Exchange Coupling in Magnetic Multilayers: A Quantum-Size Effect

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The long-wavelength oscillations observed in magnetic multilayers are explained by an indirect Ruderman-Kittel-Kasuya-Yosida–like (RKKY) exchange interaction. A perturbative theory of the RKKY-like exchange coupling between two ferromagnetic layers separated by a nonmagnetic slab is derived. The approach includes a realistic description of the multilayer one-electron states, whose wave functions satisfy matching conditions at the ferromagnetic-nonmagnetic interfaces. The quantum-size effects exhibited by the electron transmission coefficient give rise to a distinct multilayer wavelength λ, which provides the measured long periods.

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The peculiar properties exhibited by magnetic multilayers (ML's) have stimulated great effort. Long-range oscillatory exchange coupling has been observed in ML's formed by ferromagnetic transition metal slabs separated by ordinary transition or noble metal spacers [1], and recently, it has also been reported for an amorphous semiconductor spacer [2]. The coupling between successive ferromagnetic layers is a function of the spacer thickness. Oscillation periods ranging from 9 to 18 Å are found. Furthermore, antiferromagnetically coupled ML's display giant magnetoresistance [3] adding a potential technological interest to magnetic heterostructures (HS). Although the oscillatory behavior of the exchange interaction has been unambiguously associated with the Ruderman-Kittel-Kasuya-Yosida interaction (RKKY) [4], the specific mechanism responsible for the long-wavelength oscillations is not yet fully understood. The current explanations are based on the aliasing [5] and Bloch modulation effects [6], electronic interzonal transitions in extended k space [7], and spin-polarized quantum well states at the Fermi level [5,8]. In most models, the bulk energy bands of the nonmagnetic spacer are used to obtain the nesting required for the experimental periods [9]. The topology of the spacer Fermi surface [7,8] or the interference between two characteristic lengths—an extremal diameter of the Fermi surface $(2K_F)^{-1}$ and the periodicity of the spacer lattice in the growth direction [5]—generates the experimentally observed periods. We propose an alternative mechanism to explain the long-period oscillations. ML's are quasi-two-dimensional (Q2D) systems, periodic in two dimensions although translational symmetry in the ML growth direction is broken. As a consequence, ML wave functions exhibit quantum size effects, which give rise to a novel source of long-period oscillatory behavior. We present a perturbative calculation of the indirect RKKY-like exchange coupling between two ferromagnetic layers separated by a nonmagnetic slab, which takes into account the low dimensionality of the ML's. A simplified model accounts for the observed periods and provides a physical transparent interpretation of their origin.

As is well known, the coupling between isolated magnetic ions embedded in a paramagnetic metal are mediated by the conduction electrons in the manner suggested by Ruderman-Kittel [4]. The RKKY energy can be obtained from second-order perturbation theory, using the s-d Hamiltonian as the perturbation. The indirect exchange interaction that results is long ranged and oscillatory. In the RKKY calculation, the unperturbed one-electron states are described by Bloch functions periodic in the lattice. On the other hand, the actual ML electronic states are not eigenfunctions of the lattice translation operators. Electrons propagating along the ML growth direction feel potential discontinuities when moving from the ferromagnetic to the nonmagnetic slabs. Therefore, the electron wave functions, although periodic in 2D, have to satisfy matching conditions at the interfaces. Consequently, the proper one-electron states should be used in the present calculation.

For simplicity, we consider only a ML period formed by two ferromagnetic slabs separated by a nonmagnetic spacer. We assume ideal interfaces; that is, the potential is either the one-electron potential of the bulk ferromagnet or that of the nonmagnetic metal. Then, the ML period is represented by a quantum well (QW) or a quantum barrier (QB). The well or barrier character depends on the relative alignment of the constituents' one-electron potentials. The following discussion is for the QW. Extension to the QB is straightforward [10]. The QW is the simple Q2D structure that includes all the important physical ingredients required.

If the z axis is taken parallel to the ML growth direction, translational symmetry still holds in the x-y plane, and the electron motion may be considered free in x and y. In the z direction some electronic states are confined to a particular metal slab. However, the appearance of new confined states—discrete spectrum—is not the only manifestation of the QW structure. It is also clear that the bulk states of the constituent media—continuum spectrum—are modified. For extended states, that is, those with energy larger than the height of the confining barrier, their wave functions have to satisfy matching conditions at the two interfaces. As the Hamiltonian is the sum of x, y, and z contributions their eigenfunctions

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are separable. Accordingly, the true wave function of an electron of energy $E(\mathbf{K})$ moving in a QW is expressed as

$$\phi_{\mathbf{K}}(\mathbf{R}) = A f_k(z) e^{i\boldsymbol{\kappa}\cdot\boldsymbol{\rho}} u_{\mathbf{K}}(\mathbf{R}) \,, \qquad (1)$$

where A is a normalization constant, $\mathbf{K} = (\boldsymbol{\kappa}, k), \mathbf{R} = (\boldsymbol{\rho}, z), \boldsymbol{\kappa} = (kx, ky), \boldsymbol{\rho} = (x, y)$, $\boldsymbol{\kappa}$ and $\boldsymbol{\rho}$ are 2D vectors, $u_{\mathbf{K}}(\mathbf{R})$ is periodic in the lattice, and $f_k(z)$ is the solution of the 1D matching calculation. Then, the QW wave function is represented by a rapidly oscillating Bloch function modulated by an envelope function (EF). The EF varies slowly across the QW and ensures that the boundary conditions are met at the interfaces. For energies lower than the barrier height the electron motion is quantized and $f_k(z)$ is evanescent outside the well. For energies greater than the barrier all the energies are allowed. The wave vectors in both the well and the barrier are real, as those of propagating states. Each eigenvalue is twofold degenerate corresponding to two waves traveling towards increasing or decreasing z. Hence, the EF, $f_k(z)$, can be written in terms of the reflection $R(E)$ and transmission coefficients $T(E)$ across the QW. In the continuum, the quantum effects manifest themselves through an oscillatory behavior upon the energy of the transmission and reflection coefficients [11].

The band structure of transition and noble metals consists of five narrow localized d bands, crossing and hybridizing with a nearly free electron band formed from atomic s and p states. Then, their conduction electrons can be described as free electrons with effective masses, and a ML period modeled by a conventional rectangular QW. That is, the conduction electron of the two metals forming the ML are represented by a single parabolic band with constant potential V_i and effective mass $m_i^*, i = b$ for the ferromagnet and $i = w$ for the nonmagnetic metal. In this model, for energies $E > V_b$ the twofold degenerate functions $f_k(z)$ are given by

$$f_k^+(z) = \begin{cases} e^{ik_b z} + r(E)e^{-ik_b z}, & -\infty \le z \le -L, \\ t(E)e^{ik_b z}, & L \le z \le \infty, \end{cases}$$
$$f_k^-(z) = \begin{cases} t(E)e^{-ik_b z}, & -\infty \le z \le -L, \\ e^{-ik_b z} + r(E)e^{ik_b z}, & L \le z \le \infty, \end{cases} \qquad (2)$$

where $2L$ is the thickness of the potential well, $r(E)$ and $t(E)$ are the QW reflection and transmission amplitude, respectively [11], and

$$k_i = \sqrt{\frac{2m_i^*}{\hbar^2}\left(E - \frac{\hbar^2\kappa^2}{2m_i^*} - V_i\right)}.$$

According to second-order perturbation theory, at 0 K the interaction energy between two electronic spins $\mathbf{S}_i, \mathbf{S}_j$ embedded in the QW is given by

$$H(\mathbf{R}_{ij}) = -J(\mathbf{R}_{ij})\mathbf{S}_i\mathbf{S}_j, \qquad (3)$$

where the indirect exchange coupling constant is [12]

$$J(\mathbf{R}_{ij}) \simeq \frac{J_{sd}^2}{2} \sum_{\mathbf{K}<\mathbf{K}_F} \sum_{\mathbf{K}'>\mathbf{K}_F} \frac{e^{i(\boldsymbol{\kappa}-\boldsymbol{\kappa}')\rho_{ij}} f_k(z_i) f_{k'}^*(z_i) f_{k'}(z_j) f_k^*(z_j)}{E(\mathbf{K}') - E(\mathbf{K})} + \text{c.c.}, \qquad (4)$$

the summation symbol represents a sum for the discrete spectrum, and an integral for the continuum. $\mathbf{K}_F$ is the Fermi wave vector and J_{sd} is a parameter equivalent to an atomic s-d exchange integral [13]. To obtain expression (4) we come across matrix elements—integrals —involving the product of a slowly varying function of position, $f_k(z)e^{i\kappa\rho}$, and a periodic function $u_{\mathbf{K}}(\mathbf{R})$. We have assumed that the variation of the slow function across the diameter of an atom is negligible, that is, the modulation changes of the conduction electrons across the atom are neglected [3,11].

Now we consider that in the QW the atomic planes adjacent to the noble metal spacer are populated with ferromagnetically ordered magnetic moments. In the continuous limit of spin distribution, to evaluate the coupling between the two atomic planes, we start from expression (3) and integrate over the entire surface of a layer at constant $z = -L$. The interaction energy per unit area becomes independent of $\boldsymbol{\rho}$, since the surface integral is zero unless $\boldsymbol{\kappa} - \boldsymbol{\kappa}' = 0$. Then, the corresponding change in energy is calculated from

$$J_{\text{Q2D}}(z_{-LL}) \simeq \frac{J_{sd}^2}{2(2\pi)^4} \int_0^{\mathbf{K}_F} d\mathbf{K}\, [f_k^+(z_{-L}) f_k^{+*}(z_L) + f_k^-(z_{-L}) f_k^{-*}(z_L)]$$
$$\times \int_{\mathbf{K}_F}^0 d\mathbf{K}'\, \delta(\boldsymbol{\kappa} - \boldsymbol{\kappa}') \frac{[f_{k'}^{+*}(z_{-L}) f_{k'}^+(z_L) + f_{k'}^{-*}(z_{-L}) f_{k'}^-(z_L)]}{E(\mathbf{K}') - E(\mathbf{K})} + \text{c.c.}, \qquad (5)$$

where δ is the Dirac delta function. The continuous limit, in which each orbital state $\mathbf{K}$ corresponds to a volume $d\mathbf{K}/(2\pi)^3$, has been taken. The contribution from the discrete spectrum, which due to its localization in the well slab is expected to be small, has been neglected [10,14]. Then, in deriving (5) perturbation theory for degenerated states has been applied. The $\mathbf{K}'$ integral can be evaluated analytically in cylindrical coordinates, if it is assumed that the effective mass does not vary appreciably when changing from material. The restriction $\mathbf{K}' > \mathbf{K}_F$ is dictated by the exclusion principle. However, (5) can be integrated over all $\mathbf{K}'$ states, as the extra term added to the integrand is antisymmetric in $\mathbf{K}$ and $\mathbf{K}'$ and so integrates to zero [4]. After performing the $d^2\kappa'$ integral, we substitute (2) in (5) and employing the relations $|r(E)|^2 + |t(E)|^2 = 1$ and $r(E)t^*(E) + r^*(E)t(E) = 0$ [11], expression (5) can be written as

$$J_{\mathrm{Q2D}}(z_{-LL}) \simeq \frac{J_{sd}^2 2m^*}{(2\pi)^4\hbar^2} \int_0^{k_F} \left[t(k)e^{i2kL} + t^*(k)e^{-i2kL}\right] dk \int_0^{\sqrt{k_F^2-k^2}} \kappa d^2\kappa \int_0^{2\pi} d\theta_k \int_\infty^{-\infty} \frac{t(k')e^{i2k'L}}{k'^2 - k^2} dk'. \quad (6)$$

To avoid the singularities of the integrand in the k' integral, we used principal values and the Cauchy method of residues [4]. Then J_{Q2D} becomes

$$J_{\mathrm{Q2D}}(z_{-LL}) \simeq -\frac{J_{sd}^2 m^*}{2(2\pi)^2\hbar^2} \int_0^{k_F} (k_F^2 - k^2) \mathrm{Im}\, \frac{t^2(k)e^{i4kL}}{k} dk. \quad (7)$$

Besides $(k_F^2 - k^2)$, expression (7) differs from the range function of the 1D case by the transmission amplitude factor [15]. Its appearance is a direct consequence of the ML wave function properties. This is a relevant result, since the quantum effects displayed by $t(E)$ give rise to distinct features of J_{Q2D} upon the interlayer thickness. The remaining integration in k is not analytical and has to be performed numerically. The various simplifying assumptions made above are introduced to get a simple and physical transparent expression for the coupling constant. However, the calculation can be readily extended to include any Q2D model for the ML's [10].

To estimate the interlayer thickness dependence of the indirect exchange coupling J_{Q2D}, we evaluate expression (7) for a Co/Cu QW. The only parameters entering the calculation are the Fermi level, V_i and m_i^*, where $i=$ Co, Cu. We assume that the conduction electrons of both metals are described by the bulk sp bands and that $m^* = 1$. Then, when the Fermi levels are aligned, V_i is the energy of the bottom of the Co/Cu bulk sp-band referred to the Fermi level, $V_{\mathrm{Cu}} = 0.692$ Ry and $V_{\mathrm{Co}} = 0.669$ Ry [16]. The energy difference of the bulk sp band at the Γ point, $\Delta V = V_{\mathrm{Cu}} - V_{\mathrm{Co}} = 0.023$ Ry, gives the confining potential. Figure 1 presents the calculated exchange couplings versus the Cu interlayer thickness. The top curve a corresponds to J_{Q2D} obtained from expression (7), while curve b represents the coupling constant, J_{1D}, of a 1D QW. In the bottom, curve c, the calculated ordinary J_{RKKY} averaged over a plane [15], with

$$K_F = \sqrt{\frac{2m^*}{\hbar^2} V_{\mathrm{Cu}}}$$

and $m^* = 1$ is represented. The ordinary RKKY exchange interaction, curve c, shows the well known asymptotic $\sin X/X^2$ form, falls off rapidly, and has a period of 1.13 ML. This value is closed to the 1.3 ML obtained with the experimental K_F. Although, our purpose is only to give qualitative arguments, the reasonable agreement between these two values indicates the adequacy of the simplified model used to describe the bulk metals. On the other hand, both J_{Q2D} and J_{1D} do not present the characteristic RKKY-like behavior. Instead they disclose a peculiar structure. The coupling is ferromagnetic (FM) for most thicknesses, but at specific values it displays sharp antiferromagnetic (AFM) peaks. The only difference between both curves is the rippled structure with the ordinary short RKKY period, which modulates the 1D coupling constant. The modulation is not appreciable in the J_{Q2D}, since it is smoothed out by the integrals involved in the 3D case. The AFM maxima appear whenever

$$2L\sqrt{\frac{2m^*}{\hbar^2}\Delta V} = n\pi$$

or equivalently when $t(E_{\mathrm{Co}})$ is equal to 1. n is an integer and E_{Co} the bottom of the Co band. The discrete thicknesses which fulfill this equation correspond to the existence of a transmission resonance at the onset of the continuum. $t(E_{\mathrm{Co}})$ is always zero unless the resonant condition is satisfied. Then, the change of the transmission coefficient $t(E_{\mathrm{Co}})$ at each resonant energy can be identified as the cause of the AFM peaks. Furthermore the AFM period is completely determined by the confining potential. The calculated period is 6.13 ML, which compares remarkably well with the experimental one ≈ 6 ML. Furthermore, although the shape of the thickness dependence can rely on the details of the potential and could also change if the discrete spectrum is included [17], asymmetric oscillations—longer thickness intervals for FM coupling—clearly seen in Fig. 1—seem to appear in the experimental results (Fig. 4 of Ref. [18]

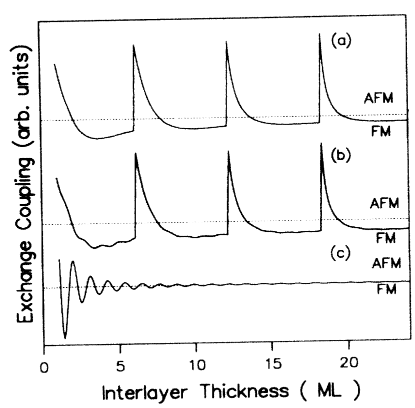

FIG. 1. Indirect exchange coupling constants versus the Cu interlayer thickness for a Co/Cu quantum well. Curve a corresponds to J_{Q2D} evaluated with the 3D ML wave function [Eq. (7)]. Curve b represents the coupling constant of J_{1D} derived from a 1D ML, while in curve c the ordinary J_{RKKY} averaged over a magnetic layer is shown [13].

and Fig. 5 of Ref. [19]). Moreover, in the present model, the strength of the AFM coupling is determined by quantum size effects on the magnetic carriers [10]. This would explain the experimental observation of AFM coupling at spacer thicknesses as large as 45 Å [18], for which the ordinary $J_{\rm RKKY}$ is almost negligible (Fig. 1, curve c). As stated above, the main object of this study was not to obtain quantitative accuracy, but to provide qualitative arguments in favor of an alternative mechanism responsible for the long-wavelength oscillation. However, the simple model described previously accounts reasonably well for the magnitude and period of the observed oscillations in the exchange coupling of magnetic ML. In the present approach the FM to AFM switch is clearly related to the quantum-size effects of the electron transmission coefficient across the nonmagnetic layer. As the thickness of this layer increases an AFM peak occurs whenever a new resonance level becomes occupied. The rate of this process is controlled by the potential discontinuity between the ML's constituent metals. Then, there is a distinct ML length

$$\lambda = \sqrt{\frac{\hbar^2\pi^2}{2m^*\Delta V}},$$

and the indirect coupling manifests the response of the conduction electrons with the characteristic wavelength λ. Since the proposed mechanism does not rely on the existence of the Fermi surface or on the periodicity of the spacer, it could also explain the observed oscillatory coupling for semiconductor spacer. However, a less crude model for the ML conduction electrons would be required to take into account the Schottky-barrier formation. In summary, we have shown that long-period oscillations in magnetic ML can be due to quantum-size effects. The boundary scattering at the spacer-magnetic film interfaces induces spatial symmetry to the electronic wave function, which give rise to long-period oscillatory exchange coupling. The above theory provides a natural explanation for the interlayer coupling in ML and open promising outlooks towards new magnetic effects in low-dimensional systems.

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