Vortexlike excitations in the heavy-fermion superconductor CeIrIn₅

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We report a systematic study of temperature- and field-dependent charge (ρ) and entropy (**S**) transport in the heavy-fermion superconductor CeIrIn₅. Its large positive thermopower S_{xx} is typical of Ce-based Kondo lattice systems, and strong electronic correlations play an important role in enhancing the Nernst signal S_{xy}. By separating the off-diagonal Peltier coefficient α_{xy} from S_{xy}, we find that α_{xy} becomes positive and greatly enhanced at temperatures well above the bulk T_c. Compared with the nonmagnetic analog LaIrIn₅, these results suggest vortexlike excitations in a precursor state to unconventional superconductivity in CeIrIn₅. This study sheds light on the similarity of heavy-fermion and cuprate superconductors and on the possibility of states not characterized by the amplitude of an order parameter.

Typically, a disorder-order phase transition is described within the context of the Ginzburg-Landau theory by an order parameter and identified by a spontaneously broken symmetry. From this point of view, a superconducting transition might be special. The order parameter of superconductivity (SC) is expressed by a complex function in the form $\Psi_s(\mathbf{r}) = |\Psi_s(\mathbf{r})|e^{i\theta(\mathbf{r})}$ [1]. Gauge symmetry is broken after phase coherence is established throughout the system. When the phase stiffness is strong, phase coherence develops *concomitantly* as Cooper pairs form, and the superconducting critical temperature T_c is mainly determined by T^{MF}, the mean-field transition temperature predicted by the BCS theory [2]. In contrast, if the superfluid density is small (e.g., in underdoped cuprates and organic superconductors), the phase stiffness is low, and the phase coherence can be destroyed by short-lived vortexlike excitations. In this situation, bulk SC cannot be realized until the phases of Cooper pairs are ordered, and T^{MF} is simply the characteristic temperature below which pairing becomes significantly *local* ($T^{MF} \gg T_c$) [3]. As learned from the cuprates, states without a well-defined order parameter emerge above T_c and include phenomena such as superconducting phase fluctuations, preformed Cooper pairs, and a pseudogap.

The CeMIn₅ (M = Co, Rh, and Ir) family of tetragonal heavy-fermion compounds is a useful platform to investigate the interplay among unconventional SC, antiferromagnetic (AFM) order and spin fluctuations in the vicinity of quantum criticality. The member CeRhIn₅ is an incommensurate antiferromagnet at ambient pressure with Néel temperature $T_N = 3.8$ K [4,5] and can be pressurized into a superconducting state with the highest $T_c \sim 2.2$ K achieved around 2.35 GPa where $T_N(p)$ extrapolates to zero [6,7]. Textured SC was observed in the region where SC and AFM coexist, characterized by vanishingly small resistivity well above the bulk T_c and the anisotropic resistive T_c [8], reminiscent of the nematic state observed in cuprates. In this pressure range, nuclear quadrupole resonance (NQR) experiments suggested the presence of a pseudogap that develops above $T_N(P)$ and extrapolates to the maximum in $T_c(P)$ [9]. Likewise, scanning tunneling spectroscopy revealed a pseudogap that coexists

with d-wave SC in CeCoIn₅ [10,11], and replacing a small amount of In by Cd induces coexisting AFM order and SC in CeCo(In$_{0.99}$Cd$_{0.01}$)₅ where again a transition to zero resistance appears well above the bulk T_c [12]. Pristine CeIrIn₅ shows filamentary SC [13,14] at atmospheric pressure with a resistive onset temperature $T_c^{on} = 1.38$ K, but a diamagnetic state appears only below $T_c^b \simeq 0.5$ K. [This is also illustrated in Fig. 1(a).] Although no direct evidence of magnetic order has yet been identified, chemical substitutions of Hg/Sn on the In site demonstrate that the SC in CeIrIn₅ is in proximity to an AFM quantum-critical point [15]. Careful magnetoresistance and Hall effect studies of CeIrIn₅ found evidence for a precursor state of unknown origin arising near 2 K in the limit of zero field [16,17]. Though the pressure dependence of the precursor state is unknown, the resistive and bulk T_c's approach each other at the maximum in a dome of bulk SC [18], suggesting the possibility that the precursor state may be competing with SC. The complex interplay among states in the CeMIn₅ superconductors bears striking similarities to the cuprates, with pure CeIrIn₅ at atmospheric pressure presenting an opportunity to more closely examine these similarities.

From electrical (ρ) and thermoelectric (**S**) transport measurements in CeIrIn₅ and a comparison to its non-4f counterpart LaIrIn₅, we identify signatures of vortexlike excitations well above T_c^{on} (T_c^b). These findings suggest the existence of a pseudogaplike state where Cooper pairs start to form locally at a temperature well above T_c^{on}, but phase coherence among pairs is destroyed by thermally activated vortexlike excitations, pointing to a common framework for the physics of such states in both heavy-fermion and cuprate superconductors [19].

Single crystalline CeIrIn₅ was grown from an indium flux method [13]. The crystal was prescreened by both resistivity and magnetic susceptibility measurements to ensure the absence of free In. Thermoelectric measurements were carried out by means of a steady-state technique. A pair of well calibrated differential Chromel-Au$_{99.93\%}$Fe$_{0.07\%}$ thermocouples was used to measure the temperature gradient. Upon a thermal gradient $-\nabla T \parallel \mathbf{x}$ and a magnetic field $\mathbf{B} \parallel \mathbf{z}$, both thermopower signal $S_{xx} = -E_x/|\nabla T|$ and Nernst signal $S_{xy} = E_y/|\nabla T|$ were collected by scanning field at fixed temperatures. The same contact geometry was also used to measure electrical resistivity (ρ_{xx}) and Hall resistivity (ρ_{yx}). Both electrical and thermal currents were applied along the

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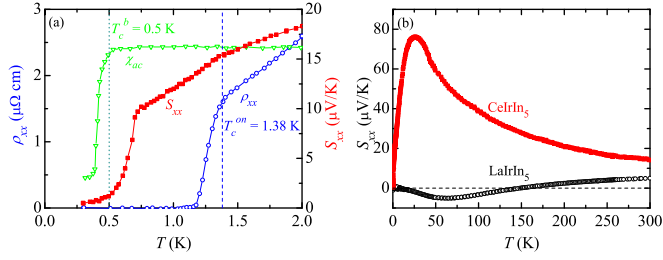[*]ykluo@lanl.gov

FIG. 1. (a) Temperature dependence of ρ_{xx} (blue), χ_{ac} (green), and S_{xx} (red) of CeIrIn$_5$, showing $T_c^{on} = 1.38$ K and $T_c^b = 0.5$ K. (b) Comparison of $S_{xx}(T)$ for CeIrIn$_5$ and LaIrIn$_5$.

a axis, and the magnetic field was parallel to **c**. The same measurements were performed on the nonmagnetic analog LaIrIn$_5$ for comparison. We adopt the sign convention as Ref. [20], which defines a *positive* Nernst signal for vortex motion [21,22].

In the presence of a temperature gradient $-\nabla T$, an electric field **E**, and a magnetic field **B**, the total current density is $\mathbf{J} = \boldsymbol{\sigma} \cdot \mathbf{E} + \boldsymbol{\alpha} \cdot (-\nabla T)$, where $\boldsymbol{\sigma}$ is the conductivity tensor and $\boldsymbol{\alpha} = \frac{\pi^2 k_B^2 T}{3q} \frac{\partial \sigma}{\partial \varepsilon}|_{\varepsilon = \varepsilon_F}$ (k_B is Boltzman constant, q is charge of carriers, and ε_F is chemical potential) is the Peltier conductivity tensor [23]. In an equilibrium state without net current, the Boltzman-Mott transport equation deduces the thermoelectric tensor

$$\mathbf{S} = \boldsymbol{\alpha} \cdot \boldsymbol{\sigma}^{-1} = \boldsymbol{\alpha} \cdot \boldsymbol{\rho}. \tag{1}$$

We start with the temperature dependence of thermopower $S_{xx}(T)$ as shown in Fig. 1(b). $S_{xx}(T)$ of LaIrIn$_5$ is positive at room temperature and changes sign near 150 K, characteristic of the expected multiband behavior [24]. In contrast, $S_{xx}(T)$ of CeIrIn$_5$ is positive in the full temperature range between 0.3 and 300 K, displaying a pronounced maximum at around 25 K with the magnitude reaching 76 μV/K. This peak in $S_{xx}(T)$ is associated with the onset of Kondo coherence [25]. These features are consistent with a Ce-based Kondo lattice in which the strong hybridization between $4f$ and conduction electrons forms a Kondo resonance with the density of states $N(\varepsilon)$ asymmetric with respect to ε_F [26,27] (see below). At low temperatures, $S_{xx}(T)$ shows a small kink at $T_c^{on} = 1.38$ K, but drops sharply at 0.7 K and tends to saturate below $T_c^b = 0.5$ K [cf. Fig. 1(a)]. Down to the lowest temperature of 0.3 K, however, $S_{xx}(T)$ still remains finite. We attribute this nonvanishing S_{xx} in the bulk superconducting state to the low T_c^b of CeIrIn$_5$: even a small temperature gradient may generate ungapped quasiparticles that contribute transport entropy.

Figure 2(a) displays an isothermal field dependence of S_{xx} at various temperatures. For all temperatures, the magnetothermopower is positive. One important feature of $S_{xx}(B)$ is a valley in the vicinity of zero field. As temperature decreases, this valley deepens and evolves into a cusp when $T \leqslant 3$ K. At 0.3 K, S_{xx} is small at $B = 0$ but recovers when the field is larger than 1.6 T. With the field dependencies of ρ_{xx} and ρ_{yx} shown in Figs. 2(b) and 2(c), respectively, it is reasonable to attribute this small transport-entropy state to a SC state. The cusp in $S_{xx}(B)$ occurring near 3 K is indicative of the loss of transport entropy well above T_c^b. The critical field recovering a normal state, however, is much smaller than that determined from
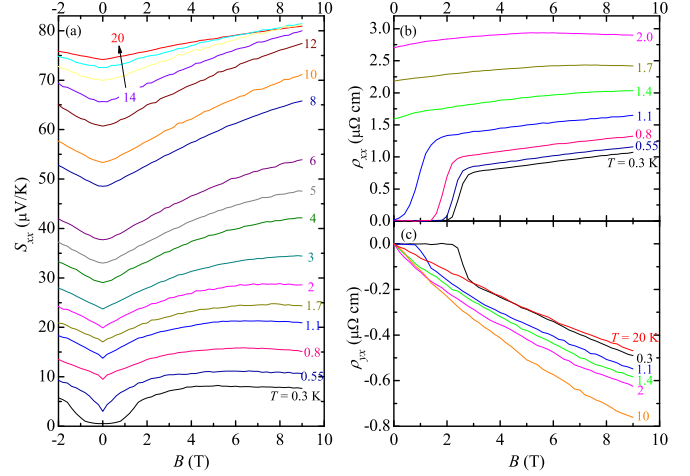

FIG. 2. (a) Field dependence of S_{xx} of CeIrIn$_5$ at selected temperatures. (b) and (c) display $\rho_{xx}(B)$ and $\rho_{yx}(B)$, respectively.

$\rho_{xx}(B)$ [Fig. 2(b)] and $\rho_{yx}(B)$ [Fig. 2(c)]. Systematic analysis of $\rho_{xx}(B)$ and $\rho_{yx}(B)$ by Nair *et al.* [16,17] showed that the modified Kohler's scaling [$\Delta\rho_{xx}(B)/\rho_{xx}(0) \propto \tan^2\theta_H$, where $\theta_H = \arctan(\rho_{yx}/\rho_{xx})$ is the Hall angle] breaks down prior to T_c^{on}, the region where we observe a large Nernst effect (see below). A similar phenomenon was observed in CeCoIn$_5$ and CeRhIn$_5$ under pressure [28], as in cuprates, and is reminiscent of a pseudogaplike precursor state [29].

In Fig. 3 we present the field dependence of the Nernst signal S_{xy}, the off-diagonal term of the thermoelectric tensor
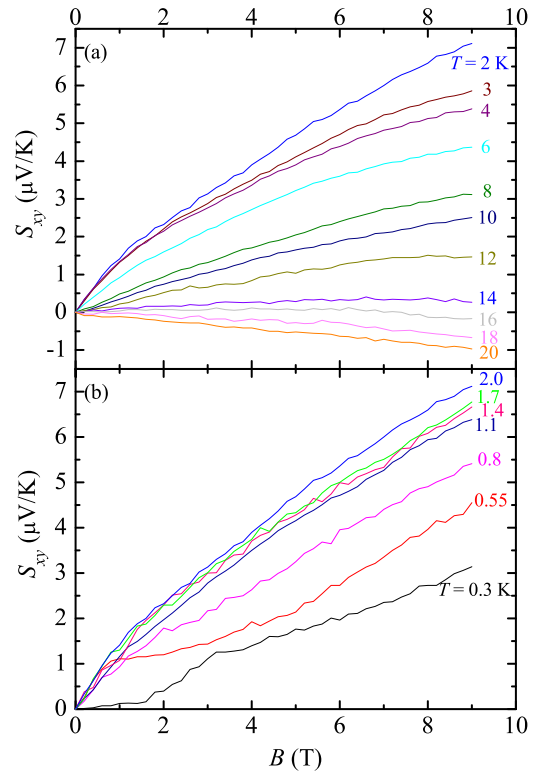

FIG. 3. Nernst signal S_{xy} of CeIrIn$_5$ as a function of B at selected temperatures. (a) $0.3 \leqslant T \leqslant 2.0$ K; (b) $2 \leqslant T \leqslant 20$ K.

S. $S_{xy}(B)$ is both negative and linear in B at 20 K. The magnitude of $S_{xy}(B)$ decreases with decreasing T and changes sign near 15 K [Fig. 3(a)]. The nonlinearity of $S_{xy}(B)$ becomes pronounced and the value of S_{xy} rapidly increases with decreasing T. At 2 K, S_{xy} reaches 7 μV/K when B is 9 T. We will see that such a large S_{xy}, even larger than that in the vortex-liquid state of cuprates [21,22], is mainly due to the Kondo effect, albeit the vortexlike excitation contribution is also non-negligible. A large Nernst effect also has been seen in other Kondo-lattice compounds, such as CeCoIn$_5$ [30–32], CeCu$_2$Si$_2$ [33], URu$_2$Si$_2$ [34], and SmB$_6$ [35]. In CeIrIn$_5$ S_{xy} starts to drop when T is lower than 2 K but remains positive down to 0.3 K, the base temperature of our measurements [Fig. 3(b)]. At 0.3 K, which is below T_c^b, $S_{xy}(B)$ increases slowly at small field but much more rapidly near 1.8 T. It is likely that this 1.8 T magnetic field defines a melting field B_m above which the vortex solid melts into a vortex-liquid state. A large number of vortices start to move in response to a temperature gradient and this results in the abrupt increase in $S_{xy}(B)$. Similar results also have been seen in other type-II superconductors, such as cuprates [21,22] and CeCoIn$_5$ [31]. This vortex-lattice melting field disappears immediately when T exceeds T_c^b, e.g., 0.55 K as shown in Fig. 3(b). This implies that a well-defined Abrikosov lattice of vortices only exists in the bulk superconducting state of CeIrIn$_5$.

Figure 4(a) shows the temperature dependence of the Nernst coefficient $\nu_N \equiv S_{xy}/B$. Here, the solid symbols are obtained at $B = 9$ T, and the open symbols represent the initial slope of $S_{xy}(B)$ as $B \to 0$. In both definitions, ν_N above T_c^{on} is large and sign changes near 15 K. It is well known that for a single-band, nonsuperconducting and nonmagnetic metal, the Nernst signal is vanishingly small, due to so-called Sondheimer cancellation [36],

$$S_{xy} = \rho_{xx}\alpha_{xy} - S_{xx}\tan\theta_H. \qquad (2)$$

A large Nernst effect has been observed in (i) multiband systems such as NbSe$_2$ [37] in which the ambipolar effect violates Sondheimer cancellation; (ii) phase slip due to vortex motion in type-II superconductors, as in underdoped cuprates [21,22]; (iii) ferromagnets such as CuCr$_2$Se$_{4-x}$Br$_x$ in which $S_{xy}(B)$ scales to magnetization $M(B)$, known as anomalous Nernst effect [38]; and (iv) Kondo-lattice systems, such as CeCu$_2$Si$_2$, in which an enhanced ν_N is determined by asymmetry of the on-site Kondo scattering rate [33].

We can exclude the anomalous Nernst effect in CeIrIn$_5$ because $S_{xy}(B)$ does not scale with the magnetization, which is essentially a linear function of B (data not shown). From the negative Hall resistivity $\rho_{yx}(B)$ shown in Fig. 2(c), we also rule out a substantive contribution from skew scattering because, as discussed in Refs. [28,39], it generates a *positive* anomalous Hall effect for Ce ions.

To study a possible multiband contribution to the Nernst signal of CeIrIn$_5$, we performed the same measurements on the non-4f counterpart LaIrIn$_5$. According to quantum oscillation measurements and density functional theory (DFT) calculations, LaIrIn$_5$ is electron-hole compensated [24,40], and a large Nernst effect is possible [37]. The Nernst signal of LaIrIn$_5$, however, is surprisingly both negative and linear in B [see Fig. 4(d) for instance], and most importantly, the Nernst coefficient remains small between 0.3 and 20 K [Fig. 4(a)].
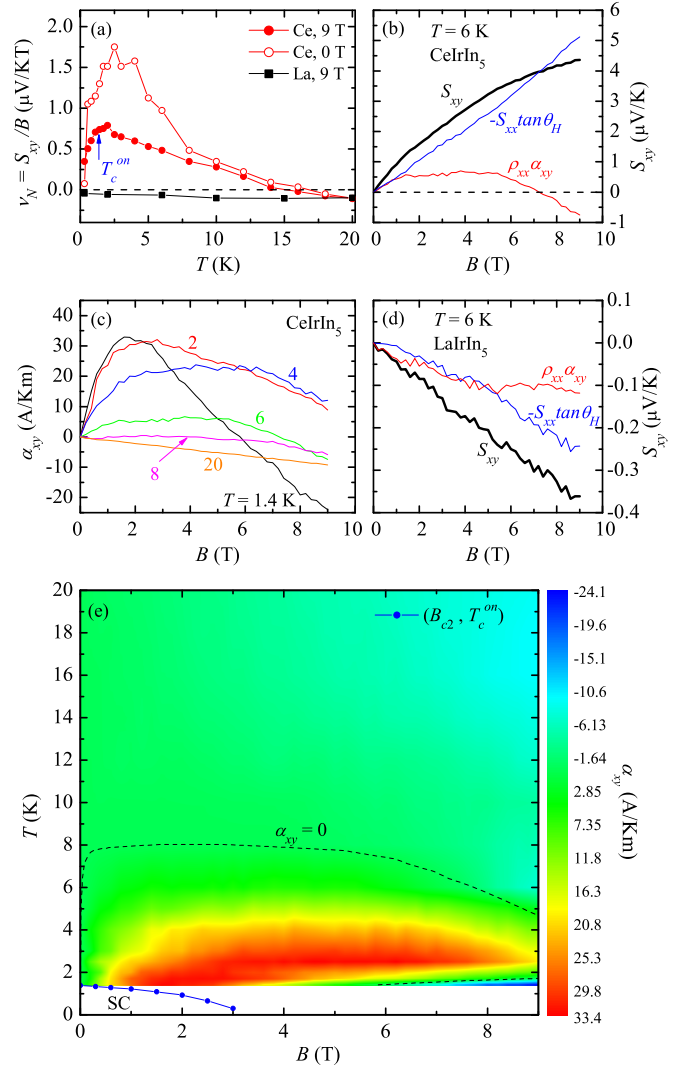

FIG. 4. (a) Temperature-dependent Nernst coefficient ν_N of LaIrIn$_5$ and CeIrIn$_5$. For CeIrIn$_5$, the open symbols are the initial slopes of $S_{xy}(B)$ as $B \to 0$. (b) and (d) show the separation of $\rho_{xx}\alpha_{xy}$ from S_{xy} at $T = 6$ K for CeIrIn$_5$ and LaIrIn$_5$, respectively. (c) Off-diagonal Peltier coefficient α_{xy} as a function of B at selected temperatures. (e) Contour plot of $\alpha_{xy}(B,T)$, with the resistively determined $B_{c2}(T)$ shown in the lower left corner. The black dashed line is the boundary where $\alpha_{xy} = 0$.

This demonstrates that a multiband effect does not play an important role in LaIrIn$_5$. Compared with LaIrIn$_5$, CeIrIn$_5$ has a somewhat larger Fermi surface due to a partially itinerant 4f band [24], electron-hole compensation is relatively unbalanced, and therefore the multiband contribution to the Nernst signal of CeIrIn$_5$ is expected to be even weaker.

To better understand the origin of a large Nernst effect in CeIrIn$_5$, we separate $\rho_{xx}\alpha_{xy}$ from the total Nernst signal S_{xy} [cf Eq. (2)]. As an example, we show S_{xy}, $\rho_{xx}\alpha_{xy}$, as well as $-S_{xx}\tan\theta_H$ at 6 K in Fig. 4(b). As seen, $-S_{xx}\tan\theta_H$ is the dominant contribution to S_{xy}. In a Kondo-lattice system, strong electronic correlations build up a resonance in the density of states near the chemical potential ε_F, and the scattering rate $(1/\tau)$ is now mainly determined by the very narrow, renormalized 4f bands, i.e., $N_f(\varepsilon)$. As a result, the

thermopower, given by Eq. (3), becomes large [41]:

$$S_{xx} \propto \frac{\partial \ln \tau}{\partial \varepsilon} \propto -\frac{\partial \ln N_f(\varepsilon)}{\partial \varepsilon}\bigg|_{\varepsilon = \varepsilon_F} \quad (3)$$

due to an asymmetric $N_f(\varepsilon)$ and is reflected in data plotted in Fig. 1(b). This asymmetry of on-site Kondo scattering also enters S_{xy} through the term $-S_{xx} \tan \theta_H$ and gives rise to the large Nernst effect in CeIrIn$_5$ and other Kondo-lattice systems as well [30,31,33,35].

We note that $-S_{xx} \tan \theta_H$ surpasses S_{xy} when B is larger than 7.3 T at 6 K, and this leads to a sign change in $\rho_{xx}\alpha_{xy}$ [Fig. 4(b)]. Figure 4(c) shows the field-dependent α_{xy} at various temperatures. Due to a large contribution from asymmetric Kondo scattering in $S_{xy}(B)$, $\alpha_{xy}(B)$ clearly differs from $S_{xy}(B)$ and, therefore, more intrinsically describes the off-diagonal thermoelectric response. $\alpha_{xy}(B)$ is negative and linear in B at 20 K. As T decreases, an anomalous positive term gradually appears on top of the negative linear background. Similar behavior was observed in CeCoIn$_5$ and was interpreted as a signature of phase-slip events caused by the passage of individual vortices [31]. To compare, we show $\rho_{xx}\alpha_{xy}$ at 6 K for LaIrIn$_5$ in Fig. 4(d). As expected, the unusual behavior is absent in LaIrIn$_5$ where there is only a small negative $\rho_{xx}\alpha_{xy}$.

It is reasonable to write α_{xy} in the form [31]

$$\alpha_{xy} = \alpha_{xy}^n + \alpha_{xy}^s, \quad (4)$$

where α_{xy}^n is the contribution from normal quasiparticles and α_{xy}^s represents an anomalous term stemming from vortex excitations. The positive $\alpha_{xy}(B)$ manifests that vortex motion dominates the quasiparticle term. We summarize these results in a contour plot of $\alpha_{xy}(B,T)$ in Fig. 4(e). Below the $\alpha_{xy} = 0$ boundary near 8 K, vortexlike excitations contribute and become most pronounced in the "island" region below 4 K. These temperature scales are qualitatively different from those in CeCoIn$_5$ in which the Nernst effect develops at very low temperature near a field-induced quantum-critical point [32]. We also note that the temperature dependence of α_{xy}^s/B in CeIrIn$_5$ cannot be reproduced even approximately by assuming that it arises from Gaussian superconducting fluctuations (data not shown) which seems successful in describing the Nernst effect for optimally doped and overdoped cuprates but not underdoped ones [42]. Taking $T_c^{on} = 1.38$ K in simulation, the calculated α_{xy}^s/B by the Gaussian model is an order of

magnitude smaller than the observed values. These findings suggest that local Cooper pairs start to form at a temperature well above T_c^{on} and that phase coherence among them is destroyed by thermally activated vortexlike excitations. We estimate the phase-order temperature (above which the phase coherence is destroyed), $T_\theta^{max} \sim 4$ K, if we adopt Emery's model [3] to CeIrIn$_5$ with lattice parameter $c = 7.515$ Å [13] and superconducting penetration depth $\lambda(0) \sim 10^4$ Å [43]. The ratio $T_\theta^{max}/T_c^b \sim 8$ (or $T_\theta^{max}/T_c^{on} \sim 2.9$) is significantly smaller than that of conventional superconductors (10^2–10^5) but is comparable to that of underdoped high-T_c cuprates (<10) [3] whose phase stiffness is soft. Perhaps not coincidentally, T_θ^{max} is comparable to the estimated zero-field temperature of a precursor state found in magnetotransport [16,17]. The filamentary nature of SC [14] also would imply a dilute superfluid density, which renders the phase fluctuations possible in CeIrIn$_5$ [3]. Finally, we note that the specific heat (C/T) of CeIrIn$_5$ deviates from a $-\log T$ dependence below ~ 2–4 K where it rolls over to a weaker (nearly constant) temperature dependence [44]. On a similar temperature scale, the ^{115}In nuclear spin-lattice relaxation rate ($1/T_1$) also shows a weak inflection at around 6 K [45]. These evolutions prior to T_c suggest the formation of a partial gap in $N(\varepsilon)$ that is in parallel with ungapped heavy quasiparticles. Whether these behaviors are the consequences of a possible pseudogap or correlated with the formation of local Cooper pairs is still an open question and requires further investigation.

Thermoelectric measurements in combination with charge transport in the heavy-fermion superconductor CeIrIn$_5$ indicate the formation of an unusual state above T_c that is reminiscent of cuprate physics. By separating the off-diagonal Peltier coefficient α_{xy} from S_{xy}, we find that α_{xy} becomes positive and greatly enhanced at temperatures well above T_c. Compared with the nonmagnetic analog LaIrIn$_5$, these results suggest vortexlike excitations in a precursor state of CeIrIn$_5$. This work sheds light on bridging the similarity between heavy-fermion and cuprate superconductors and is a step towards uncovering the mechanism of the unconventional superconductivity in the CeMIn$_5$ family compounds.

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