Structure of the Fulde-Ferrell-Larkin-Ovchinnikov state in two-dimensional superconductors

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Abstract

Nonuniform superconducting state due to strong spin magnetism is studied in two-dimensional (2D) type-II superconductors near the second order phase transition line between the normal and the superconducting states. The optimum spatial structure of the orderparameter is examined in systems with cylindrical symmetric Fermi surfaces. It is found that states with 2D structures have lower free energies than the traditional one-dimensional solutions, at low temperatures and high magnetic fields. For s-wave pairing, triangular, square, hexagonal states are favored depending on the temperature, while square states are favored at low temperatures for d-wave pairing. In these states, orderparameters have 2D structures such as square and triangular lattices.

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Nonuniform superconducting state due to a strong spin magnetic effect, which is called Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state, has often been studied, since Fulde & Ferrell and Larkin & Ovchinnikov showed that the upper critical field of a nonuniform superconducting state exceeds the Pauli paramagnetic limit (Chandrasekar-Clogston limit) under some ideal conditions. Recently, it is sometimes suggested that organic, high-\( T_c \) copper oxide, and heavy fermion superconductors can be candidates of the FFLO superconductors. This is because they have large upper critical fields so that strong spin magnetism is attainable, and also because they can be clean type-II superconductors.

In particular, the organic and the copper oxide superconductors have large critical fields, especially when orbital pair breaking effect is weaken by applying magnetic field in any direction parallel to the most conducting plane. In addition to this, some features of Fermi surface structure arising from the low dimensionality enhances the critical field of the FFLO state. Therefore, some compounds in the families of the organic and high-\( T_c \) superconductors might be good FFLO superconductors.

However, spatial oscillation of the order parameter, which characterizes the FFLO state, has not been observed yet, while very large upper critical fields were observed in some compounds such as \((\text{BEDT-TTF})_2\text{X}\) and \((\text{TMTSF})_2\text{X}\). Those critical fields might exceed the Pauli paramagnetic limit. In particular, increase of the critical field at low temperatures with positive \(\frac{d^2H_c}{dT^2}\) was observed in \((\text{TMTSF})_2\text{PF}_6\). Such behavior of the critical field is very similar to the behavior that is theoretically obtained in low dimensional FFLO superconductors. This might suggest the possibility of the FFLO state in this material or similar organic compounds, although this is not an evidence of the FFLO state. The spatial structure of the order parameter must be observed in order to prove existence or nonexistence of the FFLO state.

The spatial structure of the order parameter in the FFLO state is different from that in the traditional Abrikosov state, because their origins are quite different. The nonuniformity of the order parameter in the FFLO state is due to the displacement of the Fermi surfaces of up and down spin electrons by the Zeeman energy, not due to the orbital motion around flux.
lines. Gruenberg and Gunther examined the condition for the coexistence of these states in a three dimensional system. In the two dimensions, the Abrikosov state occurs unless the magnetic field is parallel to the conducting layer. However, when the parallel direction is approached, its Landau level index increases and then the spatial structure of the order parameter approaches to that of the FFLO state. We assume a parallel magnetic field and concentrate on the FFLO state in this paper.

In the studies so far, the spatial oscillation of the order parameter in the FFLO state is believed to be in a single direction. Larkin et al. studied an s-wave superconductor with spherical symmetric Fermi surfaces, and found that the form $\Delta(r) \propto \cos(q \cdot r)$ is the most stable solution among the periodic solutions of the gap equation expanded near the second order phase transition field. However, other cases such as systems with anisotropic Fermi surfaces and those with anisotropic pairing interactions have not been studied yet. In particular, two-dimensional (2D) model is significant when one considers the organic and copper oxide superconductors. Order parameters with spatial structures such as the square lattice and triangular lattice becomes small in larger area than the order parameter oscillating in a single direction. Thus, the states with such 2D lattice structures would gain more spin-polarization energy than the state with the one-dimensional (1D) structures, and have chance to occur at high magnetic fields. Hence, such states may occur in the 2D FFLO superconductors (including quasi-1D systems), since the critical field of the FFLO state remarkably increases at low temperatures.

In this paper, we study 2D FFLO superconductors with cylindrical symmetric Fermi surfaces. We examine s-wave pairing and d-wave pairing, the latter which is a serious candidate of the organic and high-$T_c$ superconductors. Our theory is a straightforward extension of the work by Larkin et al. to the 2D systems, finite temperatures, and anisotropic superconductivity. We consider the magnetic field parallel to the conducting layers, and neglect the orbital pair breaking effect, although weak interlayer interactions are implicitly assumed so that the mean field treatment like BCS theory is justified at low temperatures.

At first, we examine the s-wave pairing. As many authors studied, a gap function of the
generalized form

$$\Delta(r) = \Delta_q e^{i q \cdot r}$$

(1)

has the highest second order transition field. The optimum value of $|q|$ is finite for $T < T^* \approx 0.56 T_c^{(0)}$, where $T_c^{(0)}$ is the zero field transition temperature. For example, the optimum value of $|q|$ is equal to $2h/v_F$ at $T = 0$, where $h = \mu_0 H$ with the electron magnetic moment $\mu_0$. In the present case, because of the cylindrical symmetry of the system, any linear combinations

$$\Delta(r) = \sum_m \Delta_m e^{i q_m \cdot r},$$

(2)

have the same critical field, where $q_m$'s are any vectors of the optimum magnitude $|q_m| = q$ parallel to the conducting layer. However, such degeneracy is removed by the nonlinear terms in the gap equation, apart from the rotation as a whole: a particular kind of linear combinations have a lower free energy than the others, just below the critical field. Here, we have taken a finite number of $q_m$'s in the above, because we are considering periodic solutions. Near the transition point, the gap equation is expanded as

$$\log\left(\frac{T_c}{T}\right) \Delta^*_l = \sum_m [(2 - \delta_{lm}) J(\theta_{lm}) \Delta^*_m \Delta_m \Delta^*_l$$

$$+ (1 - \delta_{lm} - \delta_{l,-m}) \tilde{J}(\theta_{lm}) \Delta^*_m \Delta_m \Delta^*_l],$$

(3)

where $\theta_{lm}$ is the angle between $q_l$ and $q_m$, and

$$J(\theta_{lm}) = T \sum_n \int_{-\infty}^{\infty} d\xi \int_0^{2\pi} \frac{d\phi}{2\pi} \left(\frac{i \omega_n + \xi + h}{2}\right)^{-2}$$

$$\times \left(i \omega_n - \xi - v_F \cdot q_m + h\right)^{-1}$$

$$\times \left(i \omega_n - \xi - v_F \cdot q_l + h\right)^{-1}$$

(4)

and

$$\tilde{J}(\theta_{lm}) = T \sum_n \int_{-\infty}^{\infty} d\xi \int_0^{2\pi} \frac{d\phi}{2\pi} \left(\frac{i \omega_n + \xi + h}{2}\right)^{-1}$$

$$\times \left(i \omega_n - \xi + h - v_F \cdot q_l\right)^{-1}$$

$$\times \left(i \omega_n + \xi + h - v_F \cdot q_l + v_F \cdot q_m\right)^{-1}$$

$$\times \left(i \omega_n - \xi + h - v_F \cdot q_m\right)^{-1}.$$ 

(5)
We examine periodic solutions

(a) \( \Delta(r) = \Delta_{\text{FF}} \exp(iq \cdot r) \)

(b) \( \Delta(r) = 2\Delta_{\text{FFLO}} \cos(q \cdot r) \)

(c) \( \Delta(r) = 2\Delta_{\text{sq}}[\cos(qx) + \cos(qy)] \)

(d) \( \Delta(r) = \Delta_{\text{tri}}[\exp(iq_1 \cdot r) + \exp(iq_2 \cdot r) \]
\+ \exp(iq_3 \cdot r)] \)

(e) \( \Delta(r) = 2\Delta_{\text{hexa}}[\cos(q_1 \cdot r) + \cos(q_2 \cdot r) \]
\+ \cos(q_3 \cdot r)] \)

where \( q_1 = q(1, 0), q_2 = q(-1/2, \sqrt{3}/2), \) and \( q_3 = q(-1/2, -\sqrt{3}/2) \). We refer to the states expressed by (a), (b), (c), (d), and (e) as FF state, traditional FFLO state, square state, triangular state, and hexagonal state, respectively.

We define the factor \( a_\alpha \)

\[ |\Delta_\alpha|^2 = a_\alpha \frac{T_c - T}{T_c}, \]  

(7)

where \( \alpha \) indicates the type of state. For example, \( \alpha \) is replaced with FF, FFLO, sq, \cdots, for the states (a), (b), (c), \cdots, respectively. The free energy per unit volume is calculated by the formula

\[ \Omega - \Omega_0 = -\frac{1}{2} N(0) \frac{T_c - T}{T_c} \frac{1}{V} \int d^3r |\Delta(r)|^2. \]  

(8)

We define the factor \( b_\alpha \) by

\[ \Omega - \Omega_0 = -\frac{1}{2} N(0) b_\alpha \left[ \frac{T_c - T}{T_c} \right]^2. \]  

(9)
From the gap equation, we have

\[
\begin{align*}
\beta_{FF} &= 1/J(0) \\
\beta_{FFLO} &= 2/[J(0) + 2J(\pi)] \\
\beta_{sq} &= 4/[J(0) + 2J(\pi) + 4J(\pi/2) + 2\tilde{J}(\pi/2)] \\
\beta_{tri} &= 3/[J(0) + 4J(2\pi/3)] \\
\beta_{hexa} &= 6/[J(0) + 2J(\pi) + 4J(\pi/3) + 4J(2\pi/3) + 2\tilde{J}(\pi/3) + 2\tilde{J}(2\pi/3)].
\end{align*}
\] (10)

In the limit of \( T \to 0 \), we have \( J(0) = -1/[4h^2(1-\tilde{q}^2)^{3/2}] \) for \( \tilde{q} < 1 \), and \( J(0) = 0 \) for \( \tilde{q} > 1 \), where \( \tilde{q} = v_F q/(2\hbar) \). Since the optimum value of \( \tilde{q} \) is equal to 1 at \( T = 0 \), the expansion factor \( J(0) \) diverges. Thus, the gap equation can not be expanded in the power of \( \Delta \) in two-dimensions at \( T = 0 \). It is easily verified that there is a term proportional to \( \Delta \log \Delta \) in the asymptotic form.

For finite temperatures, we estimate \( \beta_\alpha \) numerically. The results are shown in Fig.1. It is found that the solution of the form (b), i.e., the traditional FFLO state, is optimum for \( 0.24T_c^{(0)} \lesssim T < T^* \approx 0.56T_c^{(0)} \). Below \( T \approx 0.24T_c^{(0)} \), however, other solutions have lower free energies. The triangular, square, hexagonal states are optimum for \( 0.16T_c^{(0)} \lesssim T \lesssim 0.24T_c^{(0)} \), \( 0.05T_c^{(0)} \lesssim T \lesssim 0.16T_c^{(0)} \), and \( T \lesssim 0.05T_c^{(0)} \), respectively. This result is plausible because when the temperature decreases and the magnetic field increases, node of the orderparameter in the real space becomes more favorable owing to gain in the spin-polarization energy. Our result is consistent with the result by Buzdin et al., in which the structure of the orderparameter is studied near the tricritical point.34

It is easily verified that the orderparameters of the triangular and hexagonal states have spatial structures of the triangular lattice, while that of the square state have a structure of the square lattice. The periodicity of the triangular lattice of \( |\Delta(r)| \) in the hexagonal state is twice that in the triangular state.
It is easy to extend the theory to anisotropic superconductivities. The gap function has the form

$$\Delta(\hat{p}, r) = \sum_m \Delta_m \gamma(\hat{p}) e^{i q_m \cdot r}$$

(11)

where $$\gamma(\hat{p}) = \sqrt{2}(\hat{p}_x^2 - \hat{p}_y^2)$$ for d-wave pairing. As Maki et al. studied, the angle between the optimum vector $$\mathbf{q}$$ and the x-axis is equal to $$n\pi/2$$ for low temperatures $$T/T_c(0) \lesssim 0.06$$, while $$\pi/4 + n\pi/2$$ for high temperatures $$0.06 \lesssim T/T_c(0) \lesssim 0.56$$, where $$n$$ is integer. Since there are four equivalent optimum directions of $$\mathbf{q}$$, we have three candidates to examine, i.e., (a) the FF state, (b) the traditional FFLO state, and (c) the square state.

In this case, a factor $$[\gamma(\hat{p})]^4$$ is introduced into the integrands of eq.(4) and eq.(5). Free energies of the above three states are compared in Fig.2 and Fig.3. Figure 2 shows the results for the low temperature phase, which is realized for $$T/T_c(0) \lesssim 0.06$$, while Fig.3 shows the results for the high temperature phase, which is realized for $$0.06 \lesssim T/T_c(0) \lesssim 0.56$$. In Fig.2, we find that the square state is optimum for $$T/T_c(0) \lesssim 0.06$$. In Fig.3, it is found that the square state is optimum for $$0.085 \lesssim T/T_c(0) \lesssim 0.12$$, while the traditional FFLO state is optimum for $$0.12 \lesssim T/T_c(0) \lesssim 0.56$$. At $$T/T_c(0) \approx 0.085$$, the factors $$a_{sq}$$ and $$b_{sq}$$ turn negative. Thus, the gap equation expanded up to the third order does not have any solution of this type for $$0.06 \lesssim T/T_c(0) \lesssim 0.085$$ below and near the transition temperatures. This suggests that a first order transition to the square FFLO state occurs at a field higher than the second order transition field, in this temperature region, as discussed below.

If the gap equation is simply derived from a differentiation of a free energy functional expanded in the power of $$|\Delta_{sq}|$$, the factor of $$[\Delta_{sq}]^4$$ in the free energy is negative near the second order transition point in this temperature region, since it is proportional to the inverse of $$a_{sq}$$ and that of $$b_{sq}$$. Thus, the free energy as a function of $$|\Delta_{sq}|$$ has a minimum at nonzero $$|\Delta_{sq}|$$ at the second order transition point, because there must be higher order terms such as a one proportional to $$|\Delta_{sq}|^6$$, which guarantee that the free energy increases as a function of $$|\Delta_{sq}|$$ where $$|\Delta_{sq}|$$ is large. This finite value of $$|\Delta_{sq}|$$ is the optimum solution of the gap equation at this point, and this solution must terminate at a higher field. Therefore,
it is plausible that the actual critical field is higher than the second order transition field, and the phase transition is of the first order.

A similar conjecture as the above holds for three-dimensional systems. As Larkin et al. suggested a possibility of the first order transition to the cubic state, the same possibilities for the triangular state and the square state can not be excluded, because the factors $a_\alpha$ for these states are negative in the spherical symmetric case.

Lastly, we briefly discuss orbital pair breaking effect in three-dimensional FFLO superconductors (including quasi-2D systems). For the 1D solution of the traditional FFLO state, since the vector $\mathbf{q}$ can be oriented in the direction of the magnetic field, the FFLO state can coexist with the vortex state if orbital effect is sufficiently weak. On the other hand, for the present 2D solutions, the competition with the vortex states may be a crucial problem, since the vortex functions take two out of three degrees of freedom of the coordinate space. Lebed argued that if the magnetic field is sufficiently strong, the quasi-2D system can be treated as being essentially purely 2D. In such a case, the competition mentioned above does not occur if the magnetic field is accurately parallel to the conducting plane.

In conclusion, we have examined FFLO superconductors with the cylindrical symmetric Fermi surfaces near the second order transition line, and have found that spatial structures of the 2D lattices are favored more than the 1D structure of the traditional FFLO state, for low temperatures and high magnetic fields, both for the $s$-wave pairing and the $d$-wave pairing. It is found that the traditional FFLO state, the triangular state, the square state, and the hexagonal state appear for the $s$-wave pairing as the temperature decreases, while the square states occur at low temperatures for the $d$-wave pairing. If such structures of order parameters are observed experimentally, for example, directly by the scanning tunneling microscope (STM) technique, it can be regarded as a strong evidence of existence of a nonuniform superconducting state.
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FIGURES

Fig. 1. Factors $b_\alpha$ in the free energies of the state (a)-(e) for the $s$-wave pairing.

Fig. 2. Factors $b_\alpha$ in the free energies of the state (a)-(c) for the $d$-wave pairing, when the angle between $\mathbf{q}$ and $x$-axis is equal to $n\pi/2$.

Fig. 3. Factors $b_\alpha$ in the free energies of the state (a)-(c) for the $d$-wave pairing, when the angle between $\mathbf{q}$ and $x$-axis is equal to $\pi/4 + n\pi/2$. 

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Fig. 2
Fig. 3
Fig. 1

Graph showing the transition temperature $T/T_{c}^{(0)}$ vs. the parameter $b_{a}$ with different phases: square, FFLO, triangular, hexagonal, FF.