In-plane Fulde-Ferrel-Larkin-Ovchinnikov instability in a superconductor–normal metal bilayer system under nonequilibrium quasiparticle distribution

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It is predicted that a new class of systems, superconductor/normal metal (S/N) heterostructures, can reveal the in-plane Fulde-Ferrel-Larkin-Ovchinnikov (FFLO) instability under nonequilibrium conditions at temperatures close to the critical temperature. It does not require any Zeeman interaction in the system. For S/N heterostructures under nonequilibrium distribution there is a natural easily adjustable parameter, the voltage, which can control the FFLO state. This FFLO state can be of different types: plane wave, stationary wave, and even two-dimensional structures are possible. Some types of FFLO state are accompanied by a magnetic flux, which can be observed experimentally. All the types of FFLO state can be revealed through the temperature dependence of the linear response of the system on the applied magnetic field near T_c, which strongly differs from that for the homogeneous state.

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There are two mechanisms of superconductivity destruction by a magnetic field: the orbital effect and the Zeeman interaction of electron spins with a magnetic field. Usually the orbital effect is more restrictive. However, there are several classes of systems where the orbital effect is strongly weakened (systems with large effective mass of electrons,[1,2] thin films, and layered superconductors under in-plane magnetic fields[3]) or even completely absent [superconductor/ferromagnet (S/F) heterostructures[4,5]]. Then the Zeeman interaction of electron spins with a magnetic or an exchange field is responsible for the destruction of superconductivity.

The behavior of a superconductor with a homogeneous exchange field h was studied long ago.[6–9] It was found that the homogeneous superconducting state becomes energetically unfavorable above the paramagnetic (Pauli) limit $h = \Delta_0/\sqrt{2}$, where Δ_0 is the zero-temperature superconducting gap. As predicted by Larkin and Ovchinnikov[6] and by Fulde and Ferrell,[7] in a narrow region of exchange fields exceeding this value superconductivity can appear as an inhomogeneous state with a spatially modulated Cooper-pair wave function (the FFLO state).

Now there is a growing body of experimental evidence for a FFLO phase, generated by an applied magnetic field, reported from various measurements.[10–24] However, there are not yet reported unambiguous experimental results which can be interpreted only as a fingerprint of the FFLO state.

On the other hand, it has been predicted recently[25] that the FFLO state can be realized in S/F heterostructures, where S is a singlet s-wave superconductor. Here we mean the so-called in-plane FFLO state, where the superconducting order parameter profile is modulated along the layers. It should be distinguished from the oscillations of the condensate wave function normal to the S/F interface in the ferromagnetic layer, which have been well investigated both theoretically and experimentally.[4,5,26]

In this paper we show that the in-plane FFLO state can be the most energetically favorable state in S/N heterostructures under a nonequilibrium quasiparticle distribution and propose a way to observe it. The exchange field is absent in S/N heterostructures. Correspondingly, there is no Zeeman interaction without an applied magnetic field. The transition to the FFLO state occurs due to creation of a double-step electron distribution in the bilayer. This nonequilibrium state can be reached by changing the chemical potentials of additional electrodes in opposite directions by applying a control voltage.[27,28] To the best of our knowledge, there are very few proposals for a FFLO state in nonmagnetic systems [for example, a current-driven FFLO state in two-dimensional (2D) superconductors with Fermi surface nesting,[29] in unconventional superconducting films,[30] and in nonequilibrium N/S/N heterostructures at low enough temperatures[31]). The effect considered here greatly differs from the one discussed in Ref. 31. It was demonstrated in Ref. 31 that a superconductor under a particular quasiparticle distribution is very similar to a superconductor in a uniform exchange field. Therefore, the FFLO state can be realized in this system. It is possible only at low temperatures, as is known for superconductors in a uniform exchange field.[32] Such a system is not sufficient to obtain the FFLO state at temperatures close to T_c. Here we show that two essential components, a nonequilibrium quasiparticle distribution and proximity between a superconducting film and a normal film of a particular finite width, allow us to obtain the FFLO state near T_c. The possibility to obtain the FFLO state at temperatures close to T_c is of great interest at least for two reasons: (i) We propose a way to reveal this FFLO state through the temperature dependence of its linear response to the applied magnetic field near T_c; (ii) the orbital effect of the applied magnetic field is highly nontrivial in the FFLO state: it can enhance T_c instead of suppressing it.[33]

In addition we propose an alternative way to generate the FFLO state in S/N heterostructures. It can occur due to creation of two shifted Fermi surfaces for spin-up and spin-down electrons if a spin imbalance is generated in the system.

Now we proceed with the microscopic calculations of the FFLO critical temperature of the S/N bilayer under nonequilibrium conditions. The sketch of the system is shown in Fig. 1. As we consider a nonequilibrium system, we make use of the Keldysh framework of quasiclassical theory. In our calculations we assume that (i) S is a singlet s-wave

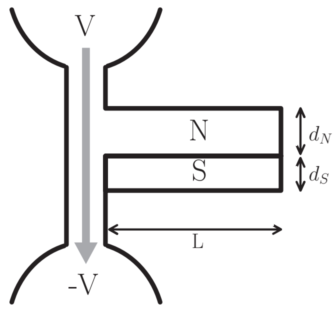

FIG. 1. Sketch of the system under consideration.

superconductor; (ii) the system is in the dirty limit, so the quasiclassical Green's function obeys Usadel equations;[34] (iii) the thickness d_S of the S layer is less than the superconducting coherence length $\xi_S = \sqrt{D_S/\Delta_0}$. This condition allows us to neglect the variations of the superconducting order parameter and the Green's functions across the S layer; (iv) we work in the vicinity of the critical temperature, so the Usadel equations can be linearized with respect to the anomalous Green's function:

$$D\nabla^2 \hat{f}^R + 2i\varepsilon \hat{f}^R + 2\pi\hat{\Delta} = 0. \tag{1}$$

Here $\hat{f}^R \equiv \hat{f}^R(\varepsilon,\boldsymbol{r})$ is the retarded anomalous Green's function. It depends on the quasiparticle energy ε and the coordinate vector $\boldsymbol{r} = (x,\boldsymbol{r}_\parallel)$, where x is the coordinate normal to the S/N interface and $\boldsymbol{r}_\parallel$ is parallel to the interface $[(yz)$ plane]. The hat over the anomalous Green's function means that it is a 2×2 matrix in spin space. However, here we consider the S/N system without Zeeman interaction, so the retarded and advanced components of the Green's function have the standard spin-singlet structure $\hat{f}^{R,A} = f^{R,A} i\sigma_2$, where σ_2 is the corresponding Pauli matrix. While we consider only the singlet pairing channel, the same is valid for the superconducting order parameter $\hat{\Delta} = \Delta i\sigma_2$. The spin structure can appear only in the distribuion function, as is described below. $D = D_{S(N)}$ stands for the diffusion constant in the superconductor (normal metal).

Equation (1) should be used with the Kupriyanov-Lukichev boundary conditions[35] at the S/N interface ($x = 0$):

$$\sigma_S \partial_x f_S^R = \sigma_N \partial_x f_N^R = g_{NS}\big(f_S^R - f_N^R\big)\big|_{x=0}, \tag{2}$$

where $\sigma_{S(N)}$ stands for the conductivity of the S (N) layer and g_{NS} is the conductance of the S/N interface. The boundary conditions at the ends of the bilayer are $\partial_x f_S^R|_{x=d_S} = \partial_x f_N^R|_{x=-d_N} = 0$.

In the FFLO state the superconducting order parameter and the anomalous Green's function are spatially modulated. We assume that $\Delta(\boldsymbol{r}) = \Delta \exp(i\boldsymbol{k}\boldsymbol{r}_\parallel)$ and $f(\boldsymbol{r}) = f(x)\exp(i\boldsymbol{k}\boldsymbol{r}_\parallel)$. It is worth noting here that this plane wave is not the only possible type of spatially modulated FFLO state which is allowed in the system. There can also be stationary wave states modulated as $\cos(\boldsymbol{k}\boldsymbol{r}_\parallel)$ and also 2D modulated structures. However, it can be shown that the critical temperature of all these states is the same and depends only on the absolute value of the modulating vector $\boldsymbol{k}$. Further choice of the most energetically favorable configuration is determined by the nonlinear terms in the Usadel equation, which are neglected now. So, while we are interested only in the instability point and the critical temperature of the corresponding FFLO state, we can consider the simplest type of modulation.

Substituting the modulated Green's function into the Usadel equation, we obtain the anomalous Green's functions in the S and N layers:

$$f_S = \frac{i\pi\Delta}{\varepsilon + iD_S k^2/2 + \frac{i g_{NS} D_S \lambda \tanh[\lambda d_N]}{2\sigma_S d_s(\lambda \tanh[\lambda d_N] + g_{NS}/\sigma_N)}}, \tag{3}$$

$$f_N(x) = \frac{(g_{NS}/\sigma_N)\cosh[\lambda(x+d_N)]}{\lambda \sinh[\lambda d_N] + (g_{NS}/\sigma_N)\cosh[\lambda d_N]} f_S, \tag{4}$$

where $\lambda^2 = k^2 - 2i\varepsilon/D_N$.

The critical temperature of the bilayer should be determined from the self-consistency equation

$$\Delta = \int_{-\omega_D}^{\omega_D} \frac{d\varepsilon}{4\pi} \Lambda \mathrm{Im}\left[f_S^R\right](\varphi_\uparrow + \varphi_\downarrow), \tag{5}$$

where ω_D is the cutoff energy, Λ is the dimensionless coupling constant, and $\varphi_{\uparrow,\downarrow}$ is the distribution function for spin-up (-down) quasiparticles. In order to generate the FFLO state we need

$$\varphi_\uparrow + \varphi_\downarrow = \tanh\frac{\varepsilon - eV}{2T} + \tanh\frac{\varepsilon + eV}{2T}. \tag{6}$$

This quasiparticle distribution can be reached in the bilayer in two different ways. (i) The bilayer can be attached to two additional electrodes with a voltage applied between them. We assume that the bilayer length L is shorter than the energy relaxation length. Then the energy distribution of the electrons in the bilayer is given by the superposition of the Fermi-Dirac distributions of the reservoirs[27,28] and $\varphi_\uparrow = \varphi_\downarrow$. (ii) If an electric current is injected into the bilayer through a ferromagnet, spin imbalance is generated at the interface between the ferromagnet and the nonmagnetic region. This is the so-called Aronov gap.[36,37] It provides the conversion (by spin-relaxation processes) of the spin-polarized current, injected from the ferromagnet, into the non-spin-polarized current, because only the non-spin-polarized current can flow through nonferromagnetic material. The value of the Aronov gap at the interface with the ferromagnet can be estimated as $eV \sim ePj_{inj}\rho l_s$, where P is the degree of spin polarization in the ferromagnet, j_{inj} is the density of the current injected from the ferromagnet, ρ is the resistivity of the normal metal, and l_s is the spin-relaxation length in it. The spin relaxation length is usually large in normal metals, so we can assume that our bilayer is shorter than l_s and, consequently, the spin imbalance is spatially constant in it. In this case $\varphi_{\uparrow(\downarrow)} = \tanh[(\varepsilon \mp eV)/2T]$.

The critical temperature of the S/N bilayer as a function of the modulation vector k is represented in Fig. 2. Different curves correspond to different values of the applied voltage eV. The curves of most physical interest are in the region of small k and a narrow interval of eV close to eV_c [see Fig. 2(b)]. The critical voltage eV_c corresponds to the complete destruction of homogeneous superconductivity in our bilayer. It is seen from Fig. 2(b) that if eV is close enough to eV_c, the critical temperature of the FFLO state is higher than T_c of the homogeneous state. That is, the FFLO state is energetically more favorable. The optimal values of the modulation vector k_{opt}, corresponding to the maximal T_c, are marked by points.

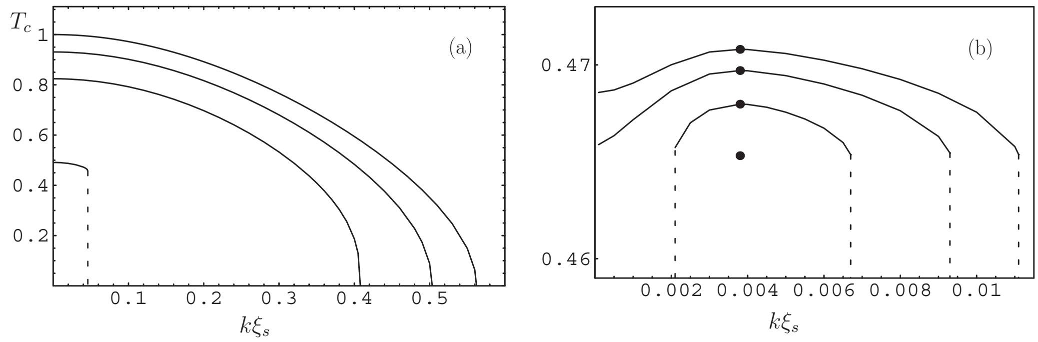

FIG. 2. (a) Dependence of the S/N bilayer critical temperature vs the modulation vector k for different values of eV. Points mark $k = k_{\mathrm{opt}}$. In units of $T_c(eV = 0, k = 0)$, $eV = 0, 0.556, 0.833, 1.111$ from top to bottom. (b) Enlarged region of (a), corresponding to small k. $eV = 1.11201, 1.11241, 1.112443, 1.112457$ from top to bottom. The other parameters are $\sigma_S/\sigma_N = 0.5$, $\xi_S g_{NS}/\sigma_S = 1.0$, $D_S/D_N = 0.04$, $d_N = 6.0\xi_S$, $d_S = 0.8\xi_S$ (Ref. 38).

A more detailed analysis shows that for the system under consideration the mean-field T_c is higher for a finite k than for $k = 0$ at any eV. Does this mean that the S/N bilayer should be in the FFLO state even in equilibrium (at $eV = 0$)? In order to analyze this question we plot in Fig. 3 the difference between the critical temperature of the FFLO state corresponding to k_{opt} and the critical temperature of the homogeneous state $\delta T_c/T_c = [T_c(k_{\mathrm{opt}}) - T_c(k = 0)]/T_c$ vs eV. As is seen from Fig. 3, $\delta T_c/T_c$ is very small for a wide range of eV and grows sharply only in the narrow region near eV_c. We have estimated that for small enough voltage biases $\delta T_c/T_c$ does not considerably exceed the Ginzburg number $\mathrm{Gi}_{2D} \sim 0.1/(k_F^2 l d) \approx 10^{-4}$–$10^{-3}$. So we cannot conclude on the basis of our mean-field analysis whether the FFLO state or the homogeneous state is more energetically favorable in this voltage range. However, in the narrow region of eV near eV_c (estimated width ~0.1–1 μV) $\delta T_c/T_c$ exceeds Gi_{2D} at least by an order of magnitude. So for this voltage region the FFLO state is indeed more favorable.

In addition, there is a narrow voltage region $eV > eV_c$ where homogeneous superconductivity is completely destroyed, but the FFLO state survives [see Fig. 2(b), where the bottom curve corresponds to $eV > eV_c$].

It is worth noting here that in order to observe the FFLO state the number of inelastic scatterers should be very small in the system. They can be described by adding an imaginary part Γ to the quasiparticle energy $\varepsilon \to \varepsilon + i\Gamma$. Then the condition $\Gamma < D_S k_{\mathrm{opt}}^2/2$ should be fulfilled.

The plane-wave state $\Delta \propto \exp(i\boldsymbol{k}\boldsymbol{r}_{||})$ can, in principle, carry a supercurrent in the bilayer plane. It is interesting to calculate this supercurrent. The corresponding expression for the supercurrent density takes the form

$$j^{(0)}(x) = \frac{\sigma(x)}{4\pi^2 e}\boldsymbol{k}\int_{-\infty}^{\infty} d\varepsilon \mathrm{Im}\big\{f^{(0)^2}(x)\big\}(\varphi_\uparrow + \varphi_\downarrow), \qquad (7)$$

where $\sigma(x) = \sigma_{S(N)}$ in the S (N) layer, and $f^{(0)}(x)$ is the solution of the linearized Usadel equation, expressed by Eqs. (3) and (4). The superscript (0) means that the value is calculated in the absence of the magnetic field. It is well known[7] that for a homogeneous system the true ground state corresponds to zero current density. For our bilayer system this statement is valid for the total current, integrated over the bilayer width $\int_{-d_N}^{d_S} dx j(x) = 0$. It can be shown by straightforward calculations that this is valid simultaneously with $\partial T_c/\partial k^2 = 0$, that is, at $k = k_{\mathrm{opt}}$. Vanishing of the total current means that the supercurrent mainly flows in the opposite directions in the N and S regions of the bilayer. This results in the appearance of a magnetic flux, which can be a hallmark of $\exp(i\boldsymbol{k}\boldsymbol{r}_{||})$ in the bilayer. This flux is plotted in Fig. 4 vs eV. The spatial profile of the corresponding magnetic field is shown in the inset to Fig. 4. However, for a state proportional to $\cos(i\boldsymbol{k}\boldsymbol{r}_{||})$ the supercurrent density $j(x) = 0$ locally for a given x. Consequently, this state is not

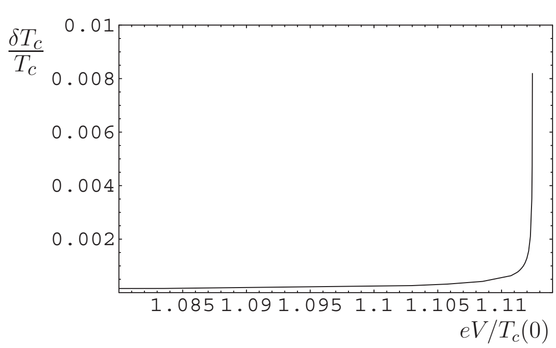

FIG. 3. Dependence of $\delta T_c/T_c = [T_c(k_{\mathrm{opt}}) - T_c(k = 0)]/T_c$ on eV. The parameters of the system are the same as in Fig. 2.

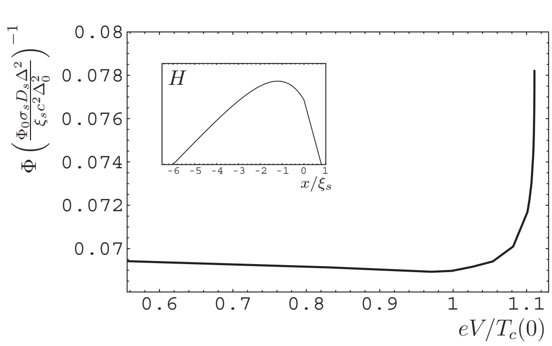

FIG. 4. Magnetic flux per unit length along $\boldsymbol{k}$, generated in the plane-wave FFLO state of the S/N bilayer, vs eV. Inset: The spatial profile of the corresponding magnetic field.

accompanied by nonzero supercurrents and cannot be detected by the corresponding magnetic flux.

Now we turn to the calculation of the Meissner response of the bilayer in the limit of a weak magnetic field $\boldsymbol{H}$, applied in the plane of the bilayer. As shown in Ref. 25, the transition of S/F hybrid structures to the in-plane FFLO state is accompanied by vanishing of the Meissner effect. This is connected to the fact that the Meissner response of a S/F bilayer in the homogeneous state can become paramagnetic, and such a structure is unstable with respect to the formation of the FFLO state.[25] The homogeneous state of our nonmagnetic S/N bilayer never exhibits the paramagnetic Meissner response. So vanishing of the Meissner response cannot be a hallmark of the in-plane FFLO state in our system. However, we have found other features of the linear response which are typical for an in-plane FFLO state in heterostructures.

We choose the vector potential $\boldsymbol{A} = (0, H_z x, -H_y x)$ to be parallel to the yz plane. For the considered FFLO state proportional to $\exp(i\boldsymbol{k}\boldsymbol{r}_\parallel)$ the contribution to the electric current density linear in magnetic field takes the form

$$j^{(1)}(x) = \frac{\sigma(x)}{2\pi^2 e}\int_{-\infty}^{\infty} d\varepsilon \Bigg[\boldsymbol{k}\,\mathrm{Im}\{f^{(1)}(x) f^{(0)}(x)\} - \frac{e}{c}\left(\boldsymbol{A} - \frac{c}{2e}\delta \boldsymbol{k}^{(1)}\right)\mathrm{Im}\{f^{(0)2}(x)\}\Bigg](\varphi_\uparrow + \varphi_\downarrow), \quad (8)$$

where the vector potential is taken in the gauge-invariant form $\boldsymbol{A} - \frac{c}{2e}\delta\boldsymbol{k}^{(1)}$. $f^{(1)}(x)$ is the linear correction to the anomalous Green's function. It is worth noting that in the homogeneous state $f^{(1)}(x)$ is zero in the gauge $\mathrm{div}\boldsymbol{A} = 0$. This is because $\mathrm{div}\boldsymbol{A}$ is the only possible first-order scalar function of $\boldsymbol{A}$. In the FFLO state $f^{(1)}(x) \propto \boldsymbol{k}\boldsymbol{A}'_x$. Therefore, in general, the linear response of the heterostructure in the FFLO state can be anisotropic with respect to the direction of the applied magnetic field.[39]

The full expression for $f^{(1)}(x)$ takes the form

$$f^{(1)}(x) = \frac{\Delta^{(1)}}{\Delta^{(0)}} f^{(0)}(x) + F_S^{(1)}, \quad (9)$$

where

$$F_S^{(1)} = \frac{(2e/c)(iD_S/d_S)}{E}\left[f_S^{(0)}\int_0^{d_S} \boldsymbol{k}[\boldsymbol{A}(x) - \delta\boldsymbol{k}^{(1)}]dx + \frac{\frac{g_{NS}}{\sigma_S}\int_{-d_N}^{0}\boldsymbol{k}[\boldsymbol{A}(x)-\delta\boldsymbol{k}^{(1)}]f_N^{(0)}(x)\frac{\cosh[\lambda(x+d_N)]}{\cosh[\lambda d_N]}dx}{\lambda\tanh[\lambda d_N] + g_{NS}/\sigma_N}\right] \quad (10)$$

and

$$E = \varepsilon + iD_S k^2/2 + \frac{i g_{NS} D_S \lambda \tanh[\lambda d_N]}{2\sigma_S d_s(\lambda\tanh[\lambda d_N] + g_{NS}/\sigma_N)}. \quad (11)$$

The correction $\Delta^{(1)}$ to the superconducting order parameter linear in magnetic field can be obtained from the self-consistency equation (5). The expression for $\Delta^{(1)}$ takes the form

$$\Delta^{(1)} = \frac{\int_{-\omega_D}^{\omega_D}\frac{d\varepsilon}{4\pi}\Lambda\,\mathrm{Im}\big[F_S^{(1)}\big](\varphi_\uparrow + \varphi_\downarrow)}{1 - \int_{-\omega_D}^{\omega_D}\frac{d\varepsilon}{4}\Lambda\,\mathrm{Re}\,[1/E]\,(\varphi_\uparrow+\varphi_\downarrow)}. \quad (12)$$

The denominator of Eq. (12) vanishes at $T = T_c$ because it is just the equation for calculating T_c at zero applied field. Consequently, for temperatures close to T_c the linear correction $\Delta^{(1)} \propto \Delta^{(0)}/(T_c - T)$. Therefore, the main contribution to $f^{(1)}(x)$ is given by the first term $f_\Delta^{(1)}(x) \propto \Delta^{(1)}$ in Eq. (9). In the state proportional to $\cos(\boldsymbol{k}\boldsymbol{r}_\parallel)$ the leading contribution to the Meissner current takes the same form (it is only twice larger). Certainly, this behavior is violated extremely close to T_c, where $\Delta^{(1)}$ becomes of the order of $\Delta^{(0)}$ and our linear approximation fails.

Therefore, it follows from Eq. (8) that the Meissner response of the S/N bilayer system in the FFLO state would exhibit a nontrivial temperature dependence. While in the homogeneous state the Meissner current $j^{(1)}(T) \propto \Delta^2 \propto (T_c - T)$ if the temperature is near T_c, in the FFLO state the leading contribution to $j^{(1)}(T) \propto \Delta^{(1)}\Delta^{(0)}$ and does not depend on temperature. In fact, this means that there are two possibilities: (i) The temperature dependence of the Meissner response near T_c in the FFLO state will be indeed nontrivial or (ii) T_c itself is shifted by the magnetic field in the FFLO state in the linear approximation, but the temperature dependence of the Meissner response can be of standard type. At the same time T_c of the homogeneous system does not depend on the applied magnetic field in the linear approximation. Which of the possibilities is realized in the particular system depends on what type of FFLO state is more stable in the system (plane wave, stationary wave, etc.). In any case, near T_c the behavior of the linear response of the system to the applied magnetic field in the FFLO state strongly differs from the behavior of the same system in the homogeneous state.

Anisotropy of $\Delta^{(1)}$ with respect to the mutual direction of the applied magnetic field and the modulation vector $\boldsymbol{k}$ is also clearly seen from Eqs. (12) and (10). This, in turn, leads to the corresponding anisotropy of the Meissner response.

In conclusion, we have shown that the in-plane FFLO state can be stabilized in a S/N bilayer under a nonequilibrium quasiparticle distribution for temperatures close to T_c. Its existence does not require any Zeeman interaction in the system. In general, this FFLO state can be of different types: plane wave, stationary wave, and, even, 2D structures are possible. The plane-wave state is accompanied by an internal magnetic flux. For all types of FFLO state near T_c the temperature dependence of the linear response of the system to the applied magnetic field should be strongly nontrivial.

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