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Modulated vortex lattice in high fields and gap nodes

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The mean field vortex phase diagram of a quasi-two-dimensional superconductor with a nodal d -wave pairing and with strong Pauli spin depairing is studied in the parallel field case in order to examine the effect of gap nodes on the stability of a Fulde-Ferrell-Larkin-Ovchinnikov- (FFLO-) like vortex lattice. We find through a heuristic argument and a model calculation with a fourfold anisotropic Fermi surface that the FFLO-like state is relatively suppressed as the field approaches a nodal direction. When taking account of available experimental results together, the present result strongly suggests that the pairing symmetry of CeCoIn₅ should be of d_{xy} type.

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In a recent paper¹ (denoted as I hereafter), we examined the vortex phase diagram of quasi-two-dimensional (Q2D) type II superconductors with strong Pauli paramagnetic (spin) depairing by focusing on the $\mathbf{H} \parallel c$ case with a field $\mathbf{H}$ perpendicular to the superconducting layers. In contrast to earlier work^{2,3} taking account of both the orbital and spin depairing effects of the magnetic field in the clean limit, the orbital depairing was incorporated fully and *nonperturbatively* there,¹ and two results opposite to those suggested previously^{2,3} were found. First of all, the mean field (MF) transition at the $H_{c2}(T)$ line changes from the familiar second-order one to a first-order (MF-FOT) one⁴⁻⁶ at a higher temperature T^* than the region in which a Fulde-Ferrell-Larkin-Ovchinnikov- (FFLO-) like^{7,8} modulated vortex lattice may appear. This feature is consistent with data for CeCoIn₅ in $\mathbf{H} \parallel c$.^{4-6,9} Second, a *second-order* transition curve $H_{\text{FFLO}}(T)$ between such a FFLO-like and ordinary vortex lattices remarkably *decreases* upon cooling. Interestingly, these two results are also consistent with more recent data for CeCoIn₅, suggesting a structural transition to a FFLO state, in $\mathbf{H} \perp c$.⁹⁻¹² A recent ultrasound measurement¹¹ also shows that the suggested FFLO state is, as we argued in I, a kind of vortex lattice. However, it should be further examined theoretically whether this qualitative agreement with the data in $\mathbf{H} \perp c$ is justified or not.

In this paper, the results of the application of the analysis in I to a model for the $\mathbf{H} \perp c$ case are reported. By including the contributions, neglected in previous work,¹⁻³ from the non-Gaussian ($|\Delta(\mathbf{r})|^4$ and $|\Delta(\mathbf{r})|^6$) terms of the Ginzburg-Landau (GL) free energy to the spatial gradient parallel to $\mathbf{H}$, where $\Delta(\mathbf{r})$ is the pair field, we find that the relative position between T^* and the H_{FFLO} line is qualitatively the same as in the $\mathbf{H} \parallel c$ case¹ as long as a spin depairing strength realistic in bulk superconductors is used; and that, at least close to H_{FFLO} , the LO state^{3,8} with periodic nodal planes perpendicular to $\mathbf{H}$ of $|\Delta|$ is more stable than the FF state^{3,7} composed of a phase modulation keeping $|\Delta|$ fixed.

Special attention is paid in this paper to the noticeable in-plane angular dependence of the FFLO curve $H_{\text{FFLO}}(T)$

found in specific heat⁹ and magnetization¹² data for CeCoIn₅: The observed FFLO curve in $H \parallel [110]$ lies at higher temperatures than that in $H \parallel [100]$. This H_{FFLO} anisotropy is much more remarkable⁹ than that of $H_{c2}(T)$ and may give decisive information about the fourfold anisotropy of the gap function. As long as the *in-plane* Fermi velocity anisotropy is negligible, it is heuristically predicted by the following simple argument that a gap anisotropy results in a H_{FFLO} anisotropy: Near the gap nodes where the superconducting gap $\Delta_{\mathbf{k}}$ is small, the coherence length $\xi_{\mathbf{k}} \approx \hbar v_F / \Delta_{\mathbf{k}}$ defined locally in the $\mathbf{k}$ space is longer.¹³ The orbital limiting field $H_{\text{orb}}(0)$ is inversely proportional to the square of the averaged coherence length in the plane perpendicular to $\mathbf{H}$ and hence is minimal when $\mathbf{H}$ is directed along the fourfold symmetric gap nodes (or minima). Since a higher H_{orb} will lead to a relatively stronger effect of spin depairing, the FFLO curve and T^* , induced by the spin depairing, are expected to lie at higher temperatures when $\mathbf{H}$ is located along a gap maximum. If we compare the expected H_{FFLO} anisotropy with the observations^{9,12} in CeCoIn₅, we inevitably reach the conclusion that, in agreement not with the original argument⁴ favoring a $d_{x^2-y^2}$ pairing just as in high- T_c cuprates but with a recent report on low- H specific heat data,¹⁴ a node (or minimum) of the gap function of CeCoIn₅ is located along the $[100]$ direction, implying a d_{xy} pairing state. Below, we will show how this conclusion is reinforced through a microscopic derivation of $H_{\text{FFLO}}(T)$ taking account of a possible in-plane fourfold anisotropy of the Fermi surface (FS). The present result might require a serious change in the picture of the pairing mechanism of CeCoIn₅ based upon similarities of the normal state properties, including the presence of antiferromagnetic fluctuation, to the high- T_c cuprates.¹⁵

First, let us sketch an outline of the MF analysis¹ for $\mathbf{H} \parallel c$. Throughout this paper, we assume $\mathbf{H} = H\hat{x}$ and the d -wave gap function $w_{\phi} = \sqrt{2} \cos(2\phi)$ or $\sqrt{2} \sin(2\phi)$, where ϕ is the azimuthal angle in the a - b plane. Within the lowest ($N=0$) Landau level (LL), the GL free energy density in the MF approximation takes the form

$$\begin{aligned}\mathcal{F}_{\text{MF}} = N(0) & \left[a_0(Q) \langle |\Delta_Q^{(0)}|^2 \rangle + \frac{V_4(Q)}{2} \langle |\Delta_Q^{(0)}|^4 \rangle \right. \\ & \left. + \frac{V_6(Q)}{3} \langle |\Delta_Q^{(0)}|^6 \rangle \right] \\ & \simeq c_0 + c_2 Q^2 + c_4 Q^4.\end{aligned}\quad (1)$$

The essential part of the MF analysis in I is to derive the coefficients, a_0 , V_4 , V_6 , c_2 , and c_4 by starting from the weak-coupling BCS model with a Zeeman (Pauli paramagnetic) term. Here, $N(0)$ is the averaged density of states (DOS) at the Fermi level, and $\langle \rangle$ is the spatial average on y and z . $\Delta(\mathbf{r})$ was expanded in terms of the LLs as $\Delta(\mathbf{r}) = \sum_{N \geq 0} \Delta_Q^{(N)}(y, z) u_Q(x)$, and the higher LLs were neglected above. For the LO (FF) state, $u_Q(x)$ takes the form $\cos(Qx)$ [$\exp(iQx)$]. A Q2D FS with a circular form in the y - z plane was assumed, although in-plane anisotropy will conveniently be included as the ϕ dependence of the Fermi velocity and DOS [see Eq. (5) below]. For an example, $a_0(Q)$ is, after performing the $\mathbf{k}$ integrals and introducing a parameter integral, expressed by

$$\begin{aligned}N(0)a_0(Q) = & \left\langle u_Q^*(x) \left(\frac{1}{|g|} - 2\pi T \int_0^\infty d\rho \frac{\cos(2\mu_0 H \rho)}{\sinh(2\pi T \rho)} \right. \right. \\ & \left. \left. \times g^{(0)}(\rho, -i\partial_x) \right) u_Q(x) \right\rangle_x,\end{aligned}\quad (2)$$

where $\langle \rangle_x$ denotes the spatial average on x , $\mu_0 H$ is the Zeeman energy, and $N(0)|g|$ is the dimensionless pairing interaction strength. The function $g^{(0)}(\rho, -i\partial_x)$ has the form

$$g^{(0)}(\rho, -i\partial_x) = N(0) \exp(-\rho^2 v_F^2 / 4r_H^2) \cos(-i\rho v_F \partial_x), \quad (3)$$

where r_H is the magnetic length and v_F the Fermi velocity. The extension a_N of a_0 to the N th LL is given by multiplying Eq. (3) by $\mathcal{L}_N(\rho^2 v_F^2 / 2r_H^2)$, if just terms diagonal with respect to the LLs are kept, where $\mathcal{L}_N(x)$ the N th Laguerre polynomial. The coefficients $V_4(Q)$ and $V_6(Q)$ are derived in a similar manner to above. The coefficients c_2 and c_4 arise from the Q dependences of a_0 , V_4 , and V_6 .

The onset T^* of the MF-FOT at H_{c2} is determined by $V_4(0)=0$ irrespective of the details of higher-order non-Gaussian terms of the GL free energy, while $H_{\text{FFLO}}(T)$ is defined by $c_2=0$ under the condition $c_4>0$. We have verified that the latter condition is always satisfied throughout the computations in the present work, so that the resulting $H_{\text{FFLO}}(T)$ is a second-order transition line. If the effective strength of spin depairing $\mu_0 H_{\text{orb}}^{2D} / (2\pi k_B T_{c0})$ is of order unity or larger, a phase diagram derived numerically in this manner includes an $H_{\text{FFLO}}(T)$ line decreasing upon cooling, where $\mu_0 H$ is the Zeeman energy, and H_{orb}^{2D} is the orbital limiting field in the 2D limit. In Ref. 1 where the V_4 and V_6 contributions to c_2 were neglected, the LO and FF states had the same H_{FFLO} line, while we find that the instability of the straight vortex lattice leading to the LO vortex state^{3,8} occurs at a slightly higher temperature than that to the FF state.^{3,7} Hence, at least close to $H_{\text{FFLO}}(T)$, the LO state becomes the ground state in $H_{\text{FFLO}} < H < H_{c2}$. Further, we find that the V_6

contribution to c_2 is quantitatively negligible, while the H_{FFLO} line is pushed down by the corresponding V_4 contribution to a lower-temperature region in which H_{c2} and the vortex state just below it are described by the $N=1$ LL. Thus, at least within the weak-coupling BCS model, a FFLO state in $\mathbf{H} \parallel c$ rarely occurs because such a $N=1$ LL vortex lattice has no FFLO-like modulation.¹ We guess that a slight specific heat anomaly⁹ in CeCoIn₅ in $\mathbf{H} \parallel c$ at low enough temperatures may be rather due to a transition between straight vortex lattices in the $N=0$ and $N=1$ LLs. A detailed study of this transition into an $N=1$ LL state will be reported elsewhere.

Now, let us turn to the $\mathbf{H} \perp c$ case. Although, in principle, the above analysis can be extended to a Q2D system with a cylindrical FS under $\mathbf{H}$ perpendicular to the cylindrical axis, we have chosen to work in an elliptic FS elongated along the $z(\parallel c)$ axis and with the dispersion relation $\varepsilon_k = \hbar^2 \sum_{j=x,y,z} \gamma_j^2 k_j^2 / (2\bar{m})$ under $\mathbf{H} \parallel \hat{x}$ in order to make numerical calculations more tractable, where $\gamma_x = \gamma_y = \gamma^{-1/2}$, and $\gamma_z = \gamma$ with $\gamma \geq 1$ and a constant $\bar{m}$. We expect the case with a moderately large γ value to *qualitatively* describe essential features in the realistic Q2D case. By isotropizing the $\mathbf{k}$ vector as $k_j = \gamma_j k_F \hat{r}_j$, where $\hat{\mathbf{r}} = (\cos \phi \sin \theta, \sin \phi \sin \theta, \cos \theta)$ is the unit vector in spherical coordinates, the velocity $\mathbf{v}$ on the FS is written as $v_j = \gamma_j^{-1} v_F \hat{r}_j$. The Jacobian $\sqrt{\gamma \sin^2 \theta + \gamma^{-2} \cos^2 \theta}$ accompanying the angular integral along the FS is exactly canceled by the angular dependence of the DOS, $N(\theta) = N(0) v_F / \sqrt{\sum_j v_j^2}$. Again, the *in-plane* (fourfold) anisotropy of the FS will first be neglected. Then, the GL free energy within the $N=0$ LL takes the form of Eq. (1), and the function $g^{(0)}(\rho, -i\partial_x)$ appearing in $a_0(Q)$ [see Eq. (3)] is replaced in the present case by

$$\begin{aligned}g_{\parallel}^{(0)}(\rho, -i\partial_x) = & \int \frac{\sin \theta d\theta d\phi}{4\pi} N(0) |w_\phi|^2 \exp(-\rho^2 \bar{v}_{yz}^2 / 4r_H^2) \\ & \times \cos(-i\rho v_x \partial_x),\end{aligned}\quad (4)$$

where $\bar{v}_{yz}^2 = \tilde{\gamma} \gamma^{-1} v_y^2 + \gamma \tilde{\gamma}^{-1} v_z^2$. The parameter $\tilde{\gamma}$ is insensitive to the uniaxial anisotropy γ but dependent on T and needs to be determined by maximizing $H_{c2}(T)$. By focusing on the low- T region, we find that $\tilde{\gamma}$ takes a value between 0.4 and 0.5 depending on the relative angle between $\mathbf{H}$ and the nearest nodal direction. Using this parameter, the anisotropy in spatial variations of $\Delta(\mathbf{r})$ within the y - z plane is given by $\gamma / \tilde{\gamma}$. Except for the modifications indicated above, the corresponding quartic and sixth-order terms of the GL free energy are derived by closely following the analysis in I. We choose $\alpha_{\parallel} = \mu_0 H_{\text{orb}}^{(\gamma=1)}(0) / k_B T_{c0}$ as a measure of the spin depairing strength in $\mathbf{H} \perp c$, where $H_{\text{orb}}^{(\gamma=1)}(0)$ is the orbital limiting field in the isotropic case.

In Fig. 1, the resulting phase diagram is shown to illustrate how the $H_{\text{FFLO}}(T)$ position depends upon the relative angle between $\mathbf{H}$ and the nodal directions. Thin solid (chain) curves are defined by $a_N(0)=0$, and the $H_{c2}(T)$ in $T > T^*$ in each case is given by each $a_0(0)=0$ line. In agreement with the heuristic argument given earlier, $H_{\text{FFLO}}(T)$ and T^* are shifted to higher temperatures as the in-plane field is directed along a gap maximum, reflecting an enhanced spin depairing

FIG. 1. H - T mean field phase diagram obtained using $\gamma=3$ and with no in-plane FS anisotropy. The transition or crossover positions in $\mathbf{H}\parallel$ gap maximum ($\parallel$ gap node or minimum) are expressed by the solid circle and filled circle (chain curves and open circle). The dotted curve and open triangle denote, respectively, the MF-FOT line and the position at which the two solid curves $a_N(0)=0$ in $N=0$ and 1 merge with each other.

in this field configuration. As in $\mathbf{H}\parallel c$, the FFLO state at least close to H_{FFLO} has the LO-like variation. By combining our numerical calculations with an analytical calculation with the orbital depairing perturbatively included, we have verified that such an in-plane H_{FFLO} anisotropy is absent without the orbital depairing (i.e., when $\alpha_{\parallel}=\infty$) and *monotonically* increases with decreasing $\alpha_{\parallel}$. In contrast, it is not easy to properly predict the corresponding anisotropy (in-plane angular dependence) of the $H_{c2}(T)$ curve. First, the depression of H_{c2} due to the spin depairing is larger as the corresponding $H_{\text{orb}}(0)$ is higher, and hence the H_{c2} magnitude may not have a monotonic $\alpha_{\parallel}$ dependence. Second, the MF-FOT line of H_{c2} is directly determined by the details of the non-Gaussian terms other than the quartic one in the GL free energy¹ and hence is quantitatively affected by our assumption of keeping the non-Gaussian terms only up to $|\Delta|^6$ in Eq. (1). Actually, the rapid increase of the MF-FOT line on cooling *just* below T^* arises due to an extremely small $V_6(0)$ near T^* and might flatten if we could numerically include the $|\Delta|^8$ and higher-order terms. In contrast, the V_6 contribution to c_2 [i.e., to $H_{\text{FFLO}}(T)$] was negligible, as in the $\mathbf{H}\parallel c$ case, consistent with the smallness of $V_6(0)$ mentioned above. We expect that the $H_{\text{FFLO}}(T)$ curve is less sensitive to the neglect of the $|\Delta|^8$ and higher-order GL terms. For these reasons, we will focus hereafter on T^* and H_{FFLO} , which directly measure the (effective) spin depairing strength. The resulting anisotropies of T^* and H_{FFLO} in Fig. 1 qualitatively agree with those of CeCoIn_5 in^{9,12} $\mathbf{H}\perp c$ if a gap node (or minimum) is located along $[100]$. As already mentioned, the MF-FOT line in the $N=0$ LL needs to lie above the corresponding $a_1(0)=0$ line in order for $H_{\text{FFLO}}(T)$ to be realized as a transition line. As Fig. 1 shows, this condition manages to be satisfied, in contrast to the $\mathbf{H}\parallel c$ case.

In order to examine how the result in Fig. 1 is affected by the *in-plane* FS anisotropy, let us next introduce it as a Fermi velocity anisotropy in a similar manner to Ref. 16:

FIG. 2. Results corresponding to Fig. 1 in the cases (a) ($|\beta|=0.2$) and (b) ($|\beta|=0.1$) defined in the text.

$$v_F \rightarrow v_F(\phi) = v_F[1 + \beta \cos(4\phi)], \quad (5)$$

where $|\beta| < 1$, accompanied by the replacement $N(0) \rightarrow N(0)v_F/v_F(\phi)$ in any angular integral [see Eq. (4)]. Apart from these replacements in our calculation, the derivation of phase diagrams is quite the same as that of Fig. 1. When $\beta > 0$ (< 0), the Fermi velocity becomes maximal (minimal) along $\hat{x}$. By combining these two cases with the two candidates $\sqrt{2} \cos(2\phi)$ and $\sqrt{2} \sin(2\phi)$ for w_ϕ , we have four different cases of the relative anisotropies under a fixed $\mathbf{H}\parallel\hat{x}$. We will classify them into two categories, (a) $w_\phi = \sqrt{2} \cos(2\phi)$ with $\beta < 0$ and $w_\phi = \sqrt{2} \sin(2\phi)$ with $\beta > 0$, and (b) $w_\phi = \sqrt{2} \cos(2\phi)$ with $\beta > 0$ and $w_\phi = \sqrt{2} \sin(2\phi)$ with $\beta < 0$. This classification is motivated by the result¹⁶ that, in the category (a), the Fermi velocity anisotropy and the pairing anisotropy favor two different orientations, competing with each other, of the square vortex lattices to be realized in fourfold anisotropic d -wave superconductors in $\mathbf{H}\parallel c$, while such a competition does not occur in (b). In Fig. 2, the resulting phase diagrams for the categories (a) and (b) are given. In the case (a), the angular dependences of H_{FFLO} and T^* are weakened by the FS anisotropy compared with those in Fig. 1, while the opposite tendency is seen in the case (b). This result can be understood by noting that the orbital depairing strength locally in the $\mathbf{k}$ space is measured in the

present case by v_y^2 in Eq. (4) (note that, in the 2D limit, v_z^2 is absent there). By focusing on the case with $\mathbf{H}$ parallel to a gap node and noting $|w_\phi|^2$ in the integrand of Eq. (4), one will notice that a nonzero $|\beta|$ tends to increase (decrease) the contributions of v_y^2 , on average, when $\beta < 0$ ($\beta > 0$). Thus, an enhanced orbital depairing in $\mathbf{H}$ parallel to a node of case (b) additionally reduces H_{FFLO} so that the difference between the two cases in Fig. 2 follows. Bearing in mind the general character of this interpretation, we believe that the results in Fig. 2 would not be qualitatively changed by a refinement of the microscopic description.

The above results commonly show an $H_{\text{FFLO}}(T)$ line shifting to higher temperatures as the in-plane field approaches a gap maximum and, compared with the data for CeCoIn_5 ,^{9,12} imply a d_{xy} state as the pairing state of this material. Although one might consider the possibility of $d_{x^2-y^2}$ pairing based on the fact that an extremely strong FS anisotropy in the case (a) may reverse the anisotropies of T^* and H_{FFLO} , such a strong FS anisotropy of the case (a) should result¹⁶ in a square vortex lattice with an orientation due to the FS anisotropy and hence contradicts not only the specific heat data¹⁴ but the observed orientation¹⁷ of the $\mathbf{H}||c$ square vortex lattice. Therefore, inclusion of the FS anisotropy *reinforces* our conclusion favoring a d_{xy} pairing, although a moderate

FS anisotropy competitive in $\mathbf{H}||c$ with the gap anisotropy [i.e., of the case (a)] is needed for quantitative understanding.

In conclusion, the mean field phase diagram of a type II superconductor with strong Pauli paramagnetic depairing and with a fourfold symmetric d -wave pairing was qualitatively studied in the parallel field case. The region in which the FFLO vortex phase appears is enlarged when the in-plane field is directed along a gap maximum. This result is reinforced by including in-plane FS anisotropies and strongly suggests a d_{xy} pairing as the best candidate for the gap function of CeCoIn_5 in spite of the electronic similarities¹⁵ to that of high- T_c cuprates. A reinterpretation of thermal conductivity data by Izawa *et al.*⁴ can be seen in Ref. 14. The present theory should be applicable to examining the pairing state of other materials, such as organic material,^{18,19} showing a remarkable Pauli paramagnetic depairing.

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