Reentrant superconductivity in conical-ferromagnet/superconductor nanostructures

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We study a bilayer consisting of an ordinary superconductor and a magnet with a spiral magnetic structure of the Ho type. We use a self consistent solution of the Bogoliubov-de Gennes equations to evaluate the pair amplitude, the transition temperature, and the thermodynamic functions, namely, the free energy and entropy. We find that for a range of thicknesses of the magnetic layer the superconductivity is reentrant with temperature T: as one lowers T the system turns superconducting, and when T is further lowered it turns normal again. This behavior is reflected in the condensation free energy and the pair potential, which vanish both above the upper transition and below the lower one. The transition is strictly reentrant: the low and high temperature phases are the same. The entropy further reveals a range of temperatures where the superconducting state is less ordered than the normal one.

More than thirty years ago, reentrant superconductivity associated with magnetic ordering was first observed in the ternary rare-earth compounds ErRhB and HoMoS [1–5]. On cooling, these materials first become superconducting at a critical temperature Tc2. Upon further cooling, inhomogeneous magnetic order sets in. This ordering coexists with superconductivity[6] over a very narrow T range. This onset is nearly immediately[7] followed by that of long-range ferromagnetic order, which entails the destruction of superconductivity, at a second critical temperature Tc1. Thus, the reason for the disappearance of the superconductivity at Tc1 is essentially the presence of the magnetism. That nonuniform magnetic ordering can appear in the presence of superconductivity is consistent with the prediction made by Anderson and Suhl[8]. Reentrant superconductivity of a different kind is also found in ferromagnet/superconductor (F/S) layered heterostructures[6]. On increasing the thickness, dF, of the ferromagnet layers in such structures, while keeping the thickness of the superconductor layers constant, the superconductivity may disappear for a certain range of thickness (dF 1 < dF < dF 2) and then return for larger dF (dF > dF 2).

The purpose of this Letter is to show that superconductivity in F/S nanostructures which is reentrant with temperature can occur under some circumstances, when the magnetic structure is non-uniform. That is, for certain types of ferromagnets, the Cooper pair amplitude in such structures can be nonvanishing in a range Tc1 < T < Tc2, with Tc1 > 0. Specifically, we have found that this reentrance occurs in F/S bilayers where the magnetic order of the F layer is of the spiral type, as in Holmium[9]. The reentrance we find is very different from that in ErRhB or HoMoS. There, the high T phase is paramagnetic and the low T phase is ferromagnetic. In our case, the magnetic order remains unchanged: it is the same above Tc1, below Tc2, and in between. Reentrance occurs also[10] in some quasi one dimensional superconductors, but there the low T phase is insulating. In our case, we have strict reentrance: the lowest T and highest T phases are the same, while in the entire range in between, superconductivity and magnetism harmoniously coexist. This is unusual. Superconducting reentrance is also found in granular films[11]: it is not due to magnetism but it involves the turning on and off of the intergrain Josephson coupling. Here, we are able to evaluate the thermodynamic functions of the system as it undergoes the transitions, and from their behavior one can glimpse the reasons for the occurrence of the reentrance. The balance between the internal energy of the system and its entropy can result in a situation where the entropy of the thermodynamically stable superconducting state is higher than that of the normal state.

Extensive theoretical[6, 12–17] work indicates that the origin of dF reentrant superconductivity in F/S nanostructures can be traced to the damped oscillatory nature of the Cooper pair wave functions in ferromagnets[18, 19]. Qualitatively, when a Cooper pair enters into an F region, it decays and the electron with magnetic moment parallel to the internal exchange field h lowers its energy by an amount proportional to h, while the other electron with opposite spin raises its energy by the same amount. Then, the kinetic energy of each electron changes and as a result[18] the Cooper pair entering into an F region acquires a spatially dependent phase in the F layer. This propagating character of the Cooper pair leads to interference between the transmitted pairing wave function through the F/S interface and the reflected wave from the opposite surface of the ferromagnet. Experimentally, the reentrant behavior of superconductivity with dF has been observed and confirmed in Nb/Cu1−xNi, bilayers and Fe/V/Fe trilayers[20–22]. However, in the work we present here, reentrance occurs with temperature, rather than just with geometry. Thus, although it is already known[23] that the nonuniform Ho structure has strong effects in the S/F proximity phenomena, no T reentrance results have been predicted or observed.

In the rest of this paper, we will first review our methods as applied to Holmium/superconductor (Ho/S) structures and then discuss the microscopic behavior of the pair amplitudes as well as the thermodynamic quantities. The approach we use here is based on exact, self-consistent, diagonalization of the Bogoliubov-de Gennes (BdG)[24] equations for clean F/S structures. This approach not only has the virtue of being very general but is also able to describe short wavelength oscillations, which is important for small structures. The self consis-
tent methods we use to diagonalize the BdG equations have been extensively described in the literature (see e.g. Ref. 25 and references therein) and details will not be given here, except where crucial.

The geometry of the Ho/S system we consider is depicted schematically in Fig. 1. The y axis is normal to the layers. The system is assumed to be infinite in the x-z plane and has a total length d in the y direction. The S layer in our assumed Ho/S system is a conventional s-wave superconductor with thicknesses $d_S$ and a Ho layer of thickness $d_F$. As in previous work, the magnetic structure is described via a local exchange field $h$, (see text). The system is assumed to be infinite in the x-z plane and y is normal to the interfaces.

FIG. 1. (Color online) Schematic of the ferromagnet (Ho) - superconductor (S) bilayer studied. The conical ferromagnet has a spiral magnetic structure described by an exchange field $h$, (see text). The system is assumed to be infinite in the $x-z$ plane and $y$ is normal to the interfaces.

To recast the magnetic structure described by an exchange field $h$ schematically in Fig. 1, and references therein) and details will not be given here, except where crucial. The system is assumed to be infinite in the $x-z$ plane and $y$ is normal to the interfaces.

As in previous work, the magnetic structure is described via a local exchange field $h$ which in this case is of the form:

\[
h = h_0 \left[ \cos \theta \hat{x} + \sin \theta \left( \frac{\pi}{\alpha} \hat{z} + \frac{\pi}{\alpha} \hat{x} \right) \right],
\]

where for Ho we have $\theta = 4\pi/9$ and $\alpha = \pi/6$. We will take $a$, the lattice constant, as our unit of length and assume throughout that the system is below the temperature (21 K) at which, $\theta$ switches from $\pi/2$ to $4\pi/9$, i.e. Ho becomes ferromagnetic.

The effective Hamiltonian, $H_{eff}$, that we use to model our Ho/S system takes the form

\[
H_{eff} = \int d^3 r \left\{ \sum_{\alpha} \psi^\dagger_{\alpha}(r) \left[ -\frac{\nabla^2}{2m^*} - E_F \right] \psi_{\alpha}(r) + \frac{1}{2} \sum_{\alpha, \beta} (i\sigma_i)_{\alpha\beta} \Delta(r) \psi^\dagger_{\alpha}(r) \psi^\dagger_{\beta}(r) + h.c. \right\} - \sum_{\alpha, \beta} \psi^\dagger_{\alpha}(r) (h \cdot \sigma) \psi_{\beta}(r),
\]

(1)

where $\Delta(r)$ is the usual singlet pair potential; $\psi_{\alpha}$ and $\psi^\dagger_{\alpha}$ are the creation and annihilation operators with spin $\alpha$ respectively; $E_F$ is the Fermi energy and $\sigma$ are the Pauli matrices. To recast the $H_{eff}$ into diagonal form, we apply a generalized Bogoliubov transformation, $\psi_{\alpha}(r) = \sum_n [u_{n\alpha}(r) \gamma_n + v^\dagger_{n\alpha}(r) \gamma^\dagger_n]$, where the quantum number $n$ enumerates the quasiparticle ($u_{n\alpha}$) and quasihole ($v^\dagger_{n\alpha}$) spinors. The $\gamma_n$ and $\gamma^\dagger_n$ are the Bogoliubov quasiparticle annihilation and creation operators respectively. By making use of the commutation relations, $[H_{eff}, \gamma_n] = -\epsilon_n \gamma_n$ and $[H_{eff}, \gamma^\dagger_n] = \epsilon_n \gamma^\dagger_n$, one obtains the BdG equations in matrix form. In the geometry chosen, the dependence of the wavefunctions on the $x$ and $z$ variables leads to an obvious phase factor that can be canceled out. This results in a set of quasi one dimensional problems of the form:

\[
\begin{pmatrix}
H_e - h_z & -h_x + ih_y \\ -h_x - ih_y & H_e + h_z
\end{pmatrix}
\begin{pmatrix}
\alpha \\ \Delta
\end{pmatrix}
= \begin{pmatrix}
0 \\ \Delta
\end{pmatrix},
\]

where $H_e \equiv -(1/2m^*)(\partial^2/\partial y^2) + \epsilon_1 - E_f$, with $\epsilon_1$ being the kinetic energy associated with the transverse direction. Thus the spatial dependence of the amplitudes is only on $y$. The exchange field $h(y)$ in Ho is nonvanishing only in the F region and precesses as given above (see also Fig. 1). The pair potential must be determined self-consistently by solving the BdG equations together with the condition,

\[
\Delta(y) = \frac{g(y)}{2} \sum_n \left[ u_{n\uparrow}(y) v^\dagger_{n\downarrow}(y) - u_{n\downarrow}(y) v^\dagger_{n\uparrow}(y) \right] \tanh(\frac{\epsilon_n}{2T}),
\]

(3)

where $T$ is the temperature, and $g(y)$ is the usual BCS coupling constant associated with a contact potential that exists only in the S region. The prime on the sum implies that only states corresponding to positive energies below the “Debye” cutoff $\omega_D$ are included. The self consistent diagonalization is achieved as in the previous work mentioned above, the only difference being that the matrices to be diagonalized are in this case unavoidably complex.

From the self consistent results one can evaluate immediately the pair amplitudes and, as explained below, the thermodynamic quantities. The transition temperatures can be most conveniently evaluated by a linearization method[26]. Near the transition temperature, the equation for $\Delta$ can be written as $\Delta_i = \sum_q J_{iq} \Delta_q$, where $\Delta_i$ are the expansion coefficients with respect to the orthonormal basis, $\phi_i(y) = \sqrt{2q} d \sin(i\pi y/d)$, and $J_{iq}$ is given as $J_{iq} \equiv \left( J^u_{iq} + J^\uparrow_{iq} \right) / 2$, where

\[
J^u_{iq} = \gamma \int d\epsilon_i \sum_n \tanh(\frac{\epsilon_n^0}{2T}) \sum_m F^u_{nm} \frac{F^\star_{nm} G^u_{nm}}{\epsilon_m^0 - \epsilon_n^0},
\]

(4)

\[
J^\uparrow_{iq} = \gamma \int d\epsilon_i \sum_n \tanh(\frac{\epsilon_n^0}{2T}) \sum_m G^\uparrow_{nm} \frac{F^\star_{nm} F^\star_{nm}}{\epsilon_m^0 - \epsilon_n^0}.
\]

(5)

Here $\gamma = \gamma_0 / 4\pi D$ with $\gamma_0$ being the dimensionless coupling constant in S; $D$ is the total dimensionless thickness of the structure, $\Delta \equiv k_F d$, and $k_F$ is the Fermi wavevector in
S. We take $k_{fs} = 1/\mu$; $\epsilon_n^{(v),0}$ are unperturbed particle (hole) energies; and $F_{unm} = \pi \sqrt{2d} \sum_{\mu \nu} (u^0_{n\mu} u^0_{n\nu} - u^0_{n\nu} u^0_{n\mu}) K_{unm}$, $G_{unm} = \pi \sqrt{2d} \sum_{\mu \nu} (v^0_{n\mu} v^0_{n\nu} - v^0_{n\nu} v^0_{n\mu}) K_{unm}$, where $K_{unm} \equiv \int^d_0 dy g(y) \phi_i(y) \phi_n(y)$. The $u^0_{n\mu}$ and $v^0_{n\mu}$ are the expansion coefficients of the unperturbed ($\Delta = 0$) particle (hole) amplitudes in terms of the basis set.

This linearization method is easily used to evaluate the transition temperature. As explained in Ref. 26, one simply has to find the largest eigenvalue, $\lambda$, of the matrix $J_u$ and see if it is greater or smaller than unity: in each case one is, respectively, in the superconducting or the normal state. The transition temperatures are those at which the largest eigenvalue changes from greater to smaller than unity: one finds $T_c$ by evaluating $\lambda$ as a function of $T$. In the usual case $\lambda$ is smaller than unity when $T$ is larger than $T_c$. In a reentrant case with superconductivity in the range $T_{c1} < T < T_{c2}$, we find $T_{c1}$ by increasing $T$ from zero until $\lambda > 1$ and $T_{c2}$ by decreasing $T$ from above $T_{c2}$ until $\lambda > 1$.

In all results given here, the thickness of the S layer is fixed at $d_S = (3/2)\xi_0$, where $\xi_0$ is the usual BCS coherence length in S. We take $\xi_0 = 100k_{fs}^{-1}$, and vary $d_F$. The magnitude of $h$ is 0.15$E_F$. Results for the transition temperature, normalized to the bulk transition temperature $T_c^0$ of S, are shown in Fig. 2, plotted as a function of $D_F \equiv d_F k_{fs}$. In the inset, we see that the overall behavior of $T_c$ consists of the expected damped oscillations with approximately the $D_F$ periodicity of the spiral magnetic structure (twelve, in our units). The main plot shows in more detail the structure near the first minimum. There we see also a lower small dome-shape plot (blue stars) with a maximum at $D_F \approx 4.5$. The system is in the normal phase inside the dome and, at constant $D_F$, it is in the superconducting phase between the two curves. In the $D_F$ range including the dome, the system, upon cooling, first becomes superconducting at a higher temperature $T_{c2}$, and with further cooling, returns to the normal phase at a lower temperature $T_{c1}$.

In Fig. 3 we display additional direct evidence confirming the existence of the reentrant behavior and showing its properties. All results in the figure are for a system in the reentrant region, with $D_F = 4.3$, and are plotted vs. $T/T_c^0$. We consider first (main plot, red triangles, left vertical scale), the Cooper pair amplitude $F(y)$ defined by $\Delta(y) \equiv g(y) F(y)$ (see Eqn. (3)). The quantity shown is $F(y = \xi_0)$, normalized to its bulk value in S, at a position one coherence length inside S. This amplitude vanishes below $T_{c1}$ and above $T_{c2}$, with the values of $T_{c1}$ and $T_{c2}$ agreeing with those previously found: we can see from Fig. 2, $T_{c1} \approx 0.07T_0^0$ and $T_{c2} \approx 0.47T_0^0$ at $D_F = 4.3$. The continuity of the pair amplitude at $T_{c1}$ and $T_{c2}$ also indicates that the transitions are of second order.

In the rest of Fig. 3 the thermodynamics of the transitions, which follows from the free energy, is shown. Using a standard formalism [26, 27], we calculated $F_S$, the free energy of the whole system in the self consistent state, and $F_N$, the normal state ($\Delta = 0$) free energy. The normalized condensation free energy $\Delta f(\approx (F_S - F_N)/(2E_0))$ is the condensation energy of bulk S material at $T = 0$ is then plotted in the main part of Fig. 3 (blue squares, right scale). Both $F_S$ and $F_N$ are monotonic and have negative curvature with $T$ as required by thermodynamics, but their difference is nonmonotonic. Although $\Delta f$ is small compared to its bulk value, we can still identify the two transition temperatures $T_{c1}$ and $T_{c2}$ from this.
plot. Their values are again in agreement, within numerical uncertainty, with those found from the pair amplitudes and from direct calculation. The system is in the superconducting state when the $T$ falls in the range $T_{c1} < T < T_{c2}$. As $T_{c1}$ is approached from above or $T_{c2}$ from below, the solution with $\Delta \neq 0$ disappears (as seen in the amplitude plot, (red) triangles), and the two free energies coincide: this is just what happens in ordinary BCS theory as the transition is approached from below. The minimum condensation free energy occurs at $T_m \approx 0.32 T_0$ which coincides with the location of the maximum pair amplitude. We also evaluated the entropy in the normal and superconducting states via textbook formulas. The normalized\cite{26} entropy difference for the same case is shown in the inset of Fig. 3. It confirms that the system indeed undergoes second order phase transitions at both $T_{c1}$ and $T_{c2}$. Unlike in a bulk superconductor, or in non-reentrant structures\cite{26}, there is now a range of $T$ ($T_{c1} < T < T_m$) where the superconducting state is less ordered than the normal one, and the entropy helps maintain the superconductivity.

What is the physics behind this $T$ reentrance? For F/S bilayers with a uniform ferromagnet, the superconductivity disappears for a certain range of $d_F$. This disappearance is due to the oscillating Cooper pair amplitude. Now, the spiral magnetization in Ho introduces an oscillating magnetic order. Both the oscillating Cooper pair amplitude. The superconducting state becomes then the higher entropy phase: the roles of the N and S phases are thus reversed, the pair potential begins to decrease, and this leads inexorably to the lower transition at $T_{c1}$, and to the reentrance into the same N phase.

We have already seen above the clear differences between this entropy competition driven situation and other singlet superconducting $T$ reentrance cases associated with field induced situations. Temperature reentrance involving long range magnetic order has been long known to occur in spin glasses\cite{29}, but the lowest $T$ and high $T$ phases (spin glass and paramagnetic respectively) are not the same. Somewhat similar but even more complicated situations occur in liquid crystals and may be a general property of frustrated systems. But a survey would take us too far afield.

To our knowledge, this effect has not been searched for. The predicted range of $T$ needed, down to about 0.1 $T_0$ should pose no difficulty. The best course should be to fabricate samples of varying $d_F$, verify the $T_c$ oscillations (see Fig. 2 inset) and then search for reentrance for $d_F$ near a minimum of the $T_c$ vs $d_F$ curve, where the phenomenon is predicted to occur. (This is possibly because such minima are associated with fragility of the superconducting state). It has proved experimentally feasible\cite{31} to study the $T$ induced $0-\pi$ state transitions in S/F/S trilayers, which are related to a different effect\cite{26} also involving nontrivial pairing correlations. Thus, that difficulties in sample making are not insurmountable.

In conclusion, we predict that F/S bilayers with an inhomogeneous conical magnetization will exhibit reentrant superconductivity with $T$, in addition to $d_F$. This superconductivity exists for $T_{c1} < T < T_{c2}$ with nonzero $T_{c1}$ under some conditions. We have shown clear evidence for this by self consistently determining the critical temperature-thickness phase diagram, and the $T$ dependence of the pair amplitude. The thermodynamics were investigated via the free energy, revealing a range of temperatures in which the normal state is lower in entropy than the superconducting state.

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