Fulde-Ferrell-Larkin-Ovchinnikov State in the Absence of a Magnetic Field

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We propose that in a system with pocket Fermi surfaces, a pairing state with a finite total momentum \( q_{\text{tot}} \) like the Fulde-Ferrell-Larkin-Ovchinnikov state can be stabilized even without a magnetic field. When a pair is composed of electrons on a pocket Fermi surface whose center is not located at \( \Gamma \) point, the pair inevitably has finite \( q_{\text{tot}} \). To investigate this possibility, we consider a two-orbital model on a square lattice that can realize pocket Fermi surfaces and we apply fluctuation exchange approximation. Then, by changing the electron number \( n \) per site, we indeed find that such superconducting states with finite \( q_{\text{tot}} \) are stabilized when the system has pocket Fermi surfaces.

KEYWORDS: FFLO state, pocket Fermi surfaces, orbital degree of freedom, fluctuation exchange approximation

The pairing states of superconductivity are classified on the basis of symmetries that satisfy the Pauli principle.\(^1\) Among the pairing states, the \( s \)-wave state is the simplest and is called conventional superconductivity. The other superconducting states are anisotropic (unconventional) ones. We can categorize the superconducting states into spin-singlet and spin-triplet states. The spin-singlet states have even parity, e.g., \( s \)-wave, under the inversion of space due to the Pauli principle. The spin-triplet states have odd parity, and in these states, the spin degree of freedom is active and many phases can occur in a system.

If we can introduce an additional degree of freedom into a system, we can find exotic superconducting states that are beyond the ordinary categorization. For this purpose, the superconductivity in systems with an orbital degree of freedom, which is not considered in the above discussion on superconducting symmetry, has been studied recently. For example, the effects of the orbital degree of freedom on superconductivity have been studied for a multi-orbital system, it describes a system with a rather high orbital symmetry, that is, the Fermi surfaces of these orbitals are exactly the same. To discuss more realistic situations, we should improve the two-orbital Hubbard model. In particular, we should include the effects of orbital anisotropy, for example, orbital-dependent hopping integrals and the transformation property of orbitals. These properties are important for magnetism in \( d \)-electron systems\(^6\) and \( f \)-electron systems\(^7\) and they should also be important for superconductivity. Among the above-mentioned properties, the multi-Fermi-surface nature due to the orbital-dependent hopping is important, since a pair on a Fermi surface can interact with a pair on another Fermi surface, which may provide a new pairing mechanism.

Among possible multi-Fermi-surface cases, a system with pocket Fermi surfaces, as shown in Fig. 1, is interesting for superconductivity. In such a system, a pair on a Fermi surface has a finite total momentum \( q_{\text{tot}} \) like the Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state\(^{10,11}\) even without a magnetic field. When the electrons comprised in this pair are scattered to another Fermi surface by fluctuations such as spin fluctuations and can form a pair on that Fermi surface, such a pairing state with finite \( q_{\text{tot}} \) can be stabilized.

In this Letter, in order to explicitly discuss the possibility of such exotic superconductivity, we consider an \( e_g \) orbital model on a square lattice as an example. This model can easily realize pocket Fermi surfaces even if we consider only nearest-neighbor hopping. To investigate the possible superconducting states in this model, we apply FLEX approximation that has been extended to multi-orbital models\(^5,12-16\) Then, we show that such pairing states with a finite total momentum like the FFLO state are stabilized in a system with pocket Fermi surfaces.

![Pocket Fermi surfaces. A pair with a total momentum \( q_{\text{tot}} \) is composed of electrons with \( k \) and \(-k + q_{\text{tot}}\) which are on the same Fermi surface. This pair can be scattered to another Fermi surface.](attachment:fig1.png)
We consider a tight-binding model for $c_σ$ orbitals given by
\begin{equation}
H = \sum_{k,\tau,\tau',\sigma} \epsilon_{k\tau\sigma} c_{k\tau\sigma}^\dagger c_{k\tau'\sigma} + U \sum_{i,\tau} n_{i\tau\uparrow} n_{i\tau\downarrow} + U' \sum_{i,\sigma,\sigma'} n_{i\sigma\uparrow} n_{i\sigma'\downarrow} + J \sum_{i,\sigma,\sigma'} c_{i\sigma\uparrow}^\dagger c_{i\sigma'\downarrow} c_{i\sigma'\uparrow} c_{i\sigma\downarrow} + J' \sum_{i,\sigma,\sigma'} c_{i\sigma\uparrow}^\dagger c_{i\sigma'\downarrow} c_{i\sigma'\uparrow} c_{i\sigma\downarrow},
\end{equation}
where $c_{i\sigma}$ is the annihilation operator of the electron at site $i$ with orbital $\sigma$ ($= \uparrow$ or $\downarrow$), $\epsilon_{k\tau\sigma}$ is its Fourier transform, $n_{i\tau\sigma} = c_{i\tau\sigma}^\dagger c_{i\tau\sigma}$, and $n_{i\tau} = \sum_\sigma n_{i\tau\sigma}$. The coupling constants $U$, $U'$, $J$, and $J'$ denote the intra-orbital Coulomb, inter-orbital Coulomb, exchange, and pair-hopping interactions, respectively. In this study, we use the relations $U = U' + J + J'$ and $J = J'$.

We consider the hopping integrals for the nearest-neighbor sites on a square lattice, and the coefficients of the kinetic energy terms in Eq. (1) are generally given by
\begin{align*}
\epsilon_{k11} &= \frac{1}{2} \left[ 3(ddd) + (dd\delta) \right](\cos k_x + \cos k_y), \\
\epsilon_{k22} &= \frac{1}{2} \left[ (ddd) + 3(dd\delta) \right](\cos k_x + \cos k_y), \quad \text{and} \\
\epsilon_{k12} &= \epsilon_{k21} = -\sqrt{3} \left[ (ddd) - (dd\delta) \right](\cos k_x - \cos k_y),
\end{align*}
where $(ddd)$ and $(dd\delta)$ are Slater-Koster integrals, and the lattice constant is set to unity.\(^{18}\)

To realize pocket Fermi surfaces in a system such as that shown in Fig. 1, we set $(ddd) = -(dd\delta) = \frac{1}{2} \epsilon_{\text{lat}}$. Figure 2 shows the Fermi surfaces for several electron numbers $n$ per site. Due to the electron-hole symmetry, it is enough to consider $0 \leq n \leq 2$. For $n = 2$, the Fermi surfaces disappear in this model. Pocket Fermi surfaces are realized for $n \geq 1.25$. In this system, if an electron with $-k$ is on a Fermi surface, the electron with $-k + Q$ is on another Fermi surface where $Q = (\pi, \pi)$, as shown in Fig. 2 for $n = 1.75$ as an example. Then, it is possible to form a pair with the total momentum $q_{\text{tot}} = Q$ by electrons with $k$ and $-k + Q$. Thus, in this study, we consider superconducting states with $q_{\text{tot}} = Q$ in addition to the ordinary superconducting states with $q_{\text{tot}} = (0, 0)$.

The Green’s function for the present two-orbital model is expressed by a $2 \times 2$ matrix. In the normal phase, the Dyson-Gorkov equation for the Green’s function in matrix form is given by
\begin{equation}
G(k) = G^{(0)}(k) + G^{(0)}(k)\Sigma(k)G(k),
\end{equation}
where $k = (k, \epsilon_n)$ and $\epsilon_n = (2n + 1)\pi T$ is the Matsubara frequency for fermions with an integer $n$ and a temperature $T$. The non-interacting Green’s function is given by $G^{(0)}(k, \epsilon_n) = |\epsilon_n - \epsilon_k + \mu|^{-1}$, where $\mu$ is the chemical potential. In the FLEX approximation, the self-energy is given by
\begin{equation}
\Sigma_{\tau\tau'}(k) = \frac{T}{N} \sum_{q,\tau_1,\tau_2} V_{\tau_1\tau_2}(q) G_{\tau_2\tau}(k - q),
\end{equation}
where $N$ is the number of lattice sites, $q = (q, \omega_n)$, and $\omega_n = 2n\pi T$ is the Matsubara frequency for bosons with an integer $m$. The matrix $V(q)$ is written as
\begin{equation}
V(q) = \frac{3}{2} [U^s \chi^s(q) U^s - U^s \chi^\tau(q) U^\tau / 2 + U^\tau] + \frac{1}{2} [U^c \chi^s(q) U^c - U^c \chi^\tau(q) U^\tau / 2 - U^\tau],
\end{equation}
where the matrix elements of $U^s$ and $U^c$ are given by $U_{11;11}^s = U_{22;22}^s = U_{11;11}^c = U_{22;22}^c = U$, $U_{11;12}^s = U_{22;12}^s = U_{12;11}^s = U_{21;21}^s = U$, $U_{11;12}^c - U_{12;11}^c = U_{21;21}^c = U_{21;12}^c = U_{22;12}^c = U_{12;21}^c = U$; the other matrix elements are zero. The susceptibilities $\chi^s(q)$ and $\chi^\tau(q)$ are given by $\chi^s(q) = \chi^\tau(q)[1 - U^s \chi^\tau(q)^{-1}]$ and $\chi^\tau(q) = \chi^\tau(q)[1 - U^c \chi^\tau(q)^{-1}]$, respectively, in matrix forms. The matrix elements of $\chi^\tau(q)$ are defined by $\chi_{\tau_1\tau_2}(q) = -\frac{1}{N} \sum k G_{\tau_1\tau_2}(k + q) G_{\tau_2\tau_1}(k)$. We solve Eqs. (2)–(4) self-consistently. Then, we can calculate response functions; for example, the spin susceptibility in the FLEX approximation is given by $\chi = 2 \Sigma_{\tau\tau'} \chi_{\tau\tau'}(q)$.

The linearized gap equation for the anomalous self-energy $\phi_{\tau\tau'}(k; q_{\text{tot}})$ is given by
\begin{equation}
\phi_{\tau\tau'}(k; q_{\text{tot}}) = -\frac{T}{N} \sum_{k',\tau_1,\tau_2,\tau_3,\tau_4} V^S_{\tau_1\tau_2}(k - k') \times G_{\tau_2\tau_3}(k') \phi_{\tau_3\tau_4}(k'; q_{\text{tot}}) G_{\tau_4\tau_1}(k' - k + q_{\text{tot}} - i\epsilon_n),
\end{equation}
where $\epsilon_n$ is the annihilation operator of the electron
\begin{equation}
V^\text{singlet}(q) = [3V^\tau(q) - V^\sigma(q)] / 2,
\end{equation}

\begin{figure}
\centering
\includegraphics{fig2}
\caption{Fermi surfaces of the model for $(dd\delta) = -(dd\delta)$ for several electron numbers $n$ per site.}
\end{figure}
\[ V^{\text{triplet}}(q) = \frac{[-V^s(q) - V^c(q)]}{2}, \]

where \( V^s(q) = [U_s^\pi \chi^s(q) U^s_s + U^s_s/2] \) and \( V^c(q) = [U^c \chi^c(q) U^c - U^c/2] \).

Before presenting the calculation results, we discuss plausible candidates for the pairing symmetry from Eqs. (5)–(7). First, in this discussion, we ignore the orbital indices in the above equations for simplicity. In this model, we find that only the spin susceptibility becomes large. Then, we expect an anisotropic superconducting state for the spin-singlet channel, since \( \chi^{\text{singlet}}(q) \) becomes large and has a positive value and the anomalous self-energy has to change its sign in the \( k \) space, as understood from Eq. (5). For the spin-triplet channel, an \( s \)-wave state is favorable, since \( V^{\text{triplet}}(q) \) becomes negative and the anomalous self-energy does not have to change its sign. The above consideration is appropriate for orbital-parallel components, i.e., \( \phi_{11}(k; \mathbf{q}_{\text{tot}}) \) and \( \phi_{22}(k; \mathbf{q}_{\text{tot}}) \), as long as the Pauli principle is satisfied. On the other hand, for orbital-antiparallel components \( \phi_{12}(k; \mathbf{q}_{\text{tot}}) \) and \( \phi_{21}(k; \mathbf{q}_{\text{tot}}) \), orbital indices transform following the \( d_{x^2-y^2} \) symmetry under symmetry operations of the lattice, and thus, the anomalous self-energy belonging to the \( d_{x^2-y^2} \) symmetry does not have to change its sign in the \( k \) space. In other words, the symmetry in the \( k \) space of a \( d_{x^2-y^2} \)-wave state for the orbital-antiparallel components is the same as that of an \( s \)-wave state for the orbital-parallel components. Thus, spin-triplet \( d_{x^2-y^2} \)-wave states and spin-singlet states other than those with the \( d_{x^2-y^2} \) symmetry are favorable for the orbital-antiparallel components. Here, we note that orbital states mix in general. However, it is expected that when the Hund’s rule coupling \( J \) is large and the inter-orbital Coulomb interaction \( U’ \) is small, the orbital-antiparallel components become dominant.

Here, we show the results for a \( 64 \times 64 \) lattice. In this study, we fix the value of the intra-orbital Coulomb interaction \( U = 6t \) and vary \( J (= J’ \) ). Then, the inter-orbital Coulomb interaction is given by \( U’ = U - 2J \). In Fig. 3(a), we show the \( n \) dependence of the static spin susceptibility \( \chi(q) = \chi(q; \omega_n = 0) \) at \( q = q_{\text{max}} \) for \( J = 0, t, \) and \( 2t \) at \( T = 0.005t \), where \( q_{\text{max}} \) is defined as the wave vector at which \( \chi(q) \) becomes the largest. For comparison purposes, we also show \( \chi(q_{\text{max}}) \) for the non-interacting system. The spin susceptibility is enhanced by the interactions, in particular, by the Hund’s rule coupling. On the other hand, \( q_{\text{max}} \) does not depend so much on the interactions as shown in Fig. 3(b). This fact indicates that the characteristic wave vector \( q_{\text{max}} \) is almost determined by the property of the non-interacting system, i.e., the Fermi-surface structure.

As shown in Fig. 3(b), the characteristic wave vector \( q_{\text{max}} \) takes various values depending on \( n \) around \( n = 0.7 - 1.5 \), since the Fermi surface structure changes around this area, as shown in Fig. 2. e.g., the Fermi surfaces consist of electron sheets for \( n = 1 \), while they consist of hole pockets for \( n = 1.25 \). Thus, several fluctuations exist in this region, and a \( d_{x^2-y^2} \)-wave spin-triplet state with dominant orbital-antiparallel components may appear if such fluctuations in a wide \( q \) region are available. However, around \( n = 0.7 - 1.1 \), the spin fluctuations are very strong for \( J = 2t \) and thus magnetic order should occur. For \( n \geq 1.5 \), we expect the appearance of a \( p \)-wave spin-singlet state because of the following reason. For the purpose of explanation, we show the Fermi surfaces for \( n = 1.75 \) in Fig. 4. The characteristic wave vector \( q_{\text{max}} \) connects electrons at \( k = q_{\text{max}}/2 \) and \( -k \). Thus, electrons around \( k \) and \(-k + Q \) comprising a pair on a Fermi surface can be scattered to another Fermi surface by the spin fluctuations and can comprise a pair with momenta around \(-k \) and \( k + Q \). These pairs are the space inversions of each other, and to utilize such fluctuations, the \( p \)-wave spin-singlet state is favorable since the anomalous self-energy changes its sign under the in-
conductivity, we cannot find any superconducting phase creasing the Hund’s rule coupling. With regard to super- the orbital-parallel components should be odd in fre- antiparallel components because of the Pauli principle. The orbital-parallel components should be odd in fre- nger states allowed in the model and for antiferromagnetic states as functions of $n$ for $J = t$ and $2t$. Figure 5(b) shows the highest transition temperatures for $J = 2t$ around $n = 1.6$. The antiferromagnetic transition temperatures are determined by $\chi(q_{\text{max}}) = 70/t$, and the system is always in the normal phase for $J = 0$ at $T \geq 0.005t$. The antiferromagnetic phase extends by increasing the Hund’s rule coupling. With regard to superconductivity, we cannot find any superconducting phase within $T \geq 0.005t$ for $J = t$. For $J = 2t$, a $p$-wave spin-singlet state with $q_{\text{tot}} = Q$ appears, as is expected from $\chi(0) \sim 1.75$ where the antiferromagnetic transition temperature tends to zero. The $d_{x^2-y^2}$-wave spin-triplet state with $q_{\text{tot}} = Q$ appears in a wider region $n \geq 1.3-1.7$, since the $d_{x^2-y^2}$-wave spin-triplet state is insensitive to the value of the characteristic wave vector $q_{\text{max}}$, similar to the $s$-wave state in the two-orbital Hubbard model. It should be noted that these superconducting states with finite $q_{\text{tot}}$ appear only in the region where the system has pocket Fermi surfaces.

To summarize, we have pointed out that a pairing state with a finite total momentum like the FFLO state is possible for a system with pocket Fermi surfaces. As an example, we have studied a model for $e_g$ orbitals by applying FLEX approximation. Then, we have found that the $p$-wave spin-singlet and $d_{x^2-y^2}$-wave spin-triplet states with $q_{\text{tot}} = Q = (\pi, \pi)$ are realized at electron number $n$ where the system has pocket Fermi surfaces. The mechanism to stabilize the superconducting states with finite $q_{\text{tot}}$ can be applied to systems other than the $e_g$ orbital system as long as such pocket Fermi surfaces exist.

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Fig. 5. (a) Transition temperatures for $U = 6t$. The solid triangles and squares denote antiferromagnetic (AF) transition tem-Fig. 4, at $n \approx 1.75$ where the antiferromagnetic transition poeratures for $J = t$ and $2t$, respectively. The open circles and triangles denote superconducting transition temperatures for the $d_{x^2-y^2}$-wave spin-triplet state and $p$-wave spin-singlet state, respectively, for $J = 2t$. In both superconducting states, Cooper pairs have the total momentum $Q = (\pi, \pi)$. (b) Transition temperatures around $n = 1.6$ for $J = 2t$. peratures are determined by $\chi$.

version $k \rightarrow -k$. These $d_{x^2-y^2}$-wave spin-triplet and $p$-wave spin-singlet states should be dominated by orbital-antiparallel components because of the Pauli principle. The orbital-parallel components should be odd in fre- Figure 5(a) shows the highest transition temperatures among transition temperatures for all superconducting states allowed in the model and for antiferromagnetic states as functions of $n$ for $J = t$ and $2t$. Figure 5(b) shows the highest transition temperatures for $J = 2t$ around $n = 1.6$. The antiferromagnetic transition temperatures are determined by $\chi(q_{\text{max}}) = 70/t$, and the system is always in the normal phase for $J = 0$ at $T \geq 0.005t$. The antiferromagnetic phase extends by increasing the Hund’s rule coupling. With regard to superconductivity, we cannot find any superconducting phase within $T \geq 0.005t$ for $J = t$. For $J = 2t$, a $p$-wave spin-singlet state with $q_{\text{tot}} = Q$ appears, as is expected from

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