Indirect RKKY exchange and magnetic states of layered ferromagnet–superconductor structures

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The long-range antiferromagnetic contribution to RKKY exchange between localized spins which stems from the transition of a metal to a superconducting state strengthens toward the surface of a bulk sample or as the dimensionality of the sample is lowered. Several versions of a mutual accommodation of superconductivity and ferromagnetism in layered ferromagnet–superconductor structures are discussed.

The question of the mechanism for the interlayer exchange coupling in superlattices and F/S multilayer structures formed by an alternation of ferromagnetic (F) and superconducting (S) layers is a key to an understanding of effects stemming from the coexistence and mutual accommodation of two competing types of long-range order. Such phenomena as the slight suppression of superconductivity in EuO/V multilayer structures [1] and the crossover from a 2D behavior to a 3D behavior in Fe/V superlattices[2] with increasing temperature or decreasing thickness of the S layers cannot be explained on the sole basis of a π-phase nature of the superconductivity[3] in these structures. Furthermore, the existence of an internal field, which causes a splitting of the BCS peak in the density of states of quasiparticles in EuO/Al (Ref. 4) and EuS/Al (Ref. 5) F/S contacts and a saturation of it in a magnetic field can be explained in terms of a nonuniform magnetic order induced in the ferromagnetic film by the superconducting substrate. An indirect Ruderman–Kittel–Kasuya–Yosida (RKKY) exchange through conduction electrons of the superconducting layers could serve as the mechanism responsible for a long-range coupling between localized spins belonging to the same F/S boundary and between localized spins of neighboring F/S boundaries in superlattices. In this letter we examine the behavior of the RKKY potential as a function of the distance between localized spins and also as a function of the relative arrangement of these spins with respect to the superconductor–insulator interfaces for samples of several configurations: a half-space, a plate, and a wire. We propose a model for exchange interactions between localized spins in ferromagnetic-insulator–superconductor F/S systems. We use this model to discuss various possibilities for the striking of a compromise between superconductivity and ferromagnetism.

The behavior of the RKKY exchange integral as a function of the distance between localized spins $\mathbf{S}_\mathbf{r}$ and $\mathbf{S}_{\mathbf{r}'}$ is determined[6] by the spatial dispersion of the spin susceptibility of conduction electrons, $\chi(\mathbf{r},\mathbf{r}')$. The indirect-exchange Hamiltonian is

$$H_{ex}=-1/4I^2\chi(\mathbf{r},\mathbf{r}')(S_\mathbf{r}S_{\mathbf{r}'}), \qquad (1)$$

where I is the s-d exchange integral, and we assume $\hbar = k_B = \mu_B = 1$ everywhere. In the normal phase, the function $\chi_n(\mathbf{r},\mathbf{r}')$ exhibits characteristic Friedel oscillations, and the integral of this function over the entire space gives us a uniform Pauli susceptibility.

It was shown in Ref. 7 that the local spin polarization corresponding to the normal phase in a dirty superconductor is cancelled by the long-range increment of antiferromagnetic sign. This additional contribution to RKKY exchange arises from the exclusion of the contribution of paired electrons from the uniform spin polarization. The superconducting increment in the susceptibility, $\delta\chi_s(\mathbf{r},\mathbf{r}')$, can be written

$$\delta\chi_s(\mathbf{r},\mathbf{r}') = \chi_s(\mathbf{r},\mathbf{r}') - \chi_n(\mathbf{r},\mathbf{r}') = -2T\sum_{\omega} \Lambda_s(\mathbf{r},\mathbf{r}',\omega). \tag{2}$$

For the infinite dirty superconductor discussed in Ref. 7, the two-particle correlation function $\Lambda_s(\mathbf{r},\mathbf{r}',\omega)$ is given by the following expression in the hydrodynamic limit, i.e., at distances $R = |\mathbf{r}-\mathbf{r}'|$ greater than the mean free path l:

$$\Lambda_s(\mathbf{r},\mathbf{r}',\omega) = \frac{N(0)\Delta^2}{2DR(\omega^2+\Delta^2)}\exp(-R/\xi_\omega). \tag{3}$$

Here $N(0)$ is the density of states of conduction electrons at the Fermi surface; Δ is the superconducting order parameter; $D = vl/3$ is the diffusion coefficient; $\xi_\omega = [D/2(\omega^2+\Delta^2)^{1/2}]^{1/2}$ is the range of the correlation function $\Lambda_s(\mathbf{r},\mathbf{r}',\omega)$, which depends on the frequency $\omega = \pi T(2n+1)$; T is the temperature; and $n = 0, \pm 1, \pm 2, \ldots$. Since the uniform spin polarization must vanish at $T=0$ in a superconductor, we can derive a sum rule for $\Lambda_s(\mathbf{r},\mathbf{r}',\omega)$ by analogy with Ref. 8. In the case of a uniform superconductor with a coordinate-independent order parameter Δ and a density of states $N(0)$, this sum rule is

$$\int d^3r'\,\Lambda_s(\mathbf{r},\mathbf{r}',\omega) = \pi N(0)\Delta^2/(\omega^2+\Delta^2)^{3/2}. \tag{4}$$

To find the long-range part of the RKKY exchange in the case of superconductors which are spatially bounded by a surface σ, we can write $\Lambda_s(\mathbf{r},\mathbf{r}',\omega)$ as the solution of a boundary-value problem. It can be shown that expression (3) is a solution of a differential equation of the diffusion type,

$$[2(\omega^2+\Delta^2)^{1/2} - D\nabla_{\mathbf{r}}^2]\Lambda_s(\mathbf{r},\mathbf{r}',\omega) = 2\pi N(0)[\Delta^2/(\omega^2+\Delta^2)]\delta(\mathbf{r}-\mathbf{r}'). \tag{5}$$

Boundary conditions on this equation are found by integrating (5) over d^3r and imposing sum rule (4). The results are

$$D\mathbf{n}\nabla_{\mathbf{r}}\Lambda_s(\mathbf{r},\mathbf{r}',\omega)|_\sigma = 0, \tag{6}$$

where $\mathbf{n}$ is the normal to the superconductor–vacuum (–insulator) interface σ. Physically, Eq. (6) means there is no flux of Cooper pairs across the surface of the superconductor.

Solving (5) along with (6) under the assumption that the density of states $N(0)$ and the order parameter Δ are constant, aside from vanishing abruptly at the surface of the superconductor,[9] σ, we can find the coordinate dependence of the correlation function

$\Lambda_s(\mathbf{r},\mathbf{r}',\omega)$ for all geometric configurations of practical interest. In the present letter we consider three such configurations: a superconducting half-space, a plate, and a wire.

In the case of a superconducting half-space, $z,z' \geqslant 0$, we have

$$\Lambda_s(\mathbf{r},\mathbf{r}',\omega)=\frac{\pi N(0)\Delta^2}{\omega^2+\Delta^2}\int\frac{d^2q_\perp \exp[i\mathbf{q}_\perp(\boldsymbol{\rho}-\boldsymbol{\rho}')]}{(2\pi)^2Dk}[e^{-k|z-z'|}+e^{-k(z+z')}], \tag{7}$$

where $k^2=q_\perp^2+\xi_\omega^{-2}$, $\mathbf{q}_\perp=\mathbf{i}q_x+\mathbf{j}q_y$, and $\boldsymbol{\rho}=\mathbf{i}x+\mathbf{j}y$. Interestingly, the interaction of a pair of spins at the surface of the superconductor ($z=z'=0$) is twice as strong as in the interior at $z=z'>\xi$, where $\xi=\xi_{\omega_0}=[D/2(\pi^2T^2+\Delta^2)^{1/2}]^{1/2}$ is the coherence length of the superconductor, where $\Lambda_s(\mathbf{r},\mathbf{r}',\omega)$ is given by expression (3). The elastic reflection of Cooper pairs from the surface of the superconductor thus leads to a sort of interference with amplification.

For a superconducting plate of thickness L, i.e., for $0\leqslant z,\ z'\leqslant L$, where $L\gg l$, we find

$$\Lambda_s(\mathbf{r},\mathbf{r}',\omega)=\frac{2\pi N(0)\Delta^2}{\omega^2+\Delta^2}\int\frac{d^2q_\perp \exp[i\mathbf{q}_\perp(\rho-\rho')]}{(2\pi)^2Dk\sinh(kL)}\cosh(kz)\cosh[k(z'-L)], \tag{8}$$

for $z<z'$. For $z>z'$, in contrast, we need to interchange these two variables in (8). It is not difficult to see that in the case of a bulk plate ($L>\xi$) the antiferromagnetic coupling between local spins at each surface is twice as strong as in the interior. In the limit $L\to\infty$ we find expression (7) for a half-space from (8). However, the result in (8) is illustrated most clearly in the quasi-2D situation, in which the film thickness L is much smaller than the coherence length ξ. In this case the interaction between localized spins is essentially independent of the variables z and z', being determined exclusively by the projection of the radius vector $\mathbf{R}$ onto the $z=0$ plane, i.e., by the quantity $R_\perp|\boldsymbol{\rho}-\boldsymbol{\rho}'|$:

$$\Lambda_s(\mathbf{r},\mathbf{r}',\omega)=\frac{N(0)\Delta^2}{DL(\omega^2+\Delta^2)}K_0(R_\perp/\xi_\omega). \tag{9}$$

In this case, according to the asymptotic expressions for the Macdonald function,

$$K_0(R_\perp/\xi_\omega)\propto\ln(\xi_\omega/R_\perp),\quad R_\perp<\xi_\omega,$$

$$K_0(R_\perp/\xi_\omega)\propto(\xi_\omega/R_\perp)^{1/2}\exp(-R_\perp/\xi_\omega),\quad R_\perp>\xi_\omega,$$

the power-law decay of the RKKY potential with increasing distance between the localized spins is slower than in the 3D case, (3), and the antiferromagnetic correlations of the localized spins are intensified at length scales $R_\perp\leqslant\xi$. From the physical standpoint, this modification of the RKKY exchange is apparently caused by a "flattening" of the wave function of the Cooper pairs in the quasi-2D film, without a change in the phase volume which they occupy.

For a superconducting wire of radius L, i.e., for $0\leqslant\rho,\ \rho'\leqslant L$ ($L\gg l$), we have

$$\Lambda_s(\mathbf{r},\mathbf{r}',\omega)=\frac{4N(0)\Delta^2}{D(\omega^2+\Delta^2)}\sum_{m=0}^{\infty}\frac{\cos m\varphi}{1+3\delta_{m,0}}\int_{-\infty}^{\infty}\frac{dq_\parallel \exp[iq_\parallel(z-z')]}{2\pi I_m'(kL)}$$

$$\times I_m(k\rho)[K_m(k\rho')I_m'(kL)-I_m(k\rho')K_m'(kL)] \tag{10}$$

for $\rho<\rho'$. For the case $\rho>\rho'$, we need to interchange these two variables. In addition, the quantity φ in (10) is the angle between the radius vectors $\boldsymbol{\rho}$ and $\boldsymbol{\rho}'$; $q_{\|}=q_z$; $k^2=q_{\|}^2+\xi_\omega^{-2}$ and $I_m(x)$, $K_m(x)$ and $I_m'(x)$, $K_m'(x)$ are modified Bessel functions of the first and second kinds and their derivatives. In the quasi-1D case, with a wire radius $L\ll\xi$, expression (10) simplifies substantially:

$$\Lambda_s(\mathbf{r},\mathbf{r}',\omega)=\frac{N(0)\Delta^2}{\omega^2+\Delta^2}\frac{\xi_\omega}{DL^2}\exp(-|z-z'|/\xi_\omega). \qquad (11)$$

The energy of the antiferromagnetic interaction between localized spins at distances $|z=z'|\leqslant\xi$ is essentially constant.

Antiferromagnetic correlations of localized spins induced by the transition of the metal to the superconducting state are thus intensified toward the surface of a bulk dirty superconductor or as the dimensionality of the sample is lowered. It is easy to verify by direct integration that all the results above (7)–(11), satisfy sum rule (4), which ensures that the uniform spin susceptibility χ_s of the superconductor is zero at $T=0$.

The results derived here can be used to formulate a simple model for exchange interactions in F/S superlattices and contacts—a model which is suitable for samples with the geometric configurations which we have discussed here. This model incorporates, in addition to the direct exchange of nearest neighbors along the ferromagnetic layers, the possibility that localized spins at F/S interfaces also interact with each other indirectly by virtue of a long-range RKKY exchange through conduction electrons of the superconducting layers. The latter interaction might be caused by an effective s-d (f) exchange I, which would result from a virtual transition of electrons from the superconductor into the insulator, and vice versa, by virtue of an overlap of the corresponding wave functions at the F/S interface. Working in this model, one can show that, under certain conditions, there can be—in addition to the coexistence of ferromagnetism and superconductivity, a mutual accommodation of these phenomena when the superconducting substrate induces a sinusoidal modulation in the spin structure of the F film. This accommodation results in a substantial averaging of the exchange field which destroys Cooper pairs in the substrate. It thus leads to a decrease in the splitting of the BCS peak in the density of states of the quasiparticles which is observed in tunneling experiments with EuO/Al (Ref. 4) and EuS/Al (Ref. 5) F/S contacts. A magnetic field parallel to the plane of the contact destroys Cooper pairs by virtue of an orbital effect and thus causes a progressive lowering of the energy of the RKKY antiferromagnetic exchange. It drives the F/S contact into a phase of a coexistence with a uniform exchange field. The splitting which results leads to saturation. A further increase in the magnetic field leads to a first-order transition to a normal ferromagnetic state, in accordance with Refs. 4 and 5.

An antiferromagnetic superconducting layered phase in which local spins in each of the F layers are ordered ferromagnetically, while the magnetizations of neighboring layers are antiparallel, may arise by virtue of RKKY exchange in F/S superlattices. Another version of the mutual accommodation has a cryptoferromagnetic superconducting layered state in which the phases of sinusoidally modulated structures of localized spins in neighboring F layers are displaced by an amount π (this is a π-phase magnetism). In each case the exchange polarization induced in the superconducting layers ($L<\xi$) of the localized spins of one ferromagnetic layer is neutralized almost completely by the polarization of the opposite sign induced by the neighboring F layer. This self-induced cancellation of the exchange field could explain the fairly weak suppression of superconductivity in EuO/V multilayer structures.[1] The 3D behavior of F/S superlattices described above gives way to a quasi-1D behavior when the period ($q_{\perp}^{-1}$) of the modulation of the magnetic order in the F layers becomes smaller than the thickness of the superconducting layers, L. In this case, we see from (8) that the RKKY exchange becomes a surface effect even under the condition $L \ll \xi$, and the superlattice breaks up into a system of weakly coupled S/F/S sandwiches.

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