Theory of the exchange coupling in magnetic metallic multilayers

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Existing theories of the oscillatory exchange coupling in magnetic metallic multilayers are reconciled within a common approach. It is shown that Ruderman-Kittel-Kasuya-Yosida-type theories and full-confinement quantum-well results are obtained as limiting cases. Numerically useful and model-independent closed-form expressions for the exchange coupling across a nonmagnetic spacer are obtained in terms of the one-electron propagators. The theory can be extended to deal with nonmagnetic alloy spacers and it is argued that the existence of disorder in the spacer may lead to significant reduction of the interlayer coupling.

Magnetic coupling between ferromagnetic metallic layers separated by a nonmagnetic metallic spacer layer has been observed in several systems. It has been found¹ that such a coupling oscillates as a function of the spacer thickness, between ferromagnetic and antiferromagnetic, and that its amplitude decays asymptotically as $1/N^2$, where N is the number of atomic planes of the spacer material. For a given value of N the energy of the magnetic configuration per unit area can be written as^{2,3}

$$\Omega(\theta) = \Omega_0 - J_1 \cos(\theta) - J_2 \cos^2(\theta), \quad (1)$$

neglecting higher powers of $\cos(\theta)$, where θ is the angle between the magnetization vectors of the ferromagnetic layers, and J_1 and J_2 are the bilinear and biquadratic coupling terms, respectively.

Different theoretical approaches have been used to calculate the coupling between the ferromagnetic layers: theories based on total-energy calculations,^{4,5} and Ruderman-Kittel-Kasuya-Yosida (RKKY) -type theories.^{6,7} In the first approach the coupling is obtained as the difference between the energies of the antiferromagnetic ($\theta = \pi$) and ferromagnetic ($\theta = 0$) configurations, whereas in the RKKY-like theories second-order perturbation theory is used to determine the bilinear term J_1 . In fact, the two approaches are complementary rather than competing. Generally, the difference in energies between the ferromagnetic and antiferromagnetic configurations is relatively small, which makes total-energy calculations rather difficult, especially at large spacer thicknesses. On the other hand, as a second-order perturbation theory, RKKY is not expected to provide a good description of the coupling for thin spacer thicknesses. RKKY has been very successful⁷ in predicting the period of oscillations for noble-metal spacers, but the phase and strength of the coupling usually depend on a rather arbitrary choice of parameters. The theory and model calculations based on the size quantization of the magnetic carrier energy levels by the exchange field in the ferromagnetic layers explain several of the observed features.^{5,8} However, at least in their original formulation, they apply to transition-metal spacers only, whereas RKKY is most appropriate to noble- and simple-metal spacers. Both approaches^{5,7-9} relate the period of oscillations of the interlayer coupling to extrema of the Fermi surface of the spacer material, while the amplitude and phase of oscillations depend mostly on the matching be-

tween the spacer and the magnetic metals.¹⁰

More recently, significant attempts have been made^{10,11} to develop a unified nonperturbative theory which can treat transition-, noble-, and simple-metal spacers within the same scheme.

The experimental observation^{2,3} that, under certain circumstances, the in-plane magnetization directions of successive ferromagnetic layers may align themselves perpendicularly, has put in evidence the importance of the biquadratic term. Slonczewski¹² proposed initially that the biquadratic exchange term arises as an effect due to interface roughness. Subsequently, however, an intrinsic mechanism for the biquadratic exchange was proposed by Edwards *et al.*¹³ and later considered by others.^{11,14,15}

Here we present a general formulation of the interlayer coupling. Model-independent closed-form expressions for the change in energy $\Delta\Omega(\theta) = \Omega(\theta) - \Omega(0)$ as well as for the bilinear and intrinsic biquadratic exchange terms are obtained in terms of the one-electron propagators. These expressions are suitable for first-principles calculations because they avoid large numerical cancellations. It is shown that RKKY theory for J_1 is recovered when terms beyond second order in the local exchange field in the ferromagnetic layers are neglected. On the other hand, the complete confinement model of Edwards *et al.*⁵ is obtained when the exchange field in the magnetic layers goes to infinity. On the basis of the present results, the strength of the interlayer coupling through a nonmagnetic dilute alloy spacer is predicted to decay exponentially as a function of the impurity concentration.

For simplicity we consider a system consisting of two parallel ferromagnetic atomic planes embedded in a nonmagnetic material. The generalization to thicker ferromagnetic layers is straightforward. We label the two ferromagnetic planes by 0 and n , so that the number of nonmagnetic spacer planes in between them is $N = n - 1$. The energy necessary to rotate the magnetization direction in the plane n relative to the one in the plane 0, at a temperature T , maybe obtained from the thermodynamical potential^{5,8}

$$\Omega = -\left(\frac{1}{\beta}\right) \int d\omega \ln\{1 + e^{\beta(\mu - \omega)}\} \times \left(-\frac{1}{\pi} \text{Im Tr} \sum_i [G_{ii}(\omega)]\right), \quad (2)$$

where $\beta = 1/k_B T$, μ is the chemical potential, $[G_{ii}(\omega)]$ is the Green function for electrons with up and down spins at site i , and Tr stands for the trace over spin and atomic orbital indices. The electron-electron interaction within the two ferromagnetic planes is described by an effective local exchange field, acting on the magnetic carriers, which is parallel to the local magnetization direction. Thus, the rotation of the magnetization in plane n corresponds to a change in the direction of the local exchange field. The resulting variation of the local electron potential in plane n is given by

$$[V(\theta)] = -\{[\cos(\theta) - 1]\sigma_z + \sin(\theta)\sigma_x\} \otimes [V_{\text{ex}}], \quad (3)$$

where θ is the angle of rotation, σ_z and σ_x are the usual Pauli matrices, and $[V_{\text{ex}}]$ is a diagonal matrix in orbital indices representing the strength of the local exchange potential. $\theta = 0$ corresponds to the ferromagnetic alignment of the magnetization of the two magnetic planes. The change $\Delta\Omega(\theta)$ in the thermodynamic potential due to the rotation is related to the change $[\Delta G]$ in the one-electron Green function. In view of the translational symmetry of the system parallel to the magnetic planes we may write

$$\Delta\Omega(\theta) = \left(\frac{1}{\pi\beta}\right) \sum_{\mathbf{q}} \int d\omega \ln\{1 + e^{\beta(\mu-\omega)}\} \times \text{Im} \sum_l \text{Tr}[\Delta G_{ll}(\mathbf{q}, \omega)], \quad (4)$$

where $\mathbf{q}$ is a wave vector parallel to the planes and belonging to the two-dimensional Brillouin zone (BZ), and l is a plane index. To simplify the notation we omit the arguments ω and $\mathbf{q}$ in the equations below. Since the perturbing potential is localized in plane n we have

$$[\Delta G_{ll}(\theta)] = [G_{ln}^0][T_{nn}(\theta)][G_{nl}^0], \quad (5)$$

where $[G^0] = [G(\theta = 0)]$ is the Green function of the ferromagnetic configuration, and

$$[T_{nn}(\theta)] = [V(\theta)]\{[I] - [G_{nn}^0][V(\theta)]\}^{-1}, \quad (6)$$

where $[I]$ is the identity matrix in the orbital and spin indices. The nonzero matrix elements of $[V(\theta)]$ and $[T(\theta)]$ involve those spin orbitals on which the exchange field acts. It follows that

$$\Delta\Omega(\theta) = \frac{1}{\pi} \sum_{\mathbf{q}} \int d\omega f(\omega) \text{Im} \text{Tr} \ln\{[I] - [G_{nn}^0(\mathbf{q}, \omega)][V(\theta)]\}, \quad (7)$$

where $f(\omega)$ is the Fermi-dirac distribution function. This equation can be simplified by noting that

$$\begin{aligned} \text{Tr} \ln\{[I] - [G_{nn}^0][V(\theta)]\} \\ = \text{tr} \ln\{[1] + [\cos(\theta) - 1]([G_{nn}^{0\uparrow}] - [G_{nn}^{0\downarrow}]) \\ + 2[G_{nn}^{0\uparrow}][V_{\text{ex}}][G_{nn}^{0\downarrow}][V_{\text{ex}}]\}, \end{aligned} \quad (8)$$

where tr stands for the trace over orbital indices only, $[G^{0\sigma}]$ is the propagator for electrons with spin σ in the ferromagnetic configuration, and $[1]$ is the identity matrix in orbital indices. Then, using Dyson's equation to relate

$[G_{nn}^{0\uparrow}]$ and $[G_{nn}^{0\downarrow}]$ we end up with the following general expression for $\Delta\Omega(\theta)$:

$$\begin{aligned} \Delta\Omega(\theta) = \frac{1}{\pi} \sum_{\mathbf{q}} \int d\omega f(\omega) \text{Im} \text{tr} \ln\{[1] - 2[\cos(\theta) - 1] \\ \times [G_{n0}^{0\uparrow}(\mathbf{q}, \omega)][V_{\text{ex}}][G_{0n}^{0\downarrow}(\mathbf{q}, \omega)][V_{\text{ex}}]\}. \end{aligned} \quad (9)$$

This equation explicitly relates the change $\Delta\Omega(\theta)$ to the $\uparrow$ and $\downarrow$ spin electron propagators between the magnetic planes, indicating that it is the propagation of electrons through the nonmagnetic spacer which gives rise to the coupling between the magnetic planes. The sensitivity of the coupling on the electronic structure of the spacer is therefore evident. In addition, it is worth stressing that once the model for the electronic structure of the system is chosen, and the propagators are determined accordingly, Eq. (9) provides a general way of calculating the coupling through a noninteracting electron system without any additional parameter or assumption.

Recently, Lang *et al.*¹⁶ deduced an expression for the change in energy between the ferromagnetic and antiferromagnetic alignments of the magnetic layers which resembles Eq. (9). However, they have considered the energy change of each configuration with respect to the bulk spacer system. Consequently, their expression is given by the difference between two terms, each involving t matrices for the magnetic planes which include both the exchange and the on-site potential differences between the magnetic and spacer materials. We, on the other hand, start from the ferromagnetic configuration and calculate the change in energy to rotate just the exchange field in plane n with respect to the parallel alignment configuration. The contributions from the on-site potential differences between the two materials are already included in our ferromagnetic reference state. Both results avoid the subtraction of large energy contributions. Equation (9), however, eliminates unnecessary cancellations, making numerical calculations of the coupling more efficient.

A closed-form and model-independent expression for the torque $\tau(\theta) = -\partial\Delta\Omega(\theta)/\partial\theta$ necessary for rotating the magnetization in plane n can be easily obtained by differentiating Eq. (9) with respect to θ . This establishes a direct connection between the present approach and the torque method used by Slonczewski^{12,17} and Hathaway and Cullen.¹⁴

For small angle deviations we find from Eq. (9) that $\Delta\Omega(\theta) \cong D\theta^2$, where

$$\begin{aligned} D = \frac{1}{\pi} \sum_{\mathbf{q}} \int d\omega f(\omega) \text{Im} \text{tr} \{ [G_{n0}^{0\uparrow}(\mathbf{q}, \omega)][V_{\text{ex}}] \\ \times [G_{0n}^{0\downarrow}(\mathbf{q}, \omega)][V_{\text{ex}}] \} \end{aligned} \quad (10)$$

gives the stiffness of the ferromagnetic configuration. When the magnetic materials are transition metals the splitting of the up- and down-spin d bands is approximately rigid. In this case, $[V_{\text{ex}}]$ has nonzero elements V_{ex} associated with the d orbitals only, and Eq. (10) becomes

$$D = -V_{\text{ex}}^2 \text{tr}[\chi_{0n}(\mathbf{q}' = \mathbf{0}, \omega' = 0)], \quad (11)$$

where tr refers to the trace over d orbitals, and

$$[\chi_{0n}(\mathbf{q}' = \mathbf{0}, \omega' = 0)] = -\frac{1}{\pi} \sum_{\mathbf{q}} \int d\omega f(\omega) \text{Im}\{[G_{n0}^{0\uparrow}(\mathbf{q}, \omega)] \times [G_{0n}^{0\downarrow}(\mathbf{q}, \omega)]\} \quad (12)$$

is the static and uniform (within the planes) transverse Hartree-Fock susceptibility of the system. As the spacer thickness varies χ_{0n} oscillates as a function of n and so does D . Careful measurements of spin-wave excitations

$$J_1 = \frac{2}{\pi} \sum_{\mathbf{q}} \int d\omega f(\omega) \text{Im} \text{tr}([G_{n0}^{0\uparrow}(\mathbf{q}, \omega)][V_{\text{ex}}][G_{0n}^{0\downarrow}(\mathbf{q}, \omega)][V_{\text{ex}}]\{[1] + 2[G_{n0}^{0\uparrow}(\mathbf{q}, \omega)][V_{\text{ex}}][G_{0n}^{0\downarrow}(\mathbf{q}, \omega)][V_{\text{ex}}]\}^{-1}), \quad (13)$$

and

$$J_2 = \frac{2}{\pi} \sum_{\mathbf{q}} \int d\omega f(\omega) \text{Im} \text{tr}([G_{n0}^{0\uparrow}(\mathbf{q}, \omega)][V_{\text{ex}}][G_{0n}^{0\downarrow}(\mathbf{q}, \omega)][V_{\text{ex}}]\{[1] + 2[G_{n0}^{0\uparrow}(\mathbf{q}, \omega)][V_{\text{ex}}][G_{0n}^{0\downarrow}(\mathbf{q}, \omega)][V_{\text{ex}}]\}^{-1})^2). \quad (14)$$

These expressions are model independent, and similarly to Eq. (9) they involve quantities directly obtainable from electronic structure calculations.

To lowest order in V_{ex} and in the case of a rigid exchange splitting the equation for J_1 reduces to

$$J_1 = -2V_{\text{ex}}^2 \text{tr}[\chi_{0n}^0(\mathbf{q}' = \mathbf{0}, \omega' = 0)], \quad (15)$$

where χ_{0n}^0 is the noninteracting susceptibility given by Eq. (12), calculated with $V_{\text{ex}} = 0$. This is essentially the RKKY result. Nevertheless, it is worth mentioning that in RKKY-type theories it is the bulk spacer susceptibility which is usually used in the calculation^{6,7} of J_1 . Such an approximation neglects one-electron potential differences between the magnetic and the spacer materials, which may be important even when $V_{\text{ex}} = 0$, especially for thin spacers or when discussing the dependence of J_1 on the thickness of the magnetic layers. This shows that Eq. (6) of Ref. 16, obtained from a second-order expansion, is basically equivalent to a RKKY-like theory, however, with an additional approximation because they treat not only the exchange but also the on-site potential differences in second order.

In the electron confinement model originally proposed by Edwards *et al.*,⁵ an appropriate but opposite limit ($V_{\text{ex}} \rightarrow \infty$) is taken such that in ferromagnetic configuration the down-spin electrons are free to move throughout the system, whereas the up-spin electrons are completely confined. In such a case, we obtain from Eq. (9) that

$$\Delta\Omega(\pi) = \frac{1}{\pi} \sum_{\mathbf{q}} \int d\omega f(\omega) \text{Im} \text{tr}\{\ln[G_{00}^B(\mathbf{q}, \omega)] - \ln[G_{nn}^S(\mathbf{q}, \omega)]\}, \quad (16)$$

where $[G_{00}^B]$ is the bulk spacer local Green function and $[G_{nn}^S]$ is the local Green function in the $(n-1)$ th plane below the surface of a semi-infinite spacer material, which is given by

$$[G_{nn}^S] = [G_{00}^B] - [G_{n0}^B][G_{00}^B]^{-1}[G_{0n}^B]. \quad (17)$$

It can be shown that Eq. (16) gives exactly the oscillatory part of Ω_{tot} obtained by Edwards *et al.*⁵ from slab calculations. From Eq. (17) it is evident that the coupling, in this limiting case, is totally determined by the

in the direction perpendicular to the layers in superlattices should show these oscillations. A stable ferromagnetic alignment between the layers leads to positive values of $\Delta\Omega(\theta)$ and D , whereas a negative value of D indicates an instability of the ferromagnetic alignment against small-angle deviations of the magnetizations.

By expanding the logarithm in Eq. (9) in powers of $\cos(\theta)$ and keeping terms up to $\cos^2(\theta)$ we obtain $\Delta\Omega(\theta)$ in the same form as in Eq. (1), with J_1 and J_2 given by

bulk electronic structure of the spacer material. Also, Eq. (16) explicitly states that the oscillations of $\Delta\Omega$ as a function of n come from the oscillations in $[G_{nn}^S]$ which are associated with quantum interferences due to reflections of the electronic waves at the surface.

The connection between RKKY-type theories and the quantum-well model of Edwards *et al.*⁵ is thus analytically established and supports the numerical results recently obtained by van Schilfgaarde and Harrison.¹⁸

The expressions for the coupling derived here are very convenient for numerical calculations because they avoid the problem of large cancellations. Nevertheless, the evaluation of $\Delta\Omega(\theta)$ still requires numerical care, particularly for large values of n . In such a case, it is necessary to obtain the one-electron propagators between the magnetic planes and perform the BZ and energy integrations very accurately. To illustrate this point we consider a simple cubic (sc) one-orbital tight-binding model with hopping restricted to nearest-neighbor sites and intra-atomic Coulomb interaction $U \neq 0$ in the planes 0 and n only. For each parallel $\mathbf{q}$ vector, G_{0n}^σ can be related to the propagator g_{0n} corresponding to $V_{\text{ex}} = 0$. We assume that the magnetic layers have (010) orientation, and take the origin of energies at the center of the bulk spacer band and choose the energy scale such that the hopping parameter $t = \frac{1}{2}$. In this case the imaginary part of g_{0n} is nonvanishing only when $\varepsilon(\mathbf{q}) - 1 \leq \omega \leq \varepsilon(\mathbf{q}) + 1$, where $\varepsilon(\mathbf{q}) = \cos(q_x a) + \cos(q_z a)$ is the two-dimensional band structure within the planes. In this case one has

$$g_{0n}(\mathbf{q}, \omega) = (-i)^{n+1} e^{i\alpha} / \cos(\alpha), \quad (18)$$

with α given by $\sin(\alpha) = \omega - \varepsilon(\mathbf{q})$. Usually, in electronic structure calculations a small imaginary part η is added to the energy ω . It is clear that such a procedure amounts to adding an imaginary part to the angle α . As a consequence, g_{0n} acquires an artificial decaying factor which exponentially dumps the coupling for sufficiently large values of N . The magnitude of η is fixed according to the number of $\mathbf{q}$ points used in the BZ integration. Therefore, in order to obtain the correct asymptotic behavior of the coupling it is necessary to work with very small values of η and very fine mesh in the BZ. These remarks apply also to a parameter-free RKKY-type calculation of the coupling based on Eq. (15).

The fact that the expressions derived here are given in

terms of one-electron propagators, allows the theory to be extended to deal with nonmagnetic alloy spacers. In such cases one would have to determine the propagators in terms of the alloy self-energy $\Sigma(\omega)$. The imaginary part of Σ is associated with the finite lifetime of quasi-particle states, resulting from the incoherent electronic scattering by the disordered alloy potential. For a binary alloy $A_{1-x}B_x$ in the dilute limit, i.e., $x \ll 1$, it is well known that Σ is approximately given by

$$\Sigma(\omega) \simeq V_A + x\Sigma^{(1)}(\omega), \quad (19)$$

where V_A is the potential of the A material and $\Sigma^{(1)}$ is the scattering matrix of a single B atom in the A host. Thus, for a dilute alloy spacer we expect the propagator G_{0n} to behave as

$$G_{0n} \sim e^{in\alpha'} e^{-xn\xi}, \quad (20)$$

where α' is real and ξ is related to $\text{Im}\Sigma^{(1)}(\omega)$. Therefore, in the dilute limit, the strength of the coupling should decay exponentially with the alloy composition x for a given alloy spacer thickness, indicating that information about the electron potentials on the magnetic layers is lost when electrons propagate through the disordered spacer. A reduction of the coupling as a function of x was recently observed in Co/Cu_{1-x}Ni_x multilayers.¹⁹ Experimentally, however, the disorder effect in concentrated alloys has to be separated from other effects such as the band filling, where the addition of impurities gradually changes the spacer Fermi surface thereby affecting the period and strength of the coupling, and the band mismatch effect¹⁰ between the spacer and the magnetic layers. Nevertheless, according to the above argument, any kind of disorder in the spacer (including roughness at the interfaces) leads to a reduction in the amplitude of the coupling. Moreover, depending on the degree of disorder, the reduction may be very strong.

Finally, to test the numerical usefulness of the present approach we have applied it to the simple tight-binding model described above, assuming a typical value of $V_{\text{ex}} = 0.2$, and choosing the origin of energy at the center of the spacer band. In the energy scale adopted here the bulk spacer bandwidth is equal to 6. We have calculated both J_1 and J_2 as a function of n for $E_F = 0$ and $E_F = 2.5$. For these values of E_F we found that the amplitude of J_2 is much smaller than that of J_1 , indicating that the in-

FIG. 1. Dependence of the bilinear (J_1) and intrinsic biquadratic (J_2) exchange couplings on spacer thickness. N is the number of atomic planes in the spacer. (a) $E_F = 0$. (b) $E_F = 2.5$.

trinsic mechanism possibly cannot solely account for the large biquadratic contribution observed by Fuss *et al.*² A comparison between Eqs. (13) and (14) shows that the intrinsic biquadratic exchange term J_2 has half the basic period of J_1 in accordance with what has been deduced by Edwards *et al.*¹³ using a free-electron gas model for the spacer. As shown in Fig. 1, J_1 oscillates with a period of two monolayers for $E_F = 0$, and with a period of three monolayers for $E_F = 2.5$. The corresponding periods of J_2 would be 1 and 1.5 monolayers, respectively, if the spacer thickness varied continuously. However, because it changes by integer numbers of atomic planes only, we do not observe oscillations in J_2 for $E_F = 0$, and an effective period of three monolayers is seen for $E_F = 2.5$.

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