Magnetic Exchange Coupling in Ferromagnet/Superconductor/Ferromagnet Multilayers

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The possibility of magnetic exchange coupling between two ferromagnets (F) separated by a superconductor (S) spacer is analyzed using the functional integral method. For this coupling to happen three basic conditions need to be satisfied. First, an indirect exchange coupling between the ferromagnets must exist when the superconductor is in its normal state. Second, superconductivity must not be destroyed due to the proximity to ferromagnetic boundaries. Third, roughness of the F/S interfaces must be small. The appearance of the superconducting gap causes a reduction of the indirect exchange coupling existent in the normal state. This reduction is temperature dependent, being weaker near the critical temperature and stronger at zero temperature. [S0031-9007(97)03941-0]

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The problem of magnetic coupling in ferromagnetic metal/normal metal multilayers has received considerable attention in recent years, both on the experimental side [1] and on the theoretical side [2]. The main features that have emerged from these experiments were associated with an indirect exchange coupling between the ferromagnetic layers via the normal metal host. An oscillatory coupling as a function of the thickness of the normal metal spacer was ubiquitously observed in several multilayered systems [3]. The prevailing experimental evidence indicates that the exchange coupling with metal spacers is short ranged, i.e., the magnetic coupling can be observed only across a layer of thicknesses 10 to 130 Å [4]. Thus, the key question, for both theory and experiment, is the following. Can such a thin metallic layer survive pair breaking effects of the ferromagnetic layers from both sides and yet remain superconducting? It is the purpose of this paper to show that appropriate choices of superconductor and ferromagnet lead to the survival of superconductivity and to new effects on the magnetic coupling. Presently there are no experimentally known multilayered systems that show magnetic coupling both above and below T_c. The main reasons for that are that the conditions that need to be satisfied are difficult to achieve. The desirable superconductor should have *high* critical temperature and *short* coherence length, while the desirable ferromagnet should be *metallic* with *not so large* pair breaking effects. Furthermore, the desirable F/S interfaces should be atomically flat and well lattice matched to avoid the effects of roughness and strain. Thus, ideal systems to study are high T_c superconductor/colossal magnetoresistance ferromagnet multilayers like $Nd_{2-x}Ce_xCuO_4/La_{1-y}Sr_yMnO_3$ or $Nd_{2-x}Ce_xCuO_4/La_{3-y}Sr_yMnO_7$ (for the *s*-wave case) and $YBa_2Cu_3O_{7-\delta}/La_{1-y}Sr_yMnO_3$ or $YBa_2Cu_3O_{7-\delta}/La_{3-y}Sr_yMn_2O_7$ (for the *d*-wave case).

If the superconducting metal, in its normal state, allows an indirect exchange coupling between ferromagnetic layers, it is valid to ask the following questions. First, how does the presence of ferromagnetic layers affect the superconductivity of the spacer? Second, does anything dramatic happen to the magnetic coupling when the system is cooled through the superconducting transition temperature of the spacer? Third, what happens to the magnetic coupling at very low temperatures when the superconductivity of the spacer is well established? Fourth, what are the effects of interfacial roughness? These questions are the central topic of this paper and they are intended to establish the conditions under which magnetic coupling in $F/S/F$ multilayers should exist.

The recent published literature on F/S multilayers has focused mostly on the changes of the critical temperature T_c of the superconductor [5,6] as a function of thickness of the ferromagnetic layers. The experimental reports have been mixed. In the cases of Nb/Gd multilayers [7] the observation of a nonmonotonic T_c has been attributed either to a change in the underlying pair breaking mechanism with increasing thickness of the Gd layer (Strunk *et al.* [6]) or to evidence for the predicted π-phase coupling [7] as a function of thickness of the Gd layer (Jiang *et al.* [7]). More recently, there were experimental attempts to observe magnetic coupling in $F/S/F$ multilayer systems $Fe_4N/NbN/Fe_4N$, Ni/Nb/Ni, and GdN/NbN/GdN [8]. Unfortunately these experimental attempts have failed. No magnetic coupling was observed even when the superconductor was in its normal state, thus indicating that the effects of interfacial roughness and strains in these systems may be strong, unlike in the more usual multilayers [1,3,4].

The only theoretical work relevant to these questions that has been published so far, to the best knowledge of the author, is the pioneering work of Sipr and Gyorffy [9]. They have analyzed *numerically* the possibility of magnetic coupling through a superconductor at zero temperature without solving for the gap equation self-consistently. The work presented here distinguishes itself from the work of Sipr and Gyorffy in the following ways. The present work shines light on the *temperature* dependence of the magnetic coupling through the superconductor (both at $T \approx 0$ and $T \approx T_c$), while solving the gap equation self-consistently in the asymptotic limit of large spacer thicknesses. The results obtained here are mostly *analytical* in contrast with the *numerical* work of Sipr and Gyorffy [9].

For the purpose of answering the questions above, the system chosen here is a trilayer consisting of two identical ferromagnets of thickness d_f separated by a superconductor of thickness d_s. The ferromagnetic layer is labeled the F layer, and the trilayer system will be referred to as $F/S/F$, when the spacer is superconducting (S layer) and $F/N/F$, when the spacer is in its normal state (N layer). The underlying assumptions are as follows. It is assumed that $d_f \gg d_s$, such the ferromagnetic layers can be treated as semi-infinite. When the spacer is in its normal state (N layer), it is assumed that an indirect exchange coupling exists between the ferromagnetic layers (F layers). It is further assumed that the critical temperature T_c of the superconductor is much smaller than the Curie temperature T_f of the ferromagnet, such that fluctuation effects on the magnetism are negligible. Furthermore, the F layers are ferromagnetic metals, the superconductor is assumed to be s-wave BCS type, and the F/S interface is assumed to be smooth, i.e., atomically flat.

Under all these assumptions, the Hamiltonian density is written as

$$H = H_S + H_F\,, \tag{1}$$

where $H_S = \bar{\Psi}_\alpha(r)\{[K + U(\mathbf{r})]\delta_{\alpha\gamma}\}\Psi_\gamma(r) + V$, and $H_{SF} = \bar{\Psi}_\alpha(r)\{(H_c)_{\alpha\gamma}\}\Psi_\gamma(r)$. The indices α and γ indicate spin components and repeated indices indicate summation. The kinetic energy term is $K = (i\nabla)^2/2m - \mu$, while the potential energy is $U(\mathbf{r}) = U_0 F(r)$, where $F(r) = [\Theta(x - d_s/2) + \Theta(-x - d_s/2)]$. Here U_0 can be positive or negative. The only exchange interaction considered in the ferromagnetic layers is between the spins of itinerant electrons, but a real space representation of the exchange interaction is used given that the $F/S/F$ system is inhomogeneous. Thus, the exchange interaction $(H_c)_{\alpha\gamma}$ is more transparently written as

$$(H_c)_{\alpha\gamma} = -[(\sigma_x)_{\alpha\gamma}h_x(r) + (\sigma_y)_{\alpha\gamma}h_y(r) + (\sigma_z)_{\alpha\gamma}h_z(r)],$$

where $h_i(r) = \int dr'\, J_i(r,r')S_i(r')F(r)$ is an effective exchange field felt only within the boundaries of the ferromagnet. Here the spin variable $S_i(r') = \bar{\Psi}_\mu(r') \times (\sigma_i)_{\mu\nu}\Psi_\nu(r')$. For $T_f \gg T_c$, and $|h_z| \gg \max(|h_y|, |h_x|)$, the F layers have negligible magnetization fluctuations at low T and an easy axis along the z axis. Thus, $h_z(r)$ can be replaced by its average value $\langle h_z(r)\rangle = J\langle S_z\rangle F(r)$. The potential energy is $V = -g\bar{\Psi}(r)_\uparrow\bar{\Psi}(r)_\downarrow\Psi(r)_\downarrow \times \Psi(r)_\uparrow[1 - F(r)]$ resulting from a delta function contact attraction. This attractive interaction leads to an s-wave superconductor.

To estimate the change in T_c of the superconductor one needs to worry only about the *restricted* partition function Z at temperature $T = \beta^{-1}$, which can be written as a functional integral [10] with action $S = \int_0^\beta d\tau \int d\mathbf{r} \times [\bar{\Psi}_\alpha(r)\partial_\tau\Psi_\gamma(r) - H_S - H_{SF}]$, where $r = (\mathbf{r},\tau)$ and $\hbar = k_B = 1$. The Hubbard-Stratonovich pair field $\Delta(\mathbf{r},\tau)$ is introduced and upon integration over the fermionic variables the partition function $Z = \int D\Delta D\bar{\Delta} \times \exp(-S_{\rm eff}[\Delta,\bar{\Delta}])$ is obtained. The effective action is $S_{\rm eff} = \int_0^\beta d\tau\{\beta|\Delta(r)|^2/g - \mathrm{Tr}\ln\beta G^{-1}\}/\beta$ and the inverse Nambu propagator is $G^{-1} = -\partial_\tau - [K + U(\mathbf{r}) + (H_c)_\sigma]\sigma_z + \Delta(r)\sigma^+ + \bar{\Delta}(r)\sigma^-$, where $\sigma^\pm = (\sigma_z \pm i\sigma_y)/2$, with σ_j being the Pauli matrices.

In the limit $k_{F_s}d_s \gg 1$, take the volume $V = L_xL_yL_z$, and define the Fermi momentum via the relation $E_F = k_{F_s}^2/2m = (3\pi^2 n)^{2/3}/2m$. Thus, choose $L_x = d_s$ and $\min\{L_y, L_z\} \gg d_s$, take the asymptotic limit $k_{F_s}d_s \gg 1$, and perform an expansion of $S_{\rm eff}$ in powers Δ to obtain the static linearized Ginzburg-Landau equation

$$\epsilon\Delta(x) - \xi_{GL}^2\frac{d^2}{dx^2}\Delta(x) = 0 \tag{2}$$

with boundary conditions $[d\Delta(x)/dx]_{x=\pm ds/2} = \mp\Delta(x)/b$ at the F/S interfaces. The coefficient b is the extrapolation length $k_{F_s}b \approx \lambda[E_F/2T_c][1 + \sqrt{1 - U_0/E_F}]/\sqrt{1 - U_0/E_F}$, with $\lambda = 1 - [1/4] \times (J\langle S_z\rangle/\Lambda)^2$, when $E_F > U_0$ and $|J\langle S_z\rangle| \ll \Lambda = \min\{2\pi T_c, (E_F - U_0)\}$.

The coefficients appearing in (2) are $\epsilon = [T - T_c]/T_c$ and $k_{F_s}\xi_{GL} = \alpha[E_F/T_c]$, where the numerical coefficient is $\alpha = [2e^{-2\gamma}/\pi]\sqrt{7\zeta(3)/48}$. The solution of (2) is $\Delta(x) = \Delta_0\cos(kx)$ inside the superconductor. In the limit $k_{F_s}d_s \gg 1$, the parameter $k \approx \pi/d_{\rm eff}$. This leads to a correction to the critical temperature of the bulk superconductor

$$\epsilon = -\left(\frac{\pi\xi_{GL}}{d_{\rm eff}}\right)^2, \tag{3}$$

where $d_{\rm eff} = (d_s + 2b)$ is the *effective* length of the superconductor. The suppression of T_c from the bulk value is small provided that $\pi\xi_{GL} \ll d_{\rm eff}$ and thus superconductivity survives. A strong suppression of T_c happens when $\pi\xi_{GL} \sim d_{\rm eff}$. Notice that the choices of the superconductor and the ferromagnetic metal are very important in order to have a weak suppression of T_c. The choice of the spacer should be a superconductor with a *high* bulk T_c and a *short* coherence length $k_{F_s}\xi_{GL}$. For the ferromagnet, it is important to be a *metal* ($E_F > U_0$), with $T_f \gg T_c$, but with *not so strong* pair breaking effects, i.e., $\alpha_{\rm pb} = |J\langle S_z\rangle|/2\pi T_c \ll 1$. To illustrate this point, take the ideal case of $Nd_{2-x}Ce_xCuO_4/La_{1-y}Sr_yMnO_3$ multilayers, where the bulk T_c of $Nd_{2-x}Ce_xCuO_4$ is $T_c \approx 30$ K, and $\xi_{GL} \approx 10$ Å [11], and the ferromagnet $La_{1-y}Sr_yMnO_3$ with $T_f \approx 300$ K and $J \approx 10$ K [12]. Ignoring anisotropy and complex spin structure effects, the pair breaking parameter $\alpha_{\rm pb} \approx 0.027$, and thus Eq. (3) can still be used to estimate the reduction of T_c due to the ferromagnetic boundaries. A modest

$E_F \approx 10^4$ K with $k_F^{-1} \approx 0.38$ Å leads to an extrapolation length $b \approx 60$ Å. The effective length of the superconductor becomes $d_{\rm eff} = d_{\rm s} + 2b$, and for $d_{\rm s} = 10$ Å, the reduction in T_c from its bulk value $T_c(\infty)$ is only 6%. Thus, for spacer thickness in the range $d_{\rm s} = 10$ Å and $d_{\rm s} = 130$ Å (where magnetic coupling is observed in many systems [4]), the suppression of T_c is even smaller and superconductivity survives.

In this case, the exchange coupling H_c appearing in (1) may be treated as a perturbation. Define the inverse Nambu propagator $G^{-1} = \mathcal{G}^{-1} + (H_c)_\sigma \sigma_z$, where an explicit separation between the inverse propagator $\mathcal{G}^{-1} = -\partial_\tau - [K + U(\mathbf{r})]\sigma_z + \Delta(r)\sigma^+ + \bar{\Delta}(r)\sigma^-$ (in the absence of ferromagnetic boundaries) and the exchange contribution $(H_c)_\sigma \sigma_z$ (due to the ferromagnetic boundaries) is made. This separation is very useful because the effective action $S_{\rm eff}[\Delta, \bar{\Delta}]$ can now be written as

$$S_{\rm eff} = \int_0^\beta d\tau \left\{ \frac{|\Delta(r)|^2}{g} - \frac{1}{\beta} \,\mathrm{Tr} \ln \beta \mathcal{G}^{-1} - H_{\rm eff} \right\}. \tag{4}$$

Minimization of the effective action without $H_{\rm eff}$, i.e., $\delta S_{\rm eff}(H_{\rm eff} = 0)/\delta \Delta^*(\mathbf{r}) = 0$ leads to the saddle point gap equation $\Delta(\mathbf{r})/g = \mathrm{Tr}[G\sigma^-]/\beta$, which can be used to compute the effective Hamiltonian $H_{\rm eff} \approx -\mathrm{Tr}[(H_c)_\sigma \sigma_z \mathcal{G}]^2/2\beta$. The effective magnetic Hamiltonian $H_{\rm eff}$ across the superconductor is

$$H_{\rm eff} = -\frac{E_J}{\mathcal{N}_{3D}} \sum_{nm,k_\perp} \int \int_{\partial\Omega} dx_1 dx_2 \chi_{nm}(k_\perp), \tag{5}$$

where the energy scale $E_J = \frac{1}{2} J_+ J_- \langle S_z \rangle_+ \langle S_z \rangle_- \mathcal{N}_{3D} V_s$ contains the density of states of the bulk superconductor $\mathcal{N}_{3D}$. The domain $\partial\Omega$ includes both ferromagnets. The indices $\pm$ refer to the F layers. The matrix element $\chi_{nm}(k_\perp) = 2D_{nm}(x_1, x_2)Q_{nm}(k_\perp)$ defines the *nonlocal* "susceptibility" of the system which appears under the double integration over x_1 and x_2. The weighting factor $D_{nm}(x_1, x_2) = w_n(x_1)w_n(x_2)w_m(x_1)w_m(x_2)$ contains the eigenstates $w_n(x)$, i.e., $H_e(x, k_\perp)w_n(x) = \xi_n(k_\perp)w_n(x)$, of the one dimensional Hamiltonian $H_e(x, k_\perp) = -[1/2m]\partial^2/\partial x^2 - \mu_{\rm eff} + U(x)$, where the *effective* chemical potential $\mu_{\rm eff} = \mu - k_\perp^2/2m$ with $k_\perp^2 = k_y^2 + k_z^2$. The additional term

$$Q_{nm}(k_\perp) = C_T^{nm} T_{nm} + C_P^{nm} P_{nm}$$

contains the coherence factor $C_T^{nm} = [p_{u_n} p_{u_m} + p_{v_n} p_{v_m}]^2$ and thermal factor $T_{nm} = [f(\epsilon_m) - f(\epsilon_n)]/[\epsilon_m - \epsilon_n]$ in the quasiparticle-quasihole channel, and the coherence factor $C_P^{nm} = [p_{u_n} p_{v_m} - p_{u_m} p_{v_n}]^2$ and the thermal factor $P_{nm} = [f(\epsilon_m) + f(\epsilon_n) - 1]/[\epsilon_m + \epsilon_n]$ in the quasiparticle-quasiparticle channel. The coefficients p_{u_n} and p_{v_n} defined, respectively, by $|p_{u_n}|^2 = [1 + \xi_n/\epsilon_n]/2$ and $|p_{v_n}|^2 = [1 - \xi_n/\epsilon_n]/2$, reflect the simplification $\Delta \approx \langle \Delta(x) \rangle$. This simplification occurs only when $\Delta(x)$ can be approximated by its spatially averaged value $\langle \Delta(x) \rangle = g \sum_{n,k_\perp} \langle w_n^2(x) \rangle \{1 - 2f[\epsilon_n(k_\perp)]\}$, thus leading to the approximate eigenenergies $\epsilon_n^2 \approx \xi_n^2 + \Delta^2$.

Assume that $E_F \gg \max\{\Delta, T\}$. In the asymptotic limit $k_{F_s} d_{\rm s} \gg 1$, the single particle wave functions $w_n(x)$ are standing waves, the discrete quantum number n becomes a continuous *momentum* index, and the sums over the indices (n, m) become integrals. Notice that the momentum $k_\perp$ is conserved across the interface. At low temperatures ($\Delta/T \gg 1$) the form of the coupling is

$$H_{\rm eff} = -E_J \frac{\mathcal{F}}{2\pi^2} \frac{\cos(2k_{F_s} d_{\rm s})}{(2k_{F_s} d_{\rm s})^2} \exp(-k_{F_s} d_{\rm s} \Delta/E_F). \tag{6}$$

While at temperatures close to $T_c (\Delta/T \ll 1)$, the magnetic coupling becomes

$$H_{\rm eff} = -E_J \frac{\mathcal{F}}{2\pi^2} \frac{\cos(2k_{F_s} d_{\rm s})}{(2k_{F_s} d_{\rm s})^2} \left[1 - \frac{2}{3\pi^2} (\Delta/E_F)^2 \right]. \tag{7}$$

It is important to analyze the qualitative features of the previous expressions. First, notice that the period of oscillation of the magnetic coupling across the superconductor is entirely controlled by the *pseudo*-Fermi momentum of the superconductor k_{F_s} defined above. The appearance of superconductivity does not introduce any new periods. This is expected since no new momentum modulation in the spin degrees of freedom occurs, through the appearance of the superconducting gap, at length scales comparable to $k_{F_s}^{-1}$. At low temperatures ($\Delta/T \gg 1$) there is an energy cost to be paid. Almost all of the electrons that were easily polarized in the normal state of the superconductor are now paired into singlets. As a result the spin polarization of the superconductor is costly, i.e., when summing over all intermediate states (virtual quasiparticle states) there is a minimum energy required: the superconducting gap [13]. The gap introduces a new length scale ξ_{GL} which controls the decay of the coupling. Notice that $\Delta/E_F \approx 0.15/k_{F_s} \xi_{GL}$ in the asymptotic limit $k_{F_s} d_{\rm s} \gg 1$. At temperatures close to T_c, when ($\Delta/T \ll 1$), there is nearly no additional decay caused by the superconducting gap. The gap is so small that intermediate quasiparticle states are strongly thermally populated. Near T_c, these intermediate quasiparticle states resemble the normal state eigenfunctions, except for the presence of a small superfluid density controlled by Δ. As a result, only an overall reduction of the prefactor of the oscillations appears. This reduction is controlled by $\Delta/E_F = 0.26(1 - T/T_c)^{1/2}/k_{F_s} \xi_{GL}$, which vanishes at $T = T_c$. Second, notice that the asymptotic decay is proportional to $(k_{F_s} d_{\rm s})^{-2}$ instead of the usual RKKY decay $(k_F r)^{-3}$ for magnetic impurities. The change in the form of the decay can be viewed as a geometrical effect since the magnetic "impurities" are now two semi-infinite ferromagnets and as a result the magnetic coupling has to be more effective. Third, since the potential U_0 labels the different types of ferromagnets, it is important to analyze the dependence of $H_{\rm eff}$ on U_0/E_F. In Eqs. (6) and (7), $\mathcal{F} = \gamma_0/(\gamma_0 + 1)^2$, where $\gamma_0 = \sqrt{1 - U_0/E_F}$. The amplitude of the coupling is gradually reduced from its maximal value at $U_0 = 0$ until it vanishes at $U_0 = E_F$. This reduction can be interpreted as the disappearance of spin dependent states at the Fermi energy of the ferromagnets as $U_0/E_F \ll 1$ increases to $U_0/E_F \approx 1$; thus it is essentially a density of states effect. Fourth, the energy scale E_J is proportional to the product $J_+ J_- \langle S_z \rangle_+ \langle S_z \rangle_-$, and thus depends both on the strength of the exchange coupling of the itinerant electrons (J_+, J_-) and on the magnetization of the ferromagnets $(\langle S_z \rangle_+, \langle S_z \rangle_-)$.

The effects of roughness on the critical temperature of the superconductor are negligible provided that the length scale of the roughness fluctuations $\ell_r \ll \xi_{GL}$. On the other hand, the effects of roughness on the magnetic coupling are very important, since ℓ_r can be easily of the order of the magnetic oscillation period $\ell_p = \pi/k_F$, and thus average out the oscillatory behavior. These studies have been performed experimentally in Fe/Cr/Fe multilayers [14], and also theoretically for several F/N/F multilayers [15]. The case of F/S/F multilayers is not so different, i.e., roughness is also expected to affect the magnetic coupling in two basic ways. The first one is that the magnetic coupling must be averaged over thickness fluctuations of the superconductor film. The second way is that the magnetic coupling is affected by lateral fluctuations, which break translational invariance, and thus conservation of momentum parallel to the F/S interface. Only the first case is discussed here. Assuming that the thickness fluctuations are Gaussian around the mean value thickness $\bar{d}_s$ with variance σ, then in the limit that $\bar{d}_s \gg \sigma$ the only modification in Eqs. (6) and (7) consist in replacing the thickness d_s by the average thickness $\bar{d}_s$ and replacing $\cos(2k_{F_s} d_s)$ by $\exp[-(2k_{F_s}\sigma)^2/2][\cos(2k_{F_s}\bar{d}_s) + 2(2k_{F_s}\sigma)(\sigma/\bar{d}_s)\sin(2k_{F_s}\bar{d}_s)]$. Thus, provided that $\sigma < \sigma^* = \ell_p/\pi\sqrt{2}$, the magnetic coupling is not dramatically reduced. For instance, if $\ell_p = 10$ Å, then for $\sigma < \sigma^* = 2.25$ Å magnetic coupling should still be observed.

To conclude, it is worth emphasizing that magnetic coupling through a superconductor may be observable with present experimental resolution, provided that appropriate choices for the ferromagnet and the superconductor are made. It is also important to emphasize that magnetic coupling should already exist when the superconductor is in its normal state. In addition, the optimal experimental configuration should involve multilayered structures with thin ferromagnets and thin superconductors, although the systems discussed here referred only to trilayers with semi-infinite ferromagnets. Ideal systems to study experimentally should consist of a high temperature, short coherence length superconductor and a metallic ferromagnet with not so large pair breaking effects. Furthermore, the layers should be well lattice matched, and have smooth F/S interfaces. Systems that are particularly tempting to investigate, both theoretically and experimentally, consist of multilayers of high T_c cuprate superconductors/colossal magnetoresistance ferromagnets.

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