

Transition from the vortex state to the Fulde–Ferrell–Larkin–Ovchinnikov state in quasi-two-dimensional superconductors

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We examine the coexistence of the vortex state and the Fulde–Ferrell–Larkin–Ovchinnikov (FFLO) state in quasi-two-dimensional type-II superconductors and the transition from this mixed state to the pure FFLO state when the Maki parameter α increases. The pure FFLO state, which is characterized by Cooper pairs having finite center-of-mass momenta $\mathbf{q} \neq \mathbf{0}$, occurs in the two-dimensional limit when the magnetic field is parallel to the conducting plane. The vectors \mathbf{q} are determined from the Fermi-surface structure and pairing anisotropy, and become finite below a temperature T^* . In quasi-two-dimensions, because of the orbital pair-breaking effect, a mixed state characterized by (n, q_{\parallel}) occurs, where n and q_{\parallel} denote the Landau-level index of the vortex state and the wave number of the additional FFLO modulation along the magnetic field. We obtain the α dependence of the upper critical field by numerical calculations. The upper critical field exhibits a cascade curve in the H - T phase diagram. It is analytically shown that n diverges in the two-dimensional limit $\alpha \rightarrow \infty$ below T^* . In this limit, the upper critical-field equation for the mixed state reduces to that for the FFLO state. A relation between n of the mixed state and q_{\perp} of the pure FFLO state is obtained, where q_{\perp} denotes the component of \mathbf{q} perpendicular to the magnetic field. It is found that the pure FFLO state is nothing but the vortex state with infinitely large n as is known in two-dimensional superconductors in a tilted magnetic field. The vortex state with large n can be regarded as the FFLO state with nonzero q_{\perp} in three dimensions.

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I. INTRODUCTION

In type-II superconductors, an applied magnetic field destroys the superconductivity by two kinds of pair-breaking effects: the orbital magnetic and Pauli paramagnetic pair-breaking effects.¹ We can define the pure orbital limit H_{c20} and the Pauli paramagnetic limit H_p , which are theoretically obtained by taking into account only the orbital effect and paramagnetic effect, respectively. The Maki parameter $\alpha \equiv \sqrt{2}H_{c20}/H_p$ expresses the strength ratio of the two pair-breaking effects.

In conventional metal superconductors, the orbital pair-breaking effect dominates the system ($H_{c20} \ll H_p$) because of the large Fermi velocity. Partial destruction of the superconductivity due to the orbital effect creates vortexes, which form a lattice below the upper critical field and causes the order parameter to become inhomogeneous.

In contrast, in purely Pauli limited superconductors, another type of nonuniform superconductivity has been proposed by Fulde and Ferrell² and Larkin and Ovchinnikov.³ In magnetic fields, the Fermi surfaces of the up- and down-spin electrons are displaced due to the Zeeman energy. If the up- and down-spin electrons on the displaced Fermi surfaces form Cooper pairs, they will have a finite center-of-mass momentum. The superconducting state of such Cooper pairs is called the Fulde–Ferrell–Larkin–Ovchinnikov (FFLO) state. It is easily verified that a finite center-of-mass momentum results in spatial modulations and nodes of the order parameter in real space. As a result, spin-polarization energy is gained by the depaired electrons near the nodes while condensation energy is lost.

Therefore, a necessary condition for the occurrence of the FFLO state is that the superconductivity survives in high fields such that $\mu_e H \sim \Delta_0$, for which the loss of condensation

energy due to modulation of the order parameter is compensated by a gain in polarization energy. Here, μ_e and Δ_0 denote the electron magnetic moment and the zero-field energy gap of the superconductivity. This condition can be expressed as $H_{c2} \sim H_p$, where H_{c2} denotes the upper critical field because $H_p \sim \Delta_0$. Therefore, the orbital pair-breaking effect needs to be very weak for the FFLO state to occur.

This is one reason why the FFLO state has not been observed in conventional metal superconductors. Gruenberg and Gunther found that the FFLO state occurs in isotopic superconductors only when the Maki parameter α is large ($\alpha \gtrsim 1.8$).⁴ Such a large Maki parameter is usually difficult to realize in alloy type-II superconductors, although it is achievable in some exotic superconductors, such as organic superconductors,⁵ heavy fermion superconductors,⁶ and oxide superconductors because of their narrow electron bands, large effective masses, and quasi-two-dimensionality.

Because of the orbital pair-breaking effect and the formation of the vortex lattice state, the dependence of the order parameter on the spatial coordinates perpendicular to the magnetic field is described by a superposition of Abrikosov functions. Therefore, there may be additional modulations due to the FFLO state only in the direction parallel to the magnetic field, as Gruenberg and Gunther have proposed.⁴ We examined the coexistence of the vortex states with higher Landau-level indexes n and the FFLO state in d -wave superconductors in a previous paper. We obtained phase diagrams when the Pauli paramagnetic effect is negligible and also when both the orbital and Pauli paramagnetic effects are taken into account.⁷ We demonstrated that higher Landau-level states and mixed states are stabilized in some cases. Buzdin and Brison also demonstrated that higher Landau-level solutions occur in isotropic systems when $\alpha \gtrsim 9$.⁸ Recently, Adachi and Ikeda have examined a mixed state in a

model of a quasi-two-dimensional heavy fermion superconductor CeCoIn₅.⁹

Bulaevskii examined film superconductors at $T=0$ in tilted magnetic fields and found that cascade transitions occur between states with different n 's when the direction of the magnetic field changes.¹⁰ It was shown that the upper critical field tends to approach that of the FFLO state when the magnetic field approaches the parallel direction. Buzdin and Brison also obtained cascade transitions at finite temperatures in two- and three-dimensional isotropic superconductors.^{8,11} We extended Bulaevskii's theory to finite temperatures in s - and d -wave superconductors.¹² We also reproduced the cascade transitions by solving the gap equation and clarified the behavior in the limit of a parallel field. We analytically demonstrated that the Landau-level index n of the vortex state diverges in the temperatures region where the FFLO state occurs in the limit. As a result, the envelope of the cascade transition lines approaches the FFLO critical field when the field orientation approaches the parallel direction. The vortex state is continuously reduced to the pure FFLO state. We obtained a relation between n and the FFLO vector \mathbf{q} that connects the vortex states and the FFLO state in the limit. The present study extends our previous theory to the mixed state in three dimensions.

Near critical fields, the order parameter of the pure FFLO state can be expressed by a linear combination of exponential functions $\exp[i\mathbf{q}_m \cdot \mathbf{r}]$, where \mathbf{q}_m are degenerate FFLO wave vectors with optimum values determined from the structures of the Fermi surfaces, the pairing interactions, and the temperature. The free energies of such states were compared in a three-dimensional isotropic system by Larkin and Ovchinnikov,³ Matsuo *et al.*,¹³ Bowers and Rajagopal,¹⁴ and Mora and Combescot,¹⁵ and in two-dimensional systems by us¹⁶ and Mora and Combescot.¹⁷ Larkin and Ovchinnikov³ showed that the state expressed by $\Delta(\mathbf{r}) \sim \cos(\mathbf{q} \cdot \mathbf{r})$ has a lower free energy than the state $\Delta(\mathbf{r}) \sim \exp[i\mathbf{q} \cdot \mathbf{r}]$ proposed by Fulde and Ferrell. We have shown that the square, triangular, and hexagonal states have lower free energies than the state $\Delta(\mathbf{r}) \sim \cos(\mathbf{q} \cdot \mathbf{r})$ at low temperatures in two dimensions.¹⁶ Mora and Combescot have shown that states described by many cosine functions have lower free energies than the state $\Delta(\mathbf{r}) \sim \cos(\mathbf{q} \cdot \mathbf{r})$ in three dimensions when the first-order phase transition is taken into account.¹⁵ The possibility of a first-order transition was also addressed by Larkin and Ovchinnikov.³

When the magnetic field is not oriented in the optimum direction of \mathbf{q} of the pure FFLO state or when more than two \mathbf{q}_m 's contribute to the linear combination for the pure FFLO state, it may appear that the mixed state is not reduced to the pure FFLO state in the limit $\alpha \rightarrow \infty$ because in the mixed state \mathbf{q} can have a nonzero component only in the direction of the magnetic field. In actuality, however, the Landau-level index n of the mixed state diverges and the components of \mathbf{q} perpendicular to the magnetic field are realized.¹⁸ This behavior is analogous to that two-dimensional systems in a tilted magnetic field. In the present paper, we demonstrate this behavior in quasi-two-dimensional systems by an analytical proof and specific numerical calculations.

For simplicity, we adopt an effective-mass model assuming that the first-order transition is suppressed to demonstrate

the continuity between the mixed state and the pure FFLO state. In actuality, in the effective-mass model with very large α , the first-order transition will occur at a slightly higher field than the second-order transition.¹⁵ In this case, the second-order transition curve below the first-order transition curve is regarded as that of the metastable transition, which can be realized when the system is supercooled.

Recently, we have obtained a result that also suggests that the Landau-level index n increases with α in an anisotropic Ginzburg-Landau model near the tricritical point.¹⁹ Comparing the phase diagrams with and without the orbital effect, we have found that areas of the mixed states with $n > 0$ in the former phase diagrams correspond to areas of the pure FFLO state with $q_{\perp} \neq 0$ in the latter phase diagrams.

In Sec. II, we present our formulation. In Secs. III and IV, we examine the pure FFLO state and the mixed state, respectively. In Sec. IV, we show the transition from the pure FFLO state to the mixed state and the limit as $\alpha \rightarrow \infty$. In Sec. V, we show the numerical results for finite temperatures. In Sec. VI, we summarize and discuss the results.

II. FORMULATION

We examine a model described by the Hamiltonian

$$H = H_0 + H_m + H' \quad (1)$$

with

$$H_0 = \sum_{\mu\sigma} \int d^3\mathbf{r} \psi_{\sigma}^{\dagger}(\mathbf{r}) \frac{1}{2m_{\mu}} \left(-i\hbar \frac{\partial}{\partial x_{\mu}} - \frac{e}{c} A_{\mu} \right)^2 \psi_{\sigma}(\mathbf{r}), \quad (2)$$

$$H_m = \sum_{\sigma} \int d^3\mathbf{r} \sigma h \psi_{\sigma}^{\dagger}(\mathbf{r}) \psi_{\sigma}(\mathbf{r}), \quad (3)$$

$$H' = \int d^3\mathbf{r} \int d^3\mathbf{r}' \psi_{\uparrow}^{\dagger}(\mathbf{r}) \psi_{\uparrow}(\mathbf{r}) V(\mathbf{r} - \mathbf{r}') \psi_{\downarrow}^{\dagger}(\mathbf{r}') \psi_{\downarrow}(\mathbf{r}'). \quad (4)$$

Here, we have defined the effective masses $m_1 = m_x$, $m_2 = m_y$, $m_3 = m_z$, Zeeman field $h = \mu_e |\mathbf{H}|$, and vector potential \mathbf{A} , where μ_e and \mathbf{H} denote the magnitude of the electron magnetic moment and the magnetic field $\mathbf{H} = \text{rot } \mathbf{A}$. We consider pairing interactions of the form

$$V(\mathbf{p}, \mathbf{p}') = g_{\alpha} \gamma_{\alpha}(\hat{\mathbf{p}}) \gamma_{\alpha}(\hat{\mathbf{p}}'), \quad (5)$$

where the suffix α expresses a symmetry.

In the effective-mass model of Eq. (2), it is convenient to define $\tilde{\mathbf{r}} = (\tilde{x}_1, \tilde{x}_2, \tilde{x}_3)$ by a scale transformation

$$\sqrt{m_{\mu}} x_{\mu} \equiv \sqrt{\tilde{m}} \tilde{x}_{\mu} \quad (6)$$

with $\tilde{m} = (m_x m_y m_z)^{1/3}$. Then, Eq. (2) is written as

$$H_0 = \sum_{\mu\sigma} \int d^3\mathbf{r} \psi_{\sigma}^{\dagger}(\mathbf{r}) \frac{1}{2\tilde{m}} \left(-i\hbar \frac{\partial}{\partial \tilde{x}_{\mu}} - \frac{e}{c} \tilde{A}_{\mu} \right)^2 \psi_{\sigma}(\mathbf{r}), \quad (7)$$

where we have defined

$$\tilde{A}_\mu \equiv \sqrt{\frac{\tilde{m}}{m_\mu}} A_\mu. \quad (8)$$

We also define $\tilde{\mathbf{p}} = (\tilde{p}_1, \tilde{p}_2, \tilde{p}_3)$ with $\tilde{p}_\mu = (\tilde{m}/m_\mu)^{1/2} p_\mu$ so that $\mathbf{r} \cdot \mathbf{p} = \tilde{\mathbf{r}} \cdot \tilde{\mathbf{p}}$. Then, the Fermi surface in $\tilde{\mathbf{p}}$ space becomes spherically symmetric, and we can define a constant Fermi momentum \tilde{p}_F and Fermi velocity $\tilde{\mathbf{v}}_F = \tilde{v}_F \tilde{\mathbf{p}} / |\tilde{\mathbf{p}}|$ with a constant magnitude $\tilde{v}_F = \tilde{p}_F / \tilde{m}$, for the scaled momentum $\tilde{\mathbf{p}}$.

Now, we derive the gap equation. The calculation is a straightforward extension of previous studies.^{7,12,20,21} Near the second-order phase transition, the gap function has the form

$$\Delta(\mathbf{r}, \mathbf{p}) = \Delta_\alpha(\mathbf{r}) \gamma_\alpha(\mathbf{p}) \quad (9)$$

and the gap equation is linearized as

$$-\log\left(\frac{T}{T_c^{(0)}}\right) \Delta_\alpha(\mathbf{r}) = \pi T \int_0^\infty dt \frac{1}{\sinh(\pi T t)} \int \frac{d\Omega_{\tilde{\mathbf{p}}'}}{4\pi} [\gamma_\alpha(\tilde{\mathbf{p}}')]^2 \times \left\{ 1 - \cos\left[t\left(h - \frac{1}{2}\tilde{\mathbf{v}}_F' \cdot \tilde{\mathbf{\Pi}}\right)\right] \right\} \Delta_\alpha(\mathbf{r}), \quad (10)$$

where $\mathbf{v}'_F = \tilde{v}_F \tilde{\mathbf{p}}' / |\tilde{\mathbf{p}}'|$ and $\tilde{\mathbf{\Pi}} = (\tilde{\Pi}_1, \tilde{\Pi}_2, \tilde{\Pi}_3)$ with

$$\tilde{\Pi}_\mu = -i\hbar \frac{\partial}{\partial \tilde{x}_\mu} - \frac{2e}{c} \tilde{A}_\mu. \quad (11)$$

The upper critical field H_{c2} is the highest $|\mathbf{H}|$ among those that give a nontrivial solution of $\Delta(\mathbf{r})$. We note that Eq. (10) is the same as that of a system with a spherically symmetric Fermi surface except that the argument $\tilde{\mathbf{p}}' = \mathbf{p}' / |\mathbf{p}'|$ of $\gamma_\alpha(\tilde{\mathbf{p}}')$ is different from the integral variable $\hat{\mathbf{p}}' = \tilde{\mathbf{p}}' / |\tilde{\mathbf{p}}'| = \tilde{\mathbf{p}}' / \tilde{p}_F$. Therefore, when $\gamma_\alpha(\tilde{\mathbf{p}}')$ is constant, it can easily be verified that the mass anisotropy does not affect the upper critical-field equation except that the vector potential is scaled as described in Eq. (8) since $\hat{\mathbf{p}}'$ is only an integral variable. In contrast, for anisotropic superconductors, the mass anisotropy affects the upper critical-field equation through the deformation of $\gamma_\alpha(\tilde{\mathbf{p}}')$ when it is expressed in $\tilde{\mathbf{p}}$ space.²¹

III. PURE FFLO STATE

In this section, we briefly review the case in which the orbital pair-breaking effect is negligible. In this case, we can set $\mathbf{A} = \mathbf{0}$ in Eqs. (2) and (11). Equation (10) has a solution of the form

$$\Delta(\mathbf{r}) \propto \exp[i\tilde{\mathbf{q}} \cdot \tilde{\mathbf{r}}/\hbar] \quad (12)$$

and is reduced to

$$-\log\left(\frac{T}{T_c^{(0)}}\right) = \pi T \int_0^\infty dt \frac{1}{\sinh(\pi T t)} \times \int \frac{d\Omega_{\tilde{\mathbf{p}}'}}{4\pi} [\gamma_\alpha(\tilde{\mathbf{p}}')]^2 \times \left\{ 1 - \cos\left[t\left(h - \frac{1}{2}\tilde{\mathbf{v}}_F' \cdot \tilde{\mathbf{q}}\right)\right] \right\}. \quad (13)$$

For example, for s -wave pairing, $\gamma_s(\mathbf{p}) = 1$, there is an infinite degeneracy with respect to the direction of $\tilde{\mathbf{q}}$, although the magnitude $|\tilde{\mathbf{q}}|$ is uniquely determined so that the critical field is maximized. For non- s -wave pairing, both the direction and the magnitude of $\tilde{\mathbf{q}}$ are optimized. Depending on the symmetries of $\gamma_\alpha(\mathbf{p})$ and the Fermi surface, and the temperature, there may be 2, 4, 8, 16, \dots -fold degeneracies with respect to the direction of $\tilde{\mathbf{q}}$. We write the optimum $\tilde{\mathbf{q}}$'s as $\tilde{\mathbf{q}}_m$ with $m = 1, 2, \dots, M$. Below and near the upper critical field, the order parameter is expressed by a linear combination,

$$\Delta(\mathbf{r}) = \sum_m \Delta_m e^{i\tilde{\mathbf{q}}_m \cdot \tilde{\mathbf{r}}}. \quad (14)$$

Among the states of this form, the physical state is that with the lowest free energy. Every degenerate $\tilde{\mathbf{q}}_m$ does not necessarily appear in the linear combination of the physical state. The most well-known form is that expressed by the linear combination of $e^{i\tilde{\mathbf{q}} \cdot \tilde{\mathbf{r}}}$ and $e^{-i\tilde{\mathbf{q}} \cdot \tilde{\mathbf{r}}}$, i.e., $\Delta(\mathbf{r}) \propto \cos(\tilde{\mathbf{q}} \cdot \tilde{\mathbf{r}})$. For the second-order transition and s -wave pairing, this state is the physical state in the effective-mass model.

Here, we note that $\tilde{\mathbf{q}}_m$ are not necessarily parallel to the magnetic field when the orbital effect is negligible. The number and directions of the optimum $\tilde{\mathbf{q}}_m$ that contribute to the physical state depend on the structures of the Fermi surface and the pairing interactions,^{22,23} and the temperature.^{12,24–26} In the present effective-mass model, since the electron dispersion becomes isotropic in $\tilde{\mathbf{p}}$ space as seen in Eq. (13), the Fermi-surface anisotropy does not remove the infinite degeneracy of the optimum $\tilde{\mathbf{q}}_m$ for s -wave pairing.

IV. MIXED STATE

In this section, we take into account both the orbital and paramagnetic pair-breaking effects. After deriving the upper critical-field equation, we consider the limit of a weak orbital effect.

For a magnetic field $\mathbf{H} = (0, 0, H)$, we define $\mathbf{A} = (-Hy, 0, 0)$ with an appropriate gauge. The scale transformation described above gives $\tilde{A}_x = -\tilde{H}\tilde{y}$ with $\tilde{H} = (\tilde{m} / \sqrt{m_x m_y})H$. We define boson operators by

$$\tilde{\eta} = \frac{\tilde{\xi}_H}{\sqrt{2}} (\tilde{\Pi}_x - i\tilde{\Pi}_y), \quad (15)$$

$$\tilde{\eta}^\dagger = \frac{\tilde{\xi}_H}{\sqrt{2}} (\tilde{\Pi}_x + i\tilde{\Pi}_y)$$

with

$$\tilde{\xi}_H = \sqrt{\frac{c}{2|e|\tilde{H}}} = \left[\frac{m_x m_y}{m_z^2} \right]^{1/12} \xi_H, \quad (16)$$

where $\xi_H = \sqrt{c/2|e|H} = \sqrt{\Phi_0/2\pi H}$, which is on the order of the BCS coherence length ξ_0 when $H \sim H_{c2}$. The operator $\tilde{\mathbf{v}}_F' \cdot \tilde{\mathbf{\Pi}}$ that appears in Eq. (10), can be rewritten as

$$\tilde{v}_F \cdot \tilde{\mathbf{P}} = \frac{1}{\sqrt{2\xi_H}} \tilde{v}_F \sin \tilde{\theta}' (e^{i\tilde{\varphi}'} \tilde{\eta} + e^{-i\tilde{\varphi}'} \tilde{\eta}^\dagger) - i\hbar \tilde{v}_F \cos \tilde{\theta}' \frac{\partial}{\partial \tilde{z}}, \quad (17)$$

where $\tilde{\theta}'$ and $\tilde{\varphi}'$ denote the polar coordinates when \tilde{z} axis is the polar axis.

Because Eq. (10) can be regarded as an eigenequation with an eigenvalue $-\log(T/T_c^{(0)})$ and eigenfunction $\Delta_\alpha(\mathbf{r})$, our problem is reduced to find the eigenfunctions with the highest eigenvalue. The solutions can be written in the form

$$\Delta(\mathbf{r}) = \bar{\Delta}(\tilde{x}, \tilde{y}) \exp[i\tilde{q}_z \tilde{z}/\hbar] \quad (18)$$

with $\partial/\partial \tilde{z}$ in Eq. (17) replaced by $i\tilde{q}_z/\hbar$. The gap Eq. (10) can be rewritten as

$$\begin{aligned} & -\log\left(\frac{T}{T_c^{(0)}}\right) \phi(\tilde{x}, \tilde{y}) \\ &= \pi T \int_0^\infty dt \frac{1}{\sinh(\pi T t)} \int \frac{d\Omega_{\tilde{p}'}}{4\pi} [\gamma_\alpha(\hat{\mathbf{p}}')]^2 \\ & \times \left\{ 1 - \cos \left[t \left(h - \frac{1}{2} \tilde{v}_F \tilde{q}_z \cos \tilde{\theta}' - \hat{\xi} \right) \right] \right\} \phi(\tilde{x}, \tilde{y}), \quad (19) \end{aligned}$$

where we have defined

$$\hat{\xi} = \frac{\tilde{v}_F \sin \tilde{\theta}'}{2\sqrt{2\xi_H}} (e^{i\tilde{\varphi}'} \tilde{\eta} + e^{-i\tilde{\varphi}'} \tilde{\eta}^\dagger). \quad (20)$$

It is convenient to expand the function $\phi(\tilde{x}, \tilde{y})$ in terms of Abrikosov functions $\phi_n^{(k)}(\tilde{x}, \tilde{y})$ defined by

$$\phi_n^{(k)}(\tilde{x}, \tilde{y}) = \frac{1}{\sqrt{n!}} (\tilde{\eta}^\dagger)^n \phi_0^{(k)}(\tilde{x}, \tilde{y}), \quad (21)$$

where $n=0, 1, 2, 3, \dots$ are the Landau-level indexes, k is an arbitrary wave number, and $\phi_0^{(k)}$ is the solution of

$$\tilde{\eta} \phi_0^{(k)}(\tilde{x}, \tilde{y}) = 0, \quad (22)$$

which is expressed as

$$\phi_0^{(k)}(\tilde{x}, \tilde{y}) = C e^{ik\tilde{x}} \exp\left[-\frac{(\tilde{y}-\tilde{y}_k)^2}{2\xi_H^2}\right] \quad (23)$$

with $\tilde{y}_k \equiv k\xi_H^2$ and a normalization constant C . The function $\phi_n^{(k)}(\tilde{x}, \tilde{y})$ is expressed as

$$\phi_n^{(k)}(\tilde{x}, \tilde{y}) = (-1)^n C e^{ik\tilde{x}} H_n \left[\sqrt{2} \frac{\tilde{y}-\tilde{y}_k}{\xi_H} \right] \exp\left[-\frac{(\tilde{y}-\tilde{y}_k)^2}{2\xi_H^2}\right] \quad (24)$$

in terms of the Hermite polynomial. The operators $\tilde{\eta}$ and $\tilde{\eta}^\dagger$ and the Abrikosov functions $\phi_n^{(k)}$ satisfy the relations

$$\begin{aligned} \tilde{\eta}^\dagger \phi_n^{(k)}(\tilde{x}, \tilde{y}) &= \sqrt{n+1} \phi_{n+1}^{(k)}(\tilde{x}, \tilde{y}), \\ \tilde{\eta} \phi_n^{(k)}(\tilde{x}, \tilde{y}) &= \sqrt{n} \phi_{n-1}^{(k)}(\tilde{x}, \tilde{y}). \end{aligned} \quad (25)$$

If we expand the eigenfunctions as

$$\Delta(\mathbf{r}) = \sum_{n=0}^{\infty} \Delta_n \phi_n^{(k)}(\tilde{x}, \tilde{y}) \exp[i\tilde{q}_z \tilde{z}/\hbar], \quad (26)$$

the gap Eq. (10) can be written as a matrix equation for the eigenvector with the vector elements $\Delta_0, \Delta_1, \Delta_2, \dots$ as

$$-\log\left(\frac{T}{T_c^{(0)}}\right) \Delta_n = \sum_{n'} D_{nn'} \Delta_{n'} \quad (27)$$

with

$$\begin{aligned} D_{nn'} &= \pi T \int_0^\infty dt \frac{1}{\sinh(\pi T t)} \int \frac{d\Omega_{\tilde{p}'}}{4\pi} [\gamma_\alpha(\hat{\mathbf{p}}')]^2 \int d\tilde{x} d\tilde{y} \phi_n^{(k)}(\tilde{x}, \tilde{y}) \\ & \times \left\{ 1 - \cos \left[t \left(h - \frac{1}{2} \tilde{v}_F \tilde{q}_z \cos \tilde{\theta}' - \hat{\xi} \right) \right] \right\} \phi_{n'}^{(k)}(\tilde{x}, \tilde{y}). \end{aligned} \quad (28)$$

When $h=0$, for s -wave pairing, the solution with $n=0$ and $q_z=0$ gives the highest upper critical field. In general, Abrikosov functions with different n 's can be mixed.

The magnetic field $|\mathbf{H}|$ appears in the Hamiltonians H_0 and H_m . The field $|\mathbf{H}|$ that originates from \mathbf{A} in H_0 is responsible for the orbital effect, and appears in the gap equation as a dimensionless parameter $a_m |\mathbf{H}|$ with the coefficient a_m defined by

$$a_m \equiv \frac{\tilde{m}}{\sqrt{m_x m_y}} \frac{2|e|}{c} \left(\frac{v_F}{2\pi T_c^{(0)}} \right)^2. \quad (29)$$

In contrast, the field $|\mathbf{H}|$ included in the Zeeman field h in H_m is responsible for the Pauli paramagnetic pair-breaking effect. For this field, it is convenient to define the dimensionless parameter $\mu_e |\mathbf{H}| / (2\pi T_c^{(0)})$. The relative strength of the Pauli paramagnetic pair-breaking effect to that of the orbital effect is expressed by the ratio of their dimensionless parameters,

$$z_m = \frac{\mu_e |\mathbf{H}| / (2\pi T_c^{(0)})}{a_m |\mathbf{H}|} = \frac{\mu_e}{2\pi T_c^{(0)} a_m}. \quad (30)$$

The parameter z_m is proportional to the Maki parameter α . For example, for s -wave pairing, numerical calculations give $a_m H_{c20} \approx 1.0372$ and $\mu_e H_P / \Delta_0 \approx 0.707107$, and hence $\alpha \approx 7.39 \times z_m$.

If we define \bar{a}_m and \bar{z}_m for the isotropic system by

$$\begin{aligned} \bar{a}_m &\equiv (2|e|/c)(v_F/2\pi T_c^{(0)})^2, \\ \bar{z}_m &= \mu_e / (2\pi T_c^{(0)} \bar{a}_m), \end{aligned} \quad (31)$$

we obtain

$$z_m = \left(\frac{m_x m_y}{m_z^2} \right)^{1/6} \bar{z}_m, \quad \alpha = \left(\frac{m_x m_y}{m_z^2} \right)^{1/6} \bar{\alpha} \quad (32)$$

with the Maki parameter of the isotropic system $\bar{\alpha} \equiv \sqrt{2} \tilde{H}_{c20} / H_P$. For the magnetic field parallel to the z axis, when $m_x \gg m_z$ we obtain $z_m \gg \bar{z}_m$, i.e., $\alpha \gg \bar{\alpha}$, which means that the system is strongly Pauli paramagnetic limited.

V. TRANSITION FROM PURE FFLO STATE TO MIXED STATE

Now, we consider a quasi-two-dimensional system such that $m_x \gg m_y, m_z$. In this case, because we have $\tilde{\xi}_H \gg \xi_H \sim \xi_0$ from Eq. (16), we may omit $\hat{\zeta} \sim (e^{i\tilde{\varphi}'} \tilde{\eta} + e^{-i\tilde{\varphi}'} \tilde{\eta}^\dagger) / \tilde{\xi}_H$ in Eq. (28) for finite n and n' . Therefore, if we can truncate the summation over n in Eq. (26), the upper critical-field equation is reduced to

$$-\log\left(\frac{T}{T_c^{(0)}}\right) = \pi T \int_0^\infty dt \frac{1}{\sinh(\pi T t)} \int \frac{d\Omega_{\tilde{\mathbf{p}}'}}{4\pi} [\gamma_\alpha(\tilde{\mathbf{p}}')]^2 \times \left\{ 1 - \cos \left[t \left(h - \frac{1}{2} \tilde{v}_F \tilde{q}_z \cos \tilde{\theta}' \right) \right] \right\} \quad (33)$$

in the limit $\tilde{\xi}_H \rightarrow \infty$, which coincides with the upper critical-field equation for the pure FFLO state expressed by Eq. (12) with $\tilde{\mathbf{q}} \equiv (0, 0, \tilde{q}_z)$. The magnitude $|\tilde{\mathbf{q}}| = |\tilde{q}_z|$ should be optimized so that the upper critical field is maximized. We write the optimum value as q_0 . For example, for s -wave pairing, it is known that $q_0 \approx 1.2 \times 2h / \tilde{v}_F$ at $T=0$. Thus, we have only two states with the highest upper critical field, which have the FFLO vectors $\tilde{\mathbf{q}} = (0, 0, \pm q_0)$. This contradicts the fact that there are more than two $e^{i\tilde{\mathbf{q}}_m \tilde{\mathbf{r}}/\hbar}$ in exactly two-dimensional systems as mentioned below Eq. (12). Furthermore, below the state expressed by a linear combination of more than two $e^{i\tilde{\mathbf{q}}_m \tilde{\mathbf{r}}/\hbar}$ at low temperatures.¹⁴⁻¹⁶

This contradiction is due to the assumption that infinitely large n 's are negligible in the two-dimensional limit. In the limit $m_x \gg m_y, m_z$, we need to consider Abrikosov functions with infinitely large Landau-level indexes n . For large n 's, the states with $n \pm 1$ can be approximated by the state with n , which means that $\phi_{n \pm 1} \approx e^{\pm i\varphi_0} \phi_n$ in Eq. (25), where $e^{\pm i\varphi_0}$ are arbitrary phase factors. Therefore, we may write

$$\tilde{\eta} = \sqrt{n} e^{-i\varphi_0}, \quad \tilde{\eta}^\dagger = \sqrt{n} e^{i\varphi_0} \quad (34)$$

in the gap equation. This procedure is analogous to those in the theory of Bose condensation and in two-dimensional type-II superconductors in a tilted magnetic field.¹² Using Eq. (34) we obtain

$$\hat{\zeta} = \frac{\sqrt{n} \tilde{v}_F \sin \tilde{\theta}'}{\sqrt{2} \tilde{\xi}_H} \cos(\tilde{\varphi}' - \varphi_0). \quad (35)$$

Thus, the gap equation (19) can be rewritten for $m_x \gg m_y, m_z$ as

$$-\log\left(\frac{T}{T_c^{(0)}}\right) \phi(\tilde{x}, \tilde{y}) = \pi T \int_0^\infty dt \frac{1}{\sinh(\pi T t)} \int \frac{d\Omega_{\tilde{\mathbf{p}}'}}{4\pi} [\gamma_\alpha(\tilde{\mathbf{p}}')]^2 \times \left\{ 1 - \cos \left[t \left(h - \frac{1}{2} \tilde{v}_F' \cdot \tilde{\mathbf{q}} \right) \right] \right\} \phi(\tilde{x}, \tilde{y}), \quad (36)$$

where

$$\tilde{\mathbf{q}} = (\tilde{q}_x, \tilde{q}_y, \tilde{q}_z) = (\tilde{q}_\perp \cos \varphi_0, \tilde{q}_\perp \sin \varphi_0, \tilde{q}_z) \quad (37)$$

with

$$\tilde{q}_\perp = \sqrt{\tilde{q}_x^2 + \tilde{q}_y^2} = \frac{\sqrt{2n}}{\tilde{\xi}_H} = \sqrt{\frac{2n\bar{\alpha}}{\alpha}} \frac{1}{\xi_H}. \quad (38)$$

If we write the optimum n for each fixed α as $n(\alpha)$, we obtain

$$\tilde{q}_\perp = \lim_{\alpha \rightarrow \infty} \sqrt{\frac{2n(\alpha)\bar{\alpha}}{\alpha \xi_H^2}}. \quad (39)$$

If $\tilde{q}_\perp \neq 0$ in the limit $\alpha \rightarrow \infty$, $n(\alpha)$ must diverge like α , i.e., $n(\alpha) \sim \alpha$. Equation (36) coincides with the upper critical-field equation of the pure FFLO state with $\tilde{\mathbf{q}}$.

Equation (39) is an essential equation that connects the pure FFLO state with the optimum \mathbf{q} in the two-dimensional limit and the mixed state with the optimum n and \tilde{q}_z in quasi-two-dimensions. From Eqs. (38) and (39), we can see that if there are more than two optimum $\tilde{\mathbf{q}}$'s in the two-dimensional limit, there must be mixed states with different n 's with close upper critical fields in quasi-two-dimensions where $m_x \gg m_y, m_z$.

For example, a d -wave superconductor with

$$\gamma_{d_{y^2-z^2}}(\tilde{\mathbf{p}}) \propto \tilde{p}_y^2 - \tilde{p}_z^2 \quad (40)$$

at low temperatures exhibits a degeneracy in the solutions of Eq. (36) with $\tilde{\mathbf{q}} = (0, \pm q_0, 0)$ and $(0, 0, \pm q_0)$ in the limit $m_x \rightarrow \infty$. From Eqs. (37) and (39), we find that when $m_x \gg m_y, m_z$, the mixed states $\Delta(\mathbf{r}) \approx \Delta_n \phi_n^{(k)}(\tilde{x}, \tilde{y}) e^{i\tilde{\mathbf{q}} \cdot \tilde{\mathbf{r}}/\hbar}$ with $(n, \tilde{q}_z) \approx (q_0^2 \tilde{\xi}_H^2 / 2, 0)$ and those with $(n, \tilde{q}_z) = (0, \pm q_0)$ have upper critical fields close to each other.

VI. s -WAVE PAIRING

In this section, we consider an s -wave superconductor as an example. Since $\gamma_s(\mathbf{p}) = 1$, the gap Eq. (10) is identical to that of the isotropic model except that the vector potential \mathbf{A} is scaled as Eq. (7) and the eigenfunctions are distorted. Because the Zeeman field h is not scaled, in contrast to the vector potential \mathbf{A} , the Maki parameter α changes from $\bar{\alpha}$ as expressed in Eq. (32).

We expand the gap Eq. (19) with respect to the operators $\tilde{\eta}$ and obtain the eigenfunctions,

$$\Delta(\mathbf{r}) = \Delta_n \phi_n^{(k)}(\tilde{x}, \tilde{y}) \exp[i\tilde{q}_z \tilde{z}/\hbar], \quad (41)$$

which are indexed by n and \tilde{q}_z . The upper critical-field equation is decoupled into those for each eigenfunction as

$$-\log\left(\frac{T}{T_c^{(0)}}\right) = \pi T \int_0^\infty dt \frac{1}{\sinh(\pi T t)} \int_0^{\pi/2} \sin \theta d\theta \times \left[1 - \cos(ht) \cos \left[\frac{1}{2} \tilde{q}_z \tilde{v}_F t \cos \theta \right] \right] \times \exp \left[-\frac{\tilde{v}_F^2}{16 \tilde{\xi}_H^2} t^2 \sin^2 \theta \right] \sum_{m=0}^n (-1)^m \times \left[\frac{1}{8 \tilde{\xi}_H^2} \tilde{v}_F^2 t^2 \cos^2 \theta \right]^m \frac{n!}{(m!)^2 (n-m)!}. \quad (42)$$

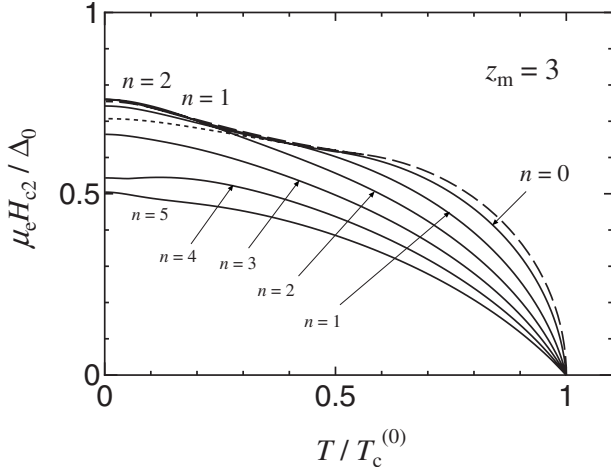


FIG. 1. Temperature dependences of the upper critical fields for $z_m=3$. The solid curves show the upper critical fields for $n=0, 1, 2, 3, 4, 5$. At each temperature, the state that gives the highest critical field is realized as a physical state and the other states are suppressed. The broken and dotted curves show the upper critical fields in the two-dimensional limit and that with the assumption of $\mathbf{q}=\mathbf{0}$, respectively.

The physical upper critical field $H_{c2}(T)$ is the highest solution of H among the solutions of Eq. (42) at each fixed T . In other words, the parameters n and \tilde{q}_z are optimized so that H_{c2} is maximized.

In systems with $\mathbf{A}=\mathbf{0}$, the pure FFLO state occurs as described in Sec. III at low temperatures. When the system is isotropic, there is infinite degeneracy with respect to the direction of \mathbf{q} . We express the \mathbf{q} 's that give the maximum FFLO critical field as $\mathbf{q}=(q_0 \sin \theta \cos \varphi, q_0 \sin \theta \sin \varphi, q_0 \cos \theta)$ with an optimum value of q_0 and arbitrary θ and φ .

In an anisotropic system with $m_x \gg m_y, m_z$, the effective Maki parameter becomes large so that $\alpha \gg \bar{\alpha}$ from Eq. (32) and $\tilde{\xi}_H \gg \xi_H$ from Eq. (16). When m_x is very large, we can make the approximation, $|\mathbf{q}| \approx q_0$. Therefore, from Eqs. (37) and (38), we obtain the optimum value of \tilde{q}_z as

$$\tilde{q}_z = \pm \sqrt{q_0^2 - \frac{2n}{\xi_H^2}} = \pm \sqrt{q_0^2 - \frac{2n\bar{\alpha}}{\alpha\xi_H^2}} \quad (43)$$

for a given n . If we consider that n is always finite for the limit $\alpha \rightarrow \infty$, Eq. (43) is reduced to $\tilde{q}_z = \pm q_0$ and the H_{c2} Eq. (42) is reduced to that of $n=0$. Therefore, when m_x is very large, states with any finite n have upper critical fields very close to that of the state with $n=0$ and $\tilde{q}_z = \pm q_0$. However, because the index n needs to be optimized for each situation, it can be infinitely large. For the state with infinitely large n , such that $n \propto \alpha \xi_H^2$, Eq. (43) results in $|\tilde{q}_z| < q_0$ and $\tilde{q}_z \neq 0$.

This can be also verified by numerical calculations. Figure 1 shows the temperature dependences of the critical fields of states of various n . At each temperature, the state with the highest critical field is physical. It is found that vortex states with $n=0, 1, 2$ occur depending on the temperature, and that the envelope is very close to the curve of the

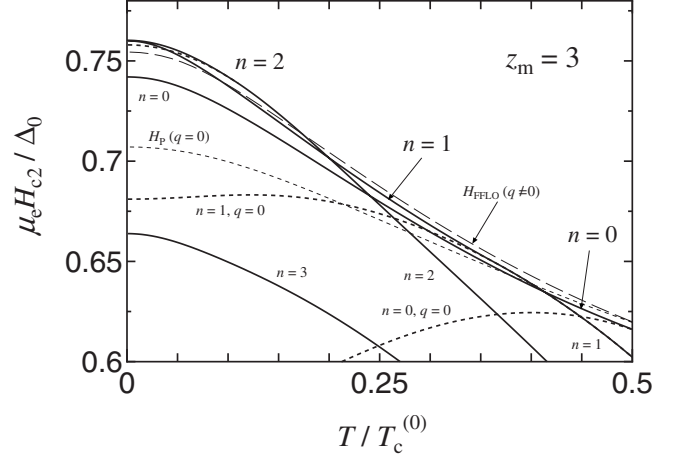


FIG. 2. Temperature dependences of the upper critical fields for $z_m=3$ at low temperature. The solid curves show the upper critical fields for $n=0, 1, 2, 3$. The dotted curves show the field when $\mathbf{q}=\mathbf{0}$ is assumed. The thin broken and dotted curves show the upper critical field in the two-dimensional limit and that with the assumption of $\mathbf{q}=\mathbf{0}$, respectively.

two-dimensional limit. It is also found that the wave vector \mathbf{q} becomes nonzero below $T \approx 0.51 \times T_c^{(0)}$.

Figure 2 shows the behavior of the critical field for each n at low temperatures. We obtain $q_z \neq 0$ below the temperature at which the solid and dotted curves branch off. Interestingly, below $T \approx 0.18 \times T_c^{(0)}$, the upper critical field of the mixed state with $n=2$ and $\tilde{q}_z \neq 0$ exceeds that in the limit $z_m \rightarrow \infty$. We will discuss this later.

Figure 3 shows the z_m ($\propto \alpha$) dependences of the upper critical fields H_{c2} for $n=0, 1, \dots, 8$. It is found that the upper critical fields of all the mixed states with $n \neq 0$ tend to approach that of the mixed state with $n=0$, and slightly exceed it, where z_m is large. At each z_m , the highest upper critical field among those with $n=0, 1, 2, \dots$ is the physical upper critical field. It is found that n of the physical state increases as z_m and the physical critical field is larger than the critical field of the $n=0$ state for $z_m \geq 1.3$.

Figure 4 shows the behavior of H_{c2} for large z_m . After reaching maxima, all the critical fields for $n \neq 0$ decrease and

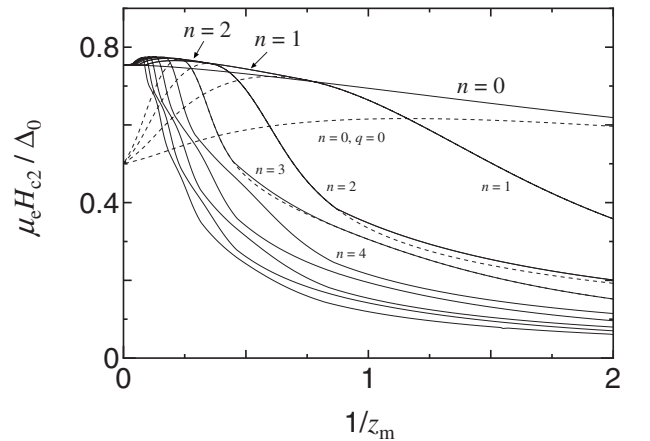


FIG. 3. z_m dependences of the upper critical fields for $n=0, 1, 2, \dots, 8$ at $T/T_c=0.01$. The dotted curves show those obtained when $\mathbf{q}=\mathbf{0}$ is assumed. $\alpha \approx 7.39 \times z_m$.

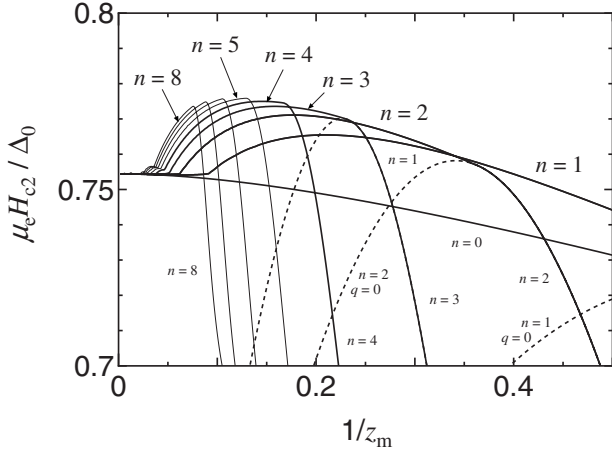


FIG. 4. z_m dependences of the upper critical fields for $n = 0, 1, 2, \dots, 8$ at $T/T_c = 0.01$ in the large z_m region. The dotted curves show those obtained when $q = 0$ is assumed. The thick and thin curves show the results for $n = 0, 1, 2, 3, 4$ and for $n = 5, 6, 7, 8$, respectively. $\alpha \approx 7.39 \times z_m$.

converge to the curve for $n = 0$ in the limit $z_m \rightarrow \infty$. Within the present theory, the physical upper critical field (i.e., the highest field at each z_m) decreases as z_m increases for very large $z_m \geq 7.7$ (i.e., $1/z_m \lesssim 0.13$). This result seems inconsistent with the naive expectation that reducing the orbital effect (for example, by increasing v_F) should reduce the pair-breaking effect. Presumably, for such a large z_m , a first-order phase transition to a mixed state must occur at a critical field higher than that obtained here, and the resultant critical field must increase monotonically with z_m .

VII. SUMMARY AND DISCUSSION

We have examined quasi-two-dimensional type-II superconductors and the two-dimensional limit. When $\mathbf{H} \parallel [0, 0, 1]$, the effective Maki parameter α is proportional to $(m_x m_y / m_z^2)^{1/6}$. Therefore, when $m_x \gg m_z$, the orbital pair-

breaking effect becomes weak and the superconductivity survives up to a higher field, where the FFLO state is favored.

In the mixed state, FFLO modulation occurs in the direction of the magnetic field and is smoothly reduced to that of the pure FFLO state in the two-dimensional limit ($m_x/m_z \rightarrow 0$) when the directions of the magnetic field and \mathbf{q} of the pure FFLO state coincide. In contrast, when their directions differ, it may appear that the mixed state is not reduced to the pure FFLO state continuously. However, in actuality, modulation perpendicular to the magnetic field is realized in large n vortex states. The physical origin of the order-parameter modulation in vortex states with higher Landau levels is the spin-polarization energy as in the pure FFLO state. The mixed states with optimum n have upper critical fields close to that of the pure FFLO state. Hence, a cascade transition occurs when α is large, which is analogous to the exactly two-dimensional system in a tilted magnetic field.^{10-12,27-30} In the two-dimensional limit $m_x \rightarrow \infty$, the Landau-level index n increases as $n \propto \alpha \propto m_x^{1/6}$. As a result, the mixed state indexed by (n, q_{\parallel}) (or the pure vortex state indexed by n when $q_{\parallel} = 0$) is continuously reduced to the pure FFLO state with $\mathbf{q} = (q_{\parallel}, \mathbf{q}_{\perp})$.

The relations (37) and (39) connect the mixed states of Eq. (41) and the pure FFLO state in the two-dimensional limit. The pure FFLO state with $q_{\perp} \neq 0$ corresponds to the mixed state with $n \propto \alpha \rightarrow \infty$, and the upper critical fields of the mixed states converges to that of the pure FFLO state. This behavior has also been confirmed by numerical calculations.

From these results, we can conclude that the FFLO state obtained in a theoretical model without orbital effects may emerge as the vortex states with higher Landau-level indexes in real materials where orbital effects are inevitable. In particular, the order-parameter modulation due to the higher Landau-level index indicates FFLO modulation perpendicular to the magnetic field. We have demonstrated the behavior in quasi-two-dimensional system by taking the two-dimensional limit.

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