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Resonances in the Field-Angle-Resolved Thermal Conductivity of $CeCoIn_5$

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The thermal conductivity measurement in a rotating magnetic field is a powerful probe of the structure of the superconducting energy gap. We present high-precision measurements of the low-temperature thermal conductivity in the unconventional heavy-fermion superconductor $CeCoIn_5$, with the heat current $\boldsymbol{J}$ along the nodal [110] direction of its $d_{x^2-y^2}$ order parameter and the magnetic field up to 7 Tesla rotating in the *ab* plane. In contrast to the smooth oscillations found previously for $\boldsymbol{J} \parallel [100]$, we observe a sharp resonance-like peak in thermal conductivity when the magnetic field is also in the [110] direction, parallel to the heat current. We explain this peak qualitatively via a model of the heat transport in a *d*-wave superconductor. In addition, we observed two smaller but also very sharp peaks in the thermal conductivity for the field directions at angles $\Theta \approx \pm 33^\circ$ with respect to $\boldsymbol{J}$. The origin of the observed resonances at $\Theta \approx \pm 33^\circ$ at present defies theoretical explanation. The challenge of uncovering their source will dictate exploring theoretically more complex models, which might include, *e.g.*, fine details of the Fermi surface, Andreev bound vortex core states, a secondary superconducting order parameter, and the existence of gaps in spin and charge excitations.

$CeCoIn_5$ is one of the most studied heavy-fermion superconductors, with a wealth of phenomena at forefront areas of condensed-matter physics, such as unconventional superconductivity with $d_{x^2-y^2}$ symmetry, a field-induced quantum critical point (QCP), and intertwined orders in its superconducting state. It is a Pauli-limited superconductor with the Maki parameter α, which characterizes the relative strength of the Pauli limiting versus orbital limiting [1], of 4.5 [2]. At low temperature ($T \lessapprox 1$ K) and high magnetic field, the superconducting transition itself becomes first order, consistent with predictions for a Pauli-limited superconductor [3, 4]. Unique to $CeCoIn_5$, there is a magnetic spin-density-wave (SDW) order in the high-field and low-temperature corner of the superconducting state [5, 6], where the possibility of a spatially inhomogeneous superconducting FFLO state was raised, as such state is expected on theoretical grounds for the Pauli-limited superconductor with $\alpha > 1.8$ [7].

Thermal conductivity measurements are a powerful probe of a superconducting state [8]. Because a superconducting condensate does not carry heat and only the normal quasiparticles are responsible for thermal transport, thermal conductivity provides insight into the character of a superconducting gap. In particular, thermal conductivity measurements in a rotating magnetic field resolve the symmetry of the superconducting order parameter [9–11]. The dependence of thermal conductivity on the direction of magnetic field has several origins. One is the so-called Volovik effect, a Doppler shift of the quasiparticle energies in the presence of supercurrent flow around a vortex core, which leads to an increase in the normal quasiparticle density of states. Another is quasiparticle scattering off the vortex cores, which results in a two-fold oscillation of thermal conductivity as a function of the angle Θ between the magnetic field and the direction of the heat current. The Volovik effect has a particularly pronounced effect on quasiparticle states around nodes of the superconducting energy gap, where the Doppler shift of energy is comparable to the superconducting energy gap and, therefore, can lead to a dramatic increase in the quasiparticle density of states. It is this sensitivity of the magnetic field response to the presence of nodes (via the Volovik effect), that was instrumental in identifying the *d*-wave symmetry of the superconducting gap in $CeCoIn_5$ [10]. The *d*-wave nodes manifested themselves in a four-fold oscillation in the thermal conductivity of $CeCoIn_5$ as the magnetic field was rotated within the *ab* plane and with the heat current along the antinodal [100] direction.

Thermal transport as a function of the direction of the magnetic field in a *d*-wave superconductor was calculated by Vorontsov and Vekhter [11–14]. When the magnetic field points along a node of a *d*-wave gap (see Fig. 1), only the two nodes with quasiparticle momentum $\boldsymbol{k}$ perpendicular to the magnetic field are Doppler-shift active, with the maximal shift and the correspondingly largest gain in the density of states. When the field is along an antinode, all four nodes are active but with a smaller Doppler shift. The resulting four-fold oscillation in the density of states (the change from node to antinode every 45°) leads to a four-fold oscillation of the thermal conductivity. However, the balance in the competition between the two maximally Doppler-active and the four less active nodes is subtle and depends on the details of the Fermi surface, temperature, and magnetic field. In fact, the four-fold terms in the thermal conductivity and the specific heat can have either a maximum or a minimum for the field direction along the nodes, depending on where, within the superconducting phase of the field-temperature (H–T) plane, the angle-resolved measurement is performed [11].

The measurements by Izawa *et al.* [10] were performed with the heat current flowing along the antinodal direction, [100]. Their thermal conductivity data were well represented by a sum of two-fold and four-fold components. For the magnetic fields and the temperatures chosen for their measurements, the thermal conductivity maxima occurred when the field was along the gap nodes, with the angle between the field and the direction of the heat current $\Theta = 45^\circ$. When more realistic Fermi surfaces were taken into accounted [14], the theory successfully reconciled the apparent contradiction between the thermal conductivity [10] and the specific heat [15] studies of $CeCoIn_5$ in identifying the type of the *d*-wave order parameter. It also explained a switch between maximum and minimum of these quantities in $CeCoIn_5$ for a field in the nodal direction, depending on the position in the H-T space [14]. This theory was also successfully applied to other systems, such as an iron-based superconductor $A_yFe_2Se_2$ [16].

In this Letter, we present results of low-temperature thermal conductivity measurements in $CeCoIn_5$ for magnetic field up to 7 Tesla with the heat current along the nodal direction, $\boldsymbol{J} \parallel$ [110]. Overall, some of the features observed for $\boldsymbol{J} \parallel$ [100] (such as maxima for the magnetic field in the nodal directions) are still present in our data. In addition, though, we observe sharp resonance-like peaks in the thermal conductivity that are the subject of this Letter.

The sample and experimental apparatus are the same as those for the high-field measurements reported elsewhere [17]. Figure 1 shows the orientation of the heat current against the nodes of the $d_{x^2-y^2}$-wave superconducting order parameter and the rotating magnetic field. In order to apply the heat current along the nodal direction, a single crystal of $CeCoIn_5$ was polished into a rectangular shape ($2.5\times0.5\times0.2\,mm^3$) with the longest edge (the direction of the heat flow) along the [110] direction. The standard one-heater and two-thermometer steady-state method was used to measure thermal conductivity. The experiments were performed in a dilution refrigerator coupled with a superconducting magnet and an Attocube piezoelectric rotator with a horizontal axis of rotation [18]. One end of the sample was rigidly attached to the sample holder on the rotation stage so that the magnetic field always laid within the *ab* plane of the sample during rotation.

Field-angle sweeps of the thermal conductivity at 1 and 3 Tesla are displayed in Fig. 2 and Fig. 3, respectively. These fields are well below $H_{c2}(0)$. General trends are in accord with previous measurements [10]. As we measure the angle with respect to the heat current, the maxima at low temperatures are shifted by 45° compared to the case of $\boldsymbol{J} \parallel$ [100] and appear when the field again aligns with the nodes of the $d_{x^2-y^2}$ order parameter, now at zero and ±90°. Vorontsov and Vekhter showed that the four-fold terms in the thermal conductivity for both $d_{x^2-y^2}$ and d_{xy} with the heat current along [100] have maxima when the field is along the nodes in the

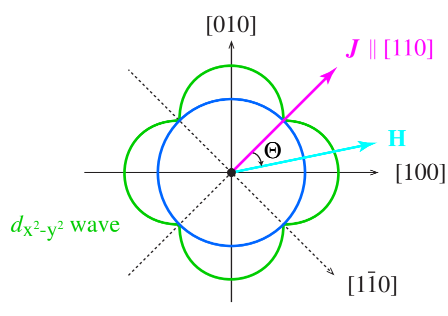

FIG. 1. Schematic diagram of the thermal conductivity measurement on $CeCoIn_5$. The heat current $\boldsymbol{J}$ (magenta arrow) was applied along a node of the $d_{x^2-y^2}$ order parameter (green curve). The angle Θ is between the magnetic field **H**(cyan arrow) and the direction of the heat current. The blue circle represents the normal Fermi surface.

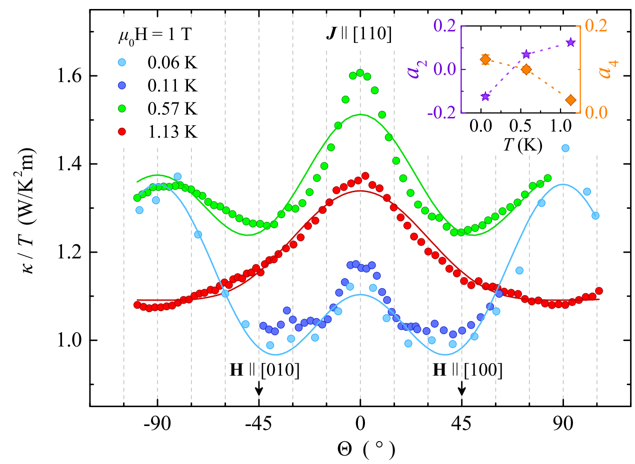

FIG. 2. Field-angle dependent thermal conductivity at different temperatures and with a fixed 1-T magnetic field. Solid lines represent fits to the equation $f(\theta) = a_0 + a_2\cos(2\theta) + a_4\cos(4\theta)$ and the coefficients are plotted in the inset.

majority of the superconducting phase [11]. Because the calculations were done for a cylindrically symmetric Fermi surface, the d_{xy} case with $\boldsymbol{J}$ along [100] is equivalent to the case of $d_{x^2-y^2}$ with $\boldsymbol{J} \parallel$ [110], and, therefore, our experimental results should be compared to the calculations for a d_{xy} order parameter with $\boldsymbol{J}$ along [100] (Fig. 9 and the right panel of Fig. 10 in Ref. [11]).

In Fig. 2, we fit the 1-T data to a combination of two- and four-fold oscillation terms, $f(\theta) = a_0 + a_2\cos(2\theta) + a_4\cos(4\theta)$. The evolution of the fitting parameters provides an important test of previous calculations of thermal conductivity in $CeCoIn_5$ [11, 14]. In particular, the coefficient of the two-fold term, a_2, changes sign around 0.3 K, whereas the coefficient of the four-fold term, a_4, remains positive but tends to zero at higher temperature. The behavior of a_4 reflects the dominance of the region of the positive value of a_4 within the model calculations for $CeCoIn_5$, shown in Fig. 5(b2) of Ref. [14]. A change of sign of a_2 was anticipated theoretically as well, see Fig. 10 (right panel) in Ref. [11], at a temperature close to $0.15 \times T_c \approx 0.3$ K. This consistency demonstrates our good overall understanding of magneto-thermal transport in *d*-wave superconductors.

There are two distinct features in our data that were not observed or anticipated previously. The first is the sharpness of the zero-angle peak versus the roundness of the 90° peaks. The second is the appearance of two additional peaks at $\Theta \approx \pm33°$ (clearly visible for the 0.11-K and 0.57-K data in Fig. 2 and 3) positioned symmetrically about $\boldsymbol{J}$.

The fits in Fig. 2 do not reproduce the sharp features around $\Theta = 0°$ found at low temperatures. The data at 3 T, shown in Fig. 3, reveal that the zero-angle anomaly sharpens in a singular fashion with reduced temperature. This anomaly has the flavor of a resonance when all three — the magnetic field, the node, and the direction of the heat flow — are parallel to each

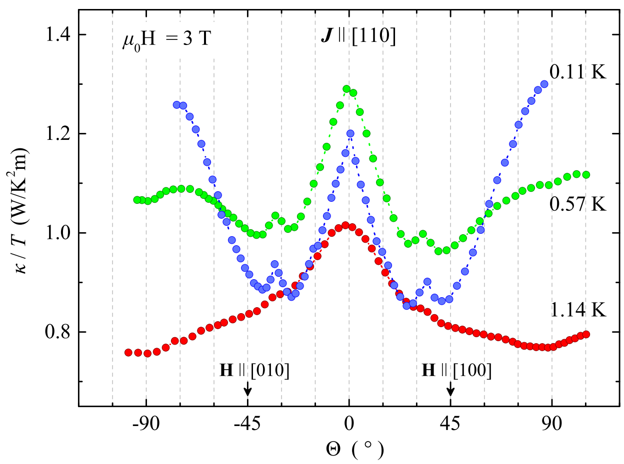

FIG. 3. Field-angle dependent thermal conductivity at three different temperatures and with a fixed 3-T magnetic field.

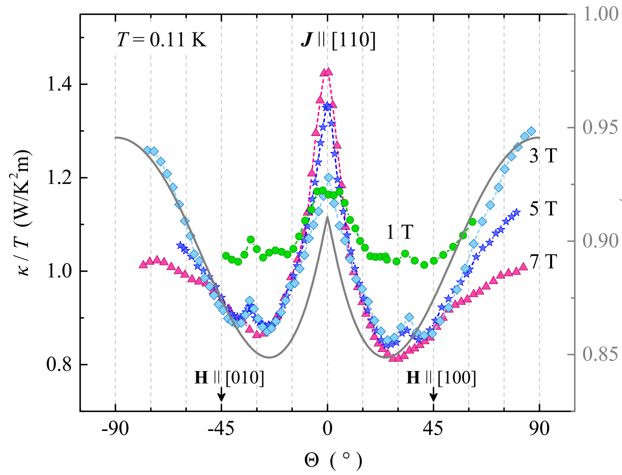

FIG. 4. Field-angle dependent thermal conductivity at 0.11 K for several magnetic fields between 1 and 7 T. The grey curve is a calculation based on the model of Ref. [11], see Supplemental Material.

other. When the field is swept through the node at $\Theta = 0°$, the contribution from the two perpendicular nodes ([$1\bar{1}0$] and [$\bar{1}10$], see Fig. 1) should be rather flat. The sharpness of the zero-angle peak must be due then to nodes along the heat current $\boldsymbol{J}$. The Volovik effect (the rise in the density of states) is minimal for quasiparticles in these nodes (the Doppler shift is zero for $\boldsymbol{k} \parallel [110] \parallel \mathbf{H}$). The origin of the peak, therefore, must be due to the increasing mean free path of nodal quasiparticles when $\mathbf{H} \parallel \boldsymbol{J} \parallel$ *node*. Note that no sharp peak for either $\mathbf{H} \parallel \boldsymbol{J}$, or $\mathbf{H} \parallel$ *node* was found in the measurement by Izawa *et al.* [10], with $\boldsymbol{J} \parallel [100]$. Similarly, theoretical calculations (see Fig. 9 in Ref. [11]) did not reveal the presence of a sharp peak at zero-angle ($\phi = 0°$ in their notation).

Figure 4 presents the thermal conductivity data at 0.11 K for different magnetic fields. With increasing magnetic field, the zero-angle peak grows, perhaps due to the increasing number of core states. The thermal conductivity at ±90°, when the field is perpendicular to the heat current, drops when the field is increased from 3 to 7 Tesla, in spite of the increasing density of the quasiparticle states along the nodes parallel to the heat current (due to the Volovik effect). This decrease of thermal conductivity at ±90° with magnetic field must be due to an even faster rise of the quasiparticle scattering off the vortices.

The model calculations, although successful at demonstrating the origin of the singular shape of the $\Theta = 0°$ peak, fail to reproduce the evolution of κ/T at $\Theta = \pm 90°$ and at $\Theta = 0°$ with field (see Fig. S1 in Supplemental Material). A possible additional contribution to the zero-angle resonance may be Andreev bound core states (Vorontsov, private communication), which are not included in the current theory. Perhaps the structure of the core states and their contribution to the heat transport are uniquely different for the field along nodes. Also, Pauli limiting is strong in $CeCoIn_5$ (but not considered fully in the model) and was invoked theoretically to explain several experimental observations, such as the anomalous form factor of the vortex lattice [19–22] and the appearance of the SDW order (the Q phase) in the low-temperature high-field corner of the superconducting phase [5, 6, 22, 23]. An enhanced quasiparticle density of states due to Pauli limiting, however, was taken into account [14] when spin-orbit scattering was neglected. The main effect of the Pauli limiting with this approximation was a suppression of the superconducting energy gap, which is consistent with our model calculations that required a strong suppression of the superconducting energy gap to reproduce the zero-angle resonance.

Currently, there is no theory that accounts for the sharp peaks in thermal conductivity found at $\Theta^* \approx \pm 33°$. Figure 5 zooms in the regions around the peaks. The Θ^* peaks are very robust, with the amplitude peaking roughly around 3 T. As Figs. 3-5 show, the positions of the peaksa are very close to being field and temperature independent: Θ^* moves less than one degree between 3 T and 5 T at 0.11 K.

Could an anomaly in the Fermi surface lead to a resonance-like feature in the thermal conductivity at Θ^*? One might expect a Fermi surface of tetragonal $CeCoIn_5$ to be four-fold degenerate, with two more peaks at $\pm(90 - 33)°$ within the range of Θ explored. However, the heat current and vortex lattice break the four-fold symmetry, leaving only the two-fold symmetry intact. We can rule out experimental artifacts that may lead to such anomalies, such as thermometer calibrations, which were established in detail ex-situ. The sample is rigidly glued to the sample holder, and therefore no movement of the sample can be the source of the Θ^* anomalies. A potential movement of the thermometers during rotation also cannot explain our observation, as such a movement must quickly reverse itself when Θ^* is passed. The symmetrical positions of the peaks and their observation at a variety of fields and temperatures also strengthen the argument against an artifact as a source of the anomalies.

One possibility is the existence of a secondary, time-reversal symmetry-preserving superconducting order param-

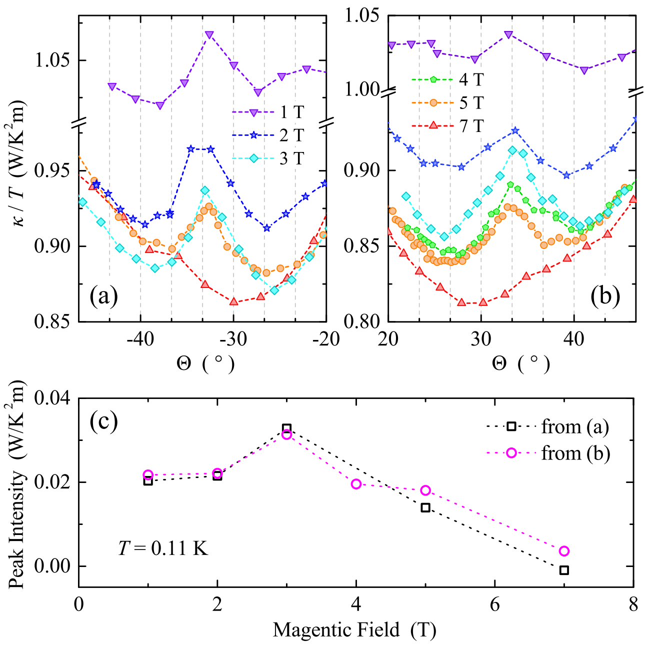

FIG. 5. Expanded view of the anomalies around (a) -33° and (b) 33°. (c) The intensities of the peaks at $\Theta \approx \pm 33°$ as a function of magnetic field. The detailed procedures of extracting the peak intensities are described in Supplemental Material.

eter, such as $d + p$ wave, that might shift the positions of the two nodes perpendicular to the heat current to $\Theta^* = \pm 33°$. The magnetic field would be aligned then with the nodes at Θ^*, but the heat current would be away from the field/nodes.

Another possibility for the peaks at ±33° is a transition in the structure of the vortex lattice. Such a transition should be hysteretic; however, there is no detectable hysteresis in the thermal conductivity. Furthermore, a neutron scattering study found the same vortex structure for field **H** ∥ [100] and **H** ∥ [110] below 7.5 T [24]. Therefore, the anomaly at ±33° is unlikely to be caused by a vortex lattice transition.

Like other d-wave superconductors, superconductivity in $CeCoIn_5$ develops out of a normal state in which scanning tunneling spectroscopy reveals the presence of a pseudogap [25, 26], and below T_c a well-defined spin resonance emerges that gaps spin excitations at or very near the nodal directions [27, 28]. Both of these gaps could have a non-trivial influence on the thermal conductivity. It is reasonable that the spin gap suppresses spin scattering and increases the mean free path of quasiparticles at gap nodes. Further, the original measurements of Izawa *et al.* show that a four-fold modulation of the thermal conductivity persists above T_c in the temperature range where the pseudogap in charge degrees of freedom appears [10], suggesting that it also may have d-wave symmetry. In a magnetic field perpendicular to [001], the lower energy mode of the Zeeman-split spin resonance decreases from 0.6 meV and extrapolates to zero energy at a field near 11 T [28, 29]; whereas, the pseudogap persists above $H_{c2}(0)$ for **H** ∥ [001]. Currently, we do not know how the pseudogap evolves with field perpendicular to [001]; nevertheless, both the spin gap and pseudogap persist in the field and temperature ranges of our thermal conductivity measurements. How these gaps might cooperate or interfere to produce a response in thermal conductivity remains an interesting open question.

The thermal conductivity of $CeCoIn_5$, for the heat current along the nodal direction of the d-wave superconducting gap in a rotating magnetic field, displays several striking features. The sharp singular peak for **H** ∥ ***J*** (and a d-wave node) can be accounted for (at least qualitatively) by the existing theory of thermal transport in unconventional superconductors. Including the roles of a spin gap and pseudogap might account quantitatively for this singular peak. Two additional anomalous sharp peaks at $\Theta^* \approx \pm 33°$ are an enigma and lie outside of current theoretical understanding. They might be an exotic combination of several effects: Andreev bound core states, secondary order parameter, subtle features in the Fermi surface, or gaps in the spin and charge sectors. This discovery points clearly to the need for increasingly sophisticated theories of thermal magnetotransport in unconventional superconductors.

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