKKY arise because our theory is non-perturbative, and hence non-linear, and it also treats the interference effect of the two ferromagnetic layers. Both these features missing in RKKY are especially important for small and even moderate thicknesses of the spacer layer.

Acknowledgments

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