Hybrid Gap Structure of the Heavy-Fermion Superconductor CeIrIn₅

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The thermal conductivity κ of the heavy-fermion superconductor CeIrIn₅ was measured as a function of temperature down to $T_c/8$, for current directions parallel $(J \parallel c)$ and perpendicular $(J \parallel a)$ to the tetragonal *c* axis. For $J \parallel a$, a sizable residual linear term κ_0/T is observed, as previously, which confirms the presence of line nodes in the superconducting gap. For $J \parallel c$, on the other hand, $\kappa/T \rightarrow 0$ as $T \rightarrow 0$. The resulting precipitous decline in the anisotropy ratio κ_c/κ_a at low temperature rules out a gap structure with line nodes running along the *c* axis, such as the *d*-wave state favored for CeCoIn₅, and instead points to a hybrid gap of E_g symmetry.

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The discovery of magnetically-mediated superconductivity in the heavy-fermion material CeIn₃ [1] has attracted considerable attention as a possible archetype for unconventional pairing. However, the fact that the superconductivity in this material only exists under pressure makes it difficult to characterize it experimentally. Fortunately, the closely related family of $CeMIn_5$ (M = Co, Ir, Rh) compounds offers an ideal testing ground for investigating the role that dimensionality, magnetic order and fluctuations play in determining the strength and symmetry of the superconducting state [2,3]. In CeCoIn₅, the observation of a fourfold anisotropy in thermal conductivity [4] and specific heat [5] on rotation of a magnetic field in the basal tetragonal plane points to a *d*-wave gap (presumably of $d_{x^2-y^2}$ symmetry [6]). A number of theoretical models propose a $d_{x^2-y^2}$ state [7,8], analogous to that realized in cuprates. Since the calculated band structure [9] and measured Fermi surface [10] of CeCoIn₅ and CeIrIn₅ are similar, and properties like the specific heat [11] and the spin-lattice relaxation rate in the nuclear quadrupolar resonance study [12,13] exhibit the same temperature dependence, it has generally been assumed that the two superconductors have the same pairing state, even though their transition temperature T_c differs by a factor of 6. However, because recent evidence suggests that the phase diagram of CeMIn₅ may contain more than one superconducting phase [14,15], it has now become crucial to pin down the pairing symmetry of CeIrIn₅.

One of the most conclusive ways to determine the pairing symmetry of a superconductor is to map out its gap structure. A powerful approach to probe the gap structure and locate the position of nodes around the Fermi surface is to measure quasiparticle heat transport as a function of direction. In this Letter, we report a study of heat transport in CeIrIn₅ ($T_c = 0.4$ K) down to $T_c/8$ for current directions parallel and perpendicular to the tetragonal axis. It reveals a dramatic anisotropy as $T \rightarrow 0$, whereby lowenergy nodal quasiparticles carry heat well in the basal plane but poorly, if at all, along the *c* axis. This is incon-

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sistent with the *d*-wave states proposed for CeCoIn₅, characterized by line nodes running along the *c* axis. In fact, it eliminates all allowed (spin singlet) pairing symmetries but one, the (1, *i*) state of the E_g representation. This state has a hybrid gap structure, with a line node in the basal plane and point nodes in the *c* direction.

Single crystals of CeIrIn₅ were grown by the self-flux method [3]. Samples were cut into parallelepipeds with dimensions $\sim 4.5 \times 0.14 \times 0.045 \text{ mm}^3$ (for $J \parallel a$) and $\sim 1 \times 0.15 \times 0.086 \text{ mm}^3$ (for $J \parallel c$). Their low residual resistivity (at $T \rightarrow 0$ and $H \rightarrow 0$) attests to high purity: $\rho_{0a}(\rho_{0c}) = 0.2(0.5) \ \mu\Omega$ cm. The bulk transition temperature is $T_c = 0.38 \pm 0.02$ K and the upper critical field $H_{c2} = 0.49$ T for $H \parallel c$. The findings reported here were reproduced on additional crystals from different growth batches for both a- and c axis directions. The thermal conductivity was measured in a dilution refrigerator using a standard four-wire steady-state method. The same indium-soldered contacts were used for electrical resistivity and thermal conductivity. The typical resistance of indium contacts at low temperature was $\sim 5 \text{ m}\Omega$. The contribution of phonons to the thermal transport is entirely negligible below 1 K.

Normal state.—The thermal conductivity of CeIrIn₅ is plotted in Fig. 1 as κ/T vs *T*, for a current perpendicular $(J \parallel a)$ and parallel $(J \parallel c)$ to the *c* axis. We first concentrate on the normal state, where the electrical resistivity $\rho(T)$ was found to satisfy the Wiedemann-Franz law to better than 1% for both *a*- and *c* axis current directions, as $T \rightarrow 0$: $\kappa_N/T = L_0/\rho_0$, where $L_0 = \frac{\pi^2}{3} (\frac{k_B}{e})^2$. This shows that our measurements do not suffer from electron-phonon decoupling (see discussion in [16,17]). κ_N exhibits the temperature dependence of a Fermi liquid, $\kappa_N(T)/T =$ $1/(a + bT^2)$, with a = 8.5(19.6) K² cm/W and b =36(90) cm/W, for $J \parallel a$ ($J \parallel c$). The fact that both samples have the same (thermal) resistivity ratio, namely $\kappa/T(T \rightarrow$ $0)/\kappa/T(0.6\text{K}) = 2.4(2.6)$ for $J \parallel a$ ($J \parallel c$), indicates a similar level of impurity scattering.

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FIG. 1 (color online). Thermal conductivity κ of CeIrIn₅, plotted as κ/T vs *T*, for a heat current perpendicular (top) and parallel (bottom) to the *c* axis, in the superconducting state (SC; H = 0) and the normal state (N; H = 0.5 T). The normal state data are fitted to the Fermi-liquid dependence $\kappa_N/T = 1/(a + bT^2)$ (red lines). A linear fit to the superconducting state data for $J \parallel a$ (below $T_c/2$) is also shown (blue line).

Superconducting state. —In the top panel of Fig. 1, κ_a/T in zero field is seen to be roughly linear in *T*, extrapolating to a finite residual linear term as $T \rightarrow 0$ [18]. This is the behavior expected of a superconductor with line nodes, i.e., with a density of states that grows linearly with energy $[N(E) \propto E]$ [19,20]. In particular, theory shows that, in the $T \rightarrow 0$ limit, κ/T reaches a universal value given by [20,21]

$$\frac{\kappa_0}{T} = \frac{1}{3} \gamma_N v_F^2 \frac{a\hbar}{2\mu\Delta_0},\tag{1}$$

where γ_N is the linear term in the normal state specific heat, v_F is the Fermi velocity, Δ_0 is the gap maximum, μ is the slope of the gap at the node, and *a* is a constant of order unity whose value depends on the particular gap structure [20]. κ_0/T is called "universal" because it does not depend on impurity concentration and can therefore be used to measure the magnitude of the gap. Experimentally, universal conduction has been observed in high- T_c cuprates [22] and in Sr₂RuO₄ [23], and Eq. (1) works well in both cases.

Let us now apply Eq. (1) to CeIrIn₅. The allowed order parameter representations in tetragonal symmetry [24] are listed in Table I (for singlet pairing). Two line node topologies are possible: vertical line nodes (where the Fermi surface cuts a vertical plane, e.g. x = 0), such as in the two *d*-wave states $(d_{x^2-y^2} \text{ in } B_{1g} \text{ or } d_{xy} \text{ in } B_{2g})$, and a *horizontal* line node (where the Fermi surface cuts the z = 0 plane), such as in the hybrid gap of the $E_{g}(1, i)$ state. The simplest gap functions are $\Delta = \Delta_0 \cos 2\phi$ and $\Delta = 2\Delta_0 \cos \theta \sin \theta e^{i\phi}$, for d-wave and $E_{\rho}(1, i)$ states, respectively. The corresponding nodal structures are illustrated in Fig. 2. Let us apply Eq. (1) to such a hybrid gap function, for which a =3/2 and $\mu \equiv \mu_{\text{line}} = 2$ [21]. Using the known values of γ_N $(7300 \text{ J K}^{-2} \text{ m}^{-3} \text{ [11]}), v_F (2 \times 10^4 \text{ m/s}, \text{ in the basal})$ plane [10]), and Δ_0 (2.5 $k_B T_c$ [13]), Eq. (1) yields $\kappa_{0a}/T =$ 28 mW/K² cm. The experimental value is $\kappa_{0a}/T \simeq$ $20 \text{ mW/K}^2 \text{ cm}$ (Fig. 1), in good agreement with the theoretical estimate. While this quantitative agreement with theory is further confirmation for the presence of a line node in the gap of CeIrIn₅ [25], it actually says little about its location. Indeed, the estimate for a *d*-wave gap gives a similar value for κ_{0a}/T . The diagnostic power of thermal conductivity in determining the topology of the gap comes from its *directional* character, accessed by sending the current in distinct high-symmetry directions of the crystal. This was not done in the previous heat transport study [11].

Anisotropy.—Current along the *c* axis reveals a qualitatively different limiting behavior, namely $\kappa_c/T \rightarrow 0$ as $T \rightarrow 0$ (Fig. 1). Simple T^2 or T^3 extrapolations yield κ_{0c}/T values no greater than 1–2 mW/K² cm, an order of magnitude smaller than κ_{0a}/T . Figure 3 shows the anisotropy ratio, κ_c/κ_a , as a function of temperature, in both normal and superconducting states. In the normal state, κ_c/κ_a is virtually *T*-independent, with $\kappa_a/\kappa_c \approx$ 2.5. The anisotropy in $\rho(T)$ is similarly constant, even well beyond the Fermi-liquid T^2 regime, with $\rho_c/\rho_a \approx$ 2.7 between 1.2 and 8 K. This simply reflects the anisotropy of the Fermi velocity (or mass tensor).

The superconducting state anisotropy is strikingly different, a difference that can only come from gap anisotropy. Two distinct features are manifest: (1) a slight increase immediately below T_c and (2) a precipitous drop

TABLE I. Even-parity (spin-singlet) pair states in a tetragonal crystal with point group D_{4h} [24]. (V = vertical line node, H = horizontal line node.)

Representation	Gap	Basis function	Nodes
$\overline{A_{1g}}$	s wave	1, $(x^2 + y^2)$, z^2	none
A_{2g}	g wave	$xy(x^2-y^2)$	V
B_{1g}	$d_{x^2-y^2}$	$x^2 - y^2$	V
B_{2g}	d_{xy}	xy	V
$E_{\rho}(1,0)$		xz	V + H
$\vec{E_{g}}(1, 1)$		(x + y)z	V + H
$\check{E_g}(1,i)$	hybrid	(x + iy)z	H + points



FIG. 2 (color online). Typical gap structures in tetragonal symmetry. Left: $d_{x^2-y^2}$ gap on a (warped) cylindrical Fermi surface, with four vertical line nodes running along the *c* axis. Right: hybrid gap on a spherical Fermi surface, with one horizontal line node in the basal plane and two point nodes along the *c* axis.

below $T \simeq T_c/3$. These two features combine to produce a broad peak centered at $T \simeq T_c/2$. We attribute the first feature to an *anisotropic* suppression of inelastic scattering, brought about as electrons pair up (anisotropically) and cease to participate in the electron-electron scattering responsible for the bT^2 term in κ_N/T .

The second feature is directly diagnostic of the nodal structure, as it comes from low-energy quasiparticles. The factor of ~3 drop in κ_c/κ_a between $T_c/3$ and $T_c/8$ clearly extrapolates to a very small value as $T \rightarrow 0$. This reveals a qualitative a-c anisotropy in the average velocity of thermally excited nodal quasiparticles. In other words, those kstates responsible for c axis conduction in the normal state appear to be much more strongly gapped. This excludes any nodal structure for which the line nodes run along the caxis, irrespective of the shape of the Fermi surface. Indeed, such vertical line nodes would simply reproduce the underlying anisotropy of v_F , and κ_c/κ_a would basically mimic the normal state anisotropy. This expectation, confirmed by calculations [26], is illustrated in the inset of Fig. 3 (horizontal blue line). A modulation of the gap along the c axis, whereby $\Delta_0 = \Delta_0(\theta)$, can produce some additional anisotropy in the superconducting state, but this is typically modest and weakly T dependent [26]. More importantly, it would never bring to nearly zero for $J \parallel c(\theta = 0)$ the residual linear term present for $J \parallel a(\theta = \pi/2)$.

By excluding vertical line nodes in the gap of CeIrIn₅, our study eliminates all allowed representations for the order parameter, except one: the two-component E_g representation (see Table I). In particular, both *d*-wave states are ruled out: $d_{x^2-y^2}$ and d_{xy} , respectively, in B_{1g} and B_{2g} symmetry. Of the three states allowed in the E_g representation, only the (1, *i*) state is generically free of vertical line nodes. Its typical (x + iy)z dependence produces a hybrid



FIG. 3 (color online). Temperature dependence of the anisotropy ratio κ_c/κ_a , in the normal state (*N*) and in the SC. The dotted line is the ratio of the two normal state fit lines in Fig. 1 and the solid line is a linear fit to the superconducting state data below $T_c/3$. Inset: calculated anisotropy (normalized at T_c) for the gaps shown in Fig. 2, namely *d*-wave [26] and hybrid [19].

gap, which possesses, in addition to the line node in the basal plane (z = 0), point nodes along the $z \parallel c$ direction, at x = y = 0 (see Fig. 2). Note that this state breaks time-reversal symmetry, and will therefore spontaneously generate an internal magnetic moment around impurities. μ SR measurements on CeIrIn₅ have not detected such moments [27], possibly because the associated fields are too small in these high purity samples.

We now consider whether our data are compatible with another special feature of the $E_g(1, i)$ state: the *c* axis point nodes of its hybrid gap. These are *linear* point nodes, i.e., $\Delta(\theta) \propto \theta$, such that $N(E) \propto E^2$, which implies that κ_0/T in the *c* direction is *not* universal. The calculation for this state [19] (inset in Fig. 3) on a spherical Fermi surface yields a residual anisotropy κ_c/κ_a that is 20% of the normal state anisotropy at $T \rightarrow 0$. This is roughly compatible with the data. The main test for the presence of point nodes will be found in the response to doping, which is currently under study [25].

We note that the anisotropy of heat conduction was also measured in CeCoIn₅ [17], but the presence of unpaired electrons in that material produces an unexpectedly large and *isotropic* residual linear term which totally masks any anisotropy that might come from the coexisting nodal quasiparticles. It should be emphasized that the lack of a sizable residual linear term in the *c* axis data reported here rules out the possibility of such uncondensed electrons in CeIrIn₅.

In conclusion, the comparison of in-plane, κ_a , and interplane, κ_c , thermal conductivity of CeIrIn₅ as $T \rightarrow 0$ reveals a large anisotropy in the superconducting gap, inconsistent with vertical line nodes (along the c axis). This eliminates all but one of the pairing states allowed in D_{4h} symmetry, including the *d*-wave state proposed for the closely related compound CeCoIn₅. This leaves as sole candidate for CeIrIn₅ the (1, i) state of the E_g representation, also a prime candidate for the superconductor UPt_3 [19–21]. The $T \rightarrow 0$ value of κ/T in both high-symmetry directions is in good quantitative agreement with calculations for this state. This, therefore, points to superconducting order parameters of different symmetry in the two isostructural members of the CeMIn₅ family of nearly magnetic heavy-fermion metals. It will be interesting to examine how these differences might arise from the respective magnetic fluctuation spectra. Direct experimental confirmation of the presence of c axis point nodes and broken time-reversal symmetry, both implied by the $E_g(1, i)$ state, is called for.

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- [1] N.D. Mathur et al., Nature (London) **394**, 39 (1998).
- [2] C. Petrovic *et al.*, J. Phys. Condens. Matter **13**, L337 (2001).
- [3] C. Petrovic et al., Europhys. Lett. 53, 354 (2001).
- [4] K. Izawa et al., Phys. Rev. Lett. 87, 057002 (2001).
- [5] H. Aoki et al., J. Phys. Condens. Matter 16, L13 (2004).
- [6] A. Vorontsov et al., Phys. Rev. Lett. 96, 237001 (2006).
- [7] T. Takimoto et al., Phys. Rev. B 69, 104504 (2004).
- [8] K. Tanaka et al., J. Phys. Soc. Jpn. 75, 024713 (2006).
- [9] T. Maehira et al., J. Phys. Soc. Jpn. 72, 854 (2003).
- [10] Y. Haga et al., Phys. Rev. B 63, 060503(R) (2001).
- [11] R. Movshovich et al., Phys. Rev. Lett. 86, 5152 (2001).
- [12] Y. Kohori et al., Phys. Rev. B 64, 134526 (2001).
- [13] G.Q. Zheng et al., Phys. Rev. Lett. 86, 4664 (2001).
- [14] M. Nicklas et al., Phys. Rev. B 70, 020505(R) (2004).
- [15] S. Kawasaki et al., Phys. Rev. Lett. 94, 037007 (2005).
- [16] M.F. Smith et al., Phys. Rev. B 71, 014506 (2005).
- [17] M.A. Tanatar et al., Phys. Rev. Lett. 95, 067002 (2005).
- [18] A residual linear term of notably smaller magnitude was found in Ref. [11], possibly due to electron-phonon decoupling [16,17].
- [19] A. Fledderjohann *et al.*, Solid State Commun. **94**, 163 (1995).
- [20] M.J. Graf et al., Phys. Rev. B 53, 15147 (1996).
- [21] M.J. Graf et al., J. Low Temp. Phys. 102, 367 (1996).
- [22] L. Taillefer et al., Phys. Rev. Lett. 79, 483 (1997).
- [23] M. Suzuki et al., Phys. Rev. Lett. 88, 227004 (2002).
- [24] C.C. Tsuei et al., Rev. Mod. Phys. 72, 969 (2000).
- [25] Measurements on La-doped samples confirm the universal character of κ_a (to be published).
- [26] I. Vekhter and A. Vorontsov, Phys. Rev. B 75, 094512 (2007).
- [27] W. Higemoto et al., J. Phys. Soc. Jpn. 71, 1023 (2002).