Mesoscopic mechanism of exchange interaction in magnetic multilayers

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We discuss a mesoscopic mechanism of exchange interaction in ferromagnet-nonferromagnet-ferromagnet metallic multilayers. We show that in the case when the thickness of the nonferromagnetic metal layer is larger than the electron mean free path, the relative orientation of magnetizations in the ferromagnets is perpendicular. The exchange energy between ferromagnets decays with the thickness of the nonferromagnetic metal layer as a power law.

I. INTRODUCTION

Both the experiment and the theory of ferromagnetnonferromagnet-ferromagnet metallic multilayers have attracted a lot of attention. $1-6$ An example of such a structure consisting of two ferromagnetic metal films separated by a nonferromagnetic metallic film is shown in Fig. 1. In the case when the thickness *L* of the nonferromagnetic metal is much smaller than the electron scattering mean free path *l* the sign of the exchange interaction energy between the ferromagnets' magnetizations oscillates as a function of *L* with a period of order of the Fermi wavelength. As a result the magnetic structure of the system oscillates between ferromagnetic and antiferromagnetic orientations of the ferromagnets' magnetizations. $1-6$ The explanation of this phenomenon is based on the fact that the interlayer exchange energy is due to the Ruderman-Kittel interaction between electron spins in different ferromagnets.

At low temperatures the exchange energy between two localized spins $\langle J(r, r')\rangle$ averaged over the scattering potential configurations decays exponentially with $|\mathbf{r}-\mathbf{r}'|$ (Ref. 7) for $|r-r'|\gg l$. Here *r* and *r'* are coordinates of spins and brackets $\langle \rangle$ stand for averaging over realizations of the scattering potential in the metal and the ferromagnets. Recent experiments on ferromagnet-nonferromagnet-ferromagnet metallic multilayers⁸ imply, however, that the exchange energy between the ferromagnets does not decay exponentially at $L \ge l$ and that the relative orientation of the ferromagnet's magnetizations is perpendicular independently of *L*. Phenomenologically, this situation can be described by an effective energy per unit area

$$
E = -J^{0}(m_1^{0} \cdot m_2^{0}) + B[(m_1^{0} \cdot m_2^{0})^2 - 1]
$$
 (1)

in the case $2B \ge |J^0|$. Here m_1^0 and m_2^0 are unit vectors parallel to spatially averaged magnetizations of the ferromagnetic films. Indices 1,2 indicate the first and the second ferromagnetic film, respectively, J^0 and $B(\theta)$ are bilinear and biquadratic coupling coefficients. In general, $B(\theta)$ is a function of the angle θ between m_1^0 and m_2^0 .

In this paper we discuss a theory of this phenomenon. It has been shown in Refs. 9–11 that the exponential decay of the average $\langle J(r,r')\rangle$ is connected to the fact that $J(r,r')$ has a random sign at large *L*. The modulus of the exchange interaction decays with *L* as a power law.

We can introduce a local exchange energy $J(\rho)$ between the ferromagnets as an average of $J(r,r')$ over a ferromagnet surface area of order of L^2 . Here ρ is the coordinate along the films. Since the interfilm's exchange energy $J(\rho)$ is small compared to the exchange energy inside the ferromagnetic films, the spatial dependence of the magnetizations can be neglected on the scale of order of *L*. According to Slonczewski,^{12,13} the biquadratic term proportional to *B* in Eq. (1) can originate from the existence of spatial fluctuations of the sign of the exchange interaction $J(\rho) = \langle J \rangle$ $+\delta J(\rho)$ along the layers. The fluctuations of $J(\rho)$ cause fluctuations of directions of magnetizations. The energy associated with spatial fluctuations of the magnetization direction can be represented as

$$
E[J(\rho), m_i(\rho)]
$$

= $-\int d^2 \rho J(\rho) [m_1(\rho) \cdot m_2(\rho)]$
+ $\alpha d \int d^2 \rho \bigg[\frac{\partial m_1(\rho)}{\partial \rho} \cdot \frac{\partial m_1(\rho)}{\partial \rho} + \frac{\partial m_2(\rho)}{\partial \rho} \cdot \frac{\partial m_2(\rho)}{\partial \rho} \bigg],$ (2)

where the first term corresponds to the interfilm exchange energy, the second term is associated with the gradients of magnetizations inside the films, *d* is the thickness of the ferromagnetic films thickness, and α is an intralayer ferromagnetic stiffness.

FIG. 1. A schematic picture of the ferromagnet (F) -normal metal (N)-ferromagnet system.

In the case when $\delta J(\rho)$ \gg $\langle J \rangle$ and $J(\rho)$ has a random sign, the energy $E[J, m_i(\rho)]$ has a minimum at a sample-specific realization $m_i(\rho) = m_i^0 + \delta m_i(\rho; [\delta J])$ with $m_1^0 \perp m_2^0$ (Refs. 12) and 14 and

$$
B = \frac{B_0}{\alpha d} G(\theta),\tag{3}
$$

$$
G = \int d^2 \rho \langle \delta J(\rho) \delta J(0) \rangle.
$$
 (4)

Here B_0 is a number of order unity.¹⁴

Let us consider the case when $J(\rho)$ has a random sign due to mesoscopic fluctuations of Ruderman-Kittel oscillations inside the metal. $9-11$ We assume that the thickness of ferromagnetic films d is larger than the spin diffusion length L_s $= \sqrt{D/\omega_s}$ and that the thermal coherence length $L_T = \sqrt{D/T}$ is larger than *d* and *L*. The latter inequality allows us to neglect the temperature dependence of *B*. Here *D* is the diffusion constant of conducting electrons, which is assumed to be the same in the ferromagnetic and nonferromagnetic parts of the sample, and ω_s is the exchange spin splitting energy in the ferromagnets. We will show that in the case $L_s \leq L$,

$$
G = \gamma(\theta) \left(\frac{E_c}{L}\right)^2 \tag{5}
$$

while in the case $L_s > L$,

$$
G = \gamma_1(\theta) \left(\frac{\omega_s}{L_s}\right)^2.
$$
 (6)

Here γ and γ_1 are smooth functions of θ of order unity and $E_c = D/\pi L^2$ is the Thouless energy. Qualitatively, Eqs. (5) and (6) can be understood as follows: In the case $|\mathbf{r}-\mathbf{r}'|\ge l$ the random oscillations of $J(r, r')$ exhibit long-range sign correlations.¹⁵ In the case $L_s \ll L$ these long-range correlations should be cut off at a length of the order of *L*. As a result, the amplitude of fluctuations of the exchange energy averaged over the area of order of L^2 is of order E_c ; and they are δ correlated at distances larger than *L*. This leads to Eq. (5) .

In the opposite limit $L_s \ge L$ the cut off length is L_s . The amplitude of fluctuations of the exchange energy averaged over the area of order L_s^2 is of order ω_s . This leads to Eq. (6) , which is independent of L .

At large nonferromagnetic metal thicknesses $L \ge l$ the average $J^0 \sim \exp(-L/l)$ decays exponentially⁷ and the biqua-

FIG. 2. Diagrams for calculation of the correlation function $\langle \delta N(\mu_1 ,\theta_1) \delta N(\mu_2 ,\theta_2) \rangle$. Solid lines correspond to electron Green's functions and dashed lines correspond to the correlation function of the scattering potential $\langle U(\mathbf{r})U(\mathbf{r}')\rangle$.

dratic contribution described by Eqs. (5) and (6) dominates exchange energy between the ferromagnetic films.

II. DERIVATION OF RESULTS

To derive the results presented above we describe the exchange energy splitting in the ferromagnet with the help of an effective Hamiltonian

$$
H = H_0 + h(r; \theta) \sigma.
$$
 (7)

Here H_0 is the Hamiltonian of the free-electron gas in a random potential $U(r)$, $h(r, \theta) \equiv \omega_s m(r, \theta)$ is the effective magnetic field which is acting only on electron spins, $m(r, \theta)$ denotes the unit vector parallel to the magnetic moment in the ferromagnets in the case when the angle between m_1^0 and m_2^0 is θ , and $\boldsymbol{\sigma} = {\sigma_x, \sigma_y, \sigma_z}$ is the Pauli matrices vector. We assume the following correlation properties of the random potential: $\langle U(\mathbf{r})\rangle = 0$ and $\langle U(\mathbf{r})U(\mathbf{r}')\rangle$ $= [1/(2 \pi \nu_0 \tau)] \delta(r - r')$. Here ν_0 is the density of states at the Fermi level, τ is the mean free scattering time of electrons.

To get the correlation function $\langle \delta J(0)\delta J(\rho)\rangle$ we consider sample specific fluctuations of the thermodynamic potential $\Omega(\theta)$ of the electrons as a function of θ ,

$$
\Omega(\theta) = \langle \Omega(\theta) \rangle + \delta \Omega(\theta). \tag{8}
$$

Using the identity $d\delta\Omega(\theta)/d\theta = \int d^2\theta \delta J(\rho)$ we get

$$
G(\theta) = \left\langle \left(\frac{d \,\delta\Omega}{d\,\theta}\right)^2 \right\rangle. \tag{9}
$$

In the case of noninteracting electrons we can express the thermodynamic potential as $\Omega = \int_0^{\mu} d\mu N(\mu)$, where $N(\mu)$ is the number of electrons at a given chemical potential μ . Then, the correlation function of fluctuations of the thermodynamic potential has the form

$$
\langle \delta\Omega(\theta_1)\delta\Omega(\theta_2)\rangle = \int_0^\mu d\mu_1 d\mu_2 \langle \delta N(\mu_1, \theta_1)\delta N(\mu_2, \theta_2)\rangle.
$$
\n(10)

To calculate it we use the usual diagram technique for averaging over configurations of the disordered potential.¹⁶ Diagrams for the correlation function of the number of electrons are shown in Fig. 2. As a result we have

$$
\langle \delta N(\mu_1, \theta_1) \delta N(\mu_2, \theta_2) \rangle
$$

\n
$$
= \frac{2}{\pi} T \sum_{\omega_n > 0} \omega_n \text{Re} \int_V d^3 r d^3 r' D_{\alpha\beta}^{\gamma\nu}(r, r'; \omega_n) D_{\beta\alpha}^{\gamma\gamma}(r', r; \omega_n)
$$

\n
$$
= \frac{2}{\pi} T \sum_{\omega_n > 0} \text{Re} \int \frac{d^2 q}{(2\pi)^2}
$$

\n
$$
\times \sum_m \frac{\omega_n}{[E_m(\theta_1, \theta_2) + Dq^2 + \omega_n + i(\mu_1 - \mu_2)]^2}, \quad (11)
$$

where $\omega_n = 2 \pi nT$ is the Matsubara frequency, $n=1,2...$ and $\alpha, \beta, \gamma, \mu$ are spin indices. Diffusion propagators $D_{\alpha\beta}^{\gamma\nu}(r,r';\omega_n)$ obey the equation

$$
\{[-D\triangle + \omega_n + i(\mu_1 - \mu_2)]\delta_{\alpha\xi}\delta_{\gamma\mu} + i[\boldsymbol{h}(\boldsymbol{r};\theta_1)\boldsymbol{\sigma}_{\gamma\mu}\delta_{\alpha\xi} \n-\boldsymbol{h}(\boldsymbol{r};\theta_2)\boldsymbol{\sigma}_{\alpha\xi}\delta_{\gamma\mu}]\}D_{\xi\beta}^{\mu\nu}(\boldsymbol{r},\boldsymbol{r}';\omega_n) = \delta(\boldsymbol{r}-\boldsymbol{r}')\delta_{\gamma,\nu}\delta_{\alpha,\beta}.
$$
\n(12)

The second equality in Eq. (11) is the representation in terms of eigenvalues of Eq. (12) . In the case of the system geometry shown in Fig. 1, the eigenvalues are equal to Dq^2 $+E_m(\theta_1, \theta_2)$. Here the spectrum $E_m(\theta_1, \theta_2)$ is determined by the equation

$$
\left(-D \,\delta_{\alpha\xi} \delta_{\gamma\mu} \frac{d^2}{dz^2} + i[\,\boldsymbol{h}(\boldsymbol{r};\theta_1) \,\boldsymbol{\sigma}_{\gamma\mu} \delta_{\alpha\xi} - \boldsymbol{h}(\boldsymbol{r};\theta_2) \,\boldsymbol{\sigma}_{\alpha\xi} \delta_{\gamma\mu}]\right) \times \Psi_m(z;\mu,\xi) = E_m \Psi_m(z;\gamma,\alpha). \tag{13}
$$

To calculate Eq. (11) we use following equalities

$$
\int \frac{d^2 q}{(2\pi)^2} \sum_{m} [E_m + Dq^2 + \omega_n + i(\mu_1 - \mu_2)]^{-2}
$$

= $-\frac{d}{d\omega_n} \int \frac{d^2 q}{(2\pi)^2} \frac{1}{4\pi i}$
 $\times \int_c dp \frac{1}{\frac{Dp^2}{L^2} + Dq^2 + \omega_n + i(\mu_1 - \mu_2)} \frac{d \ln \det(p)}{dp}$
= $\frac{1}{8\pi D} \frac{d}{d\omega_n} \ln[\det(ip_0) \det(-ip_0)].$ (14)

Here det(p) = $\Pi_m(p - E_m)$ is the spectral determinant of Eq. (13), and $p_0 = \sqrt{(L^2/D)} [\omega_n + i(\mu_1 - \mu_2)]$. In Eq. (14) the integration contour C runs around zeros of $det(p)$. Let us note that although Eqs. (10) and (11) are formally divergent, their contribution to $\langle (d \delta\Omega/d\theta)^2 \rangle$ is finite.

Let us consider the case $L_s \ll L_d$ when results do not depend on ω _s. To define boundary conditions for Eq. (13) it is convenient to introduce operators

$$
S_{\pm} = \frac{1}{2} [1 \pm (m(z; \theta_1) \sigma_1) [m(z; \theta_2) \sigma_2] \}.
$$
 (15)

Then the boundary conditions for $\Psi_m(z; \gamma, \alpha)$ are: $(d/dz)S_+\Psi_m=0$ at $z=\pm(d+L/2)$ and $S_-\Psi_m=0$ at $L/2$ $\langle \vert z \vert \langle d \vert + L/2 \rangle$. As a result the solution of the eigenvalue problem Eq. (13) gives the following spectral determinant:

$$
\det(ip) = \left\{ \sinh p \sinh(1 + 2d/L)p
$$

$$
+ [1 - \cos(\theta_1 - \theta_2)] \frac{1 + \cosh(2dp/L)}{4} \right\}
$$

$$
\times \left\{ 1 + \cosh(2 + 2d/L)p - [1 + \cos(\theta_1 + \theta_2)] \right\}
$$

$$
\times \frac{1 + \cosh(2dp/L)}{2} \right\}.
$$
(16)

Integrating over the chemical potentials, summing over Matsubara frequencies, and considering the case $L_T = \sqrt{D/T}$ $\geq L$;*d* we get

$$
G = \frac{S}{8L^2} E_c^2 \int_0^\infty dp \, p^5 \Phi(p)
$$

$$
\times \left[1 - \frac{1}{2} \frac{d}{d\theta} \left(\frac{\sin 2\theta}{1 + \Phi(p)} \frac{1 - \cos 2\theta}{4} \right) \right], \quad (17)
$$

where

$$
\Phi(p) = \frac{1 + \cosh 2dp/L}{\sinh p \sinh(1 + 2d/L)p}.
$$
 (18)

The main contribution to the integral in Eq. (17) is from region the $p > 1$. In this case $\Phi(p) < 1$ and the expression in the brackets in Eq. (17) can be approximated as $\sin^2 \theta$ with a precision of order unity. The case when $L_s \ge L$ can be studied in the same way giving Eq. (6) .

III. CONCLUSION

We have shown that the biquadratic part of the exchange energy between the ferromagnetic films, which originates from mesoscopic fluctuations, decays as a power law of the nonferromagnetic metal thickness L [see Eqs. (5) and (6)]. Since the average exchange energy decays exponentially with *L*, the exchange energy has a random sign. This leads to the perpendicular relative orientation of magnetizations of the ferromagnetic films.

Let us estimate the magnitude of the exchange interaction between the magnetizations of the ferromagnetic films. Assuming that $L = 6 \times 10^{-7}$ cm, $d=1.5 \times 10^{-7}$ cm, $D \sim 1$ -10 cm²/sec, $\omega_s = 10^3$ K, and $\alpha \sim 2 \times 10^{-6}$ erg/cm we get an estimate $L_s \sim 10^{-7}$ cm and $B(\theta) \sim (10^{-3} - 10^{-1})$ \times (erg/cm²). We think that the theory presented above can be relevant for the experiment 8 where biquadratic coupling magnitude was of order 10^{-3} erg/cm².

ACKNOWLEDGMENTS

The work of B.S. was supported by the Division of Material Sciences, U.S. National Science Foundation under Contract No. DMR-9625370 and by U.S.-Israeli Binational Science Foundation Grant No. 94-00243.

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