Theory of intrinsic and thermally induced interlayer magnetic coupling between ferromagnetic films separated by an insulating layer

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A formalism developed previously to study the interlayer exchange coupling between ferromagnetic layers separated by a nonmagnetic metal spacer is applied to the case of an insulating spacer. It allows a *unified* treatment of both cases (metal and nonmetal spacer), provided one introduces the concept of a *complex Fermi surface*. In contrast to the metal-spacer case, where the exchange coupling decreases with increasing temperature, the coupling across an insulating spacer is found to increase with temperature. This finding is in agreement with recent experimental observations.

The exchange coupling between ferromagnetic layers separated by a nonmagnetic metal spacer has been a subject of intense research in the past few years. This activity has been triggered by the discovery of oscillations of the interlayer exchange coupling versus spacer layer thickness.[1] The periods of oscillations have been explained successfully in terms of the spacer Fermi surface;[2] in particular, the periods predicted for noble metal spacers have been verified experimentally in numerous cases.[3]

Recently, renewed interest in this field has been brought by the observation of interlayer exchange coupling across a *nonmetallic* spacer layer (amorphous Si).[4] A striking feature is that the coupling, in contrast to the metal case, *increases* with increasing temperature.[5] Furthermore, the coupling may be induced by illumination by visible light.[6] It is important to note then in all the experiments mentioned above, the nonmetallic spacer was disordered or even amorphous.

A model for the interlayer exchange coupling at $T = 0$ across a tunneling barrier has been proposed by Slonczewski.[7] However, to our knowledge, no theory of the thermal dependence of the interlayer exchange coupling across a nonmetallic spacer has been given yet.

In the present paper, a formalism developed previously for studying the interlayer exchange coupling through a metal spacer[8] is extended to the case of a nonmetallic spacer. The treatment and the formulas are completely analogous to the metallic case, provided one introduces the concept of *complex Fermi surface*. The physical behavior of the coupling, however, differs strongly from the metal spacer case. At $T = 0$, the coupling is found to decrease exponentially with spacer thickness. The most striking result concerns the thermal dependence of the coupling: whereas the exchange coupling across a metallic spacer decreases with temperature, in the case of an insulating spacer, it is found to increase strongly with temperature. This result provides a plausible explanation for the experimental observations of thermally induced coupling.

Let us briefly summarize the formalism and the results, which have been presented previously, for the case of a metallic spacer.[8] We ascribe the interlayer coupling to the interferences of electron waves in the spacer layer, due to (spin-dependent) reflections at the paramagnetic-ferromagnetic interfaces. It is thus clear that the reflection coefficients will play a key role in the formalism. Electron states in the spacer can be propagative Bloch waves, as in a bulk material, but also *evanescent* waves, because of the finite thickness of the spacer. It must be realized that the concepts of *reflection* and *transmission coefficients* pertain to evanescent waves as well. Propagative states give rise to oscillatory contributions to the coupling, while evanescent states yield exponentially decreasing terms. Thus, both kinds of states contribute *a priori* to the coupling; their respective importance depends essentially on the nature of the states that are present at the Fermi level.

For the sake of simplicity and clarity, we shall restrict our discussion to a simple free-electron model, as sketched in Fig. 1; the results can be generalized to the case of a more complicated band structure. The spacer, of thickness D, is sandwiched between two semi-infinite ferromagnetic layers F_A and F_B, whose respective magnetizations are at an angle θ with respect to each other. The zero of potential is taken at the bottom of the majority band of the ferromagnetic layers; the potential of the minority band is given by the exchange splitting Δ,

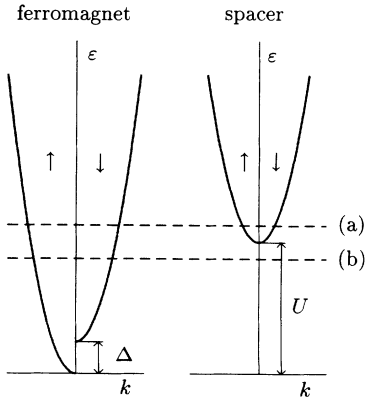

FIG. 1. Sketch of the free-electron model; the dashed line indicates the position of the Fermi level, (a) for the metallic spacer case, (b) for the insulating spacer case.

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while the potential of the spacer is U. Thus $U > \varepsilon_F$ ($U < \varepsilon_F$) corresponds to an insulating (respectively metallic) spacer.

Since the in-plane translational invariance is not broken, the in-plane component $\mathbf{k}_\parallel$ of the wave vector is a good quantum number, i.e., it is conserved under reflection and transmission. For an incident electron of wave vector $\mathbf{k}^i = (\mathbf{k}_\parallel, k_z^i)$, the wave vectors of the reflected and transmitted waves are, respectively, $\mathbf{k}^r = (\mathbf{k}_\parallel, -k_z^i)$ and $\mathbf{k}^t = (\mathbf{k}_\parallel, k_z^t)$, and the (complex) reflection amplitude is given by

$$r = \frac{k_z^i - k_z^t}{k_z^i + k_z^t} \ . \tag{1}$$

It may be seen easily that the above relation holds both for propagative waves (i.e., with k_z real) and for evanescent waves (i.e., with k_z imaginary).

The exact expression of the interlayer coupling per unit area, in terms of these reflection coefficients, is[8]

$$\begin{aligned} E_{AB}(\theta) = \frac{1}{4\pi^3}\mathrm{Im}\int d^2\mathbf{k}_\parallel \int_{-\infty}^{+\infty} d\varepsilon\, f(\varepsilon) \\ \times \ln\left[1 - 2\left(\bar{r}_A\bar{r}_B + \Delta r_A \Delta r_B \cos\theta\right) e^{iq_z D} \right. \\ \left. + \left(\bar{r}_A^2 - {\Delta r_A}^2\right)\left(\bar{r}_B^2 - {\Delta r_B}^2\right) e^{2iq_z D}\right] \ , \end{aligned} \tag{2}$$

where $q_z = k_z^r - k_z^i$, $f(\varepsilon)$ is the Fermi-Dirac function, and $\bar{r}_{A(B)}$ and $\Delta r_{A(B)}$ are, respectively, the spin-average and spin-asymmetry of the reflected amplitudes on F_A (F_B), i.e.,

$$\bar{r}_{A(B)} = \frac{r^{\uparrow}_{A(B)} + r^{\downarrow}_{A(B)}}{2} \tag{3a}$$

and

$$\Delta r_{A(B)} = \frac{r^{\uparrow}_{A(B)} - r^{\downarrow}_{A(B)}}{2} \ . \tag{3b}$$

The derivation of Eq. (2) involves integrations over k_z from $-\infty$ to $+\infty$, which are closed in the upper and lower complex half planes for the incident and reflected waves, respectively, by using the theorem of residues. There are two kinds of poles: those lying on the real axis correspond to propagative states, while those lying off the real axis correspond to evanescent states, and both kinds of states contribute on an equal footing to the coupling in Eq. (2).

In the present case the reflected amplitudes on F_A and F_B are equal, so that we can drop the corresponding indices. In Eq. (2), the sign convention is such that the state $\mathbf{k}^i = (\mathbf{k}_\parallel, k_z^i)$ is such that $\varepsilon_{\mathbf{k}^i} = \varepsilon$ and $k_z^i < 0$ in the case of a propagative wave, or $\mathrm{Im}k_z^i < 0$ for an evanescent wave. The expression of $E_{AB}(\theta)$ can be expanded in powers of $\cos\theta$ as

$$E_{AB}(\theta) = J_0 + J_1\cos\theta + J_2\cos^2\theta + \cdots \tag{4}$$

with the Heisenberg coupling constant

$$\begin{aligned} J_1 = -\frac{1}{4\pi^3}\mathrm{Im}\int d^2\mathbf{k}_\parallel \int_{-\infty}^{+\infty} d\varepsilon\, f(\varepsilon) \\ \times \frac{2\Delta r^2 e^{iq_z D}}{1 - 2\bar{r}^2 e^{iq_z D} + \left(\bar{r}^2 - \Delta r^2\right)^2 e^{2iq_z D}} \ . \end{aligned} \tag{5}$$

For the metal-spacer case, in the limit of large spacer thickness, one gets[8]

$$\begin{aligned} J_1 = \frac{1}{4\pi^2}\frac{\hbar^2 k_F^2}{mD^2}\,\mathrm{Im}\left(\Delta r^2 e^{2ik_F D}\right) \\ \times \frac{2\pi k_B T D m/\hbar^2 k_F}{\sinh\left(2\pi k_B T D m/\hbar^2 k_F\right)} \ , \end{aligned} \tag{6}$$

where $k_F = \left[2m(\varepsilon_F - U)/\hbar^2\right]^{1/2}$ is the Fermi wave vector of the spacer.

Let us now derive the corresponding result for an insulating spacer. For large thicknesses, the denominator in Eq. (5) is ≈ 1. We first perform the integration over the energy; the leading contribution arises from the neighborhood of ε_F, where the evanescent waves have the longest range, and where the Fermi-Dirac function varies rapidly. Thus, the factor Δr^2 may be kept constant, equal to its value at ε_F; the exponential factor $e^{iq_z D} = e^{-2\kappa D}$, with $\kappa = [k_\parallel^2 + 2m(U-\varepsilon)/\hbar^2]^{1/2}$, may be expanded around ε_F as

$$\exp(-2\kappa D) \approx \exp(-2\kappa_F D)\,\exp\left[(\varepsilon - \varepsilon_F)\frac{2mD}{\hbar^2\kappa_F}\right] \ , \tag{7}$$

where κ_F corresponds to $\varepsilon = \varepsilon_F$. The integration over energy is performed easily,[9] and one gets, in the limit of low temperatures ($k_B T < \hbar^2\kappa_F/mD$),

$$\begin{aligned} J_1 = -\frac{1}{4\pi^3}\,\mathrm{Im}\int d^2\mathbf{k}_\parallel\, 2\Delta r^2\,\frac{\hbar^2\kappa_F}{2mD}\,e^{-2\kappa_F D} \\ \times \frac{2\pi k_B T D m/\hbar^2\kappa_F}{\sin\left(2\pi k_B T D m/\hbar^2\kappa_F\right)} \ . \end{aligned} \tag{8}$$

Next, one performs the integration over $\mathbf{k}_\parallel$; the leading contribution arises from the neighborhood of $\mathbf{k}_\parallel = 0$, where κ_F is minimum ($\equiv \kappa_F^*$) and the sum is calculated in a standard way, by taking all factors (but the exponential) constant, equal to their value at $\mathbf{k}_\parallel = 0$, and by expanding the exponential around $\mathbf{k}_\parallel$:

$$\exp\left(-2\kappa_F D\right) \approx \exp\left(-2\kappa_F^* D\right)\exp\left(-\frac{k_x^2 + k_y^2}{\kappa_F^*}D\right) \ , \tag{9}$$

and one finally gets

$$\begin{aligned} J_1 = -\frac{1}{4\pi^2}\frac{\hbar^2{\kappa_F^*}^2}{mD^2}\,\mathrm{Im}(\Delta r^2 e^{-2\kappa_F^* D}) \\ \times \frac{2\pi k_B T D m/\hbar^2\kappa_F^*}{\sin\left(2\pi k_B T D m/\hbar^2\kappa_F^*\right)} \ . \end{aligned} \tag{10}$$

At $T = 0$, this reduces to Slonczewski's result for the interlayer exchange coupling across a tunneling barrier.[7] The sign of the coupling is determined by the argument of Δr^2; the coupling is antiferromagnetic (respectively ferromagnetic) if ${\kappa_F^*}^2 < k_F^{\uparrow}k_F^{\downarrow}$ (${\kappa_F^*}^2 > k_F^{\uparrow}k_F^{\downarrow}$), where $k_F^{\uparrow}$ ($k_F^{\downarrow}$) is the Fermi wave vector for majority spin (minority spin) electrons in the ferromagnet.

If one compares the above result with the corresponding one for the metal-spacer case, Eq. (6), one notes that the only difference is the replacement of k_F by $i\kappa_F^*$.

This striking result may be interpreted in a very simple manner provided one generalizes suitably the concept of Fermi surface: since evanescent and propagative waves contribute to the coupling on an equal footing, it is quite natural to introduce the concept of *complex Fermi surface*, by allowing k_z to have a nonzero imaginary part.[10] The complex Fermi surface is thus defined as the variety $\varepsilon(\mathbf{k}_\parallel, k_z) = \varepsilon_F$, with $\mathbf{k}_\parallel$ real and k_z complex. This adds to the usual *real* portions of the Fermi surface, complex portions, in particular, in the regions of the $\mathbf{k}_\parallel$ plane where there no real portions are found. This concept holds, not only for free electrons, but for any arbitrary band structure: this follows from the fact that Bloch's theorem holds for complex wave vectors as well.[11] One also notes in passing that the complex portions of the Fermi surface depend on the choice of the z axis, i.e., on the crystalline orientation of the layers.

The *complex Fermi surface* of free electrons is shown in Fig. 2, both for the metal and insulator cases. In the metal case, the complex Fermi surface has, in addition to the usual real sphere of radius k_F, an imaginary portion with the shape of a one-sheet revolution hyperboloid. In the insulator case, on the other hand, there is no real portion for the Fermi surface: the latter consists of an imaginary portion, with the shape of a two-sheets revolution hyperboloid; the distance between the two sheets of the hyperboloid is $2\kappa_F^*$.

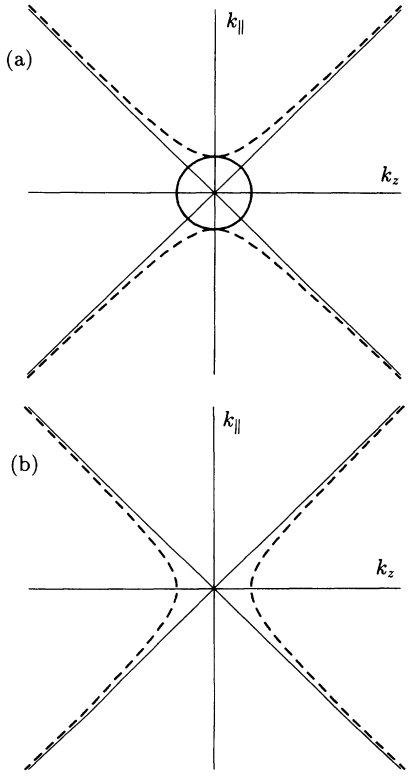

FIG. 2. Imaginary Fermi surface, for free electrons: (a) metal spacer and (b) insulating spacer. The solid (respectively dashed) line corresponds to real (imaginary) perpendicular component k_z.

The concept of complex Fermi surface allows a unified approach to the problem of interlayer exchange coupling, both for metallic and insulating spacer materials; in particular, in the limit of large spacer thickness, the coupling variation versus spacer thickness, and its temperature dependence, are determined by the extrema of the complex Fermi surface of the spacer material.

Of course, the coupling behavior is completely different in both cases. While it is oscillatory for a metal spacer, it decreases exponentially with spacer thickness in the insulator case. Another striking difference concerns the temperature dependence, which is given by the last factor in Eqs. (6) and (10). For a metal the coupling decreases with increasing temperature. On the other hand, when we consider the insulator case, replacing k_F by $i\kappa_F^*$ amounts to replacing the *hyperbolic sine* denominator by a *sine*; thus the exchange coupling *increases* with temperature for an insulating spacer. This may be understood easily: at finite temperature, the contribution of states below the Fermi level is lowered, at the expense of states above the Fermi level; since the penetration length

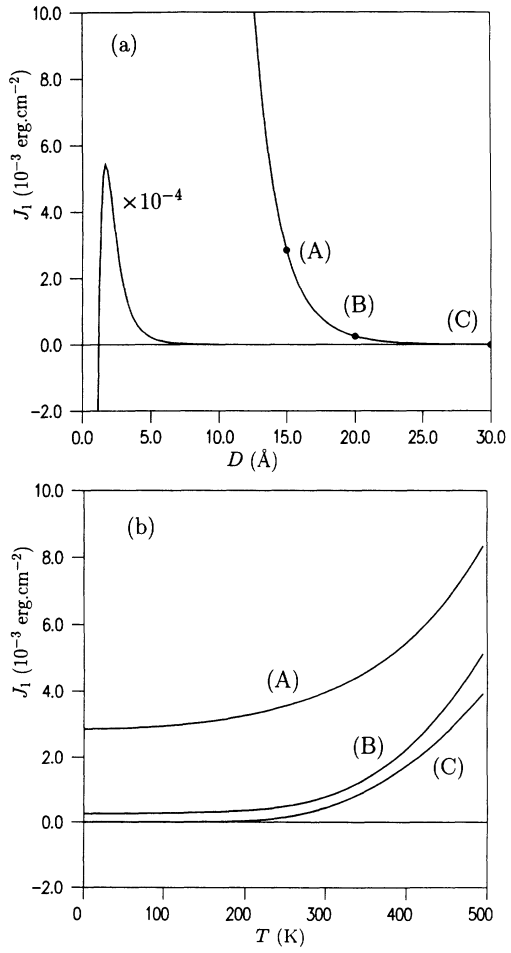

FIG. 3. Calculated interlayer exchange coupling for the free-electron model with $\varepsilon_F = 10.0$ eV, $\Delta = 1.5$ eV, and $U - \varepsilon_F = 0.1$ eV: (a) exchange coupling vs spacer thickness at $T = 0$ and (b) exchange coupling vs temperature for various spacer thicknesses, corresponding to the solid points: (A) 15.0 Å, (B) 20.0 Å, and (C) 30.0 Å.

of the latter is larger than the one of the former, the exchange coupling is thereby enhanced.

To illustrate the above results more quantitatively, we have performed numerical calculations of the exchange coupling for the free-electron model, with $\varepsilon_F = 10.0$ eV, $\Delta = 1.5$ eV, and $U - \varepsilon_F = 0.1$ eV; the calculation uses the exact expression (5), not the asymptotic result (10). The results are displayed in Fig. 3. With the above choice of parameters, the coupling at large spacer thicknesses is antiferromagnetic ($J_1 > 0$). One clearly observes the strong temperature increase of the coupling; as expected from Eq. (10), the relative thermal variation increases with increasing thickness.

One should be careful when comparing the results with experimental observations of coupling across nonmetallic spacers: indeed, as already mentioned, the latter concern materials that are disordered or even amorphous, whereas the theory presented here pertains to ordered systems; the importance of disorder for the *thermally induced* coupling remains to be clarified. Nevertheless, the finding of a positive temperature coefficient for the exchange coupling through an insulating spacer provides a plausible explanation for the experimental observations of *thermally induced* exchange coupling.[5] Note that the latter result is not restricted to the free-electron case, and may be shown to hold for any insulating spacer material. The explanation of the *photoinduced* coupling is more difficult: it probably involves localized excited states due to disorder, having a long lifetime, and which can be populated by optical pumping.

In conclusion, the general formalism of interlayer exchange coupling presented in Ref. 8, together with the concept of *complex Fermi surface*, allows a unified description of the interlayer exchange coupling across metallic and insulating spacer layers. It has been illustrated by analytic and numerical calculations for a simple free-electron model, and provides a simple tentative explanation for the recent observation of thermally induced exchange coupling across nonmetallic spacer layers.

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