Inadequacy of the asymptotic approximation for the interlayer coupling in Fe/Ag/Fe and Fe/Au/Fe (001) trilayers

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We discuss the basic assumptions underlying the stationary phase approximation to the interlayer exchange coupling between magnetic materials across nonmagnetic spacers. We show that, for temperatures and spacer thicknesses of interest, it cannot be applied to systems such as Fe/Ag/Fe and Fe/Au/Fe (001) trilayers. Such inadequacy results from particular confinement effects caused by the Fe layers in the Ag and Au extremal Fermi-surface ''neck'' states. This is of special relevance to the interpretation of experimental data. [S0163-1829(99)04738-4]

The bilinear exchange coupling J between magnetic materials across nonmagnetic metallic spacers oscillates with decaying amplitude between positive (ferromagnetic) and negative (antiferromagnetic) values, as the spacer layer thickness N is increased. For systems with crystalline layers and sharp interfaces, J can be calculated at any temperature T from an expression of the form[1–5]

$$J(N) = \sum_{\vec{k}_{||}} \int_{-\infty}^{\infty} dE \ f(E) F(E, \vec{k}_{||}, N), \qquad (1)$$

where $f(E)$ is the Fermi-Dirac distribution function, $\vec{k}_{||}$ is a wave vector parallel to the layers, $F(E, \vec{k}_{||}, N)$ is given in terms of the electronic Green functions of the system, and the sum over $\vec{k}_{||}$ is restricted to the two-dimensional Brillouin zone (BZ).

The prominent features of the oscillatory behavior of J, i.e., period, phase, and amplitude, are related to the electronic structure of the multilayered system. While numerical evaluations of Eq. (1) do not highlight this relationship, simple approximations, currently referred to as the stationary phase (SP) method, allow semianalytical expressions for J to be derived. They express the coupling asymptotically (i.e., for large values of N) as a sum of contributions coming from extremal wave vectors of the spacer Fermi surface (FS) in the direction perpendicular to the layers.[6,7,4] The oscillation period of each contribution is determined by the spacer FS, whereas the amplitude and phase are regulated also by the degree of confinement of the carriers within the spacer layer.

The stationary phase method has been successfully applied to analyze both experimental data and results of full numerical calculations of J in several systems, such as Co/Cu/Co (001) and Fe/Cu/Fe (001) multilayers.[2,4,7–9] Good agreement between total-energy calculations and the SP approximation were obtained in these systems. However, as we show here, there are cases in which this method is inadequate for spacer thicknesses and temperatures of experimental interest. This is precisely what happens in Fe/Ag/Fe (001) and Fe/Au/Fe (001) trilayers. To understand why this is so, it is instructive to review the basic assumptions of the method.

The SP method relies on the fact that, for fixed E and $\vec{k}_{||}$, the function F in the integrand of Eq. (1) oscillates with N. Thus, F can be expanded in a Fourier series in N, whose coefficients c_s and corresponding wave vector $k_\perp$ are functions of E and $\vec{k}_{||}$. The following approximations are then introduced to allow an analytical evaluation of the integrals over these variables. The first one is based on the fact that the argument of the trigonometric functions in the Fourier series is proportional to $k_\perp(E, \vec{k}_{||})N$, which, for sufficiently large N, causes very rapid oscillations as functions of $\vec{k}_{||}$. Therefore, the Fourier coefficients can be regarded as slowly varying functions of $\vec{k}_{||}$, and the dominant contributions to the integration come from the regions in $\vec{k}_{||}$ space where $k_\perp$ is stationary. These correspond to the stationary wave vectors $k_\perp^0(E)$ of the spacer constant-energy surface in the direction perpendicular to the layers. Thus, by expanding $k_\perp(E, \vec{k}_{||})$ up to second order around each extremal point $\vec{k}_{||}^0(E)$, the integration over $\vec{k}_{||}$ can be analytically performed in terms of Gaussian integrals. A similar reasoning can be used to evaluate the remaining integral over E. The integrand involves the product of $f(E)$, Fourier coefficients (FC) c_s, and trigonometric functions. Since the arguments of the latter are proportional to $k_\perp^0(E)N$, they also oscillate rapidly as functions of E for large N. It follows that the contributions from different energies tend to cancel each other, apart from those in an energy region of the order of $k_B T$ around the Fermi energy E_F, where $f(E)$ varies rapidly as well.[10] We emphasize the importance of not neglecting the energy dependence of the Fourier coefficients within this energy interval. As already pointed out,[7,4,11] the behavior of c_s with E strongly depends on the character of the multilayer electronic states in the neighborhood of $E = E_F$ and $\vec{k}_{||} = \vec{k}_{||}^0(E_F)$. In the case in which the electronic states are extended throughout the multilayer, both amplitude and phase of the corresponding FC vary slowly with E. However, when the states are

confined within the spacer layer, the energy dependence of the phase ϕ_s of c_s is pronounced and cannot be neglected. Thus, as a general procedure, both $k_\perp^0(E)$ and $\phi_s(E)$ may be linearized about E_F in the temperature range of interest, and the integral evaluated in the complex energy plane. The resulting asymptotic expression for the coupling reads

$$J(N) = \text{Im} \sum_{\vec{k}_{||}^0} \sum_{s=1}^{\infty} \frac{k_B T}{2sNa}$$
$$\times \tau |\partial_x^2 k_\perp \, \partial_y^2 k_\perp|^{-1/2} c_s(E_F, \vec{k}_{||}^0)$$
$$\times \frac{e^{2isk_\perp^0 Na}}{\sinh[2\pi k_B T(sNa(\partial k_\perp^0/\partial E) + (\partial \phi_s/\partial E))]},$$
(2)

where the sum over $\vec{k}_{||}^0$ covers all the stationary points of $k_\perp$, a is the spacer interplane distance, $\partial_x^2 k_\perp$ and $\partial_y^2 k_\perp$ are the second partial derivatives of $k_\perp$ with respect to k_x and k_y, respectively. Here, $\tau = i$ when $\partial_x^2 k_\perp$ and $\partial_y^2 k_\perp$ are both positive, $\tau = -i$ when they are both negative, and $\tau = 1$ when they have opposite signs.

It is worth stressing that, for $k_B T > (\partial \phi_s/\partial E)^{-1}$, the derivative of ϕ_s in the argument of the hyperbolic sine cannot be neglected, no matter how large N is. Indeed, for sufficiently large values of the argument of the hyperbolic sine, we have that

$$\sinh\left[2\pi k_B T\left(sNa\frac{\partial k_\perp^0}{\partial E} + \frac{\partial \phi_s}{\partial E}\right)\right]^{-1}$$
$$\simeq \exp\left[-2\pi k_B T\frac{\partial \phi_s}{\partial E}\right]\exp\left[-2\pi s k_B TNa\frac{\partial k_\perp^0}{\partial E}\right].$$
(3)

Clearly, the term in $\partial \phi_s/\partial E$ affects the amplitude of the corresponding component of the coupling, and its temperature dependence.[12]

In deriving Eq. (2), it was implicitly assumed that in the neighborhood of E_F and $\vec{k}_{||}^0(E_F)$, $c_s(E, \vec{k}_{||}^0(E))$ are analytic functions of E, and that their phases can be linearized about E_F in the energy interval of interest. This is correct in many cases, but not always. In fact, $c(E, \vec{k}_{||}^0(E))$ is nonanalytic at the energy where the character of the corresponding electronic state of the multilayer changes from extended to confined within the spacer layer. This transition becomes evident when the multilayer materials are described by free-electron models, as discussed by Mathon et al.[11] In this case, carriers with spin σ experience local potentials V^σ and V_s inside the magnetic and nonmagnetic layers, respectively. To illustrate the effect Mathon et al. assumed that, in the ferromagnetic configuration of the trilayer, there is perfect matching between $V^\downarrow$ and V_s, so that the minority-spin electrons experience no change in their local potentials when they move from the ferromagnetic layer to the nonmagnetic spacer. The majority-spin electrons, on the other hand, experience a potential well of depth $V = V^\downarrow - V^\uparrow$ inside the spacer layer. Such a quantum well may confine majority-spin electrons within the spacer layer, depending on the value of E_F, which

is determined by the number of electrons per atom in the sandwich. In this model, $k_{||}^0(E) = 0$ and, for $E \leqslant V$, the phases of the FC's are given by[11]

$$\tan\left(\frac{\phi_s}{s}\right) = 2\frac{[E(V-E)]^{1/2}}{(V-2E)}.$$
(4)

It is clear that ϕ_s is nonanalytic at the top of the well, where $\partial \phi_s/\partial E$ diverges. Thus, if $E_F \simeq V$, Eq. (2) does not hold. While this situation may seem rather special, and unlikely to occur in real systems, we argue below that something very similar actually happens in Fe/Ag/Fe and Fe/Au/Fe (001) trilayers.

The nearest-neighbor distance between atoms in both fcc Ag and Au matches the lattice constant of bulk bcc Fe to within less than 1%. This allows the growth of Fe/Ag and Fe/Au multilayers in the [001] direction with low-stress interfaces. In the stacking, the fcc Ag and Au (001) planes are rotated by 45° around the [001] direction relative to the Fe (001) planes. Within every atomic plane in the trilayer, atoms are arranged according to the same square lattice, with a common two-dimensional Brillouin zone. It is just the interplane distance that has distinct values in the magnetic and nonmagnetic materials.[13] As regards the FS's of Ag and Au, both exhibit two extrema in the [001] direction, usually called "belly" (b) and "neck" (n), whose positions in the two-dimensional BZ we label $\vec{k}_{||}^{(b)}$ and $\vec{k}_{||}^{(n)}$, respectively.[14] The long- and short-period oscillatory components of the coupling mediated by Ag and Au are associated with these two extrema.

In noble metals such as Ag and Au, the electronic states with energy $E \approx E_F$ have an essentially sp character. Hence, the confinement within the spacer of states around either the "belly" or the "neck" depends on the existence of states in Fe with the same character, energy and $\vec{k}_{||}$ (in the rotated BZ of bcc Fe). To investigate this point, we examine the band structures of fcc Ag, fcc Au, and rotated bcc Fe, as functions of $k_\perp$, for $\vec{k}_{||}^{(b)}$ and $\vec{k}_{||}^{(n)}$. Results are shown in Fig. 1 ("belly") and Fig. 2 ("neck"), where E_F is represented by horizontal dashed lines. Calculations were performed using the same tight-binding parameters as in Ref. 13.

From Fig. 1 we conclude that, in the ferromagnetic configuration, the minority-spin states with $\vec{k}_{||} = \vec{k}_{||}^{(b)}$ and $E \approx E_F$ of both Ag and Au are fully confined by the Fe layers. This is due to the corresponding minority-spin state of Fe having d character. On the other hand, the majority-spin electrons are only partially confined, since the corresponding states in Fe have some sp character owing to sp-d hybridization. It follows that the FC's $c_s(E, \vec{k}_{||}^{(b)})$ are well-behaved functions of energy in the neighborhood of E_F, allowing the "belly" contribution to the coupling to be calculated by the SP method.

The situation at the "necks," however, is quite different, as shown in Fig. 2. Here, the relevant piece of information is the behavior of majority-spin bands of Fe in the neighborhood of E_F. We find two bands, one crossing the Fermi energy twice, and another exhibiting a rather interesting behavior. The first band has strong d character for all values of $k_\perp$ up to the border of the BZ. The second one has an even stronger d character and shows almost no dispersion for $k_\perp$
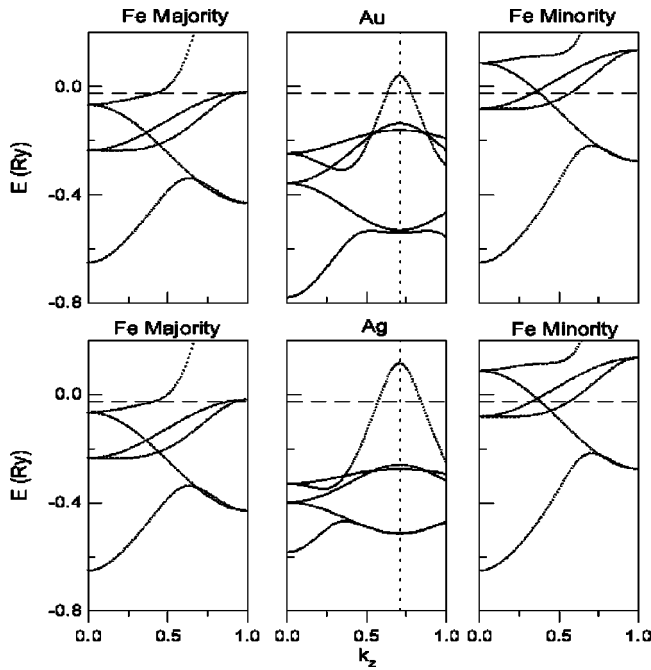
FIG. 1. Band structures of bcc Fe, fcc Ag, and fcc Au along the (100) direction ($\vec{k}_\parallel^{(b)}=0$); k_z is in units of $2\pi/a$, where a is the Fe lattice constant. Vertical and horizontal dashed lines indicate the boundary of the prismatic first Brillouin zone and the Fermi energy, respectively.

up to about π/a, where it hybridizes with the sp band and suddenly changes its character to nearly pure sp. This occurs in a quite narrow energy interval close to E_F, leading to an equally sudden change in the spatial behavior of the corresponding electronic states, going from effectively confined in
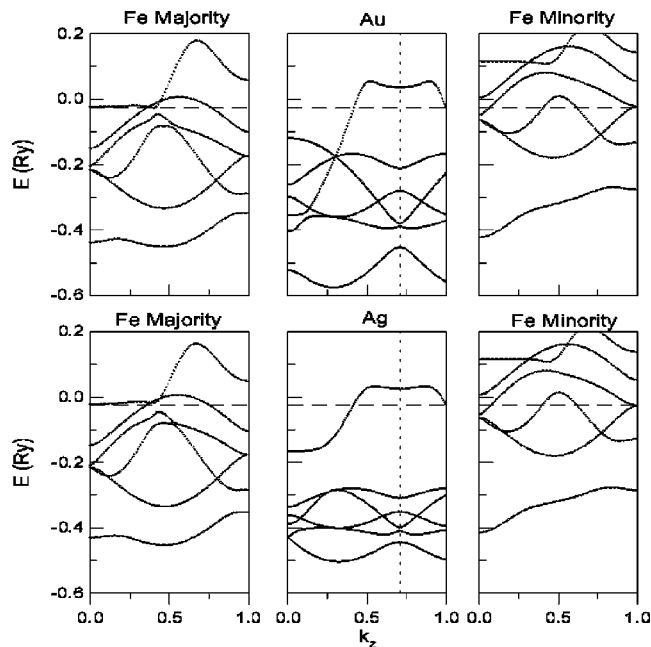

FIG. 2. Band structures of bcc Fe, fcc Ag, and fcc Au along the (100) direction ($\vec{k}_\parallel^{(n)}$); k_z is in units of $2\pi/a$, where a is the Fe lattice constant. Vertical and horizontal dashed lines indicate the boundary of the prismatic first Brillouin zone and the Fermi energy, respectively.
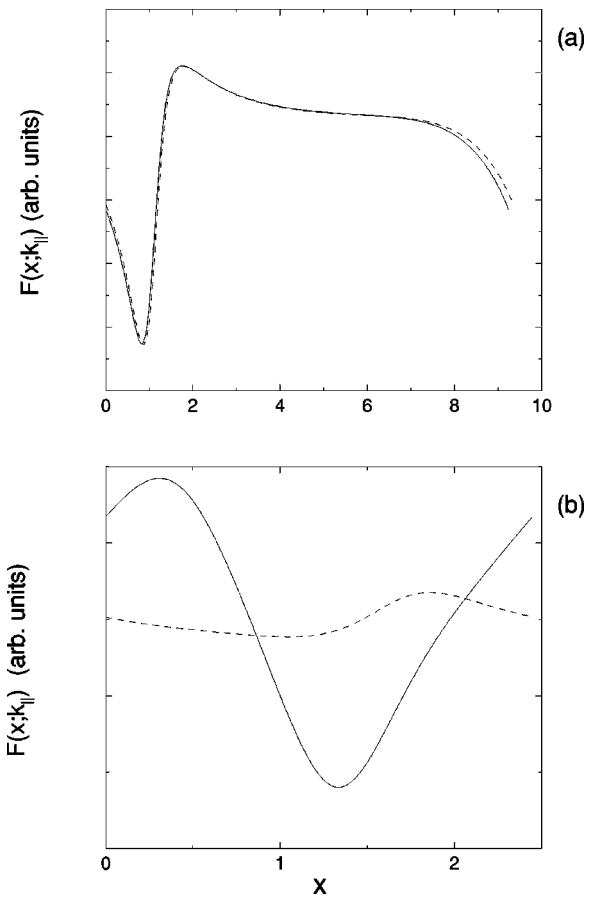

FIG. 3. Function $F(x;E,\vec{k}_\parallel^0)$ for Fe/Au/Fe (001) trilayer (see text) plotted as a function of x for $E=E_F$ (solid line) and $E=E_F+1$ mRy (dashed line) for (a) $\vec{k}_\parallel^{(b)}$ and (b) $\vec{k}_\parallel^{(n)}$.

the spacer layer to extended throughout the system. The impact of this transition on the energy dependence of the FC's is dramatic. This is well illustrated by the behavior of the curves from which the Fourier coefficients $c_s(E_F,\vec{k}_\parallel^0)$ are calculated. These curves are obtained by shifting the values of $F(N;E_F,\vec{k}_\parallel^0)$ for different spacer thicknesses N to the first period $(-\pi/k_\perp,\pi/k_\perp)$.[4] The shifted points fall on a continuous curve $F(x;E_F,\vec{k}_\parallel^0)$, which can be Fourier analyzed. Figure 3 shows results for Fe/Au/Fe (001) trilayers calculated for different energies and wave vectors $\vec{k}_\parallel^{(b)}$ (a) and $\vec{k}_\parallel^{(n)}$ (b). The dependence of the FC's on energy in each case can be appreciated by comparing the curves for E_F (full line) and $E_F+\delta E$ (dashed line), with $\delta E=1$ mRy. While the two energy curves for the ''belly'' nearly coincide, those for the ''neck'' are markedly different. This reflects the abrupt change in the character of the electronic state in the neighborhood of the neck within this very narrow energy range. In fact, for the neck the energy dependence of the phase of the FC's can only be regarded as linear in a much narrower energy interval, of the order of 1 μRy. This is a consequence of $\partial^2\phi_s/\partial^2 E|_{E_F}$ being much larger at the neck than at the belly. This restricts the validity of the asymptotic approximation in Eq. (2) for the neck contribution to J in Fe/Ag/Fe and Fe/Au/Fe (001) trilayers to temperatures not higher than $0.1\dot{K}$, which is not useful for practical purposes. Moreover, since $\partial\phi_s/\partial E|_{E_F}$ at the neck is 2–3 orders of magnitude

larger than that at the belly, even at low temperatures the asymptotic behavior of the neck contribution occurs for extremely large values of N.

In conclusion, we have discussed the basic assumptions underlying the derivation of the stationary phase approximation to the interlayer coupling and shown that, for temperatures and spacer thicknesses of interest, it cannot be applied to systems such as Fe/Ag/Fe and Fe/Au/Fe (001). The inad-

equacy of the stationary phase method for these systems results from very particular confinement effects caused by the Fe layers in the Ag and Au extremal FS neck states. This is of special relevance to the interpretation of experimental data, where it is usually assumed that the functional dependences of J on N and T are correctly given by Eq. (2). The correct procedure in cases such as those discussed here is to compare experimental results with full numerical calculations.

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[14] For both Ag and Au $\vec{k}_{||}^{(b)} = \vec{0}$, whereas $\vec{k}_{||}^{(n)}$ in the two materials occur at slightly different positions along the $\bar{\Gamma} - \bar{W}$ direction of the two-dimensional BZ.