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Temperature Controlled Fulde-Ferrell-Larkin-Ovchinnikov Instability in Superconductor-Ferromagnet Hybrids

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We show that a wide class of layered superconductor-ferromagnet (S/F) hybrids demonstrates the emergence of the Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) phase well below the superconducting transition temperature. By decreasing the temperature, one can switch the system from uniform to the FFLO state which is accompanied by the damping of the diamagnetic Meissner response down to zero and also by the sign change in the curvature of the current-velocity dependence. Our estimates show that an additional layer of the normal metal (N) covering the ferromagnet substantially softens the conditions required for the predicted FFLO instability, and for existing $S/F/N$ systems, the temperature of the transition into the FFLO phase can reach several kelvins.

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In 1964, Fulde and Ferrell [1] and Larkin and Ovchinnikov [2] theoretically showed that strong magnetic field acting on the electron spins in low-dimensional superconductors induces a peculiar nonuniform superconducting phase with the spatial modulation of the order parameter [the Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) phase]. The key ingredient for the FFLO state formation is the splitting of the Fermi surfaces for the spin-up and spin-down electrons due to the Zeeman interaction. In this case, the Cooper pair cannot be constructed from the electrons with the opposite momenta anymore, and the total momentum of the pair becomes nonzero. The resulting nonuniform profile of the superconducting gap strongly depends on the sample dimensionality and the anisotropy of the superconductor [3].

Despite the transparent physics behind the FFLO instability, its experimental observation appeared to be extremely challenging. First, one needs to deal with the low-dimensional samples or with the layered heavy-fermion compounds in order to damp the orbital effect which usually dominates over the Zeeman interaction and suppresses the superconductivity at the magnetic fields well below the FFLO instability threshold [4,5]. Second, the FFLO phase is known to be very sensitive to the disorder which is typically rather strong in thin films or layered superconductors [6,7]. As a result, the convincing evidence of the FFLO states formation in an external magnetic field has been provided only for some quasi-two-dimensional organic superconductors such as λ -(BETS)₂GaCl₄ [8], λ -(BETS)₂FeCl₄ [9], κ -(BEDT-TTF)₂Cu(NCS)₂ [10–14], and β'' -(ET)₂SF₅CH₂CF₂SO₃ [15,16]. The layered structure

of these compounds damps the orbital effect for the field orientation parallel to the layers, while the highly anisotropic Fermi surface is expected to provide additional stability for the FFLO phase [3].

Another promising possibility to realize the FFLO pairing appears in the multilayered superconductor-ferromagnet (S/F) structures where the interfaces between the layers are transparent for the electrons [17]. In such sandwiches, the splitting of the Fermi surfaces occurs due to the exchange field inside the F layer which does not produce the orbital currents. As a result, the Cooper pair wave function becomes modulated across the layers, and the FFLO phase appears. This leads to a number of unusual phenomena such as the oscillatory dependence of the critical temperature of the S/F bilayer on the F -layer thickness [18,19] or the π -junctions formation [20,21]. The rich interference physics coming from the interplay between the FFLO oscillations period and the layers thicknesses as well as the unusual spin patterns arising in such systems make them attractive for superconducting spintronics [22,23].

For more than two decades, it was believed that in S/F sandwiches the Cooper pair wave function is always modulated only in the direction perpendicular to the layers due to the in-plane system homogeneity. But recently, it was demonstrated that the spin-triplet superconducting correlations emerging in such a system favors the formation of the in-plane FFLO phase with the gap potential modulated along the layers [24]. As a result, the critical

temperature for the FFLO phase in the certain range of parameters becomes higher than the one for the uniform state, and the transition from the normal to the FFLO state occurs. Remarkably, the emergence of the in-plane FFLO phase should reveal itself through the vanishing Meissner response of the sample on the external parallel magnetic field. Experimentally, such a feature can be detected, e.g., in the surface inductance measurements on the basis of the two-coil technique [25,26] which has been recently applied to the study of the screening properties of the S/F bilayers [27,28]. The similar instabilities of the uniform state have been predicted also for a ferromagnetic cylinder covered by the superconducting shell [29–31] as well as for the planar superconductor–normal-metal (N) structures under non-equilibrium quasiparticle distribution [32,33]. However, it appeared that the experimental observation of the in-plane FFLO states in all these systems is hampered by the rigid restrictions of the required material characteristics.

In this Letter, we predict the existence of the in-plane FFLO phase well below the critical temperature in a wide class of thin-film S/F and $S/F/N$ sandwiches. The phase diagrams of such hybrids demonstrate several very unusual features, which, to our knowledge, contrast with the diagrams of the all-known systems supporting the FFLO states. Specifically, the FFLO domain can be totally isolated from the phase transition line between the normal and the uniform superconducting states. By decreasing the temperature, one can provoke the phase transition between the uniform and FFLO states which is accompanied by the vanishing Meissner response on the in-plane magnetic field and the sign change in the curvature of the current-velocity dependence. Our estimates show that the conditions required for the predicted FFLO instability are rather soft and can be fulfilled for a large number of the existing $S/F/N$ systems consisting of, e.g., the superconducting NbN, MoN, MgB₂, NbTi, TaN, or WSi layer, the ferromagnetic CuNi, PdFe, FeNi, or Gd layer, and the layer of Au, Ag, Al, or Cu as a normal metal. For such systems, the critical temperature of the transition into the modulated state can reach several kelvins, which makes them very promising for the experimental observation of the FFLO phase.

We start from the general arguments illustrating the origin of the low-temperature FFLO phase formation. Consider a thin-film S/F sandwich of thickness much smaller than the London penetration depth λ . The condition of the gauge invariance of the free-energy functional allows us to establish an equivalence of the sign change of the total magnetic response of the thin-film structure (i.e., the quantity λ^{-2} averaged across the structure) and the free-energy instability towards the formation of the state with a finite in-plane phase gradient. This general recipe is valid for arbitrary temperatures and is nicely confirmed by further direct numerical calculations of the full free energy. For temperatures T near the superconducting transition

temperature T_c , the screening parameter λ^{-2} , which determines the relation $\mathbf{j}_s = -(\lambda^{-2}/4\pi)\mathbf{A}$ between the superconducting current \mathbf{j}_s and the vector potential \mathbf{A} , can be expanded in the small parameter $\tau = (T_c - T)/T_c$,

$$\lambda^{-2} = \chi\tau + \kappa\tau^2, \quad (1)$$

where the coefficients χ and κ are temperature independent. In the absence of the F layer, the standard BCS model gives $\chi > 0$ and $\kappa < 0$. At the same time, the exchange field in the ferromagnet gives rise to the spin-triplet superconducting correlations which renormalize these coefficients. For rather large normal-state conductivity of the F layer and small thickness of the S film, the coefficient χ becomes strongly damped and can even vanish. The latter fact indicates the formation of the in-plane FFLO state at $T = T_c$ [24]. It is important that the coefficient κ should remain negative, reflecting the decrease in the number of quasiparticles when decreasing the temperature. As a result, even for $\chi > 0$, there exists a possibility for the vanishing of λ^{-2} . If the total thickness of the S/F sandwich is much smaller than the London penetration depth, the part of the free energy containing the square of the superconducting phase gradient is proportional to the λ^{-2} value averaged across the structure. Thus, for $|\kappa| \gg \chi$ in Eq. (1), the FFLO phase can emerge at the temperature T_F well below T_c : $T_F/T_c = 1 - \chi/|\kappa|$. It is exactly this FFLO instability which makes it impossible to observe the global paramagnetism predicted in Refs. [34–36]. The latter paramagnetic state just does not correspond to the free-energy minimum [37].

To provide support for the above qualitative arguments, we perform an explicit microscopic calculation of the magnetic screening parameter λ^{-2} for the dirty S/F bilayer. Our analysis is based on the nonlinear Usadel equation

$$-D(g\partial_x^2 f - f\partial_x^2 g) + 2(\omega_n + ih)f - 2\Delta = 0 \quad (2)$$

with the normalization condition

$$g^2 + ff^\dagger = 1. \quad (3)$$

Here, $g(x, \omega_n, h)$ and $f(x, \omega_n, h)$ are the normal and anomalous Green functions, respectively, $f^\dagger(x, \omega_n, h) = f^*(x, \omega_n, -h)$, $\Delta(x)$ is the superconducting gap potential in the S layer, h is the exchange field in the F layer, $\omega_n = \pi T(2n + 1)$ are the Matsubara frequencies, and D is the diffusion coefficient of the corresponding layer. The anomalous function and the gap potential also satisfy the self-consistency equation

$$\Delta \ln \frac{T}{T_{c0}} + \sum_{n=0}^{\infty} \left(\frac{\Delta}{n + 1/2} - 2\pi T \text{Re}f \right) = 0, \quad (4)$$

where T_{c0} is the critical temperature of the isolated superconducting layer. Assuming the small thickness $d_0 = (d_s + d_f) \ll \lambda$, where d_s and d_f are the thicknesses

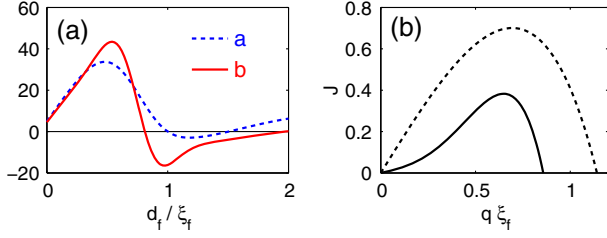


FIG. 1. (a) The coefficients $a = \alpha(2\pi T_c)^2 \lambda_c^2$ and $b = \beta(2\pi T_c)^4 \lambda_c^2$ in the expansion (6) as functions of the F -layer thickness. (b) The dependences of the supercurrent $J = j_s \lambda_c^2 T_c / (\xi_f T_{c0})$ on the superconducting velocity q for $T = 0.8 T_c(d_f)$, where $\xi_f = \sqrt{D_f / h}$ is the superconducting coherence length inside the ferromagnet, and $T_c(d_f)$ is the critical temperature corresponding to the thickness d_f of the F layer. The solid (dashed) line corresponds to $d_f = 0.91 \xi_f$ ($d_f = 0.3 \xi_f$). Here, we define $\lambda_c^2 = ec\Phi_0 d_0 / (16\pi^3 \sigma_s d_s T_c)$ and take $\xi_f = 10 \xi_s$, $\sigma_s d_s / (\sigma_f \xi_f) = 0.06$.

of the S and F layers, respectively, we may write the London screening parameter averaged over the structure thickness in the form (see, e.g., Ref. [50])

$$\lambda^{-2} = \frac{16\pi^3 T}{ec\Phi_0 d_0} \sum_{n=0}^{\infty} \int_{-d_s}^{d_f} \sigma \text{Re}(f^2) dx, \quad (5)$$

where σ is the normal-state conductivity which takes the value σ_s (σ_f) for the S (F) layer, and $\Phi_0 = \pi \hbar c / e$ is the magnetic flux quantum.

Technically, it is more convenient to rewrite the expansion (1) in terms of the small temperature-dependent gap potential $\Delta(T)$ [37]

$$\lambda^{-2} = \alpha \Delta^2(T) + \beta \Delta^4(T), \quad (6)$$

where the coefficients α and β do not depend on the temperature, and $\Delta(T)$ vanishes at $T = T_c$ and monotonically increases with decreasing T .

First, we derive the coefficients α and β . To do this, we assume that the thickness of the superconductor is small: $d_s \ll \xi_s$, where $\xi_s = \sqrt{D_s / (2\pi T_{c0})}$. This allows us to neglect the spatial variations of the gap potential across the S layer. Also, we assume that $h \gg T_{c0}$. For this limit, the coordinate dependence of the anomalous Green function has been previously calculated in Ref. [38] up to the terms $\sim O(\Delta^4)$. Substituting this expansion for f into Eq. (5) and performing the straightforward calculations, we find the analytical expressions for α and β which are presented in Ref. [37]. The typical dependences of these coefficients on the F -layer thickness are shown in Fig. 1(a). If the ratio σ_f / σ_s is large enough, the coefficient α can become negative for a certain range of d_f values which signals the formation of the FFLO state at the critical temperature. However, at the points where $\alpha = 0$, the

coefficient β is always negative. As a result, even for small positive α values, the second term in Eq. (6) fully compensates the term $\propto \Delta^2$ at a certain temperature below T_c making λ^{-2} vanish and the FFLO phase appear. This finding provides a new perspective on the experimental observation of the transitions between the uniform and FFLO phases since they can be controlled by the variation of the temperature.

Another intriguing feature associated with the low-temperature FFLO instability is the sign reversal in the nonlinear contribution to the relation between the supercurrent \mathbf{j}_s and the superconducting velocity which is proportional to the value $\mathbf{q} = \nabla\varphi - (2\pi/\Phi_0)\mathbf{A}$ (φ is the phase of the superconducting order parameter Δ). Qualitatively, this phenomenon can be understood within the Ginzburg-Landau model. Near the transition to the FFLO phase, the superconducting contribution to the density of the free energy has the form [51]

$$F = [-\alpha_0 \tau + \beta_0 q^2 + \delta_0 q^4] \Delta^2 + (\gamma_0 + \eta_0 q^2) \Delta^4, \quad (7)$$

and $j_s \propto \partial F / \partial q$. At a fixed small q , the minimization of the free energy with respect to Δ gives $\Delta^2 = [\alpha_0 \tau - \beta_0 q^2] / (2\gamma_0) + O(q^4)$. Substituting this expression into the supercurrent, we get $j_s \propto [\beta_0 \alpha_0 \tau q - \beta_0^2 q^3 + 2\delta_0 \alpha_0 \tau q^3] / \gamma_0$. Far from the FFLO phase domain, the last term in the expression for j_s is negligibly small compared to the second one, and $\partial^2 j_s / \partial q^2 < 0$. However, near the FFLO instability, the coefficient β_0 becomes damped, and the last term in j_s with $\delta_0 > 0$ produces the sign reversal in the curvature of the dependence $j_s(q)$ at small q .

To calculate the dependence $j_s(q)$ microscopically, one has to replace $\omega_n \rightarrow \omega_n + Dq^2/2$ in the Usadel equation (2). Generalizing the expressions for the coefficients α and β in Eq. (6) for $q \neq 0$ and taking into account that $j_s = (\Phi_0 / 2\pi) q \lambda^{-2}(q)$, we obtain the dependences $j_s(q)$ which are shown in Fig. 1(b). One sees that for d_f values far from the region of the FFLO instability (dashed curve), the dependence $j_s(q)$ has a standard form with the negative second derivative for all q values. However, in the vicinity of the FFLO phase (solid curve), the curvature of the function $j_s(q)$ changes its sign at small q which can be considered as a precursor of the nearby FFLO transition. Experimentally, such a change in the curvature sign should reveal itself in the third-harmonics electromagnetic response measurements [52–54].

All the described analytical results are perfectly supported by the numerical solution of the nonlinear Usadel equation [37] and the direct calculation and comparison of the free energies for the states with different modulation vectors q . The advantage of the numerical approach is its applicability to arbitrary low temperatures, in contrast to the above perturbation theory over small Δ which is limited by the condition $(T_c - T) \ll T_c$. Below, we present the numerical results obtained for the $S/F/N$ trilayers.

Previously, in Ref. [24] it was demonstrated that an additional layer of the normal metal covering the ferromagnet may produce more favorable conditions for the FFLO state formation. The key idea is to choose the thickness of the F layer to maximize the amplitude of the spin-triplet correlations at the F/N interface. Then, if the normal conductivity of the N layer is large enough, the averaged magnetic screening parameter of the sandwich becomes substantially damped favoring the FFLO instability. Here, we exploit this idea and show that for a wide class of S , F , and N compounds, which are typically used in fabrication of the π junctions or the spin valves, the emergence of the FFLO phase in the $S/F/N$ geometry occurs at the experimentally achievable parameters.

Note also that the transition to the FFLO state can be accompanied by the appearance of the in-plane local current density which should average to zero after the integration across the sandwich. The corresponding spontaneous magnetic fields can become of the order of the first critical field H_{c1} which is, of course, a quite measurable value for a variety of experimental techniques. Certainly, these spontaneous currents appear only for a particular profile of the gap function $\Delta = \Delta_0 e^{iqy}$, and further studies are necessary to clarify if this state is more energetically favorable than the sinusoidal-like gap profiles.

Note that in our calculations, we neglect the contribution of the magnetic field energy, which is for sure a valid approximation when the London penetration depth well exceeds the structure thickness.

Figure 2 shows the series of the phase diagrams of the $S/F/N$ trilayers for different thicknesses d_s of the S layer. The parts of the red curves below T_c indicate the lines of the

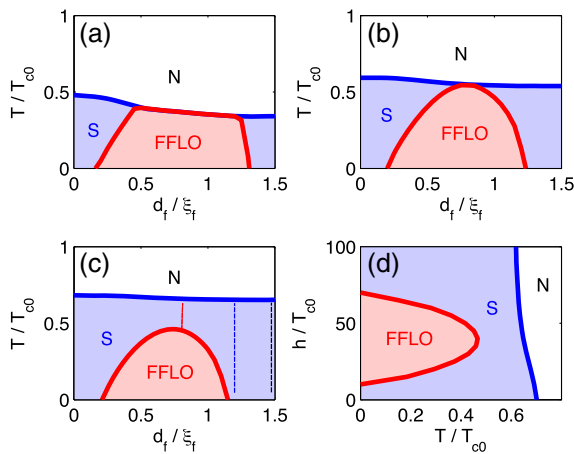


FIG. 2. (a)–(c) Phase diagrams of the $S/F/N$ sandwiches with $h/T_{c0} = 25$ and different thicknesses d_s of the S layer. The ratio d_s/ξ_0 with $\xi_0 = \sqrt{2\pi}\xi_s$ takes the values (a) 1.2, (b) 1.4, (c) 1.6. (d) Phase diagram of the $S/F/N$ system with $d_s/\xi_0 = 1.6$. In all panels, $\sigma_f/\sigma_s = 1$, $\sigma_n/\sigma_s = 150$, $d_n/\xi_0 = 1$. In panel (c), the dashed lines indicate the d_f values corresponding to the curves in Fig. 3.

type-I phase transition between the uniform and the FFLO phases. The increase of d_s results in the shrinkage of the FFLO domain, and above the certain threshold this domain can even become fully isolated from the normal state by the region corresponding to the uniform phase. The absence of the boundary between the FFLO and the normal-state regions on the h - T phase diagram [see Fig. 2(d)] contrasts (at least to our knowledge) with the phase diagrams of all previously known systems where the direct transition between the normal or FFLO phases can occur.

Finally, Fig. 3(a) demonstrates the typical dependences of the magnetic screening parameter λ^{-2} on the temperature for different thicknesses d_f of the ferromagnetic layer. In all cases, d_f is chosen in a way that at $T = T_c$ the uniform superconductivity emerges [see Fig. 2(c)]. There are three qualitatively different types of λ^{-2} behavior as the temperature decreases. The first one (black dash-dotted curve) is a monotonic increase of λ^{-2} which realizes for the systems parameters far away from the FFLO domain. In contrast, the second one (red solid curve) demonstrates the temperature-induced FFLO phase formation: when decreasing T from T_c the parameter λ^{-2} starts to grow, reaches its maximum, and then drops down to zero at the point of the FFLO instability. The third one (blue dashed curve) is realized in the intermediate parameter region. Even if the FFLO state does not emerge at any temperature, the dependence $\lambda^{-2}(T)$ can have a maximum which is very unusual for the conventional superconducting systems and serves as a precursor of the nearby FFLO domain. Also, our numerical calculations confirm the sign change in the second derivative of the current-velocity dependence $j_s(q)$ near the FFLO domain [see Fig. 3(b) and compare with Fig. 1(b)].

To sum up, we have demonstrated the in-plane FFLO instability well below the critical temperature for S/F and $S/F/N$ hybrids. Experimentally, such an instability can be

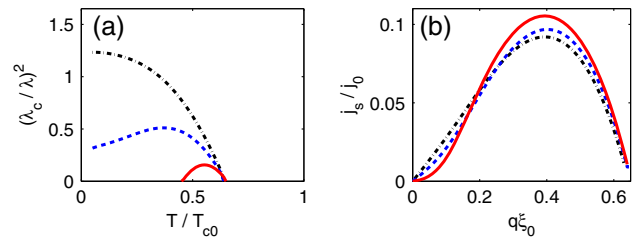


FIG. 3. (a) The dependences of the screening parameter λ^{-2} on the temperature for the $S/F/N$ sandwiches with $d_s/\xi_0 = 1.6$, the N -layer thickness $d_n/\xi_0 = 1$, and different thicknesses of the F layer. (b) The current flowing along the layers as a function of the superconducting velocity for different d_f values and $T = 0.45T_{c0}$. In both panels, the black dash-dotted, blue dashed, and red solid curves correspond to $d_f = 1.5\xi_f$, $d_f = 1.2\xi_f$, and $d_f = 0.8\xi_f$, respectively. The values ξ_0 and j_0 are defined as $\xi_0 = \sqrt{2\pi}\xi_s$ and $j_0 = \sigma_s T_c / (e\xi_0)$. Other parameters are the same as in Fig. 2.

detected by the vanishing Meissner response of the system or by the sign reversal of the third harmonics in the electromagnetic response measurements. At the same time, even outside the FFLO domain on the phase diagram, the vicinity of the FFLO instability threshold leads to the unusual nonmonotonic dependence of the magnetic screening parameter λ^{-2} on the temperature. This feature serves as a precursor of the FFLO phase formation. Remarkably, the emergence of the FFLO states in $S/F/N$ sandwiches should occur at the parameter region which can be easily achieved with the widespread materials. The combination of a superconducting NbN, TaN, or WSi layer of thickness ~ 10 nm and normal metal such as Ag, Au, or Al of thickness ~ 20 – 30 nm gives the normal conductivity ratio $\sigma_n/\sigma_s \sim 150$, which is perfect for the observation of the FFLO states (see, e.g., Fig. 2). At the same time, the relatively high critical temperature of NbN $T_{c0} \sim 10$ – 15 K makes us hope that the transition to the FFLO phase may correspond to the temperatures of the order of several kelvins. As usual, the most suitable ferromagnets are CuNi or PdFe which have relatively large coherence lengths providing a possibility to fabricate the layers with $d_f \sim \xi_f$. Thus, the $S/F/N$ sandwiches seem to provide a perfect platform for the observation of the FFLO superconducting states.

Note finally that the above findings presumably can be used to improve the design of kinetic inductance detectors of electromagnetic radiation. Indeed, changing the temperature near the critical temperature of the FFLO transition (where the Meissner screening effect vanishes), one can get rather strong and rapid changes in the kinetic inductance determined by the effective penetration depth and subsequent increase of the detector sensitivity.

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