Anisotropy of thermal conductivity and possible signature of the Fulde-Ferrell-Larkin-Ovchinnikov state in $CeCoIn_5$

C. Capan,[1] A. Bianchi,[1] R. Movshovich,[1] A. D. Christianson,[2] A. Malinowski,[1] M. F. Hundley,[1] A. Lacerda,[2] P. G. Pagliuso,[3] and J. L. Sarrao[1]

[1]*Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA*

[2]*National High Magnetic Field Laboratory, Los Alamos, New Mexico 87545, USA*

[3]*Instituto de Fisica Gleb Wataghin, UNICAMP, 13083-970, Campinas, Brazil*

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We have measured the thermal conductivity of the heavy-fermion superconductor $CeCoIn_5$ in the vicinity of the upper critical field, with the magnetic field perpendicular to the c axis. Thermal conductivity displays a discontinuous jump at the superconducting phase boundary below critical temperature $T_0 \approx 1$ K, indicating a change from a second- to first-order transition and confirming the recent results of specific heat measurements on $CeCoIn_5$. In addition, the thermal conductivity data as a function of field display a kink at a field H_k below the superconducting critical field, which closely coincides with the recently discovered anomaly in specific heat, tentatively identified with the appearance of the spatially inhomogeneous Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) superconducting state. Our results indicate that the thermal conductivity is enhanced within the FFLO state, and call for further theoretical investigations of the order parameter's real-space structure (and, in particular, the structure of vortices) and of the thermal transport within the inhomogeneous FFLO state.

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I. INTRODUCTION

Over the last several years there has been renewed interest in the spatially inhomogeneous Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state. The FFLO state was predicted as early as the mid-1960s[1,2] to occur in a clean Type II superconductor in high magnetic fields, when the Zeeman energy becomes comparable to the condensation energy. Then, Pauli limiting[3] plays an important role in defining both the superconducting critical field H_{c2} and the temperature T_0 below which the FFLO state is expected to appear.[4] Within the FFLO state, spin-up and spin-down electrons of a spin-singlet superconductor can only stay bound if the Cooper pair has a finite momentum. As a result, the FFLO state is formed with a spatially oscillating order parameter. The exact description of the corresponding phase diagram for both s- and d-wave superconductors,[5–8] as well as the stable spatial structures in two and three dimensions in the presence of vortices,[5,9–11] are subjects of intense theoretical investigations.

In spite of the straightforward nature of the theoretical prediction, the experimental observation of the FFLO state has turned out to be a difficult task. In fact, very few superconductors fulfill the necessary conditions for the formation of an FFLO state. The relative importance of the Pauli and orbital limiting can be described by the so-called Maki parameter $\alpha = \sqrt{2}(H_{c2}^0/H_p)$. H_{c2}^0 is the orbital limiting field due to the kinetic energy of the superconducting currents around the vortex cores, commonly derived from the slope of the experimentally determined H-T phase boundary at T_c, as $H_{c2}^0 = 0.7(dH_{c2}/dT)|_{T_c}$.[12] $H_p = \sqrt{2}\Delta_0/g\mu_B$ is the Pauli limiting field due to the potential energy of the electron's spin (Zeeman energy). Here Δ_0 is the zero temperature value of the superconducting gap, g is the electron's effective g factor, and μ_B is the electron's Bohr magneton.[3] Within the calculation of Ref. 4, α must be greater than 1.8 for the FFLO state to be realized.

There are several classes of materials that are traditionally thought of as potential candidates for the formation of the FFLO states. These include low-dimensional organic superconductors and heavy-fermion superconductors. The low-dimensional organic superconductors are promising, because when the field is applied within the conducting planes of a two-dimensional (2D) superconductor, the orbital limiting is suppressed entirely, as the diamagnetic screening currents can only flow within the plane. In such a case, Pauli limiting determines the critical field $H_{c2} = H_p$, the Maki parameter $\alpha = \infty$, and the FFLO state should be stabilized below the critical temperature $T_0 \approx 0.55 T_c$ for magnetic field close to H_{c2}. This straightforward prediction led to a number of experimental investigations of the superconducting properties of lower-dimensional organic superconductors. Several investigators suggested the existence of FFLO states, e.g., based on the superconducting phase diagram[13] or the magnetothermal transport properties.[14]

Heavy-fermion superconductors are also attractive because of their potentially large values of the Maki parameter. Here, the heavy electron masses lead to low Fermi velocities of the quasiparticles and, in turn, to relatively ineffective orbital limiting, or large H_{c2}^0. For this reason, heavy-fermion materials have had their deserved share of attention, and the features in the magnetization of $CeRu_2$[15] and the phase diagram of UPd_2Al_3[16] were taken as possible signatures of the FFLO states.

However, and not for the lack of effort, there is to date no accepted definitive proof of the existence of the FFLO state in any of the systems described above. In this paper we describe the magnetothermal studies of a very strong candidate to possess the FFLO state, the heavy-fermion superconductor $CeCoIn_5$.

$CeCoIn_5$ is a clean *d*-wave superconductor,[17–19] with a T_c of 2.3 K, the highest among the Ce-based heavy fermions. It exhibits a layered structure of alternating $CeIn_3$ and $CoIn_2$ planes, suggesting a possible quasi-2D electronic nature of this compound. This is supported by the experimental observation of a quasicylindrical sheet in the Fermi surface of $CeCoIn_5$ via de Haas-van Alphen studies.[20] The estimated Maki parameter α is about 3.6 for the field perpendicular to the $CeIn_3$ planes $(H \parallel c)$,[21] and close to 4.5 for the field-in-the-plane orientation $(H \perp c)$.[22] Thus, $CeCoIn_5$ is a good candidate for the formation of an FFLO state. As new results accumulate, there is growing evidence that the FFLO state may indeed be realized in $CeCoIn_5$. First, the superconducting phase transition at low temperatures becomes first order, which is manifested by the sharp specific heat anomaly at T_c in the specific heat for both $H \parallel c$[21] and $H \perp c$[23] orientations. In addition, steps in magnetostriction[21,24] and in magnetization,[25,26] and a step in the thermal conductivity for $H \parallel c$,[19] are observed. The change of the superconducting anomaly from second to first order[21] was interpreted as a realization of the Maki scenario, which attributes this change to a strong Pauli limiting effect in a Type II superconductor.[27,28] When the field was applied within the *a-b* plane $(H \perp c)$, a second anomaly in the specific heat was observed within the superconducting state,[23,29] indicating a phase transition into a new superconducting state, tentatively identified as the FFLO state in $CeCoIn_5$. In addition, steps in magnetization of $CeCoIn_5$ were observed by Radovan *et al.*,[29] and were interpreted as an indication of the multiquantum vortices expected under certain circumstances for the 2D superconductors within the FFLO state.[9–11] The validity of such an interpretation is at present under debate.[30,31] On the theoretical front, a recent analysis of a linearly increasing H_{c2} at the lowest temperatures[22] suggested that this too can be accounted for within an FFLO scenario for $CeCoIn_5$.

While these results make the FFLO scenario a very appealing one for $CeCoIn_5$, there is no clear evidence so far for spatially inhomogeneous superconductivity in the second low-temperature phase. A recent study revealed an increased penetration depth at the lower transition, which was interpreted as a decrease of the superfluid density due to the formation of the FFLO state.[32] An ultrasound investigation of the high-field state revealed the decrease of the sound velocity from that in the vortex state, which was also presented in support of the FFLO nature of that state.[33]

Here we present our results of magnetothermal transport measurements in $CeCoIn_5$ with the field applied within the $CeIn_3$ planes $(H \perp c)$. The LO structure, which emerged from early theoretical work,[2] is a collection of periodically spaced planes of nodes of the superconducting order parameter that are perpendicular to the direction of the applied field. The LO order parameter is described as $\psi(\vec{r}) = \psi_0 \cos(\vec{q}\vec{r})$, oscillating in space along the direction of vector $\vec{q} \parallel H$, as illustrated in Fig. 1. In recent years, thermal conductivity was used effectively to probe the anisotropy of the order parameter in unconventional superconductors, specifically, the structure of the nodes in *k* space. This is due to the fact that

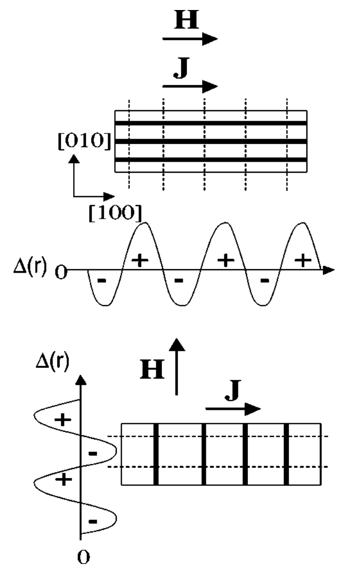

FIG. 1. Illustration of the vortex structure (solid lines) and the FFLO modulation (dashed lines) with the field parallel (top) and perpendicular (bottom) to the heat current.

normal quasiparticles, which are easily excited along the nodal directions, do carry heat, whereas the superconducting background does not.[19,34,35] The inherent anisotropy of the LO state makes thermal conductivity an attractive tool for identifying it. For the vast majority of clean superconductors, thermal conductivity drops as the sample enters the superconducting state, due to the opening of the superconducting gap over the entire Fermi surface and the resulting rapid decrease of the number of normal quasiparticles which carry heat. One would then expect the thermal conductivity within the nodal planes to be higher than in the rest of the sample. Thermal conductivity would then be larger when the heat current $\vec{Q}$ flows along the nodal planes, and perpendicular to the applied magnetic field $(\vec{Q} \perp H)$, than when the heat flow is parallel to the magnetic field $(\vec{Q} \parallel H)$. Note that this anisotropy is of the opposite sign from that due to the vortices, with higher thermal conductivity along the vortices $(\vec{Q} \parallel H)$,[36] and therefore the two contributions should be easily differentiated, especially if the contribution due to the 2D planes turns out to dominate that from the one-dimensional (1D) vortices. This picture unfortunately turns out to be too simplistic for the case of $CeCoIn_5$ and is complicated by several effects described below.

Motivated by the idea described above, we measured thermal conductivity in $CeCoIn_5$ at low temperatures, in the vicinity of the upper critical field, with the magnetic-field-oriented in plane, using a dilution refrigerator in the 20 T magnet of the NHMFL facility at LANL. The sample, a needle-like single crystal with dimensions of 2.18, 0.28, and 0.064 mm, was flux grown at LANL, as described in Ref. 17. After a chemical etch and polishing to remove the residual free indium, the sample had resistivity of 3.2 $\mu\Omega$ cm at 4.2 K and a RRR of $\rho(300\ K)/\rho(4.2\ K) = 9.4$. The experimental setup for thermal conductivity consists of a heater attached to one end of the sample, and two RuO_2 thermom-

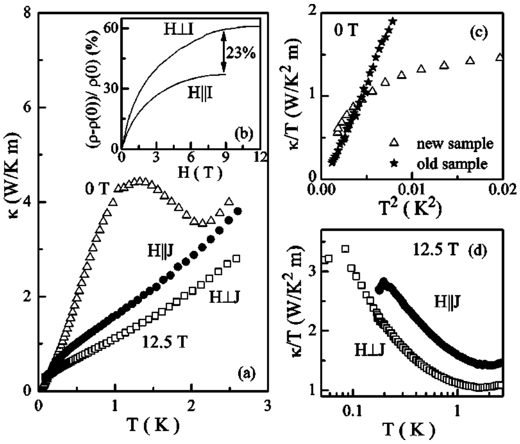

FIG. 2. (a) Thermal conductivity κ vs temperature of $CeCoIn_5$ at zero field and 12.5 T, with the field parallel (●) and perpendicular (□) to the heat current. Inset (b) In-plane magnetoresistance of $CeCoIn_5$ with field perpendicular (top curve) and parallel (bottom curve) to the current. (c) Thermal conductivity of the present sample (△) and the sample used in Ref. 18 (⋆), normalized to the value at T_c of κ=2 W/Km. (d) Thermal conductivity divided by temperature for the field of 12 T parallel (●) and perpendicular (□) to the heat current, showing diverging κ/T as $T\rightarrow 0$.

eters in thermal contact with the sample at two points along its length. A dc heat current flows along the [100] direction of the sample (along its longest dimension). The resulting temperatures of both thermometers are measured using an LR700 resistance bridge. The measurements were performed with magnetic field applied parallel and perpendicular ($H\|[010]$) to the heat current. The two thermometers were calibrated at each field against a reference thermometer placed in a field-free region.

II. THERMAL CONDUCTIVITY IN THE NORMAL STATE AND ZERO FIELD

Figure 2 shows the thermal conductivity of $CeCoIn_5$ as a function of temperature up to 2.5 K at zero field and at 12.5 T, for field oriented parallel and perpendicular to the heat current. One notices a substantial drop in thermal conductivity induced by the applied field above T_c, well as a significant difference between the two field orientations. It is possible to account for the normal-state anisotropy by considering the magnetoresistance of $CeCoIn_5$, displayed in the inset of Fig. 2. The longitudinal and transverse magnetoresistances at 2.5 K and 9 T, with an in-plane current, are 37% and 60%, respectively. This gives a difference in magnetoresistance $(\rho_{\perp}-\rho_{\|})/\rho(0)$ of 23% between the field oriented parallel and perpendicular to the electrical current. If we determine the difference of thermal conductivity between both field orientations, $(\kappa_{\|}-\kappa_{\perp})/\kappa(0)$ at 2.5 K and 12.5 T, we find a value of 21%, very close to the anisotropy of magnetoresistance. Thus, the anisotropy of the heat transport in the normal state can be accounted for by the difference in the quasiparticle scattering for the two field orientations. This is not surprising, because the quasiparticle contribution dominates the heat transport in $CeCoIn_5$.[18] Therefore, in order to highlight the differences between the superconducting states for the two orientations of the magnetic field studied, in what follows, we often present the thermal conductivity data scaled by the values in the normal state at 12.5 T.

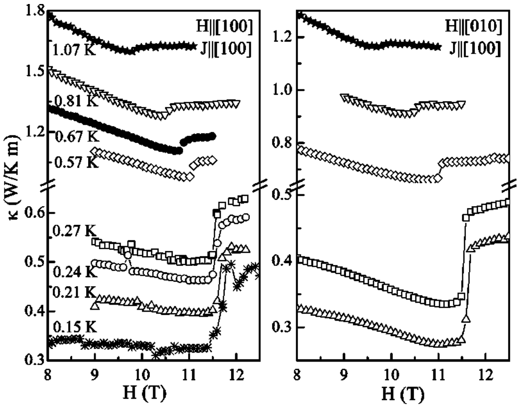

FIG. 3. Thermal conductivity vs magnetic field of $CeCoIn_5$ between 8 and 12.5 T. Left panel: $H\|J$. Right panel: $H\perp J$, (⋆) 1.07, (▽) 0.81, (●) 0.67, (◇) 0.57, (□) 0.27, (○) 0.24, (△) 0.21, and (∗) 0.15 K.

The normal-state thermal conductivity for the field of 12.5 T, just above the superconducting critical field of 12 T, is displayed in Fig. 2(d) as κ/T versus T on a log-log plot. κ/T appears to diverge, reflecting the possible presence of the quantum critical point (QCP) in $CeCoIn_5$, suggested by both specific heat and resistivity measurements.[37] A similar behavior with the QCP lying very close to the superconducting critical field was observed for $H\|[001]$.[38–40]

It is interesting to compare the zero-field thermal conductivity measurements on the present sample to the previously published data,[18] in particular the zero-temperature limit of κ/T. The zero-field data are displayed in Fig. 2(c) as κ/T versus T^2. In spite of the very large difference in the peak of κ/T below T_c, the low-temperature values below 70 mK are rather close to each other. We take this as an indication that the low-temperature behavior might be reflecting a universal, independent of the impurity concentration, limit of thermal conductivity expected for unconventional superconductors with lines of nodes in the superconducting energy gap, in accordance with the original interpretation of the low-temperature thermal transport in $CeCoIn_5$.[18]

III. THERMAL CONDUCTIVITY IN THE VORTEX STATE

The thermal conductivity data in the low-temperature high-field part of the phase diagram are displayed in Fig. 3. The transition to the normal state is marked by a pronounced jump in the thermal conductivity at the lowest temperatures. The jump in thermal conductivity confirms the first-order nature of the superconducting transition, reported previously on the basis of the specific heat measurements.[23] The first-order nature of the superconducting transition for $H\|[001]$ was deduced on the basis of the thermal conductivity measurements by Izawa *et al.*[19]

The absolute slope of the thermal conductivity versus magnetic field in the vortex state is reduced as the tempera-

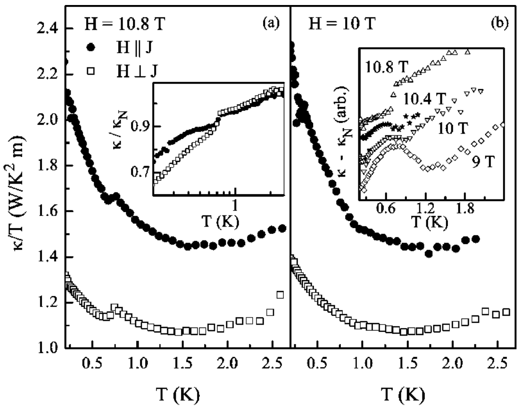

FIG. 4. Temperature dependence of thermal conductivity κ in $CeCoIn_5$ between 50 mK and 2.5 K. (●) $H \| J$; (□) $H \perp J$. (a) 10.8 T, inset: data normalized to thermal conductivity at 12.5 T in the normal state; (b) 10 T, inset: κ-κ_N vs temperature at 9 (◇), 10 (▽), 10.4 (⋆), and 10.8 T (△). The values of κ at 12.5 T have been used as κ_N, and the data for different fields have been shifted vertically for clarity.

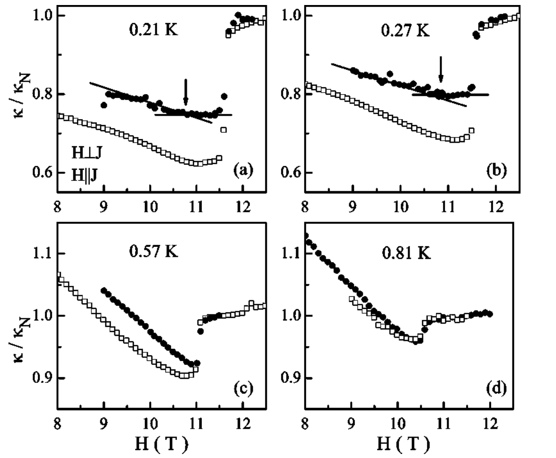

FIG. 5. Normalized thermal conductivity vs magnetic field of $CeCoIn_5$ for $H\|J$ (●) and $H \perp J$ (□). Upper left: 0.21 K, upper right: 0.27 K, lower left: 0.57 K, lower right: 0.81 K. The data for both orientations have been normalized by the corresponding normal-state values at 12.5 T. The arrows indicate the positions of the kinks for $H\|J$.

ture is lowered. This is likely a result of the competition between the increase with magnetic field of both the density of states and the quasiparticle scattering rate, due to vortices. The Volovik effect, or Doppler shift of the quasiparticle energies, results in the $\sqrt{H}$ increase of the density of states in d-wave superconductors in low magnetic fields.[41,42] As the field is increased, so is the number of scattering vortices, and this has the effect of decreasing thermal conductivity in higher magnetic field. At higher temperature, the number of quasiparticles is largely determined by temperature and the contribution from the Volovik effect loses its significance. The vortex scattering effect then dominates thermal transport, resulting in the decrease of thermal conductivity with magnetic field. On the other hand, at low temperature, the Volovik effect dominates the thermal broadening and efficiently competes with the reduction of thermal conductivity, due to the vortex scattering. In $CeCoIn_5$ this results in a slower decrease of thermal conductivity with increasing magnetic field at lower temperatures, as displayed in Fig. 3.

Thermal conductivity for both parallel and perpendicular field orientations is depicted in Fig. 4 as a function of temperature for several values of field between 50 mK and 2.5 K. The inset in Fig. 4(a) shows the data for 10.8 T, after it was normalized by the thermal conductivity at 12.5 T (in the normal state) to highlight the step-like increase of κ as the system goes into the superconducting state. The inset in Fig. 4(b) shows the data after subtraction of the normal-state 12.5-T data. The enhancement of thermal conductivity below T_c at zero field, due to an increased quasiparticle mean free path, shown in Fig. 2(a), can still be clearly resolved at 9 T. An apparent small rise in thermal conductivity at the superconducting transition at 10 and 10.4 T is due to the competition between the increase of the thermal conductivity in the normal state with increasing magnetic field and a drop in thermal conductivity as the system becomes superconducting when magnetic field is swept (see Fig. 3). When temperature is swept, the effect of the increasing κ in the normal state prevails at 10 and 10.4 T. The data for 10.8 T show a pronounced step (similar to the data for the field sweeps) associated with the first-order nature of the superconducting transition, which dominates at this field.

Figure 5 displays the thermal conductivity normalized by the value in the normal state at T_c at several temperatures for the two field orientations between 8 and 12 T. The thermal conductivity decreases with increasing field in the mixed state and increases slightly in the normal state. The normalized values of thermal conductivity for $H\|J$ orientation are higher than that for $H \perp J$. At 0.21 and 0.27 K the difference is close to 12%, and at higher temperatures it is significantly reduced (only 4% at 0.57 K and 2% at 0.81 K). This anisotropy of thermal conductivity is due to the vortex scattering of the quasiparticles. More precisely, in a semiclassical approach the scattering off the vortex is maximal when the quasiparticle velocity is perpendicular to it, resulting in a lower thermal conductivity when the field is perpendicular to the heat current. This anisotropy is naturally expected to vanish at H_{c2} when the vortices overlap.[36] The same calculation predicts that the anisotropy of κ will change sign at the $T \ll T_c$ limit, where the excitation of the quasiparticles perpendicular to the vortices (Volovik effect) becomes the dominant effect and enhances the heat current in the direction perpendicular to the magnetic field. Our data show that the anisotropy of thermal conductivity in $CeCoIn_5$ is growing to the lowest temperature measured, once again indicating the need to go to lower temperature to test the theoretical prediction. There are no similar calculations for the d-wave case. More theoretical work is needed to help us understand the magnetothermal transport in the vortex state of $CeCoIn_5$.

IV. THE SUPERCONDUCTING PHASE DIAGRAM OF $CeCoIn_5$

The upper critical field H_{c2}, determined from both the temperature and the field sweeps, is displayed in Fig. 6, to-

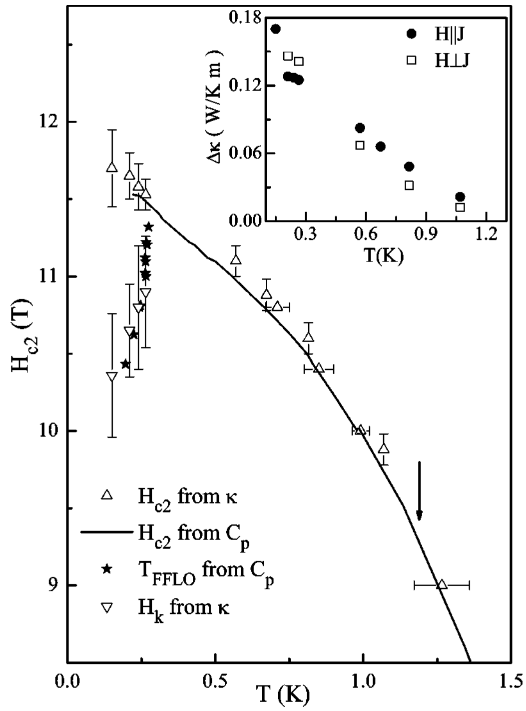

FIG. 6. Magnetic field vs temperature phase diagram of $CeCoIn_5$ deduced from thermal conductivity and specific heat measurements. (Solid line) H_{c2} and ($\star$) T_{FFLO} are deduced from the specific heat data of Ref. 23. H_{c2} ($\triangle$) and H_k (∇) are obtained from the thermal conductivity data (details are in the text). The arrow indicates the critical temperature where superconducting transition changes from second to first order. Inset: the size of the jump in κ at the superconducting transition for the field sweeps for (●) $H \| J$ and (□) $H \perp J$.

gether with the critical field deduced from the specific heat. Note that there is no difference in H_{c2} for the field parallel and perpendicular to the heat current, as expected for a tetragonal compound, since the additional in-plane anisotropy due to the *d*-wave gap is zero upon the 90° rotation. The shape of H_{c2}, as well as the first-order nature of the transition at low temperatures, is in agreement with previous reports.[21,23] The strong temperature dependence of H_{c2} at low temperatures, as opposed to the saturated behavior expected in the BCS theory, is an important observation supporting the existence of the FFLO state in $CeCoIn_5$.[22] In fact, this is a common feature for a number of other superconductors suggested to be in the Pauli-paramagnetic limit, such as UBe_4[43] and κ-$(BEDT\text{-}TTF)_2Cu(CSN)_2$.[44]

The amplitude of the jump in thermal conductivity is comparable for both field orientations and decreases almost linearly with increasing temperature, as shown in the inset of Fig. 6. This is in contrast to the sharp decrease in the temperature step, associated with the first-order transition, observed at the critical point in magnetocaloric measurements for the field along the *c* axis.[21] It would be difficult to locate precisely the critical point where the order of the superconducting phase transition changes from second to first, for $H \| J$, because the transition occurs gradually. Nevertheless, the critical point is consistent with that determined from specific heat measurements.[23]

V. FULDE-FERRELL-LARKIN-OVCHINNIKOV (FFLO) TRANSITION

An additional feature in the κ versus H curves displayed in Fig. 5 is resolved at the lowest temperatures. A kink appears in the data at a field H_k for $H \| J$, and the thermal conductivity is nearly constant between H_k and H_{c2} for temperatures below 0.27 K. Given the sharpness of the jump at H_{c2} and the first-order nature of the transition, this feature is clearly distinct from the H_{c2} anomaly. This anomaly is not present in the data at 0.57 K and above, where the thermal conductivity continues to decrease up to H_{c2}. We therefore interpret the region of the flat $\kappa(H)$ preceding the sharp jump at H_{c2} as an enhancement of thermal conductivity. Further, H_k decreases as the temperature is reduced. Figure 6 displays H_k in the H-T plane, together with the phase diagram deduced from the specific heat measurements.[23] The H_k points coincide well with the second low-temperature phase-transition line found in the specific heat below H_{c2}. The absence of such a flat portion at high temperatures, where the thermal conductivity is monotonously decreasing up to H_{c2}, and the good agreement of H_k with the specific heat anomaly, lead us to speculate that the enhancement of thermal conductivity is due to the formation of the second low-temperature superconducting state in $CeCoIn_5$, identified as a potential FFLO state.[23,29] There is a good agreement between the FFLO phase diagrams obtained by different groups with different $CeCoIn_5$ samples.[23,29,32,33] This indicates that all of these samples, grown from flux, are of high quality, with the mean-free path much larger than expected (100–1000 Å) for the modulation period of the FFLO state in $CeCoIn_5$. Features in the thermal conductivity of an organic superconductor near the superconducting critical field were also interpreted as possible signatures of the FFLO state.[14]

The enhancement of thermal conductivity between H_k and H_{c2} manifests itself clearly in the $H \| J$ data. It is more difficult to make a definitive statement for the data in the $H \perp J$ geometry since data below H_{c2} is rounded and a gradual rise is present in the data for higher temperature, outside of the FFLO phase.

The enhancement of thermal conductivity for the $H \| J$ orientation is contrary to our original simple-minded expectation, since in this geometry the nodal planes in the LO phase are perpendicular to the direction of the heat current, and therefore would not be expected to enhance thermal conductivity. There are several possible explanations of our results. Recent theoretical work[45] suggests that the lowest energy state is not a pure LO state with a single modulation wave vector $\vec{Q}$, but a modified LO state with a combination of three modulation wave vectors. If so, one would not expect an additional anisotropy with respect to the direction of the magnetic field due to FFLO nodal planes. The contribution to thermal conductivity from the nodal planes of the LO state is not *a priori* dominant over that from the vortices, and must be investigated theoretically.[46] Another scenario[47] suggests that the bottleneck for the heat transport along the field direction is the vortex cores. One would expect the structure of the vortices to be modulated by the nodal planes. The vortex core's size might increase at the nodal planes, reducing the bottleneck and leading to the enhancement of thermal conductivity. The interplay between the vortex and FFLO state was theoretically studied for 2D superconductors.[11] The resulting spatial structures can be alternating nodal planes and lines of vortices, or more intricate structures, depending on the Landau level quantization number of the order parameter. More theoretical work on the vortex structure within the FFLO state for three-dimensional (3D) superconductors, and its effect on the thermal transport in particular, is called for and should clarify whether such new structures can account for the observed enhancement of the thermal conductivity in the low-temperature superconducting state.

VI. CONCLUSION

Thermal transport is a powerful probe of a superconducting state. We performed thermal conductivity measurements to investigate the properties of both the vortex state and the second low-temperature phase of $CeCoIn_5$, with the magnetic field applied within the *a*-*b* plane of this tetragonal compound. Our data demonstrate that the superconducting phase transition becomes first order between roughly 10 T and the superconducting critical field, in accordance with previous specific heat measurements, indicating the importance of the Pauli limiting effect in $CeCoIn_5$. In addition, we observed a kink in thermal conductivity for the field parallel to the direction of the heat current, coincident with the phase transition in the second low-temperature state of $CeCoIn_5$, suggested previously to be an FFLO state.[23,29] Thermal transport within the FFLO state at present remains unexplored theoretically, and the observed enhancement of thermal conductivity within the FFLO state of $CeCoIn_5$ is puzzling. Our experimental results present a challenge for the understanding of inhomogeneous superconductivity.

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