Cooper Pairs with Broken Parity and Time-Reversal Symmetries in D-wave Superconductors

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Abstract

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 both parity and time-reversal 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 characterized by singlet-triplet mixed Copper pairs with broken time-reversal symmetry 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 Abrikosov vortex phase [3,2,1], 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 quasi-particles spectra and relatively heavy masses of quasi-particles [4]. The latter compounds are currently the most interesting and perspective superconducting materials, including organic [5], heavy fermions [6], high-\( T_c \) [7], MgB\(_2\) [8], and some other superconductors.

Superconducting orbital order parameter, \( \Delta(r_1,r_2) \), corresponding to pairing of two electrons in Cooper pair, can usually be expressed as \( \Delta(r_1,r_2) = \Delta(R)\Delta(r) \) [9,10], where external order parameter, \( \Delta(R) \), is related to motion of a center of mass of Cooper pair, \( R = (r_1 + r_2)/2 \), whereas internal order parameter, \( \Delta(r) \), describes relative motion of electrons in Cooper pair, \( r = r_1 - r_2 \). In this context, type-II superconductors in their vortex phases are characterized by broken symmetries of \( \Delta(R) \), corresponding to vortices and Meissner currents. Other important issues are symmetries of internal orbital order parameter, \( \Delta(r) \), and related spin part of order parameter, \( \Delta(\sigma_1,\sigma_2) \). To satisfy Fermi statistics, in case of singlet superconductivity (where the total spin of Cooper pair \( |S| = 0 \)), internal order parameter, \( \Delta(r) \), has to be an even function of coordinate \( r \), whereas, in case of triplet superconductivity (where \( |S| = 1 \)), \( \Delta(r) \) has to be an odd function of \( r \). In accordance with symmetry properties of \( \Delta(r) \) (or its Fourier component, \( \hat{\Delta}(k) \)), superconductors are subdivided into conventional ones [1,2] (where superconductivity is described by BCS s-wave singlet pairing) and unconventional ones [9,10] (where symmetry of \( \hat{\Delta}(k) \) is lower than underlying symmetry of crystalline lattice). At present time, 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 \( \text{Sr}_2\text{RuO}_4 \) [12], \((\text{TMTSF})_2\text{X}\) [13], ferromagnetic [14], and heavy fermion [9,10,15] superconductors. It is a common belief that magnetic field does not change internal order parameters, \( \hat{\Delta}(k) \) and \( \Delta(\sigma_1,\sigma_2) \), in conventional [1,2] and unconventional [9,10] type-II superconductors. Moreover, although Meissner currents break time-reversal symmetry of \( \Delta(R) \), internal order parameters, \( \hat{\Delta}(k) \) and \( \Delta(\sigma_1,\sigma_2) \), are believed to preserve \( t \to -t \) symmetry.

The goal of our Letter is to demonstrate that there have to exist 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 type-IV superconductor with broken \( k \to -k \) and \( t \to -t \) symmetries of internal Cooper pairs wave functions in vortex phase, provided that effective constant of triplet pairing is not exactly zero, \( g_t \neq 0 \). We show that the above mentioned theorem is an inherent property of singlet superconductivity and is due to careful account for paramagnetic
spin-splitting effects in vortex phase, which have been treated so far only for \( g_t = 0 \) [17,1].

We define type-IV superconductivity as singlet superconductivity at \( H = 0 \) which exhibits broken symmetries of internal Cooper pairs wave functions, \( \Delta(k) \) and \( \Delta(\sigma_1, \sigma_2) \), in vortex phase. In our particular case, internal order parameter in vortex phase is shown to be a mixture of a singlet d-wave component, \( \Delta_s(k) \), with a triplet component, \( i \Delta_t(k) \). Note that this order parameter breaks not only parity symmetry, \( k \rightarrow -k \), but also a time-reversal symmetry, \( t \rightarrow -t \), due to an imaginary coefficient \( i \). Below, we demonstrate that the effects of singlet-triplet coexistence are of the order of unity in a number of modern clean type-II superconductors, where the orbital upper critical fields are of the order of paramagnetic limiting fields, \( \mu_B H_{c2}(0) \sim T_c \) [18] (see Table 1). It is important that the suggested theorem is very general: it is valid even for simplest spin independent electron-electron interactions for both attractive and repulsive interactions in triplet channel. As discussed below, this theorem is based on symmetry arguments and is a consequence of broken spin symmetry (due to paramagnetic effects) and broken translational invariancy of \( \Delta(R) \) (due to the existence of vortices).

Here, we discuss how paramagnetic effects result in the appearance of a triplet component in vortex phase using qualitative arguments. We recall that, in vortex phase, external order parameter, \( \Delta(R) \), is a function of \( R \) on scale of \( \xi \), where \( \xi \) is a coherence length [1-4]. Therefore, \( \Delta(R) \) corresponds to superconducting pairing of electrons with total non-zero momenta of Cooper pairs of the order of \( |q| \sim \hbar/\xi \). 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 \( q \neq 0 \). Therefore, singlet superconductivity, which is characterized by spin order parameter \( \Delta(+,-) = -\Delta(-,+) \) [9,10], has to be mixed with a triplet component, characterized by spin order parameter \( \Delta(+,-) = \Delta(-,+) \) (i.e., \( |S| = 1 \) and \( S_y = 0 \)) (see Fig.1).

Below, we quantitatively describe superconducting pairing with internal order parameter, exhibiting broken inversion and time-reversal symmetries, in d-wave singlet superconductor with layered electron spectrum,

\[
\epsilon_0(k) = \frac{(k_x^2 + k_y^2)}{2m} + 2t_\perp \cos(k_z d), \quad \epsilon_F = \frac{mv_F^2}{2},
\]

in a magnetic field:

\[
H = (0, H, 0), \quad A = (0, 0, -Hx).
\]

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

\[
\hat{H} = \hat{H}_0 + \hat{H}_{int}, \quad \hat{H}_0 = \sum_{k,\sigma} \epsilon_\sigma(k) \ a_\sigma^+(k) \ a_\sigma(k),
\]
\[ \hat{H}_{int} = \frac{1}{2} \sum_{\mathbf{q}, \sigma} \sum_{\mathbf{k}, \mathbf{k}_1} V(\mathbf{k}, \mathbf{k}_1) a^+_\sigma(\mathbf{k} + \frac{\mathbf{q}}{2}) a^-_\sigma(-\mathbf{k} + \frac{\mathbf{q}}{2}) a^-_{-\sigma}(-\mathbf{k}_1 + \frac{\mathbf{q}}{2}) a^-_{\sigma}(\mathbf{k}_1 + \frac{\mathbf{q}}{2}), \] (3)

where \( \epsilon_\sigma(\mathbf{k}) = \epsilon_0(\mathbf{k}) - \sigma \mu BH \) (\( \sigma = \pm 1 \)), \( a^+_\sigma(\mathbf{k}) \) and \( a^-_\sigma(\mathbf{k}) \) are electron creation and annihilation operators. As usually \([9,10]\), electron-electron interactions are subdivided into singlet and triplet ones:

\[ V(\mathbf{k}, \mathbf{k}_1) = V_s(\mathbf{k}, \mathbf{k}_1) + V_t(\mathbf{k}, \mathbf{k}_1), \quad V_s(\mathbf{k}, \mathbf{k}_1) = V_s(-\mathbf{k}, -\mathbf{k}_1), \]
\[ V_t(\mathbf{k}, \mathbf{k}_1) = -V_t(-\mathbf{k}, -\mathbf{k}_1) \] (4)

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

\[ G_{\sigma,\sigma}(\mathbf{k}, \mathbf{k}_1; \tau) = -< T_\tau a_\sigma(\mathbf{k}, \tau) a^+_\sigma(\mathbf{k}_1, 0) >, \quad F_{\sigma,-\sigma}(\mathbf{k}, \mathbf{k}_1; \tau) = -< T_\tau a_\sigma(\mathbf{k}, \tau) a^-_{-\sigma}(\mathbf{k}_1, 0) >, \]
\[ F_{\sigma,\sigma}(\mathbf{k}, \mathbf{k}_1; \tau) = -< T_\tau a^+_\sigma(-\mathbf{k}, \tau) a^+_\sigma(\mathbf{k}_1, 0) >, \] (5)

as well as singlet and triplet superconducting order parameters,

\[ \Delta_s(\mathbf{k}, \mathbf{q}) = \frac{1}{2} \sum_{\mathbf{k}_1} V_s(\mathbf{k}, \mathbf{k}_1) T \sum_{\omega_n} [F_{+, -}(i\omega_n; \mathbf{k}_1 + \frac{\mathbf{q}}{2}, \mathbf{k}_1 - \frac{\mathbf{q}}{2}) - F_{-, +}(i\omega_n; \mathbf{k}_1 + \frac{\mathbf{q}}{2}, \mathbf{k}_1 - \frac{\mathbf{q}}{2})], \]
\[ \Delta_t(\mathbf{k}, \mathbf{q}) = \frac{1}{2} \sum_{\mathbf{k}_1} V_t(\mathbf{k}, \mathbf{k}_1) T \sum_{\omega_n} [F_{+, -}(i\omega_n; \mathbf{k}_1 + \frac{\mathbf{q}}{2}, \mathbf{k}_1 - \frac{\mathbf{q}}{2}) + F_{-, +}(i\omega_n; \mathbf{k}_1 + \frac{\mathbf{q}}{2}, \mathbf{k}_1 - \frac{\mathbf{q}}{2})] \] (6)

by standard ways \([9,10,19]\).

The goal of our Letter is to consider a phase transition line between metallic and singlet-triplet mixed superconducting phases in Ginzburg-Landau (GL) region, \((T_c - T)/T_c \ll 1\) \([1-4]\), where \(T_c\) is a transition temperature between metallic state and d-wave singlet phase at \(H = 0\). For this purpose, we linearize Gorkov equations \([9,10,19]\) with respect to order parameters \((6)\) and obtain the following system of linear equations \([21]\):

\[ \Delta_s(\mathbf{k}, \mathbf{q}) = \frac{1}{2} \sum_{\mathbf{k}_1} V_s(\mathbf{k}, \mathbf{k}_1) T \sum_{\omega_n} [\Delta_s(\mathbf{k}_1, \mathbf{q}) S + \Delta_t(\mathbf{k}_1, \mathbf{q}) D], \]
\[ \Delta_t(\mathbf{k}, \mathbf{q}) = \frac{1}{2} \sum_{\mathbf{k}_1} V_t(\mathbf{k}, \mathbf{k}_1) T \sum_{\omega_n} [\Delta_t(\mathbf{k}_1, \mathbf{q}) S + \Delta_s(\mathbf{k}_1, \mathbf{q}) D], \]
\[ S = G^0_+(i\omega_n, \mathbf{k}_1 + \frac{\mathbf{q}}{2}) G^0_-(-i\omega_n, -\mathbf{k}_1 + \frac{\mathbf{q}}{2}) + G^0_+(i\omega_n, \mathbf{k}_1 + \frac{\mathbf{q}}{2}) G^0_+(i\omega_n, -\mathbf{k}_1 + \frac{\mathbf{q}}{2}), \]
\[ D = G^0_+(i\omega_n, \mathbf{k}_1 + \frac{\mathbf{q}}{2}) G^0_-(-i\omega_n, -\mathbf{k}_1 + \frac{\mathbf{q}}{2}) - G^0_+(i\omega_n, \mathbf{k}_1 + \frac{\mathbf{q}}{2}) G^0_+(i\omega_n, -\mathbf{k}_1 + \frac{\mathbf{q}}{2}), \] (7)

where \( G^0_\sigma(i\omega_n, \mathbf{k}) = 1/(i\omega_n - \epsilon_\sigma(\mathbf{k})) \) is Green function of a free electron in the presence of paramagnetic effects and \(\omega_n\) is Matsubara frequency \([20]\). [Note that common Eqs.(7) directly demonstrate singlet-triplet coexistence effects in vortex phase, where \(\mathbf{q} \neq 0\) (see Fig.1)].
Below, we consider in detail an important example, coexistence of singlet $d_{x^2-y^2}$-wave and triplet $p_z$-wave order parameters, which corresponds to the following matrix elements of electron-electron interactions:

$$
\left( \begin{array}{c} V_s(k, k_1) \\ V_t(k, k_1) \end{array} \right) = -\frac{4\pi}{v_F} \left( \begin{array}{cc} g_s \cos 2\phi \cos 2\phi_1 \\ g_t \cos(\phi - \phi_1) \end{array} \right), \quad g_s > 0, \quad g_s > g_t,
$$

where $\phi$ and $\phi_1$ are polar angles corresponding to momenta $k$ and $k_1$, respectively. [Note that inequalities $g_s > 0$ and $g_s > g_t$ guarantee that singlet $d_{x^2-y^2}$-wave phase is a ground state at $H = 0$ and $T < T_c$.] After substitution of Eqs.(8) in Eqs.(7), we represent order parameters as follows, $\Delta_s(k, q) = \sqrt{2}\cos 2\phi \Delta_s(q)$ and $\Delta_t(k, q) = \sqrt{2}\cos \phi \Delta_t(q)$, and rewrite Eqs.(7) in a matrix form:

$$
\left( \begin{array}{cc} A_{ss}(q) & A_{st}(q) \\ A_{ts}(q) & A_{tt}(q) \end{array} \right) \left( \begin{array}{c} \Delta_s(q) \\ \Delta_t(q) \end{array} \right) = \left( \begin{array}{c} \Delta_s(q)/g_s \\ \Delta_t(q)/g_t \end{array} \right).
$$

We calculate matrix $\hat{A}(q)$ at $q_y = 0$ in GL region [3,4,22,9] which corresponds to its expansion as power series in small parameters, $v_Fq_x/T_c \ll 1$ and $t_{\perp}dq_z/T_c \ll 1$. As a result, we obtain:

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

with $\Omega$ being a cut-off energy. Magnetic field (2) is introduced in Eqs.(9),(10) by means of a standard quasi-classical approximation [23,22,4,3], $q_x \rightarrow -i(d/dx)$, $q_{\perp}/2 \rightarrow eA_{\perp}/c = eHx/c$, which leads to the following matrix GL equations extended to the case of triplet-singlet coexistence:

$$
\left( \begin{array}{cc} \tau + \xi^2 \frac{(d^2}{dx^2} \right) - \frac{(2\pi \xi)}{\phi_0} H^2x^2, & i\sqrt{\zeta(3)} \frac{\xi}{\sqrt{2\pi}} \left( \frac{\mu_B H}{T_c} \right) \xi \frac{d^2}{dx^2}, \\ i\sqrt{\zeta(3)} \frac{\mu_B H}{\sqrt{2\pi}} \xi \frac{d^2}{dx^2}, & \tau + \frac{q_{\perp}/gs}{gs} \xi - \frac{3\xi^2}{2} \left( \frac{d^2}{dx^2} - \frac{(2\pi \xi)}{\phi_0} H^2x^2 \right) \end{array} \right) \left( \begin{array}{c} \Delta_s(x) \\ \Delta_t(x) \end{array} \right) = 0,
$$

where $\tau = (T_c - T)/T_c \ll 1$, $\xi = \sqrt{\zeta(3)}v_F/4\sqrt{2\pi T_c}$ and $\xi = \sqrt{\zeta(3)}t_{\perp}d/2\sqrt{2\pi T_c}$ are GL coherence lengths [4,9], $\zeta(3) \approx 1.2$ is zeta Riemann function, $\phi_0$ is a flux quantum, $x$ is coordinate of a center of mass of Cooper pair. In typical case, where $gs - gt \sim gs$, Eqs. (11) have the following solutions:

$$
\left( \begin{array}{c} \Delta_s(x) \\ \Delta_t(x) \end{array} \right) = \left( \begin{array}{c} \exp \left( -\frac{\tau x^2}{2\xi^2} \right) \\ i \sqrt{\frac{\pi}{2\xi^2}} \left( \frac{q_{\perp}gs}{gs} \right) \left( \frac{\mu_B H}{T_c} \right) \sqrt{\frac{\pi}{2\xi^2}} \exp \left( -\frac{\tau x^2}{2\xi^2} \right) \end{array} \right).
$$

Eqs.(11),(12) are the main results of our Letter. They extend GL differential equation [1-4,22,9,10] and its Abrikosov solution for superconducting nucleus, $\exp(-\tau x^2/2\xi^2)$ [3,1,2],
to the case \( g_t \neq 0 \). Eqs.(11),(12) directly demonstrate that, in vortex phase, singlet order parameter always coexists with triplet one, characterized by \( |S| = 1 \) and \( S_y = 0 \) (\( \mathbf{H} \parallel \mathbf{y} \)), for arbitrary sign of effective triplet coupling constant \( g_t \). It is important that triplet component \( (12) \) breaks not only parity but also time-reversal symmetry due to the existence of non-diagonal matrix elements, proportional to \( iH \), in Eqs.(11). Indeed, \( \Delta^* t(x) \neq \Delta t(x) \) in Eq.(12), which indicates that \( t \rightarrow -t \) symmetry is broken and, thus, Cooper pairs possess some internal magnetic moments [9,10].

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 type-II superconductors, where \( \mu_B H_{c2}(0) \sim T_c \) and \( |g_s| \sim |g_t| \). Indeed triplet-singlet components ratio in Eqs. (12) at \( x = \sqrt{2}\xi_\parallel/\sqrt{\tau} \) and low temperatures, \( \tau \sim 1 \), can be estimated as \( R = \Delta_t/\Delta_s \sim i [\mu_B H_{c2}(0)/T_c] \) (see Table 1). The appearance of a triplet component, breaking time-reversal symmetry, has to change all qualitative features of vortex phases in d-wave superconductors. These include the appearance of non-zero internal magnetic moments of Cooper pairs, possible unusual topology of superconducting vortices, the appearance of spin-wave excitations, the disappearance of quasi-particles near zeros of \( d_{x^2-y^2} \)-wave gap, \( \cos 2\phi = 0 \), unusual spin susceptibility, and other non-trivial phenomena to be studied in the future. We suggest that, in clean type-II superconductors, there exist the forth critical fields, \( H_{c4}(T) \), corresponding to phase transitions (or crossovers) between Abrikosov vortex phases and some exotic vortex phases and call such materials type-IV superconductors. In conclusion, we point out that singlet-triplet mixing effects were earlier studied in He\(^3\) [24], Larkin-Ovchinnikov-Fulde-Ferrell phase [25,26], for surface superconductivity [27], and in superconductors without inversion symmetry [9,28,29].

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TABLE I: Upper critical fields, $H_{c2}(0)$ [30-33], 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     |
FIG. 1: Paramagnetic effects split electron spectra with spin up and spin 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 energy difference $|\epsilon_1^+ - \epsilon_1^-| = qv_F + 2\mu_B H$ is not equal to energy difference $|\epsilon_2^- - \epsilon_2^+| = -qv_F + 2\mu_B H$. [For simplicity, linearized one-dimensional electron spectrum, $\epsilon(k) = v_F|k|$, is shown].