Cooper Pairs with Broken Parity and Spin-Rotational Symmetries in *d*-Wave Superconductors

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Paramagnetic effects are shown to result in the appearance of a triplet component of order parameter in a vortex phase of a *d*-wave superconductor in the absence of impurities. This component, which breaks parity and spin-rotational symmetries of Cooper pairs, is expected to be of the order of unity in a number of modern superconductors such as organic, high T_c, and some others. A generic phase diagram of such type-IV superconductors, which are singlet ones at $H = 0$ and in the Meissner phase, and characterized by singlet-triplet mixed Copper pairs $\Delta_s + i\Delta_t$ with broken symmetries in a vortex phase, is discussed.

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It is well known [1,2] that type-II superconductors, where superconductivity survives at high magnetic fields $H_{c1}(T) < H < H_{c2}(T)$ as the Abrikosov vortex phase [1–3], are subdivided into two main classes. They are superconducting alloys (or dirty superconductors) [1,2] and relatively clean materials, where type-II superconductivity is due to anisotropy of their quasiparticles' spectra and relatively heavy masses of quasiparticles [4]. The latter compounds are currently the most interesting and perspective superconducting materials, including organic [5], heavy fermion [6], high T_c [7], MgB_2 [8], and some other superconductors.

Superconducting orbital order parameter $\Delta(\mathbf{r}_1, \mathbf{r}_2)$, corresponding to the pairing of two electrons in a Cooper pair, can usually be expressed as $\Delta(\mathbf{r}_1, \mathbf{r}_2) = \Delta(\mathbf{R})\Delta(\mathbf{r})$ [9,10], where the external order parameter $\Delta(\mathbf{R})$ is related to the motion of a center of mass of a Cooper pair,$\mathbf{R} = (\mathbf{r}_1 + \mathbf{r}_2)/2$, whereas the internal order parameter $\Delta(\mathbf{r})$ describes the relative motion of electrons in the Cooper pair $\mathbf{r} = \mathbf{r}_1 - \mathbf{r}_2$. In this context, type-II superconductors in their vortex phases are characterized by broken symmetries of $\Delta(\mathbf{R})$, corresponding to vortices and Meissner currents. Other important issues are symmetries of the internal orbital order parameter $\Delta(\mathbf{r})$ and the related spin part of order parameter $\Delta(\sigma_1, \sigma_2)$. To satisfy Fermi statistics, in the case of singlet superconductivity (where the total spin of a Cooper pair is $|\mathbf{S}| = 0$), the internal order parameter $\Delta(\mathbf{r})$ has to be an even function of coordinate $\mathbf{r}$, whereas, in the case of triplet superconductivity (where $|\mathbf{S}| = 1$), $\Delta(\mathbf{r})$ has to be an odd function of $\mathbf{r}$. In accordance with the symmetry properties of $\Delta(\mathbf{r})$ [or its Fourier component, $\hat{\Delta}(\mathbf{k})$], superconductors are subdivided into conventional ones [1,2] (where superconductivity is described by BCS *s*-wave singlet pairing) and unconventional ones [9,10] [where the symmetry of $\hat{\Delta}(\mathbf{k})$ is lower than the underlying symmetry of the crystalline lattice]. At present, unconventional *d*-wave singlet superconductivity is firmly established in high T_c [11] and some organic materials. There exist also several strong candidates for unconventional triplet superconducting pairing such as Sr_2RuO_4 [12], organic superconductors $(TMTSF)_2X$ [13], ferromagnetic [14], and heavy fermion [9,10,15] superconductors. It is a common belief that magnetic field does not change the internal order parameters $\hat{\Delta}(\mathbf{k})$ and $\Delta(\sigma_1, \sigma_2)$ in conventional [1,2] and unconventional [9,10] type-II superconductors.

The goal of our Letter is to demonstrate that there must be type-IV superconductors [16] which exhibit qualitatively different magnetic properties. More precisely, we suggest and prove the following theorem: each singlet type-II superconductor in the absence of impurities is actually a type-IV superconductor with broken parity $\mathbf{k} \rightarrow -\mathbf{k}$ and spin-rotational symmetries of internal Cooper pair wave functions in vortex phase, provided that the effective constant of triplet pairing is not exactly zero, $g_t \neq 0$ [17]. We show that the above-mentioned theorem is an inherent property of singlet superconductivity and is due to careful accounting for paramagnetic spin-splitting effects in a vortex phase, which have been treated so far only for $g_t = 0$ [1,2,18].

We define type-IV superconductivity as singlet superconductivity at $H = 0$ and in the Meissner phase, which exhibits broken symmetries of internal Cooper pair wave functions $\hat{\Delta}(\mathbf{k})$ and $\Delta(\sigma_1, \sigma_2)$ in vortex phase. In our particular case, the internal order parameter in the vortex phase is shown to be a mixture of a singlet *d*-wave component $\hat{\Delta}_s(\mathbf{k})$ with a triplet component $i\hat{\Delta}_t(\mathbf{k})$, where $\Delta_t(\mathbf{k})$ is an imaginary part of a complex order parameter [17]. Below, we demonstrate that the effects of singlet-triplet coexistence are expected to be of the order of unity in a number of modern strongly correlated clean type-II superconductors, where the orbital upper critical fields are of the order of the paramagnetic limiting fields [1,2,18] $\mu_B H_{c2}(0) \sim T_c$ [19] (see Table I). It is important that the suggested theorem is very general: it is valid even for the simplest spin independent electron-electron interactions for both attractive and repulsive interactions in a triplet channel.

As discussed below, the above-mentioned theorem is based only on symmetry arguments and is a consequence of broken symmetry in spin space (due to paramagnetic effects) and broken translational invariance of $\Delta(\mathbf{R})$ (due

TABLE I. Upper critical fields $H_{c2}(0)$ [20–23], transitions temperatures T_c, and triplet-singlet ratio $|R|$ are listed for some modern layered d-wave and s-wave superconductors.

	$\beta-(ET)_2AuI_2$	$\beta-(ET)_2IBr_2$	$YBa_2Cu_3O_7$	MgB_2
$H_{c2}(0)[T]$	5.5(‖)	2.4(‖)	110(⊥)	18(‖)
$\mu_B H_{c2}(0)[K]$	3.7	1.6	74	12
$T_c[K]$	4.3	2.3	85	35
R	0.85	0.7	0.85	0.4

to the existence of vortices). Therefore, our qualitative results about singlet-triplet mixed order parameter and the broken symmetries of Cooper pair internal wave functions are irrespective of a weak coupling model used in the Letter. We recall that, in the vortex phase, translational invariance of $\Delta(\mathbf{R})$ is broken and $\Delta(\mathbf{R})$ is a function of $\mathbf{R}$ on a scale of ξ, where ξ is a coherence length [1–4]. Therefore, $\Delta(R)$ corresponds to superconducting pairing of electrons with total nonzero momenta of Cooper pairs of the order of $|\mathbf{q}| \sim \hbar/\xi$. As seen from Fig. 1, a probability of pairing for electrons with spin-up and spin-down $|\Delta(+,-)|^2$ is different from that for electrons with spin-down and spin-up $|\Delta(-,+)|^2$, if $\mathbf{q} \neq 0$. Therefore, singlet superconductivity, which is characterized by the spin order parameter $\Delta(+,-) = -\Delta(-,+)$ has to be mixed with a triplet component, characterized by spin order parameter $\Delta(+,-) = \Delta(-,+)$ [9,10] (see Fig. 1). Note that, in such a triplet component, spin-rotational symmetry is broken and spins of Cooper pairs are directed perpendicular to an external magnetic field.

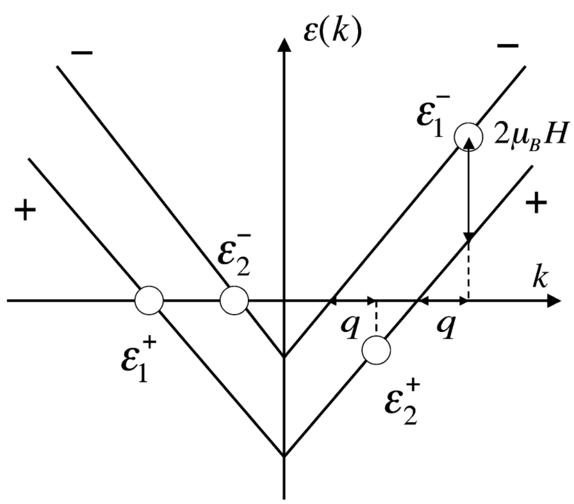

FIG. 1. Paramagnetic effects split electron spectra with spins-up and spins-down: $\epsilon^+(k) = \epsilon_0(k) - \mu_B H$ and $\epsilon^-(k) = \epsilon_0(k) + \mu_B H$, correspondingly. Two Cooper pairs with spin parts of wave functions $\Delta(+,-)$ and $\Delta(-,+)$ and equal total momenta $q \neq 0$ are characterized by different probabilities to exist, since the energy difference $|\epsilon_1^+ - \epsilon_1^-| = qv_F + 2\mu_B H$ is not equal to the energy difference $|\epsilon_2^- - \epsilon_2^+| = -qv_F + 2\mu_B H$. [For simplicity, the linearized one-dimensional electron spectrum $\epsilon_0(k) = v_F|k|$ is shown].

Here, we quantitatively describe superconducting pairing with the internal order parameter, exhibiting broken inversion and spin-rotational symmetries, in a d-wave singlet superconductor with layered electron spectrum,

$$\epsilon_0(\mathbf{k}) = (k_x^2 + k_y^2)/2m + 2t_\perp \cos(k_z d), \qquad \epsilon_F = mv_F^2/2 \tag{1}$$

in a parallel magnetic field

$$\mathbf{H} = (0, H, 0), \qquad \mathbf{A} = (0, 0, -Hx). \tag{2}$$

In this case, where electron-electron interactions do not depend on electron spins, the total Hamiltonian of electron system can be written in the form

$$\begin{aligned} \hat{H} &= \hat{H}_0 + \hat{H}_{\text{int}}, \qquad \hat{H}_0 = \sum_{\mathbf{k},\sigma} \epsilon_\sigma(\mathbf{k}) a_\sigma^+(\mathbf{k}) a_\sigma(\mathbf{k}), \\ \hat{H}_{\text{int}} &= \frac{1}{2}\sum_{\mathbf{q},\sigma}\sum_{\mathbf{k},\mathbf{k}_1} V(\mathbf{k},\mathbf{k}_1) a_\sigma^+\Big(\mathbf{k}+\frac{\mathbf{q}}{2}\Big) a_{-\sigma}^+\Big(-\mathbf{k}+\frac{\mathbf{q}}{2}\Big) \\ &\quad \times a_{-\sigma}\Big(-\mathbf{k}_1+\frac{\mathbf{q}}{2}\Big) a_\sigma\Big(\mathbf{k}_1+\frac{\mathbf{q}}{2}\Big), \end{aligned} \tag{3}$$

where $\epsilon_\sigma(\mathbf{k}) = \epsilon_0(\mathbf{k}) - \sigma\mu_B H$ $(\sigma = \pm 1)$, $a_\sigma^+(\mathbf{k})$, and $a_\sigma(\mathbf{k})$ are electron creation and annihilation operators. As usual [9,10], electron-electron interactions are subdivided into singlet and triplet ones:

$$\begin{aligned} V(\mathbf{k},\mathbf{k}_1) &= V_s(\mathbf{k},\mathbf{k}_1) + V_t(\mathbf{k},\mathbf{k}_1), \\ V_s(\mathbf{k},\mathbf{k}_1) &= V_s(-\mathbf{k},\mathbf{k}_1) = V_s(\mathbf{k},-\mathbf{k}_1), \\ V_t(\mathbf{k},\mathbf{k}_1) &= -V_t(-\mathbf{k},\mathbf{k}_1) = -V_t(\mathbf{k},-\mathbf{k}_1). \end{aligned} \tag{4}$$

We define the normal and Gorkov (anomalous) finite temperature Green functions,

$$\begin{aligned} G_{\sigma,\sigma}(\mathbf{k},\mathbf{k}_1;\tau) &= -\langle T_\tau a_\sigma(\mathbf{k},\tau) a_\sigma^+(\mathbf{k}_1,0)\rangle, \\ F_{\sigma,-\sigma}(\mathbf{k},\mathbf{k}_1;\tau) &= \langle T_\tau a_\sigma(\mathbf{k},\tau) a_{-\sigma}(-\mathbf{k}_1,0)\rangle, \\ F_{\sigma,-\sigma}^+(\mathbf{k},\mathbf{k}_1;\tau) &= \langle T_\tau a_\sigma^+(-\mathbf{k},\tau) a_{-\sigma}^+(\mathbf{k}_1,0)\rangle, \end{aligned} \tag{5}$$

as well as singlet and triplet superconducting order parameters,

$$\begin{aligned} \Delta_s(\mathbf{k},\mathbf{q}) &= -\frac{1}{2}\sum_{\mathbf{k}_1} V_s(\mathbf{k},\mathbf{k}_1) T \\ &\quad \times \sum_{\omega_n}\Big[F_{+,-}\Big(i\omega_n;\mathbf{k}_1+\frac{\mathbf{q}}{2},\mathbf{k}_1-\frac{\mathbf{q}}{2}\Big) \\ &\quad - F_{-,+}\Big(i\omega_n;\mathbf{k}_1+\frac{\mathbf{q}}{2},\mathbf{k}_1-\frac{\mathbf{q}}{2}\Big)\Big], \\ \Delta_t(\mathbf{k},\mathbf{q}) &= -\frac{1}{2}\sum_{\mathbf{k}_1} V_t(\mathbf{k},\mathbf{k}_1) T \\ &\quad \times \sum_{\omega_n}\Big[F_{+,-}\Big(i\omega_n;\mathbf{k}_1+\frac{\mathbf{q}}{2},\mathbf{k}_1-\frac{\mathbf{q}}{2}\Big) \\ &\quad + F_{-,+}\Big(i\omega_n;\mathbf{k}_1+\frac{\mathbf{q}}{2},\mathbf{k}_1-\frac{\mathbf{q}}{2}\Big)\Big], \end{aligned} \tag{6}$$

by the standard ways [9,10,24,25].

The goal of our Letter is to consider the superconducting nucleus in the vicinity of the phase transition line between metallic and singlet-triplet mixed superconducting phases in the Ginzburg-Landau (GL) region, $(T_c - T)/T_c \ll 1$ [1–4], where T_c is a transition temperature between the metallic state and the d-wave singlet phase at $H = 0$. For this purpose, we linearize the Gorkov equations [9,10,24] with respect to order parameters (6) and obtain the following system of linear equations [26]:

$$\Delta_s(\mathbf{k},\mathbf{q}) = -\frac{1}{2}\sum_{\mathbf{k}_1} V_s(\mathbf{k},\mathbf{k}_1) T \sum_{\omega_n} [\Delta_s(\mathbf{k}_1,\mathbf{q}) S + \Delta_t(\mathbf{k}_1,\mathbf{q}) D],$$

$$\Delta_t(\mathbf{k},\mathbf{q}) = -\frac{1}{2}\sum_{\mathbf{k}_1} V_t(\mathbf{k},\mathbf{k}_1) T \sum_{\omega_n} [\Delta_t(\mathbf{k}_1,\mathbf{q}) S + \Delta_s(\mathbf{k}_1,\mathbf{q}) D],$$

$$S = G^0_+\left(i\omega_n, \mathbf{k}_1 + \frac{\mathbf{q}}{2}\right) G^0_-\left(-i\omega_n, -\mathbf{k}_1 + \frac{\mathbf{q}}{2}\right) + G^0_-\left(i\omega_n, \mathbf{k}_1 + \frac{\mathbf{q}}{2}\right) G^0_+\left(-i\omega_n, -\mathbf{k}_1 + \frac{\mathbf{q}}{2}\right),$$

$$D = G^0_+\left(i\omega_n, \mathbf{k}_1 + \frac{\mathbf{q}}{2}\right) G^0_-\left(-i\omega_n, -\mathbf{k}_1 + \frac{\mathbf{q}}{2}\right) - G^0_-\left(i\omega_n, \mathbf{k}_1 + \frac{\mathbf{q}}{2}\right) G^0_+\left(-i\omega_n, -\mathbf{k}_1 + \frac{\mathbf{q}}{2}\right), \quad (7)$$

where $G^0_\sigma(i\omega_n, \mathbf{k}) = 1/(i\omega_n - \epsilon_\sigma(\mathbf{k}))$ is the Green function of a free electron in the presence of paramagnetic effects and ω_n is the Matsubara frequency [25]. [Note that common Eq. (7) directly demonstrates singlet-triplet coexistence effects in the vortex phase since $D \neq 0$ at $\mathbf{q} \neq 0$. Indeed, Eq. (7) does not have a solution when $\Delta_t(\mathbf{k},\mathbf{q}) = 0$, implying that such a triplet component must occur (see also Fig. 1)].

Below, we consider in detail an important example, the coexistence of singlet $d_{x^2-y^2}$-wave [26] and triplet p_x-wave order parameters, which correspond to the following matrix elements of electron-electron interactions:

$$\begin{pmatrix} V_s(\mathbf{k},\mathbf{k}_1) \\ V_t(\mathbf{k},\mathbf{k}_1) \end{pmatrix} = -\frac{4\pi}{v_F}\begin{pmatrix} g_s & \cos 2\phi \cos 2\phi_1 \\ g_t & \cos(\phi - \phi_1) \end{pmatrix}, \quad g_s > 0, \quad g_s > g_t, \quad (8)$$

where ϕ and ϕ_1 are polar angles corresponding to momenta $\mathbf{k}$ and $\mathbf{k}_1$, respectively. [Note that inequalities $g_s > 0$ and $g_s > g_t$ guarantee that the singlet $d_{x^2-y^2}$-wave phase is a ground state at $H = 0$ and $T < T_c$]. After substitution of Eq. (8) in Eq. (7), we represent order parameters as follows, $\Delta_s(\mathbf{k},\mathbf{q}) = \sqrt{2}\cos 2\phi \Delta_s(\mathbf{q})$ and $\Delta_t(\mathbf{k},\mathbf{q}) = \sqrt{2}\cos\phi \Delta_t(\mathbf{q})$, and rewrite Eq. (7) in a matrix form:

$$\begin{pmatrix} A_{ss}(\mathbf{q}) & A_{st}(\mathbf{q}) \\ A_{ts}(\mathbf{q}) & A_{tt}(\mathbf{q}) \end{pmatrix}\begin{pmatrix} \Delta_s(\mathbf{q}) \\ \Delta_t(\mathbf{q}) \end{pmatrix} = \begin{pmatrix} \Delta_s(\mathbf{q})/g_s \\ \Delta_t(\mathbf{q})/g_t \end{pmatrix}. \quad (9)$$

We calculate matrix $\hat{A}(\mathbf{q})$ at $q_y = 0$ in the GL region [3,4,9,27] which corresponds to its expansion as a power series in small parameters $v_F q_x / T_c \ll 1$ and $t_\perp d q_z / T_c \ll 1$. As a result, we obtain

$$\hat{A} = \begin{pmatrix} (2\pi T)\sum_{\omega_n>0}^{\Omega}[\frac{1}{\omega_n} - \frac{1}{8\omega_n^3}(v_F^2 q_x^2 + 4t_\perp^2 q_z^2 d^2)], & -\mu_B H v_F q_x (\pi T_c)\sum_{\omega_n>0}^{\infty}\frac{1}{\omega_n^3} \\ -\mu_B H v_F q_x (\pi T_c)\sum_{\omega_n>0}^{\infty}\frac{1}{\omega_n^3}, & (2\pi T)\sum_{\omega_n>0}^{\Omega}[\frac{1}{\omega_n} - \frac{1}{8\omega_n^3}(3v_F^2 q_x^2/2 + 4t_\perp^2 q_z^2 d^2)] \end{pmatrix}, \quad (10)$$

with Ω being a cutoff energy. Magnetic field (2) is introduced in Eqs. (9) and (10) by means of a standard quasiclassical approximation [3,4,27,28], $q_x \to -i(d/dx)$, $q_z/2 \to eA_z/c = eHx/c$ which leads to the following matrix GL equations extended to the case of triplet-singlet coexistence:

$$\begin{pmatrix} \tau + \xi_\parallel^2(\frac{d^2}{dx^2}) - \frac{(2\pi\xi_\perp)^2}{\phi_0^2}H^2x^2, & i\frac{\sqrt{7\zeta(3)}}{\sqrt{2}\pi}(\frac{\mu_B H}{T_c})\xi_\parallel(\frac{d}{dx}) \\ i\frac{\sqrt{7\zeta(3)}}{\sqrt{2}\pi}(\frac{\mu_B H}{T_c})\xi_\parallel(\frac{d}{dx}), & \tau + \frac{g_t - g_s}{g_t g_s} + \frac{3}{2}\xi_\parallel^2(\frac{d^2}{dx^2}) - \frac{(2\pi\xi_\perp)^2}{\phi_0^2}H^2x^2 \end{pmatrix}\begin{pmatrix} \Delta_s(x) \\ \Delta_t(x) \end{pmatrix} = 0, \quad (11)$$

where $\tau = (T_c - T)/T_c \ll 1$, $\xi_\parallel = \sqrt{7\zeta(3)}v_F/4\sqrt{2}\pi T_c$ and $\xi_\perp = \sqrt{7\zeta(3)}t_\perp d/2\sqrt{2}\pi T_c$ are GL coherence lengths [4,9], $\zeta(3) \simeq 1.2$ is the zeta Riemann function, ϕ_0 is a flux quantum, and x is coordinate of a center of mass of the Cooper pair. In typical cases, where the singlet superconducting transition temperature is not close to the triplet one ($g_s - g_t \sim g_s$, $g_s > g_t > 0$), or where the effective triplet coupling constant is repulsive ($-g_t > 0$), Eq. (11) has the following solutions:

$$\begin{pmatrix} \Delta_s(x) \\ \Delta_t(x) \end{pmatrix} = \begin{pmatrix} \exp(-\frac{\tau x^2}{2\xi_\parallel^2}) \\ i\sqrt{\tau}\frac{\sqrt{7\zeta(3)}}{\pi}(\frac{g_t g_s}{g_t - g_s})(\frac{\mu_B H}{T_c})(\frac{\sqrt{\tau}x}{\sqrt{2}\xi_\parallel})\exp(-\frac{\tau x^2}{2\xi_\parallel^2}) \end{pmatrix}. \quad (12)$$

Eqs. (11) and (12) are the main results of our Letter. They extend the GL differential equation [1–4,9,10,27] and its famous Abrikosov solution for the superconducting nucleus $\exp(-\tau x^2/2\xi_\parallel^2)$ [1–3] to the case $g_t \neq 0$. Eqs. (11) and (12) directly demonstrate that, in a vortex phase, singlet order parameter always coexists with the triplet one, characterized by $|\mathbf{S}| = 1$ and $S_y = 0$ ($\mathbf{H} \parallel \mathbf{y}$), for an arbitrary sign of effective triplet coupling constant g_t. Note that triplet component (12), breaking parity and spin-rotational symmetries, may also, in principle, beak time-reversal symmetry due to the existence of nondiagonal matrix elements, proportional to iH in Eq. (11) [17].

To summarize, the main message of the Letter is that Cooper pairs cannot be considered as unchanged elementary particles in vortex phases of modern strongly correlated strongly type-II superconductors, where $H_{c2}(0) \sim H_p$ [i.e., $\mu_B H_{c2}(0) \sim T_c$] and $|g_s| \sim |g_t|$ [29]. Indeed the triplet-singlet components ratio in Eq. (12) at $x_0 = \sqrt{2}\xi_{\|}/\sqrt{\tau}$, where x_0 is a characteristic "size" of the superconducting nucleus (12), and $(T_c - T)/T_c \sim T_c$ (i.e., $\tau \sim 1$) can be estimated as $R = \Delta_t/\Delta_s \sim i(\mu_B H_{c2}(0)/T_c)$ (see Table I). Note that the appearance of a triplet component (12), where Cooper pair spins are perpendicular to an external magnetic field, has to change all qualitative features of vortex phases in d-wave superconductors. These include the unusual topology of superconducting vortices [30], the appearance of spin-wave-like excitations, the disappearance of quasiparticles near zeros of the $d_{x^2-y^2}$-wave superconducting gap, possible unusual spin susceptibility, phase sensitive effects, and other nontrivial phenomena to be studied in the future. We suggest that, in clean type-II superconductors, there exist the fourth critical fields, $H_{c4}(T)$, corresponding to crossovers (or phase transitions) between Abrikosov vortex phases and exotic vortex phases with broken symmetries; we call such materials type-IV superconductors. In conclusion, we point out that singlet-triplet mixing effects were earlier studied in He_3 [31], Larkin-Ovchinnikov-Fulde-Ferrell phase [32,33], for surface superconductivity [34], and in superconductors without inversion symmetry [9,35,36].

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