Triplet pairing and upper critical field in the mixed state of d-wave superconductors

V. V. Kabanov  
Josef Stefan Institute 1001, Ljubljana, Slovenia

We show that an additional triplet component of the order parameter is generated in the vortex phase of the d-wave superconductor. Spatial variations of the triplet component are analyzed for a strong spin-orbit coupling. Corrections to the London equation and an unusual temperature dependence of the upper critical field, $H_{c2}(T)$, are obtained in the case of the weak spin-orbit coupling.

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There is a common belief that the copper-based high-temperature superconductors have nontrivial order parameter (OP) transforming as $B_{1g}(x^2 - y^2)$ representation of $D_{4h}$ point group \[1\]. In the framework of the BCS theory this "d-wave" symmetry is related to the 'internal' coordinate of the OP, $\mathbf{r} = \mathbf{r}_1 - \mathbf{r}_2$, assuming its decomposition as $\Delta(\mathbf{r}_1, \mathbf{r}_2) = \Delta(\mathbf{R})\Delta(\mathbf{r})$, where $\mathbf{R} = (\mathbf{r}_1 + \mathbf{r}_2)/2$ is the center-of-mass coordinate \[2\]. This symmetry of the OP implies that the spectrum of elementary excitations is gapless in certain directions so that the d-wave gap is very sensitive to an external perturbation. For example, applying a uniform magnetic field generates a secondary triplet OP \[6\]. It is generated in the mixed state of type-II superconductors \[3, 4\]. P-wave component of the OP could be also generated by a surface induced spin-orbit coupling (SOC) \[5\]. Appearance of ip component \[6\] is associated with the Lifshitz invariants (LI) in the free energy (FE) \[7\]. More recently it has been shown that the triplet OP $i\Delta_t$ is generated in the mixed state of type-II superconductors \[8\]. It was argued that this effect is a consequence of broken symmetry in the spin space due to the paramagnetic effect and a broken translational invariance. This effect was called as type-IV superconductivity \[8\]. Phenomenologically nontrivial secondary OP is generated by linear coupling of the primary OP gradients to the secondary OP. From the microscopic point of view the interaction of electrons on the Fermi surface is repulsive in the channel of secondary OP. Nevertheless, non-diagonal terms corresponding to the coupling to the primary OP leads to nonzero value of the secondary OP \[8\]. The relative amplitude of the p-wave component depends on the strength of SOC and may be of the order 10-20\% \[8\].

Previously it was shown that the superconducting current in the d-wave superconductor reduces the symmetry group and generates the secondary triplet OP \[6\]. It is possible since the symmetry allows for the presence of terms in the Ginzburg-Landau FE functional which are of the first order in gradients. Usually these terms are allowed for crystals where the inversion symmetry is broken. Inversion is the symmetry operation of $D_{4h}$ group and generation of the LI is possible if the secondary component breaks the inversion symmetry \[7, 9\]. Here we investigate the electromagnetic response in the mixed state of the type-II d-wave superconductor in the presence of LI \[6, 7\] and formulate the criteria for experimental observation of the effect.

First let us briefly discuss the symmetry properties of p-wave superconducting OP \[10\]. The triplet OP has 9 components. In the case of the strong SOC spin is coupled to the lattice and transforms together with the lattice. P-wave component of $D_{4h}$ point group is written as $\mathbf{d}(\mathbf{k}, \mathbf{R}) = (p_x(\mathbf{R})k_x + p_y(\mathbf{R})k_y)\mathbf{z}$, where $\mathbf{z}$ is a unite axial vector in the spin space and $p_x(\mathbf{R})$ and $p_y(\mathbf{R})$ play the role of the OP in the Ginzburg-Landau functional. In the case of the weak SOC the spin and the orbital parts of the OP transforms independently and $\mathbf{d}(\mathbf{k}, \mathbf{R}) = p_x(\mathbf{R})k_x + p_y(\mathbf{R})k_y$, where two vectors in the spin space $p_x(\mathbf{R}), p_y(\mathbf{R})$ play the role of the OP. In the latter case the coupling between singlet and triplet OP is possible only in the presence of the magnetic field.

**Strong SOC.** The FE of d-wave superconductor per unite of volume can be written as:

$$F = \frac{\hbar^2}{2m}[|\mathbf{D}\psi|^2 + \alpha(T - T_c)|\psi|^2 + \frac{\beta}{2}|\psi|^4 + i\eta\left[\psi^*(D_xp_x - D_yp_y) - \psi(D_x^*p_x^* - D_y^*p_y^*)\right]$$

\[1\]

$$+ \alpha_p(|p_x|^2 + |p_y|^2) + \frac{\mathbf{H}^2}{8\pi}$$

where $\psi$ and $p_x, y$ are the primary d-wave and the secondary p-wave OP, $\mathbf{D} = -i\nabla - 2e\mathbf{A}$, $\mathbf{A}$ is the vector potential for magnetic field $\mathbf{H} = \nabla \times \mathbf{A}$, $\alpha, \alpha_p, \beta > 0, \eta$ are real constants. Here we write explicitly the LI describing p-wave component. In Eq.\(1\) the additional term, quadratic in $p_x, p_y$, is written to secure $p_x = p_y = 0$ solution in the uniform state $\psi_d = const$.

Minimizing the FE for variation of $\delta\psi, \delta p_x, \delta p_y$ we obtain the following set of equations:

$$\frac{\hbar^2}{2m}\mathbf{D}^2\psi + \alpha(T - T_c)\psi + \beta|\psi|^2\psi + i\eta(D_xp_x - D_yp_y) = 0$$

\[2\]

$$p_x = \frac{i\eta}{\alpha_p}D_x\psi$$

$$p_y = \frac{-i\eta}{\alpha_p}D_y\psi$$

\[3\]
Substituting Eq.(3) to Eq.(2) we obtain standard Ginzburg-Landau equation

\[ \frac{\hbar^2}{2m^*} D^2 \psi + \alpha (T - T_c) \psi + \beta |\psi|^2 \psi = 0 \] (4)

with renormalized effective mass \((m^*)^{-1} = m^{-1} - 2\eta^2/\alpha_p\). The renormalization of the effective mass was derived in Eq.(7) of Ref.[6]. If renormalized effective mass is negative \((m^*)^{-1} < 0\) \((2\eta^2/\alpha_p > m^{-1})\) the helical phase is formed[7] and higher order gradient terms for secondary OP should be included to the FE, Eq.(1). The properties and the thermodynamics of helical phases have been studied in Ref.[7] and are not considered here. If renormalized effective mass \((m^*)^{-1} > 0\) \((2\eta^2/\alpha_p < m^{-1})\) the FE, Eq.(1), describes the formation of the secondary OP in the presence of the magnetic field or in the presence of the supercurrent[6]. Therefore we assume in the following that \((m^*)^{-1} > 0\) and higher order gradient terms are not essential in generation of the secondary OP.

Minimizing the FE for variations of the vector potential \(\delta A\) and substituting Eqs.(3,4) we obtain superconducting current:

\[ j_s = -\frac{i e}{m^*}(\psi^* \nabla \psi - \psi \nabla \psi^*) - \frac{4e^2}{m^* c} |\psi|^2 A \] (5)

Now let us calculate the OP and the properties of the mixed state in the external magnetic field. It is convenient to introduce dimensionless variables, \(\psi = \psi_0 f\), \(p_{x,y} = \psi_0 P_{x,y}\), where \(\psi_0 = \sqrt{[\alpha(T - T_c)]/\beta}\), \(r = \lambda(T) \rho\), \(\lambda(T)^2 = \frac{m^*}{m^* + \epsilon c^2 \eta^2}\), \(A = \frac{2}{\sqrt{\pi}} \xi(T) A\), where \(\xi(T)^2 = -\frac{\hbar^2}{2m^* \alpha(T - T_c)}\) and \(h = \frac{2\kappa}{\sqrt{\lambda(T)}}(\lambda(T)H).\) Separating the phase and the modulus of the OP, \(f = f_0 \exp(\phi)\), and redefining the vector potential the system of equations is reduced to the equation for the modulus of the OP \(f = f_0\) and for the magnetic field \(h[11]:\)

\[ -\frac{\nabla}{\kappa} f_0 + \frac{1}{f_0^3} (\nabla \times h)^2 = f_0 - f_0^3 \] (6)

\[ f_0^2 h = \frac{2}{f_0} \nabla f_0 \times \nabla \times h - \nabla \times \nabla \times h, \] (7)

where \(\kappa = \lambda/\xi\) is Ginzburg-Landau parameter. Amplitude of the p-wave component is expressed in terms of \(f_0\) and \(h\) as:

\[ \mathcal{P}_{x,y} = \mp \frac{i \eta}{\alpha_p \xi} \frac{i \nabla}{\kappa} - \frac{\text{curl}(h)}{f_0^3} \bigg|_{x,y} f_0 \] (8)

Therefore the problem is reduced to the standard solution of the Eqs.(6,7) and then to the calculation of the p-wave amplitudes applying Eq.(8).

Weak magnetic field \(H_{c1} \ll H \ll H_{c2}\). Single vortex. Solution of Eqs. (6,7) are known and can be written in the form[11]:

\[ f_0 = c p + \ldots \]
\[ h = h(0) - c p^2/2\kappa + \ldots \] (9)

where \(c\) is the constant of the order of 1. This expansion is valid near the vortex core \(\rho \ll 1/\kappa\). The amplitude of the p-wave component in that case is:

\[ \mathcal{P}_x = -i \mathcal{P}_y = \frac{\eta c}{\alpha_p \xi \kappa} \frac{x - iy}{\rho} \] (10)

This effect is consistent with the symmetry arguments and was briefly discussed in connection with superfluidity of \(^3\)He[12]. Near the vortex core the screening current is flowing around the vortex as well as the gradient of the modulus of the OP is finite. As it was shown in Ref.[6] currents generate \(ip\) secondary OP. On the other hand a real vector breaks the inversion symmetry and generates the real \(p\)-component of the secondary OP[5]. Far from the vortex core but in the region where the screening current is strong \(1/\kappa \ll \rho \ll 1\) \(f_0^{-2} dh/d\rho = -(\kappa \rho)^{-1}\) and \(f_0^3 = 1 - (\kappa \rho)^{-2}[11]\) the amplitude of the p-wave component are

\[ \mathcal{P}_x = -\frac{\eta}{\alpha_p \xi \kappa} \frac{ix}{\rho} \]
\[ \mathcal{P}_y = \frac{\eta}{\alpha_p \xi \kappa} \frac{iy}{\rho} \] (11)

This is again consistent with the previous results. Far from the vortex core the modulus of the OP is almost constant (gradient is small) and the screening current is strong. Therefore only \(ip\) secondary OP survives in that area[5].

**Strong magnetic field** \((H_{c2} - H)/H_{c2} \lesssim 1\) **Magnetic field** in that case is determined by the formula[11]:

\[ h = \kappa + \epsilon - \frac{f_0^2}{2\kappa} \] (12)

where \(\epsilon = \kappa \frac{H - H_{c2}}{H_{c2}}\). Spacial dependence of \(f_0\) is determined by[11]:

\[ f_0^2 = |c_0|^2 \sum_{m,n} (-1)^{mn} \exp(-i\pi n/2) \exp(-\pi(m^2 + n^2 - mn)/\sqrt{3}) \exp(3^{1/4} \pi^{1/2} \kappa i (nx + (2m - n)y)/\sqrt{3}) \] (13)

where \(|c_0|^2 = (H - H_{c2})/\beta H_{c2}\) and \(\beta = 1.1596\). Substituting Eqs.(12,13) to Eq.(8) we obtain

\[ \mathcal{P}_x = -i \mathcal{P}_y = \frac{\eta}{\alpha_p \xi \kappa} \left( \frac{\partial f_0}{\partial x} + i \frac{\partial f_0}{\partial y} \right) \] (14)

**Weak SOC.** In the strong magnetic field when Zeeman energy is larger then the energy of the SOC spin decouples from the lattice. In that case vector functions \(p_x(R), p_y(R)\) will be coupled to the magnetic field \(H\) (similar effect was considered in Ref.[13]). Therefore linear in gradients term in the FE has the following form:

\[ i\eta' \langle \nabla \times A \rangle \left[ \psi^*(D_x p_x - D_y p_y) - \psi (D_x^* p_x^* - D_y^* p_y^*) \right] \]
Here we use cartesian basis in the spin space \( \uparrow \downarrow \) and therefore the triplet OP has zero projection of the spin to the direction of the magnetic field \( \mathbf{B} \). Minimizing the FE for variation of the OP \( \delta \psi, \delta \mathbf{p}_x, \delta \mathbf{p}_y \) we obtain the following equation for \( \psi \):

\[
\frac{\hbar^2}{2m} \nabla^2 \psi + \alpha (T-T_c) \psi + \beta |\psi|^2 \psi - \frac{\eta^2}{\alpha_p} \nabla^2 (\mathbf{B} \psi) = 0, \tag{15}
\]

where \( \mathbf{p}_{x,y} = \pm i \eta D_{x,y}(\mathbf{B} \psi) \). If we assume that the magnetic field is constant and equal to the external magnetic field Eq. (15) is reduced to Eq. (4) with the field dependent effective mass \( m^*(H) = m - 2\eta^2 H^2 / \alpha_p \).

Straightforward variation with respect to vector potential \( \mathbf{A} \) gives the expression for superconducting current \( \mathbf{j}_s \):

\[
\mathbf{j}_s = -i e \left( \psi^* \nabla \psi - \psi \nabla \psi^* \right) - \frac{4\eta^2}{m^*(H) c} |\psi|^2 \mathbf{A} + \frac{2i e \eta^2 H}{\alpha_p} (\psi^* \nabla (\mathbf{B} \psi) - \psi \nabla (\mathbf{B} \psi^*)) - \frac{\eta^2}{\alpha_p} \nabla \times \mathbf{v} \times \mathbf{H}, \tag{16}
\]

To simplify this equation let us assume that the magnetic field is a constant. After simple calculations we get the formula for \( \mathbf{j}_s \),

\[
\mathbf{j}_s = -i e \left( \psi^* \nabla \psi - \psi \nabla \psi^* \right) - \frac{4\eta^2}{m^*(H) c} |\psi|^2 \mathbf{A} + \frac{\eta^2}{\alpha} \nabla \times \mathbf{H}, \tag{17}
\]

where \( \nu = -\frac{2m^*(H)}{\gamma} (\alpha (T-T_c) |\psi|^2 + \frac{\eta^2}{2} |\psi|^4) \). It is seen from this equation that the correction to the supercurrent is similar to the ordinary Hall current, where an effective electric field is proportional to the gradient of the superfluid density, \( n_s = |\psi|^2 \).

When the magnetic field increases further effective mass \( m^*(H) \) becomes small and next order terms in the gradients should be considered in the expansion of the FE density. This is important for calculation of the upper critical field \( H_{c2} \). For the calculation of \( H_{c2} \) we analyze linearized equation with constant magnetic field directed along \( \mathbf{z} \) axis. Therefore only \( z \) component of \( \mathbf{p}_{x,y} \) OP is relevant. We also write second order gradient terms for \( \psi \) component in \( \mathbf{A} \) in the FE density as \( \frac{k^2}{2m_p} (|\mathbf{D} \mathbf{p}_x|^2 + |\mathbf{D} \mathbf{p}_y|^2) \). Introducing new dimensionless variables, \( \mathcal{H} = \frac{2\pi^2 \mu^2}{\Phi_0}, \mathcal{H} = \frac{2\pi^2 \mu^2}{\Phi_0}, \mathcal{H} = \frac{2\pi^2 \mu^2}{\Phi_0} \), \( \Phi_0 \) is the flux quanta and \( x \to x \sqrt{\mathcal{H}/\xi_0} \), one obtains the following set of equations:

\[
\mathcal{H} \left( -\psi'' + x^2 \psi \right) + \tau \psi + \mathcal{H}^{3/2} \nu (p_x' + i x p_y) = 0
\]

\[
\mathcal{H} \left( -p_x'' + x^2 p_x \right) + b_p - i \nu \mathcal{H}^{3/2} \psi = 0, \tag{18}
\]

where \( \nu = \frac{\eta^2 \Phi_0}{2m_p c}, \mathcal{H} = \frac{2\pi^2 \mu^2}{\Phi_0}, b = \alpha_p c/\alpha T_c \sim 1, \tau = (T-T_c)/T_c \). Here we use Landau gauge \( \mathbf{A} = (0, Hx, 0) \) and therefore assume that all functions depend on \( x \) only. Using standard substitution:

\[
\psi = \sum s_n \exp \left( -x^2 / 2 \mathcal{H} n \right) H_n(x),
\]

\[
p_{x,y} = \sum q_{n(x,y)} \exp \left( -x^2 / 2 \mathcal{H} n \right) H_n(x), \tag{19}
\]

where \( H_n(x) \) are hermitian polynomials. After simple calculations we obtain the following algebraic equations which determines critical temperature in the magnetic field:

\[
(2n + 1) \mathcal{H} + \tau - \nu^2 \mathcal{H}^3 \left[ \frac{n + 1}{(2n + 3) \mathcal{H} + b} + \frac{n}{(2n - 1) \mathcal{H} + b} \right] = 0, \tag{20}
\]

Upper critical field is determined by the highest possible critical temperature which corresponds to \( n = 0 \) in the Eq. (20). Hence the critical transition temperature in the magnetic field is determined as

\[
T_c(\mathcal{H})/T_c = 1 - \mathcal{H} + \frac{\nu^2 \mathcal{H}^3}{3 \mathcal{H} + b}. \tag{21}
\]

There are a number of interesting observations based on Eq.(21). The presence of LI increases critical temperature of the superconductor. This effect is well known \([8]\). More interestingly, the correction to the upper critical field becomes nonlinear and leads to the positive curvature of \( \mathcal{H}_{c2} \) as the function of temperature: \( d^2 \mathcal{H}_{c2}(T)/dT^2 > 0 \). Further increase of the magnetic field may lead to the recovery of superconductivity in higher fields \([15]\). It is also consistent with the previous results \([2]\), where it was shown that increase of the critical temperature due to formation of inhomogeneous helical phase is proportional to \( \eta^2 \). Therefore, calculated upper critical field should be substantially larger then paramagnetic or Clogston limit \([8, 16]\). Generalizing this result to the weak SOC we can see that the shift of the critical temperature due to purely paramagnetic effect is proportional to \( \eta^2 \mathcal{H}^2 \). This is exactly the results for \( T_c(H) \) which follows from Eq.(21). We have to remember that in high magnetic field higher order gradient terms should be considered. The higher order orbital effect is proportional to \( \mathcal{H}^2 \) and reduce the effect of LI. If higher order orbital effects are strong enough the reentrant superconductivity in high magnetic field will be suppressed completely.

In Figure 1 we plot the temperature dependence of \( H_{c2} \). To avoid infinite increase of critical temperature in the high field we take into account higher order gradient terms in the formula Eq.(21). As a result critical field is determined by the formula:

\[
T_c(\mathcal{H})/T_c = 1 - \mathcal{H} - \frac{\gamma \mathcal{H}^2}{\mathcal{H}^2 + 3 \mathcal{H} + b}, \tag{22}
\]
where $\gamma$, $\gamma_p \sim 1$ dimensionless constants describing the effect of the higher order gradient terms in the Landau expansion. To demonstrate the temperature dependence of the critical field we choose $\gamma = 1/4$, $b = 1$, $\gamma_p = 1$ and $\nu^2 c$ is changing from 1, to 1.15. The results, presented in Fig. 1 are restricted by standard range of applicability of Ginzburg-Landau theory and the limit $T \rightarrow 0$ should be considered only as a qualitative. In that case more accurate microscopic equation should be analyzed.

To estimate the strength of the effect we refer to Gorkov’s equation formulated for the case of the strong SOC in, and in the case of paramagnetic effect in. It follows from the analysis in the strong SOC limit $\Delta_p/\Delta_d \approx \Delta_{SO}/E_f \approx \Delta_{SO}\mu_B H_{c2}/\Delta_0^2$, where $\Delta_{p,d}$ is the $p(d)$ gap, respectively, $\Delta_{SO}$ is the energy of the SOC, $E_f$ is the Fermi energy, and $\mu_B$ is the Bohr magneton. From the phenomenological consideration it follows that $\Delta_p/\Delta_d \approx \eta/\xi\alpha_p$. Therefore LI are determined by the SOC $\eta/\xi\alpha_p \approx \Delta_{SO}/E_f$. In the case of the weak SOC the estimate reads $\Delta_p/\Delta_d \approx \eta\mu_B H_{c2}/\Delta_d[8]$. In that case LI are determined by by the paramagnetic effect. Therefore at the value of the field $H \approx H_{c2}\Delta_{SO}/\Delta_d$ we expect a crossover from the strong to the weak SOC limit. The characteristic value of triplet gap was evaluated in and may be of the order of 0.8 of $\Delta_d$ in superconductors where $H_{c2}$ is close to the paramagnetic limit.

As a result we can estimate the value of the parameter $\nu^2 c \approx (H_{c2}/\Delta_0)^2 \sim 1$ in Fig. 1. Therefore we believe that the effect may be observed in materials with weak SO coupling and with high upper critical field $H_{c2}$, that exceeds paramagnetic limit. Possible candidates for this effect are organic superconductors, where highly nonlinear temperature dependence of the upper critical field is observed. On the other hand, the p-wave component of the order parameter in the mixed state of superconductors may be observed by the direct vortex imaging with the scanning tunnelling spectroscopy. This effect should be strong in materials with the strong SOC.

In conclusion we have shown that the additional triplet secondary OP is generated in the vortex (Abrikosov) phase of type II d-wave superconductor. This effect is caused by the superconducting currents and by the spatial variations of the OP in the presence of the magnetic field. According to estimates using the microscopic theory the triplet component may be as large as 80% of the primary d-wave component in the field near $H_{c2}[8]$. Nonlinear corrections in the vector potential to the London equation are derived. The upper critical field $H_{c2}(T)$ is calculated in the limit of weak SOC. It is shown that temperature dependence of $H_{c2}(T)$ is highly nonlinear near the transition to the helical phase.

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