Multi-Orbital Fulde-Ferrell-Larkin-Ovchinnikov State in SrTiO$_3$ Heterostructures

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We study the exotic superconducting states in SrTiO$_3$ heterostructures on the basis of the three-orbital model reproducing the band structure of two-dimensional electron gases. We show various Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) states induced by the broken inversion symmetry and the orbital degree of freedom. In particular, a novel orbital-dependent FFLO state is stabilized in the magnetic field along the [110]-axis. The field angle dependence of the FFLO state is clarified on the basis of the spin and orbital texture in the momentum space. It is shown that the in-plane anisotropy of the critical magnetic field is an indication of the orbital degree of freedom in Cooper pairs. The carrier density dependence of the superconducting state is also discussed.

1. Introduction

Since the discovery of two-dimensional conducting electron gases at the interface between the two band insulator perovskite oxides SrTiO$_3$/LaAlO$_3$, quantum phases in SrTiO$_3$ heterostructures have been explored extensively. The superconductivity has been found in the SrTiO$_3$/LaAlO$_3$ interface,$^2$ SrTiO$_3$ surface induced by the electric double layer transistor (EDLT),$^3$ SrTiO$_3$/LaTiO$_3$ interface,$^4$ and δ-doped SrTiO$_3$. Interestingly, these superconducting states are artificially tuned by a gate voltage.$^{3,6-10}$ The EDLT technique also realized the electric-field-induced superconductivity in KTaO$_3$, MoS$_2$, ZrNCl, and La$_{1-x}$Sr$_x$CuO$_4$. In this paper, we propose exotic superconducting states appearing in the SrTiO$_3$ heterostructures.

Superconductivity induced by the condensate of Cooper pairs having a finite center of mass momentum was proposed by Fulde and Ferrell$^{11}$ and by Larkin and Ovchinnikov$^{12}$ five decades ago,$^{13}$ although the standard BCS theory assumes zero center of mass momentum in Cooper pairs.$^{14}$ Experimental searches of the Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state found the heavy fermion superconductor CeCoIn$_5$ and organic superconductors$^{17-19}$ to be promising candidates. The FFLO state in the population imbalanced cold fermion gases$^{20}$ and in the nuclear matter$^{21}$ have also attracted interests.

For the FFLO state to be stabilized rather than the BCS state, the spin polarization must be caused by something breaking the time-reversal symmetry, such as the applied magnetic field and proximity to the ferromagnet.$^{11-13}$ The magnetic field is most easily achieved, but it simultaneously leads to the orbital depairing effect and often destabilizes the FFLO state.$^{22}$ In the above candidates for the FFLO superconductor the heavy and anisotropic effective mass of quasiparticles suppresses the orbital depairing effect and may stabilize the FFLO state. When the magnetic field is applied parallel to the conducting plane of two-dimensional electron gases, the orbital depairing effect is completely suppressed. Therefore, the FFLO state is expected to appear in the SrTiO$_3$ heterostructures.

The artificial tuning of the superconducting state with use of the gate voltage$^{3,6-10}$ may enable a novel FFLO state to emerge. For instance, the lack of the space inversion symmetry on the surface/interface allows an FFLO state beyond the paradigm of Fulde and Ferrell and of Larkin and Ovchinnikov. The Fulde-Ferrell (FF) state is the single-$Q$ condensate represented by the order parameter $\Delta(r) = \Delta e^{iqr}$ in the real space,$^{11}$ while the Larkin-Ovchinnikov (LO) state is the double-$Q$ state where $\Delta(r) = \Delta (e^{iqr} + e^{-iqr})/2 = \Delta \cos(qr)$.$^{12}$ It has been theoretically proved that the LO state rather than the FF state is thermodynamically stable. Hence, the LO state would have been realized in the above-mentioned heavy fermion and organic superconductors.$^{13}$ On the other hand, recent studies on the superconductivity lacking the inversion symmetry, that is called noncentrosymmetric superconductivity,$^{23}$ elucidated the “helical superconducting state” similar to the FF state.$^{24-29}$ Owing to the antisymmetric spin-orbit coupling appearing in the noncentrosymmetric crystals,$^{23}$ the helical state is stabilized in the low magnetic field region above $H_{c1}$ in contrast to the fact that the LO state needs a high magnetic field close to the Pauli limit to be stabilized. Ref. 28 also showed that the intermediate state between the FF and LO states, that we call “complex stripe (CS) state”, emerges from the helical state. Because the helical superconducting state is robust against the spin polarization, the coexistence with the ferromagnetic order in the SrTiO$_3$/LaAlO$_3$ interface has been discussed.$^{30}$ Indeed, the ferromagnetic order has been observed in the SrTiO$_3$/LaAlO$_3$ interface.$^{31-35}$ Although these previous theories are based on the single-band models, it has been shown that the orbital degree of freedom in $t_{2g}$ electrons plays an important role on the magnetic response of the superconductivity in the SrTiO$_3$ heterostructures.$^{36}$ Thus, a further novel FFLO state may be induced by the cooperation between the broken inversion symmetry and the orbital degree of freedom.

In this paper, we investigate the FFLO superconducting state in the SrTiO$_3$ heterostructures on the basis of the three-orbital model reproducing the band structure of two-dimensional electron gases, and examine the roles of the orbital degree of freedom and spin-orbit coupling. It is shown that the orbital degeneracy in $t_{2g}$-orbitals on the Ti ions remarkably affects the FFLO state. In particular, we obtain the following results for the multi-orbital FFLO state; (1) a rich
phase diagram involving the orbital-dependent complex stripe state, which has not been uncovered for the single-band models and (2) highly anisotropic behaviors of superconducting state with respect to the in-plane rotation of the magnetic field. We also investigate the evolution of the FFLO state by increasing the carrier density, which can be controlled by the gate voltage.3–6,10

In Sect. 2, we introduce the three-orbital tight-binding model for SrTiO heterostructures, and explain the linearized gap equation by which the superconducting instability is investigated. In Sect. 3.1, the in-plane anisotropy of the critical magnetic field is shown and attributed to the spin texture affected by the orbital degeneracy. The FFLO states in the magnetic field parallel to the [100]-axis and [110]-axis are studied in Sect. 3.2 and Sect. 3.3, respectively. The in-plane angle dependence of the superconducting state is illustrated in Sect. 3.4. The carrier density dependence of the FFLO states is discussed in Sect. 4. Finally, we summarize the results and propose experimental searches of the various FFLO states in the SrTiO heterostructures in Sect. 5.

2. Formulation

2.1 Three-orbital model for SrTiO heterostructures

We adopt a three-orbital tight-binding model for 2g orbitals on Ti ions. As shown by the experiments and electronic structure calculations, the conduction bands in SrTiO heterostructures mainly consist of the 2g orbitals. Although the electronic structure depends on the interface/surface termination, the band structure of two-dimensional electron gases is described by the one-body part of the Hamiltonian,

$$H_0 = H_{\text{kin}} + H_{\text{hyb}} + H_{\text{CEF}} + H_{\text{odd}} + H_{\text{LS}},$$

$$H_{\text{kin}} = \sum_{k \sigma} \left[ (\epsilon_m(k) - \mu) c_{m \sigma}^\dagger c_{m \sigma} + \frac{1}{2} \delta_{\sigma} \right],$$

$$H_{\text{hyb}} = \sum_{k \sigma} \sum_{s=\uparrow,\downarrow} [V(k) c_{k,1s}^\dagger c_{k,2s} + \text{h.c.}],$$

$$H_{\text{CEF}} = \Delta \sum_i n_{2s},$$

$$H_{\text{odd}} = \sum_{k \sigma} \sum_{s=\uparrow,\downarrow} [V_{s}(k) c_{k,1s}^\dagger c_{k,3s} + V_{s}(k) c_{k,2s}^\dagger c_{k,3s} + \text{h.c.}],$$

$$H_{\text{LS}} = \lambda \sum_i L_i \cdot S_i,$$

where $c_{m \sigma}$ is the annihilation operator for an electron with momentum $k$, orbital $m$, and spin $\sigma$. Here, the $(d_{xz}, d_{zy}, d_{xy})$ orbitals are denoted by the orbital index $m = (1, 2, 3)$, respectively. The first term $H_{\text{kin}}$ describes the kinetic energy of each orbital and includes the chemical potential $\mu$. The second term $H_{\text{hyb}}$ is the intersite hybridization term of $d_{xz}$ and $d_{xy}$ orbitals. The third term $H_{\text{CEF}}$ introduces the crystal electric field with tetragonal symmetry.

Since the mirror symmetry with respect to the conducting plane is broken by the interface/surface, the hybridization is allowed between the $d_{xy}$-orbital and the $(d_{xz}, d_{zy})$-orbitals and represented by the odd-parity hybridization term $H_{\text{odd}}$. Thus, the broken inversion symmetry is introduced in our model. The Rashba spin-orbit coupling appears as an effective spin-orbit coupling arising from the odd-parity hybridization term $H_{\text{odd}}$ and the LS coupling term $H_{\text{LS}}$. Although the Rashba spin-orbit coupling is often assumed phenomenologically in the theoretical models, it is microscopically derived in our model.

The following tight-binding forms are adopted in this paper.

$$\epsilon_1(k) = -2t_1 \cos k_x - 2t_2 \cos k_y,$$

$$\epsilon_2(k) = -2t_2 \cos k_x - 2t_1 \cos k_y,$$

$$\epsilon_3(k) = -2t_1 (\cos k_x + \cos k_y) - 4t_4 \cos k_x \cos k_y,$$

$$V(k) = 4t_5 \sin k_x \sin k_y,$$

$$V_\ell(k) = 2it_{\ell\text{odd}} \sin k_x,$$

$$V_\ell(k) = 2it_{\ell\text{odd}} \sin k_y.$$

The electronic structure of two-dimensional electron gases in the SrTiO/LaAlO interface is reproduced by the parameter set $(t_1, t_2, t_3, t_4, t_5, t_{\ell\text{odd}}, \lambda, \Delta) = (1, 1, 0.2, 0.4, 0.1, 0.1, 0.1, 2.45)$, where the unit of energy is chosen as $t_1 = 1$. The first principles band structure calculation obtained $t_1 \approx 300$ meV. In this paper we mostly investigate the high two-dimensional carrier density $n = 0.15 \approx 1 \times 10^{14}$ cm$^{-2}$ which has been realized in the gate-controlled SrTiO/LaAlO interface and SrTiO surface.3 We show the carrier density dependence of the superconducting state in Sect. 4 and demonstrate unusual properties in the lower carrier density region.

For the study of the superconductivity, we assume attractive interactions in the $s$-wave channel,

$$H_1 = U \sum_i \sum_{m,m'} n_{i,m\uparrow} n_{i,m\downarrow} + U' \sum_{i,m,m'} n_{i,m\uparrow} n_{i,m'\downarrow},$$

where $U$ and $U'$ describe the intraorbital and interorbital attractive interaction, respectively. The $s$-wave symmetry of the superconductivity has been evidenced by both theory and experiment. The measurement of superfluid density showed the full excitation gap in the superconducting state.50 Furthermore, it has been theoretically shown that the transition temperature around 0.3 K and its non-monotonic carrier density dependence in the SrTiO/LaAlO interface are reproduced by the $s$-wave attractive interaction mediated by the optical phonons.51 The renormalization of band structure observed by ARPES measurements has also been explained by taking into the proper electron-phonon coupling.40 Thus, the $s$-wave superconductivity is likely to occur in SrTiO heterostructures as well as in the bulk SrTiO.3 We assume $U = U'$ and choose $U$ so that the transition temperature at zero magnetic field is $T_c = 0.001$. It has been confirmed that the following results are almost independent of $U$ and the ratio $U'/U$ as long as the reasonable condition $U'/U \leq 1$ is satisfied.53 We do not take into account the pairing interaction in the odd-parity channel, and therefore, the parity mixing in Cooper pairs is ignored. This simplification is also justified, because the role of induced odd-parity Cooper pairs on the FFLO superconductivity is negligible unless the amplitude of odd-parity Cooper pairs is comparable to that of even-parity Cooper pairs.54,55 Although the superconductivity induced by the odd-parity Cooper pairs has been theoretically proposed,56,57 we do not consider such unconventional Cooper pairing which may be caused by the strong electron correlation.
The purpose of our study is to elucidate the FFLO superconducting states in the two-dimensional electron gases formed on the SrTiO$_3$ heterostructures. Thus, we consider the Zeeman coupling term which is induced by the magnetic field or by the coexisting ferromagnetic order, \( 30 \)

\[
H_Z = -\sum_k \sum_{m} \sum_{s,s'} \mu_B H \cdot \sigma_{ss'} c^\dagger_{k,ms} c_{k,ms}', \tag{14}
\]

where \( \sigma \) is the Pauli matrix and \( \mu_B \) is the Bohr magneton. When we apply the magnetic field, the destruction of superconductivity occurs through the orbital depairing effect as well as through the spin polarization induced by the Zeeman coupling term (paramagnetic depairing effect). However, the orbital depairing effect is negligible for the magnetic field parallel to the two-dimensional electron gases, although it reduces the critical magnetic field along the [001]-axis to \( H_c^e \sim 0.1 \) T which is much smaller than the Pauli limit. The ferromagnetic moment coexisting with the superconductivity is parallel to the conducting plane in the SrTiO$_3$ interface, and therefore, the proximity effect gives rise to the effective magnetic field \( H \) along the \( ab \)-plane. Thus, we assume the parallel magnetic field in the following part and ignore the orbital depairing effect. The total Hamiltonian is given by \( H = H_0 + H_t + H_Z \).

### 2.2 Linearized gap equation

We determine the instability to the superconducting state by solving the linearized mean field gap equation formulated in the following way. First, we diagonalize the one-body Hamiltonian, \( H_0 + H_Z \), using the unitary matrix

\[
\hat{U}(k) = \begin{pmatrix}
u_{1,1} & \cdots & \nu_{1,6} \\
u_{2,1} & \cdots & \nu_{2,6} \\
\vdots & \ddots & \vdots \\
\nu_{6,1} & \cdots & \nu_{6,6}
\end{pmatrix}, \tag{15}
\]

Thereby, the basis changes as \( c^\dagger_k = \Gamma_k^j U^j(k) \), where \( c^\dagger_k = (c^\dagger_{k,11}, c^\dagger_{k,21}, \cdots, c^\dagger_{k,6}) \) and \( \Gamma_k^j = (\gamma_{1,1}^j, \gamma_{1,2}^j, \cdots, \gamma_{6}^j) \). With use of the operators in the band basis, the one-body Hamiltonian is diagonalized as,

\[
H_0 + H_Z = \sum_k 6 \sum_{j=1}^6 E_j(k) \gamma_{j} \gamma_{j}, \tag{16}
\]

where \( E_j(k) \) is a quasiparticle’s energy and we take \( E_j(k) \geq E_i(k) \) for \( i > j \). The Matsubara Green functions in the spin and orbital basis are obtained as,

\[
G_{m',m}(k,i\omega) = -\int_0^\beta d\tau e^{i\omega \tau} c^\dagger_{k,m',\tau} c_{k,\tau}(0),
\]

\[
= \sum_{j=1}^6 (i\omega_j - E_j(k))^{-1} c_{m',j}(k) c^\dagger_{m,j}(k), \tag{18}
\]

where \( \omega_j \) is the Matsubara frequency.

The linearized gap equation is formulated by looking at the divergence of the T-matrix, which is defined as,

\[
\hat{T}(q) = \hat{T}_0(q) - \hat{T}(q) \hat{H}_t \hat{T}_0(q). \tag{19}
\]

The wave vector \( q \) represents the center of mass momentum of Cooper pairs. The matrix element of the irreducible T-matrix \( \hat{T}_0(q) \) is obtained as

\[
T_0^{(mn,m'n')}(q) = T \sum_{\omega_n} \sum_k \{G_{m',m}(q/2 + k, i\omega_n)G^\dagger_{m',n}(q/2 - k, -i\omega_n) - G_{m,n}(q/2 + k, i\omega_n)G^\dagger_{m',n}(q/2 - k, -i\omega_n)\}, \tag{20}
\]

where \( T \) is the temperature. When we represent the T-matrix using the basis \( \{mn\} = (11, 12, 13, 22, 23, 31, 32, 33) \), the interaction term is represented by the \( 9 \times 9 \) diagonal matrix, \( \hat{H}_t = (U_m\delta_{mn}) \) with \( U_m = U \) for \( m = 1, 5, 9 \) and \( U_m = U' \) for others. The superconducting transition occurs when the maximum eigenvalue \( \lambda_{\text{max}} \) of the matrix, \( -\hat{H}_t \hat{T}_0(q) \), is unity. The order parameter in the orbital basis is obtained by the eigenvector (\( \phi_{mn} \)) which is proportional to \( \delta_{mn} = -g \sum_k (c_{q/2+k,m}^\dagger c_{q/2-k,n}) \) with \( g = U \) for \( m = n \) and \( g = U' \) for \( m \neq n \). Thus, the superconducting state below the transition temperature \( T_c(H) \) is determined by the linearized gap equation, although the full solution of the Bogoliubov-de Gennes equation is required for studies on the superconducting state at low temperatures.

The superconducting transition is induced by the quasi-long-range order in two-dimensional systems and described by the Berezinskii-Kosterlitz-Thouless (BKT) transition.\(^{28, 59}\) Indeed, the critical behaviors of BKT transition have been observed in the SrTiO$_3$/LaAlO$_3$ interface,\(^2\),\(^7\),\(^60\) The BKT transition temperature is roughly obtained by the following equation,\(^59\)

\[
T_{\text{BKT}} = \frac{\pi}{2m^* \rho(T_{\text{BKT}})}, \tag{21}
\]

where \( \rho(T) \) is the superfluid density. Because of the large superfluid density and small transition temperature of SrTiO$_3$ heterostructures, the transition temperature obtained by the mean field theory (\( T_c \)) almost coincides with the BKT transition temperature,\(^2\) as \( 1 - T_{\text{BKT}}/T_c \ll 1 \). Thus, the superconductivity in the SrTiO$_3$ heterostructures is approximately described by the mean field theory, although the long-range order obtained by the mean field theory is interpreted as a quasi-long-range order at finite temperatures.

### 2.3 Classification of FFLO states

For the clarity of discussions, we classify various FFLO states which will appear in the following part. Although the FFLO states have been classified into the single-\( Q \) FF state\(^{11}\) and the double-\( Q \) LO state,\(^12\) the so-called helical state can emerge in the noncentrosymmetric superconductors.\(^24\)\(^-\)\(^29\) The order parameter of helical state is formally the same as that of the FF state, but the magnitude of the center of mass momentum \( |q| \sim (\alpha/E_F) \cdot (\mu_B H/T_c) \cdot \xi^2 \) is much smaller than that in the FF state \( |q| \sim \xi^{-1} \), where \( \xi \) is the coherence length. The helical state is stabilized by the asymmetric deformation of the Fermi surface arising from the antisymmetric spin-orbit coupling and the magnetic field,\(^23\) and therefore, the magnitude \( |q| \) is proportional to \( \alpha \) and \( H \). Thus, the long-period spatial modulation characterizes the helical state. The crossover from the helical state to the FF state occurs by increasing the magnetic field,\(^54\) and we classify these two states by looking at the magnitude of \( q \). Above the crossover field the CS state where \( \Delta(r) = \Delta \left( \epsilon_{qr} + \delta e^{-\epsilon_{qr}} \right) / 2 \neq \Delta \cos(qr) \) may be stabilized in the low temperature region.\(^28\) Below, we will show...
that a novel two-component CS state due to the orbital degree of freedom is stabilized in the SrTiO$_3$ heterostructures (see Sect. 3.3).

3. FFLO superconductivity

As we find that the superconducting state remarkably depends on the direction of magnetic field, we first show the anisotropic paramagnetic depairing effect in Sect. 3.1. Then, we show the FFLO superconducting states for $H \parallel [100]$ and for $H \parallel [110]$ in Sects. 3.2 and 3.3, respectively. The field angle dependence of the superconducting state for $H \parallel (\cos \theta, \sin \theta, 0)$ is discussed in Sect. 3.4.

3.1 In-plane anisotropy of paramagnetic depairing effect

Fig. 1. (Color online) The critical magnetic field of the BCS state (red dashed lines) and that of the FFLO state (black solid lines). Thick and thin lines show the normalized critical magnetic fields $\mu_B H_C/T_C$ along the [100]-axis and along the [110]-axis, respectively. We assume the carrier density $n = 0.15 \sim 1 \times 10^{15}$ cm$^{-2}$ throughout Sect. 3.

First, we show the anisotropy of paramagnetic depairing effect which arises from the orbital degeneracy in the electronic structure. We also study the thermodynamical stability of FFLO superconductivity by comparing the critical magnetic fields of the BCS state and the FFLO state. Figure 1 shows that the critical magnetic field is enhanced by allowing the Cooper pairs to have a finite center of mass momentum. For $H \parallel [100]$, the enhancement is negligible and is much smaller than the effect of canonical FFLO superconductivity on the critical magnetic field. On the other hand, the critical magnetic field along the [110]-axis is pronouncedly enhanced. Since the critical magnetic field for $H \parallel [110]$ is smaller than that for $H \parallel [100]$, a large paramagnetic depairing effect is indicated for the former.

We explain a large anisotropy of the paramagnetic depairing effect by illustrating the spin texture of spin-split bands. The spin degeneracy in the band structure is lifted by the spin-orbit coupling in the noncentrosymmetric metal. The momentum dependent spin polarization is described by the "g-vector" which is defined for the l-th band as:

$$ g_l(k) = (E_{2l}(k) - E_{2l-1}(k)) \bar{S}_{2l}^{yz}(k). \quad (22) $$

The spin polarization axis $\bar{S}_{2l}^{yz}(k) = S_{2l}^{yz}(k)/|S_{2l}^{yz}(k)|$ is obtained by calculating the averaged spin $S_{2l}^{yz}(k) = \langle \sum_{m} \sum_{s'} \sigma_{s's'} \bar{c}_{k,ms}^{\dagger} \bar{c}_{k,ms'} \rangle_{2l}$ for each momentum $k$. The arrows in Fig. 2 show the g-vector of the 1st band whose spin-split Fermi surfaces are drawn by the solid and dashed lines. We see that the g-vector is almost perpendicular to the [100]-axis in the half of the Brillouin zone, $|k_x| > |k_y|$, where the 1st band mainly consists of the $d_{x^2-y^2}$-orbital. As shown in the literature, the Cooper pairs are robust against the paramagnetic depairing effect when the spin polarization axis is perpendicular to the magnetic field. Thus, the quasi-one-dimensional superconducting state mainly induced by the $d_{x^2-y^2}$-orbital substantially avoids the paramagnetic depairing effect for $H \parallel [100]$. Indeed, a large critical magnetic field beyond the Pauli limit has been observed in the SrTiO$_3$/LaAlO$_3$ interface. On the other hand, we see that the spin polarization axis is not perpendicular to the [110]-axis in the most Brillouin zone except for a tiny region near $k_x = k_y$. Therefore, the large paramagnetic depairing effect suppresses the BCS state for $H \parallel [110]$, and it is partly avoided in the FFLO state.

The large anisotropy discussed above is attributed to the orbital degree of freedom in the $t_{2g}$ electron system. Indeed, the spin texture shown in Fig. 2 is typical for the orbitally degenerate noncentrosymmetric metal. When the crystal electric field is large enough so that the orbital degree of freedom is quenched, the often-assumed Rashba-type antisymmetric spin-orbit coupling, $g_l(k) \propto (\sin k_x - \sin k_y, 0)$, is obtained. On the other hand, in orbitally degenerate systems the g-vector dramatically changes the direction near the symmetric axis in the Brillouin zone, as actually shown in Fig. 2. Thus, the characteristic spin texture in the multi-orbital systems causes the anisotropic behaviors of superconductivity against the magnetic field. In the following subsections, we show that the FFLO states in the SrTiO$_3$ heterostructures depend on the in-plane direction of magnetic field. Interestingly, a novel FFLO state is stabilized in the magnetic field along the [110]-axis.

3.2 Non-monotonic evolution of FFLO state in $H \parallel [100]$

The superconducting state shows several crossover and the FFLO state non-monotonically changes as increasing the magnetic field along the [100]-axis. The antisymmetric spin-orbit coupling arising from the interfacial mirror symmetry breaking is of the Rashba type, and therefore, the Cooper pairs acquire a center of mass momentum perpendicular to the magnetic field. Thus, we obtain $q \parallel [010]$ except for $H = 0$. Figure 3(a) shows $q_y$ as a function of the magnetic field. We see a peak around $\mu_B H/T_C = 2.4$, although $|q_y|$...
monotonically increases with the magnetic field in the single-band models.\textsuperscript{54} The peak is associated with the dimensional crossover of the superconducting state.\textsuperscript{36} The superconductivity is mainly induced by the degenerate ($d_{x^2}$, $d_{y^2}$)-orbitals at zero magnetic field, and it crossovers to the quasi-one-dimensional superconducting state induced by the $d_{x^2-y^2}$-orbital around $\mu_B H/T_c = 2.4$. Since the paramagnetic depairing effect is suppressed in the latter as we discussed in Sect. 3.1, the center of mass momentum in Cooper pairs is decreased with increasing the magnetic field for $2.4 < \mu_B H/T_c < 5$.

The superconducting state shows another crossover from the helical state to the FF state around $\mu_B H/T_c = 5$, as indicated by the second increase of $q_y$ above $\mu_B H/T_c > 5$. Then, the sign of $q_y$ suddenly changes at $\mu_B H/T_c = 5.5$. Because the two momentum are degenerate at $\mu_B H/T_c = 5.5$, the multiple superconducting phases would appear as illustrated in Fig. 3(b). Here, it is assumed that the Cooper pairs' condensates for both momentum $q_y = q_+ = +0.0005$ and $q_y = q_- = -0.0011$ coexist below $T_c$ as in the single-band model.\textsuperscript{28} The superconducting state shows a phase transition from the single-Q FF state with $q_y > 0$ to the double-Q state, and it again changes to the single-Q FF state with $q_y < 0$ with increasing the magnetic field.

The order parameter in the double-$Q$ phase is described as,

$$\Delta_{mn}(r) = \Delta_{mn}^{(+)} e^{i\eta_{+}\cdot r} + \Delta_{mn}^{(-)} e^{i\eta_{-}\cdot r}.$$ 

Since $|q_x| \neq |q_y|$, both amplitude and phase of the order parameter are spatially inhomogeneous as in the CS state. As the sign change of $q$ does not occur in the single-orbital models,\textsuperscript{27,28,54} the multiple superconducting transitions illustrated in Fig. 3(b) are attributed to the orbital degree of freedom in $t_{2g}$-electron systems.

3.3 Orbital-dependent FFLO state in $H \parallel [110]$

Next, we investigate the FFLO state in the magnetic field along the [110]-axis. Figure 4 shows that the center of mass in Cooper pairs drastically increases above the magnetic field $\mu_B H/T_c > 1.7$. Similar behaviors have been obtained for single-band models for noncentrosymmetric superconductors\textsuperscript{54} and indicate the crossover from the helical state to the FF state. However, the superconducting state in the high magnetic field region is distinct from the conventional FF state, as we will explain below.

Figure 5 draws the center of mass momentum $q = (q_x, q_y)$ in the two-dimensional momentum space for various magnetic fields. Although the isotropic Rashba superconductor acquires a center of mass momentum perpendicular to the in-plane magnetic field, namely $q \parallel [110]$ in this case, we see the deviation of $q$ from the symmetric [110]-axis at high magnetic fields, $\mu_B H/T_c > 2.3$. The deviation is caused by the quasi-one-dimensional nature in the Fermi surface which favors the FFLO state with $q \parallel [110]$ or $q \parallel [010]$. The high-field FFLO state is determined by the competition between the Rashba spin-orbit coupling and the anisotropic Fermi surface, and the center of mass momentum is generally represented as, $q_{1,2} = q_{1,2} \pm q_1$. We here decomposed $q_1$ and $q_2$ by $q_{1,2} \parallel [110]$ and $q_{1,2} \parallel [110]$.

Because of the mirror symmetry along the [110]-axis, the superconducting state has a two-fold degeneracy with respect to the center of mass momentum, $q_1$ and $q_2$. Thus, the superconducting state is described by the two-component order parameters, $\eta = (\eta_1, \eta_2)$, where $\eta_1$ is for $q_1$ and $\eta_2$ is for $q_2$, respectively. The Ginzburg-Landau free energy is described with use of $\eta$ as,

$$F = -a_0 \left( 1 - \frac{T}{T_\text{c}(H)} \right) |\eta_1|^2 + \frac{\beta}{2} |\eta_1|^4 + \gamma |\eta_1|^2 |\eta_2|^2$$

$$+ \kappa_1 \left( |D_x \eta_1|^2 + |D_y \eta_1|^2 \right) + \kappa_2 \left( |D_x \eta_2|^2 + |D_y \eta_2|^2 \right),$$

where $D_a = -i\partial_a + 2eA_a$ is a covariant derivative. It should be noticed that the principal axis of gradient terms is different between the two component order parameters, as $(\hat{x}, \hat{y}) \neq$...
When the magnetic field is precisely applied to the ab-plane, the gradient terms do not play any role. Then, the double-Q state, where \( \eta \propto (1, e^{i\phi}) \), is stable when \( \gamma < 0 \). On the other hand, the single-Q state, \( \eta \propto (0, 1) \) or \( \eta \propto (1, 0) \), is stable otherwise.

The order parameter in the orbital basis is represented by \( \eta \) as,

\[
\Delta_{\mu r}(r) = \eta_1 \Delta_{\mu 1}^{(1)} e^{iq_1 r} + \eta_2 \Delta_{\mu 2}^{(2)} e^{iq_2 r}, \tag{25}
\]

\[
= e^{iq_1 r} \left[ \eta_1 \Delta_{\mu 1}^{(1)} e^{iq_2 r} + \eta_2 \Delta_{\mu 2}^{(2)} e^{-iq_2 r} \right], \tag{26}
\]

where \( \Delta_{\mu 1}^{(1)} \) and \( \Delta_{\mu 2}^{(2)} \) are obtained by the linearized gap equation for \( q = q_1 \) and for \( q = q_2 \), respectively. For both momentum, the dominant components are \( \Delta_{11}^{(1,2)} \) and \( \Delta_{22}^{(1,2)} \) describing the intra-orbital Cooper pairs formed by the \( d_{x^2-y^2} \) and \( d_{xy} \) orbitals, respectively. We find \( |\Delta_{11}^{(1)}| > |\Delta_{22}^{(2)}| \), because the \( d_{xy} \)-orbital (\( d_{xz} \)-orbital) favors the FFLO state in the single-Q state, respectively.

On the other hand, the order parameters are described in the double-Q state as,

\[
\Delta_{11}(r) = \Delta e^{iq_1 r} \left[ \delta e^{iq_2 r} + e^{-iq_2 r} \right], \tag{27}
\]

\[
\Delta_{22}(r) = \Delta e^{iq_2 r} \left[ e^{iq_1 r} + \delta e^{-iq_1 r} \right], \tag{28}
\]

where \( \Delta = |\Delta_{11}^{(1)}| = |\Delta_{22}^{(2)}| \), and \( \delta < 1 \). For instance, we obtain \( \delta = 0.88 \) at \( \mu_B H / T_C = 3.2 \), and it decreases with enhancing the quasi-one-dimensional property of Fermi surfaces by decreasing the tight-binding parameter \( t_3 \).

The averaged center of mass momentum in Cooper pairs depends on the orbital; \( q_{\perp} = \frac{1}{2} (q_1 + q_2) \) for the \( d_{x^2-y^2} \)-orbital while \( q_{\perp} = \frac{1}{2} (q_1 + q_2) \) for the \( d_{xy} \)-orbital. Thus, we call the double-Q state “orbital-dependent complex stripe (ODCS) state”. Analogous “layer dependent complex stripe state” has been proposed for the multilayer superconductors affected by the spin-orbit coupling.

Because the orbital polarization in the Cooper pairs costs an energy, it is expected that the double-Q state is stable rather than the single-Q state. The superconducting gap in the single-Q state is anisotropic in the Brillouin zone, and therefore, the single-Q state gains the condensation energy less than that in the double-Q state. Thus, we assume \( \gamma < 0 \) and draw the schematic phase diagram in Fig. 6. The ODCS state and the helical state is stabilized in the high magnetic field region and in the low magnetic field region, respectively.

![Fig. 5.](image-url) (Color online) The evolution of the center of mass momentum of Cooper pairs with increasing the magnetic field along the [110]-axis. Each mark is obtained at the magnetic field and temperature represented in Fig. 6. The superconducting state has a two-fold degeneracy with respect to the center of mass momentum in the high magnetic field region.

![Fig. 6.](image-url) (Color online) The phase diagram for \( H \parallel [110] \). The solid line is the critical magnetic field obtained by our calculation, and the marks correspond to those in Fig. 5. The second order phase transition line is schematically drawn by the dashed line. The ODCS state and the helical state is stabilized in the high magnetic field region and in the low magnetic field region, respectively.

### 3.4 Field angle dependence

As shown in Sects. 3.1-3.3, the paramagnetic depairing effect and the resulting FFLO state are different between \( H \parallel [100] \) and \( H \parallel [110] \) because of the orbital degree of freedom in the \( t_{2g} \)-electrons. We here clarify the field angle dependence of the superconducting state.

Figure 7(a) shows the field angle dependence of the critical magnetic field for \( H = H(\cos \theta, \sin \theta, 0) \). As we showed in Sect. 3.1, the critical magnetic field is enhanced near \( \theta = 0 \) and \( \theta = \pi/2 \), while the critical field is insensitive to \( \theta \) around \( \theta = \pi/4 \). Thus, when the field angle dependence would be observed in the measurement of critical magnetic field in the SrTiO\(_3\) heterostructures, it is indicated that the orbital degree of freedom plays an important role on the paramagnetic depairing effect.

When the magnetic field is slightly tilted from the [110]-axis, the degeneracy in the center of mass momentum \( q = q_1 \) and \( q = q_2 \) is lifted, and therefore, the single-Q FF state is stable just below the critical magnetic field. However, the ODCS state is stabilized around \( \theta = \pi/4 \) by decreasing the magnetic field as schematically drawn in Fig. 7(a). The FF state continuously changes to the helical state with decreasing
the magnetic field, as we discussed before.

Figure 7(b) shows the field angle dependence of the Cooper pairs’ momentum. For $H \parallel [100]$ and [010], the momentum $q$ is perpendicular to the magnetic field, and the magnitude $|q|$ is small because the paramagnetic depairing effect is suppressed. The magnitude grows with tilting the magnetic field to the [110]-axis. The center of mass momentum of Cooper pairs drastically changes through $\theta = \pi/4$, where two momentum $q = q_1$ and $q = q_2$ are degenerate.

4. Carrier Density Dependence of FFLO State

While we clarified the unconventional FFLO states in the high carrier density region $n = 0.15 \approx 1 \times 10^{14}$ cm$^{-2}$, we here show that they disappear in the low carrier density region. It has been shown that the superconducting state changes with increasing the carrier density from the $d_{x^2-y^2}$-orbital-induced superconductivity to the $d_{xz}/d_{yz}$-orbitals-induced superconductivity. For our choice of parameters, the crossover occurs around $n = 0.07 \approx 5 \times 10^{13}$ cm$^{-2}$. This crossover density is larger than the experimentally observed one $n \approx 2 \times 10^{13}$ cm$^{-2}$, but they are in the reasonable agreement with each other. Below the crossover density, the orbital degree of freedom is almost quenched, and therefore, the unusual properties of FFLO superconductivity discussed in Sect. 3 do not appear. Indeed, the Cooper pairs’ momentum shows a conventional growth with the magnetic field along the [100]-axis (Fig. 8(a)). For $n = 0.05 \approx 3.5 \times 10^{13}$ cm$^{-2}$, the superconducting state shows a single crossover from the helical state to the FF state as in the single-band models. Since the paramagnetic depairing effect is not suppressed as much as in the quasi-one-dimensional superconducting state for $n = 0.15 \approx 1 \times 10^{14}$ cm$^{-2}$, the magnitude of Cooper pairs’ momentum is much larger than that in the high carrier density region (compare Fig. 8(a) with Fig. 3(a)). We confirmed that the Cooper pairs’ momentum is perpendicular to the magnetic field along the [110]-axis. Thus, the ODCS state does not appear at $n = 0.05 \approx 3.5 \times 10^{13}$ cm$^{-2}$.

Near the crossover carrier density region $n = 0.1 \approx 7 \times 10^{13}$ cm$^{-2}$, the center of mass momentum in Cooper pairs changes the sign two times, corresponding to the two crossover discussed in Sect. 3.2. The $q_y$ is negative in the two-dimensional superconducting state at low magnetic fields, but that is positive in the quasi-one-dimensional superconducting state in the intermediate magnetic field region. It again changes the sign at $\mu_B H/T_c \approx 3.7$ where the helical state has changed to the FF state. Thus, the non-monotonic magnetic field dependence appears in the center of mass momentum of Cooper pairs. However, the sudden change of $q_y$ does not occur in contrast to Fig. 3(a), and therefore, the double-$Q$ CS state is not stabilized at least near $T = T_c(H)$. When we apply the magnetic field along the [110]-axis, the ODCS state is stabilized, but the critical temperature of the ODCS state [$T_{ODCS}/T_c \sim 0.1$] is much smaller than that in the high carrier density region $n = 0.15 \approx 1 \times 10^{14}$ cm$^{-2}$ [$T_{ODCS}/T_c \sim 0.5$].

Unusual properties of FFLO state are induced in the SrTiO$_3$ heterostructures in the high carrier density region above the crossover density. Although our model overestimates the unconventional FFLO states are realized above $n = 2 \times 10^{13}$ cm$^{-2}$.

![Fig. 7](https://example.com/f7.png)

**Fig. 7.** (Color online) (a) The solid line shows the critical magnetic field at $T = T_c$ as a function of $\theta$, the angle of in-plane magnetic field $H = H(\cos \theta, \sin \theta, 0)$. The red dashed line is the critical field of the BCS state. The superconducting states are schematically depicted. The ODCS state is stabilized in the shaded area. We see the helical state and the FF state below the crossover line (dash-dotted line), respectively. The double-$Q$ CS state in Fig. 3(b) is not drawn in this figure. (b) The Cooper pairs’ momentum $q = (q_x, q_y)$ at $T/T_c = 0.05$ and $H = H_{2d}(T)$ for various magnetic field directions from $\theta = 0$ to $\theta = \pi/2$. Marks in Fig. 7(b) correspond to those in Fig. 7(a). For example, the open blue circle is obtained for $\theta = \pi/2$.

![Fig. 8](https://example.com/f8.png)

**Fig. 8.** (Color online) The center of mass momentum in Cooper pairs at $T = T_c(H)$ as a function of the magnetic field along the [100]-axis. We adopt the carrier densities (a) $n = 0.05 \approx 3.5 \times 10^{13}$ cm$^{-2}$ and (b) $n = 0.1 \approx 7 \times 10^{13}$ cm$^{-2}$. We plot $q_y$ as in Fig. 3(a).
5. Summary and Discussion

We have investigated the multi-orbital FFLO superconductivity in the two-dimensional electron gases on the SrTiO\textsubscript{3} heterostructures. Owing to the broken inversion symmetry at the interface/surface, the Cooper pairs acquire a finite center of mass momentum in the magnetic field parallel to the two-dimensional conducting plane. It has been demonstrated that unconventional FFLO states emerge from the orbital degree of freedom in $d_{zx}$-orbitals in the high carrier density region.

First, for $H \parallel [100]$ the Cooper pairs’ momentum shows a non-monotonic magnetic field dependence indicating the crossover in the superconducting state. Indeed, the paramagnetic depairing effect changes the superconducting state from the two-dimensional helical state to the quasi-one-dimensional FF state. Near the crossover from the helical state to the FF state, the Cooper pairs’ momentum discontinuously changes, and the double-$Q$ CS state is stabilized below $T_c$. Second, for $H \parallel [110]$ the double-$Q$ ODCS state is stabilized in the high magnetic field region. Therein, the Cooper pairs formed by the $d_{xy}$-orbital and those by the $d_{zx}$-orbital have inequivalent center of mass momentum. These behaviors have not been indicated in the single-band models, and they are indeed attributed to the orbital degeneracy between the $(d_{xy}, d_{zx})$-orbitals. In particular, the ODCS state is a novel FFLO state in the multi-orbital system.

The unusual properties of FFLO state disappear in the low carrier density region where the superconductivity is mainly induced by the single $d_{xy}$-orbital. Thus, the high carrier density is needed to stabilize the multi-orbital FFLO state studied in this paper. Combining our results with the experimental observation,\textsuperscript{38} we expect that the high carrier density region is realized when $n > 2 \times 10^{13}$ cm$^{-2}$. The carrier density in the SrTiO\textsubscript{3} heterostructures is controlled by the gate voltage without mixing the impurities, and the high carrier density beyond $n = 1 \times 10^{14}$ cm$^{-2}$ has been brought to realization.\textsuperscript{3,6}

The experimental studies of superconducting state in the high magnetic field or in the ferromagnetic state\textsuperscript{30} are desired to clarify the various FFLO states arising from the multi-orbital nature of two-dimensional electron gases. We proposed that the large in-plane anisotropy in the critical magnetic field is an evidence for the paramagnetic depairing effect as well as the orbital degree of freedom. Thus, the anisotropy is also an indication for the FFLO superconductivity. More direct observation of the FFLO state with use of the STM and so on is of course desired. Then, our prediction for the carrier density dependence may be verified.

Finally, we discuss two ingredients which are not taken into account in our model. One is the subband structure. According to the band structure calculations,\textsuperscript{40-46} several $d_{yz}$-orbital-derived subbands cross the Fermi level, but the $d_{xy}/d_{zx}$-orbitals are quantized in the [001]-axis direction because of their light effective mass. Thus, the subband structure in the $d_{xy}/d_{zx}$-orbitals is negligible. On the other hand, taking into account the $d_{xz}$-orbital-derived subbands reduces the carrier density in the $d_{xy}/d_{zx}$-orbitals-derived band. However, our results in the high carrier density region are not altered as far as the substantial part of carriers populates in the $d_{xy}/d_{zx}$-orbitals. The other ingredient neglected so far is the disorders. Although we investigated the superconducting state in the clean limit, the SrTiO\textsubscript{3} heterostructures indeed contain substantial disorders.\textsuperscript{3,8} The disorders may alter our results on the multi-orbital FFLO state because they smear out the orbital character of the band. Furthermore, it has been shown that the disorders destabilize the FFLO state\textsuperscript{13} although the helical state is not completely eliminated.\textsuperscript{23} Therefore, it is desirable to take into account the disorders for a comparison with experiments. We leave this issue for the future study. It is also desired to fabricate the (at least locally) clean SrTiO\textsubscript{3} heterostructures in order to eliminate the disorders.

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