Reentrant Superconducting Phase 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 Bogolioubov–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.

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More than 30 years ago, reentrant superconductivity associated with magnetic ordering was first observed in the ternary rare-earth compounds $ErRh_4B_4$ and $HoMo_6S_8$ [1–5]. On cooling, these materials first become superconducting at a critical temperature T_{c2}. 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 T_{c1}. Thus, the reason for the disappearance of the superconductivity at T_{c1} 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, d_F, 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 ($d_{F1} < d_F < d_{F2}$) and then return for larger d_F ($d_F > d_{F2}$).

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 nonuniform. That is, for certain types of ferromagnets, the Cooper pair amplitude in such structures can be nonvanishing in a range $T_{c1} < T < T_{c2}$, with $T_{c1} > 0$. 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 $ErRh_4B_4$ or $HoMo_6S_8$. 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 T_{c1}, below T_{c2}, 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 d_F 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 $\mathbf{h}$ 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 d_F has been observed and confirmed in $Nb/Cu_{1-x}Ni_x$ 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 Letter, 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-consistent 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 $\mathbf{h}$ which in this case is of the form: $\mathbf{h} = h_0\{\cos\theta\hat{\mathbf{y}} + \sin\theta[\sin(\frac{\varphi y}{a})\hat{\mathbf{x}} + \cos(\frac{\varphi y}{a})\hat{\mathbf{z}}]\}$, where for Ho we have [9,23] $\theta = 4\pi/9$ and $\varphi = \pi/6$. 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, θ switches from $\pi/2$ to $4\pi/9$, i.e., Ho becomes ferromagnetic.

The effective Hamiltonian, $\mathcal{H}_{\text{eff}}$, that we use to model our Ho/S system takes the form

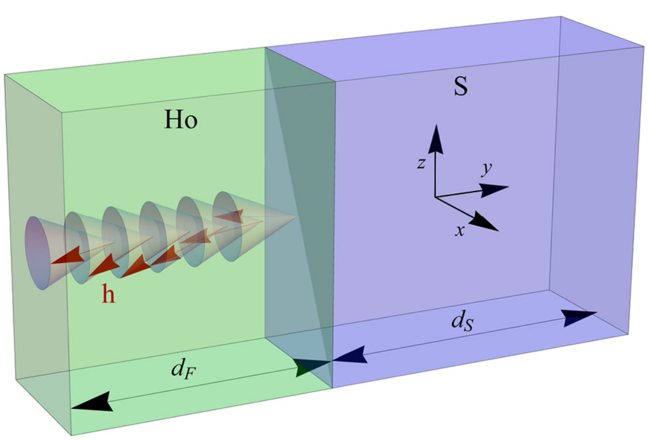

FIG. 1 (color online). Schematic of the ferromagnet (Ho)-superconductor (S) bilayer studied. The conical ferromagnet has a spiral magnetic structure described by an exchange field $\mathbf{h}$, (see text). The system is infinite in the x-z plane and y is normal to the interfaces.

$$\mathcal{H}_{\text{eff}} = \int d^3r\bigg\{\sum_{\alpha}\psi_{\alpha}^{\dagger}(\mathbf{r})\left(-\frac{\nabla^2}{2m^*} - E_F\right)\psi_{\alpha}(\mathbf{r}) + \frac{1}{2}\times\bigg[\sum_{\alpha,\beta}(i\sigma_y)_{\alpha\beta}\Delta(\mathbf{r})\psi_{\alpha}^{\dagger}(\mathbf{r})\psi_{\beta}^{\dagger}(\mathbf{r}) + \text{H.c.}\bigg] - \sum_{\alpha,\beta}\psi_{\alpha}^{\dagger}(\mathbf{r})(\mathbf{h}\cdot\boldsymbol{\sigma})\psi_{\beta}(\mathbf{r})\bigg\}, \quad (1)$$

where $\Delta(\mathbf{r})$ is the usual singlet pair potential; $\psi_{\alpha}^{\dagger}$ and ψ_{α} are the creation and annihilation operators with spin α respectively; E_F is the Fermi energy and $\boldsymbol{\sigma}$ are the Pauli matrices. To recast the $\mathcal{H}_{\text{eff}}$ into diagonal form, we apply a generalized Bogoliubov transformation, $\psi_{\alpha}(\mathbf{r}) = \sum_n[u_{n\alpha}(\mathbf{r})\gamma_n + v_{n\alpha}^*(\mathbf{r})\gamma_n^{\dagger}]$, where the quantum number n enumerates the quasiparticle ($u_{n\alpha}$) and quasihole ($v_{n\alpha}$) spinors. The γ_n and $\gamma_n^{\dagger}$ are the Bogoliubov quasiparticle annihilation and creation operators, respectively. By making use of the commutation relations, $[\mathcal{H}_{\text{eff}}, \gamma_n] = -\epsilon_n\gamma_n$ and $[\mathcal{H}_{\text{eff}}, \gamma_n^{\dagger}] = \epsilon_n\gamma_n$, one obtains the BdG equations in matrix form. In the geometry chosen, the dependence of the wave functions 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 (in y) problems of the form

$$\begin{pmatrix} H_e - h_z & -h_x + ih_y & 0 & \Delta \\ -h_x - ih_y & H_e + h_z & -\Delta & 0 \\ 0 & -\Delta^* & -H_e + h_z & h_x + ih_y \\ \Delta^* & 0 & h_x - ih_y & -H_e - h_z \end{pmatrix} \times \begin{pmatrix} u_{n\uparrow} \\ u_{n\downarrow} \\ v_{n\uparrow} \\ v_{n\downarrow} \end{pmatrix} = \epsilon_n \begin{pmatrix} u_{n\uparrow} \\ u_{n\downarrow} \\ v_{n\uparrow} \\ v_{n\downarrow} \end{pmatrix}, \quad (2)$$

where $H_e \equiv -(1/2m^*)(\partial^2/\partial y^2) + \epsilon_{\perp} - E_F$, with $\epsilon_{\perp}$ being the kinetic energy associated with the transverse direction. Thus the spatial dependence of the amplitudes is only on y. The exchange field $\mathbf{h}(y)$ 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,

$$\Delta(y) = \frac{g(y)}{2}\sum_n{}'[u_{n\uparrow}(y)v_{n\downarrow}^*(y) - u_{n\downarrow}(y)v_{n\uparrow}^*(y)]\tanh\left(\frac{\epsilon_n}{2T}\right), \quad (3)$$

where T is the temperature, and $g(y)$ is the usual BCS coupling constant g 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 ω_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 [26,27]. Near the transition temperature, the equation for Δ can be written as $\Delta_i = \sum_q J_{iq}\Delta_q$, where Δ_i are the expansion coefficients with respect to the orthonormal basis, $\phi_i(y) = \sqrt{2/d}\sin(i\pi y/d)$, and J_{iq} is given as $J_{iq} \equiv (J^u_{iq} + J^v_{iq})/2$, where

$$J^u_{iq} = \gamma \int d\epsilon_\perp \sum_n \left[\tanh\left(\frac{\epsilon_n^{u,0}}{2T}\right)\sum_m \frac{F^*_{qnm}F_{inm}}{\epsilon_n^{u,0} - \epsilon_m^{v,0}}\right], \quad (4)$$

$$J^v_{iq} = \gamma \int d\epsilon_\perp \sum_n \left[\tanh\left(\frac{\epsilon_n^{v,0}}{2T}\right)\sum_m \frac{G_{qnm}G^*_{inm}}{\epsilon_n^{v,0} - \epsilon_m^{u,0}}\right]. \quad (5)$$

Here $\gamma = (\gamma_0/4\pi D)$ with γ_0 being the dimensionless coupling constant in S; D is the total dimensionless thickness of the structure, $D \equiv k_{FS}d$, and k_{FS} is the Fermi wave vector in S. We take $k_{FS} = 1/a$; $\epsilon_n^{u(v),0}$ are unperturbed particle(hole) energies; and $F_{inm} = \pi\sqrt{2d}\sum_{pq}(u^0_{np\uparrow}u^0_{mq\downarrow} - u^0_{np\downarrow}u^0_{mq\uparrow})K_{inm}$, $G_{inm} = \pi\sqrt{2d}\sum_{pq}(v^0_{np\uparrow}v^0_{mq\downarrow} - v^0_{np\downarrow}v^0_{mq\uparrow})K_{inm}$, where $gK_{inm} \equiv \int_0^d dy g(y)\phi_i(y)\phi_n(y)\phi_m(y)$. The u^0_{np} and v^0_{mq} 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. [26,27], one simply has to find the largest eigenvalue, λ, of the matrix J_{iq} 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 λ as a function of T. In the usual case λ 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 $\lambda > 1$. We also searched for transitions in the intermediate region but none was found.

In all results given here, the thickness of the S layer is fixed at $d_S = (3/2)\xi_0$, where ξ_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 $0.15E_F$. 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, $T_{c1} \approx 0.07T_c^0$ and $T_{c2} \approx 0.47T_c^0$ 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 [26,28], we calculated F_S, the free energy of the whole system in the self-consistent state, and F_N, the normal state ($\Delta \equiv 0$) free energy. The normalized condensation free energy $\Delta f \equiv (F_S - F_N)/(2E_0)$ (E_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 Δf is small compared to its bulk value, we can still identify the two

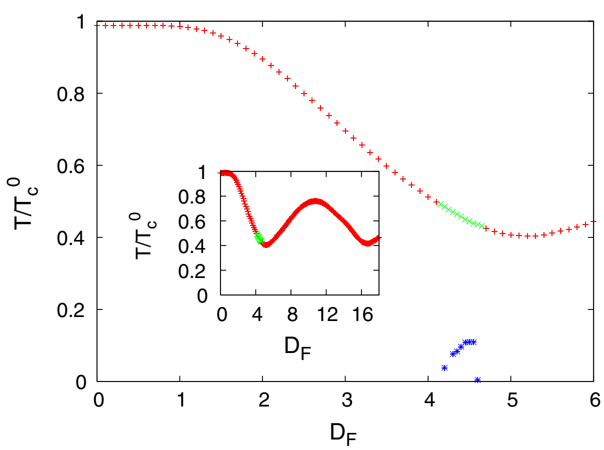

FIG. 2 (color online). Calculated transition temperature T_c, normalized by T_c^0 (see text), vs the dimensionless ferromagnet width, D_F ($\equiv d_F k_{FS}$). Main plot: The upper points [red +, green $\times$'s] are the usual critical temperature (T_{c2}), leading to the superconducting state as T is lowered. In the region $4 \lesssim D_F \lesssim 5$ [highlighted by the green $\times$'s] a second transition back to the normal state appears at the (blue) star points forming the lower "dome." The inset shows a broader range of magnet widths, revealing the overall periodicity of T_{c2}.

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 $\Delta \not\equiv 0$ 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 $T_m \approx 0.32 T_c^0$ 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 [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 nonreentrant structures [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 magnetic structure and the superconductivity are nonuniform, consistent with the prediction in Ref. [8] that superconductivity may coexist

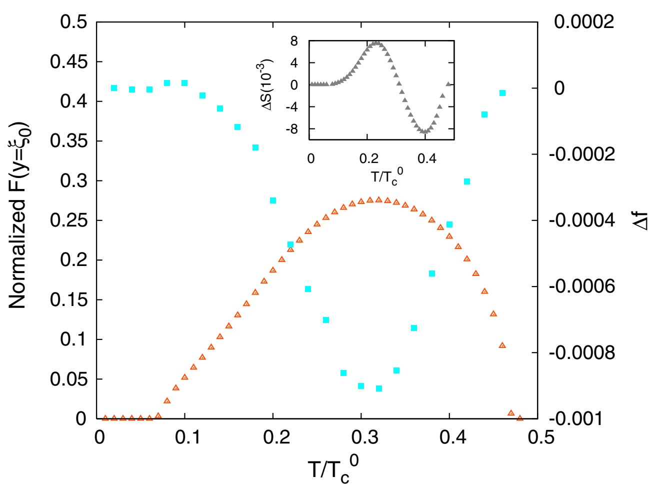

FIG. 3 (color online). Pair amplitude and thermodynamic functions. All quantities are plotted vs T/T_c^0. In the main plot, the (red) triangles and left vertical scale display the normalized (see text) singlet Cooper pair amplitude $F(Y)$, one correlation length inside S. This quantity vanishes at the upper transition temperature ($T_{c2} \approx 0.47 T_c^0$) and again at the lower transition $T_{c1} \approx 0.07 T_c^0$. The (blue) squares and right scale are the normalized (see text) condensation free energy, Δf. The vanishing of Δf at the upper and lower transitions is clearly seen. The inset shows the normalized entropy difference $\Delta S \equiv -[d\Delta f/d(T/T_c^0)]$.

with nonuniform magnetic order. Thermodynamically, we have here a subtle example of entropy-energy competition. In the range $T_m < T < T_{c2}$, Δf and ΔS behave qualitatively as they do for an ordinary [29] bulk superconductor in the region $0 < T < T_c$, (although they are much smaller). In either case ΔS vanishes at both ends of the range and has a minimum at a finite T in between. But in our case T_m is nonzero. For $T < T_m$, ΔS turns positive because of the oscillatory nature of the pair potential. 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 [30], 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 [31] 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_c^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 [32] to study the T induced 0-π state transitions in $S/F/S$ trilayers, which are related to a different effect [26] also involving nontrivial pairing correlations. Thus, difficulties in sample making are not insurmountable [33].

In conclusion, we predict that F/S bilayers with an inhomogeneous conical magnetization will exhibit reentrant superconductivity with T, in addition to d_F. Thus, 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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