Anomalous thermopower and Nernst effect in CeCoIn₅: Loss of entropy current in precursor state

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Abstract – The heavy-electron superconductor CeCoIn₅ exhibits a puzzling precursor state above its superconducting critical temperature at TC = 2.3 K. The thermopower and Nernst signal are anomalous. Below 15 K, the entropy current of the electrons undergoes a steep decrease reaching ∼0 at TC. Concurrently, the off-diagonal thermoelectric current α_{xy} is enhanced. The delicate sensitivity of the zero-entropy state to field implies phase coherence over large distances. The prominent anomalies in the thermoelectric current contrast with the relatively weak effects in the resistivity and magnetization.

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Introduction. – Several novel, interesting phenomena have been discovered in the anisotropic, heavy-electron superconductor CeCoIn₅, which has a critical transition temperature TC = 2.3 K [1]. Evidence for d-wave pairing symmetry are seen in a number of experiments [2–5]. The Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state has been suggested to exist below a temperature T ∼ 0.3 K [6]. In a magnetic field H||c (the c-axis), a crossover from non-Fermi liquid to Fermi liquid behavior has been reported [7,8]. Magnetization results [9] reveal that a weak, unusual magnetic order begins to appear ∼20 K above TC. This precursor state is mysterious and often associated with a “hidden” order parameter. Recently, Bel et al. [10] reported that a giant Nernst signal appears at ∼25 K. They reported that the sign of the Nernst signal is opposite to that expected from the vortex-Nernst signal, which has been intensively studied in cuprates [11–13].

Using thermopower, Nernst-effect and other transport measurements on high-purity CeCoIn₅, we have determined both the diagonal and off-diagonal components of the thermoelectric (Peltier) conductivity tensor α. In the precursor state at H = 0, a pronounced reduction of the thermopower reveals a remarkably steep loss of carrier entropy current. A weak field suppresses this low-entropy state. After subtraction of the thermal-Hall contribution to the Nernst signal and correcting its sign, we show that the enhanced Nernst signal correlates with the thermopower anomalies in the precursor state.

Off-diagonal thermoelectric currents. – In a solid, an applied temperature gradient −∂T drives a charge current density J = α · (−∇T). Because the total current density in the sample is zero, an internal electric field E arises to drive a counter current J' = σ · E, where σ is the conductivity tensor. The x and y components of E are observed, respectively, as the thermopower ρα = E_x/∂xT and the Nernst signal e_N = E_y/∂yT (we choose axes with −∂T || x and H || z). The thermopower, which involves balancing 2 counter charge currents ||x, is just the ratio of 2 transport quantities, i.e. S = α/σ, with α = α_{xx} and σ = σ_{xx} (we drop subscripts on diagonal quantities). However, the Nernst signal e_N is more involved.

In the simplest situation, the transverse gradient −∂yT is negligible (isothermal case). Along ŷ, we have the 2 off-diagonal currents α_{yx}(−∂yT) and σ_{yx}E_x, which must be cancelled by σE_y. In terms of the resistivity tensor ρ = σ⁻¹, we have [12]

\[ e_N = \rho \alpha_{xy} + \rho_{xy} \alpha. \]  

The Nernst signal e_N senses the sum of the off-diagonal Peltier current and the ordinary Hall current multiplied by S.

However, when the thermal Hall conductivity κ_{xy} is large (as in CeCoIn₅ below 20 K [8]), a sizeable transverse gradient −∂yT appears, which drives a charge current ||ŷ

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Fig. 1: Panel (a): the field dependence of the thermopower $S(H)$ of CeCoIn$_5$ at selected $T$ ($H || c$). The zero-field anomaly deepens rapidly as $T$ fall towards $T_c = 2.3$ K. Below $T_c$, $S(H)$ rises nearly vertically at $H_{c2}$, but continues to increase to a broad maximum (at $\sim 8T$ at 1K). Beyond the peak $S$ decreases to a plateau value. Panel (b) shows the curves below 7K in expanded scale.

via $\alpha$ (this is just the ordinary thermopower responding to $-\partial_s T$). Instead of eq. (1), we have [12]

$$e_N = \rho \alpha_{xy} - S \left[ \frac{\sigma_{xy}}{\sigma} + \frac{\kappa_{xy}}{\kappa_e} \right],$$

which expresses balancing the 3 off-diagonal currents $\sim \alpha_{xy}$, $\sim \sigma_{xy}$ and $\sim \kappa_{xy}$ by the current $\sigma E_y$. Thus to obtain $\alpha_{xy}$ from $e_N$, we should subtract the charge and thermal Hall currents. (We adopt the sign convention [12,13] that $e_N$ shares the sign of $\alpha_{xy}$, i.e. when $\alpha_{xy} > 0$ and dominant, $e_N$ is positive. Vortex flow in a superconductor gives a positive $e_N$.)

Fig. 2: The $H$ dependence of the thermoelectric conductivity $\alpha$ from 2 to 17K, inferred from $S$ and $\rho$. At low $T$, $\alpha$ displays a narrow zero-field cusp which implies an electronic state in which the qp entropy current is strongly suppressed as $T \to T_c$. The state is very sensitive to $H$. As $T \to T_c$, $\alpha$ decreases (increases) if $|H|$ is smaller (larger) than $H_{in,f}$ (arrow drawn for 3K).

Thermopower and $\alpha$. – We first describe the field dependence of $S$ and the inferred $\alpha$. At low $T$, $S$ displays a very interesting field dependence (fig. 1a). Above $\sim 15$K, $S$ is nearly insensitive to $H$ aside from a weak cusp-like anomaly at $H = 0$. With decreasing $T$, the anomaly deepens and imparts a strong $H$ dependence that extends to $\sim 12T$. In the interval 3–7K (Panel b), the zero-$H$ cusp is bracketed by a steep $H$-linear dependence (at 3K, $S$ increases 14-fold between $H = 0$ and 12T). Below $T_c$, $S$ rises nearly vertically from zero at $H_{c2}$ (the upper critical field), attains a broad maximum (8.1T at 1K), and settles down to a plateau value at large $H$.

Using the magnetoresistance $\rho$ vs. $H$ from ref. [8], we have converted the $S$-$H$ curves into $\alpha$ vs. $H$ (fig. 2). At 17K, $\alpha$ displays the weak $H^2$ dependence characteristic of the normal-state thermoelectric conductivity $\alpha^n$ given by the Mott expression $\alpha^n = \frac{k_B^2 T^2}{\hbar^2} \left[ \frac{\partial \sigma}{\partial \epsilon} \right]_{\mu}$, where $k_B$ is Boltzmann’s constant, $\epsilon$ the elemental charge, $\epsilon$ the energy and $\mu$ the Fermi level.

However, as $T \to 3$K, the minima in $\alpha$ deepen to cusps even sharper than those in $S$. Below 15K, the curve of $\alpha$ displays an inflexion point at a field $H_{in,f}$ where $\partial^2 \alpha / \partial H^2 = 0$ ($H_{in,f}$ increases from 3.7T at 3K to 9.4T)
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Fig. 3: The observed Nernst signal $e_N$ vs. $H$ in CeCoIn$_5$ from $T = 9$ to 19 K (Panel a) and from 0.75 to 8 K (Panel b). Below $T_c$, $e_N$ undergoes a sharp jump from zero at $H_{c2}$. Throughout, $e_N$ includes a significant contribution from a current driven by $\kappa_{xy}$. The sign is always positive (see eq. (2)).

At 9 K. The inflexion field reveals 2 factors that pull $\alpha$ in opposite directions. As $T$ decreases, $\alpha$ increases if $|H| > H_{inf}$, but decreases for $|H| < H_{inf}$. The cusp anomaly is dominant for $|H| < H_{inf}$. Further, the field scale defining the cusp sharpness ($\sim 100$ G at 3 K) suggests that a weak $H$ suppresses long-range phase coherence of the electronic state at low $T$ (see below).

A notable feature is that the anomaly in $\alpha$ is much larger than that in $\rho$. For e.g., within the interval of the cusp ($H = 0 \rightarrow H_{inf}$), $\alpha$ increases more than five-fold at 3 K, whereas the cusp in $\rho$ constitutes only 20% of the total resistivity [8]. The unusually large cusps in $S$ (fig. 1b) reflect this huge discrepancy. We return to these features after describing the Nernst results.

Nernst and $\alpha_{xy}$. – In ref. [10], an enhanced Nernst signal was reported below $\sim 25$ K, but with a sign opposite to that of the vortex-Nernst effect [11–13]. Our measurements of $e_N$ (fig. 3) are nominally consistent in magnitude with ref. [10]. However, we find that the sign of $e_N$ is positive (after some correspondence [14], the authors report the sign as positive as well [15]).

Using $S$ and the tensors $\sigma$ and $\kappa$ measured in ref. [8], we now use eq. (2) to extract $\alpha_{xy}$. The separate contributions are shown in fig. 4a at $T = 4$ K. The curve of $e_N$ is the sum of $\rho\alpha_{xy}$ and the augmented Hall term $-S(\frac{\kappa_{xy}}{\sigma_{xy}} + \frac{\kappa_{xy}}{\sigma_{xy}})$. We note that, in CeCoIn$_5$, $S > 0$ (hole-like), while $\sigma_{xy} < 0$ and $\kappa_{xy} < 0$ (electron-like). Roughly $\frac{1}{3}$ of the Hall term derives from the current due to $\kappa_{xy}$ (dashed line).

The derived field profiles of $\alpha_{xy}$ are highly instructive (fig. 4b). At the high-$T$ end (17 and 19 K), the curves are $H$-linear, consistent with the off-diagonal thermoelectric response of quasi-particles (qp) in moderate fields. We identify this as the qp background term, atop of which a positive contribution to $\alpha_{xy}$ emerges as $T \rightarrow T_c$. Referring back to $e_N$ in fig. 3, we may now see that, at temperatures above 15 K, the “enhanced” Nernst signal with pronounced curvature merely reflects the large charge and thermal Hall currents $\sigma_{xy}$ and $\kappa_{xy}$. These contribute to

Fig. 4: Panel (a): separation of the observed Nernst curve $e_N$ at 4 K into the off-diagonal Peltier term $\rho\alpha_{xy}$ and the charge- and-thermal Hall term $-S(\frac{\kappa_{xy}}{\sigma_{xy}} + \frac{\kappa_{xy}}{\sigma_{xy}})$. As shown by the dashed curve, 30–40% of the latter derives from $\kappa_{xy}$. $S > 0$ (hole-like), while $\sigma_{xy} < 0$ and $\kappa_{xy} < 0$ (electron-like). Panel (b): the curves of $\alpha_{xy}$ vs. $H$ derived from $e_N$ using eq. (2). At 17 and 19 K, $\alpha_{xy}$ is nearly $H$-linear, consistent with a qp origin. The plotted quantity is the sum of 2 terms (eq. (3)). The broadly peaked profile at lower $T$ (<15 K) is the anomalous term $\alpha_{sxy}$. At the lowest $T$, the negative qp term $\alpha_{sxy}$ pulls $\alpha_{xy}$ to large negative values (−100 A/mK at 12 T and 2 K).
The pattern of the curves in fig. 4b suggests that \( \alpha_{xy} \) is comprised of 2 terms, viz.

\[
\alpha_{xy} = \alpha_{xy}^n + \alpha_{xy}^s,
\]

where \( \alpha_{xy}^n \) is the negative qp term and \( \alpha_{xy}^s \) the positive anomalous term. Below 15 K, the latter swells rapidly with a characteristic field profile that peaks at relatively low fields (2-3 T) and then decays slowly at large \( H \). The \( T \) dependence of \( \alpha_{xy} \) at a fixed \( H \) (3 T) is shown in fig. 5a. It is apparent that the anomalous term appears as a positive contribution on top of a negative, \( H \)-linear background.

In fig. 4b, the curves below 5 K exhibit a steep decrease to large negative values at high fields. As reported in ref. [8], measurements of \( \kappa_{xy} \) and \( \sigma_{xy} \) reveal that, below 6 K, the electronic mean-free-path \( \ell \) is sharply enhanced in large \( H \) because of field-suppression of scattering by spin disorder [8]. We identify the high-field changes in \( \alpha_{xy} \) with strong enhancement of the qp term \( \alpha_{xy}^n \), which scales like \( \ell^2 \) (\( \alpha_{xy}^n \) satisfies the Mott expression as \( \alpha^n \), with \( \sigma \) replaced by \( \sigma_{xy} \)). The increase in \( \ell \) strongly enhances \( \alpha_{xy}^s \), which is intrinsically negative, as we noted at 17–19 K. At 2 K, the qp contribution is so large that it pulls \( \alpha_{xy} \) to very large negative values (−100 A/mK at 12 T). Hence, at high fields, the total off-diagonal term \( \alpha_{xy} \) is dominated by the steep growth of the negative qp term, but at low fields, it is dominated by the positive anomalous term. This is just the 2 disparate trends separated by \( H_{nf} \) in the curves of \( \alpha \) (fig. 2), but now observed in the off-diagonal channel.

**Loss of entropy current.** – We now return to the implications of the thermopower. Figure 5a shows the \( T \) dependence of \( \alpha(T, H) \) at several fixed \( H \). Starting at 20 K, the zero-field curve \( \alpha(T, 0) \) initially decreases with a modest slope, but below 15 K, it accelerates to fall steeply to zero close to \( T_c \).

There are 2 unusual aspects of \( \alpha(T, H) \). First, in conventional superconductors, \( \alpha \) (and \( S \)) display a step-like decrease to zero at \( T_c \), whereas \( \alpha \) here is already strongly suppressed above \( T_c \). By Onsager reciprocity, \( \alpha = \tilde{\alpha}/T \), where \( \tilde{\alpha} \) defines the heat current \( \mathbf{J}_h \) produced by \( \mathbf{E} \) (with \( \nabla T = 0 \)), viz. \( \mathbf{J}_h = \alpha \mathbf{E} \), so that we may regard \( J_s = \alpha E \) as the entropy current density generated by \( \mathbf{E} \), so \( S \) is the entropy transported per unit charge (a modern treatment is given in ref. [16]). In this view, the sharp decrease of \( \alpha(T, 0) \) in the broad 13-K interval above \( T_c \) (fig. 5a) implies a loss of entropy of the carriers starting at \( \sim 15 \) K, and going to completion very close to \( T_c \).

Secondly, as shown in fig. 2, the zero-field behavior is very field sensitive. At 3 K, the cusp is rounded in a very weak field (100 G) and nearly completely suppressed at \( H_{nf} (3.7 \) T). A field of 100 G is equivalent to a magnetic length \( \sqrt{\hbar/eB} \sim 2600 \) Å. It is unlikely that field sensitivity on such long lengths can be explained semi-classically. Instead, it is indicative of field decoherence of an electronic wave function that retains phase coherence over large distances. Two examples displaying such weak-field sensitivity are the suppression of weak Anderson localization, via dephasing of paths related by time-reversal symmetry, and field suppression of phase coherence in the fluctuation regime of superconductors.

Within our accuracy, the onset temperature of \( \alpha_{xy}^s \) (fig. 5b) is identical with that of the cusp anomaly in \( \alpha \) (fig. 2). The actual anomalous contribution detected in the Nernst signal shares the same origin as the anomalous term in the thermopower; both appear below 15 K rather than 25 K. An important feature, however, is that \( \alpha \) is much more field sensitive than \( \alpha_{xy}^s \), as may be seen by comparing figs. 2 and 4b. The latter survives to fields larger than 6 T whereas the cusp in \( \alpha \) is affected by very weak \( H \).
Magnetization. – In analogy with the cuprates [12,13], an appealing candidate for the precursor state above $T_c$ would seem to be the vortex-liquid scenario in which the Cooper-pair condensate lacks phase rigidity on long length scales because of spontaneous (anti)vortices. The scenario may account for the steep fall of $\alpha$ as $T \rightarrow T_c$ because regions of the sample in which the condensate retains phase coherence have reduced entropy. In a field, vortices inserted by $H$ lead to rapid phase decoherence of these regions to produce the steep increase observed in $\alpha$. Moreover, the flow of vortices in the applied gradient generates a large, positive Nernst signal as observed. Insofar as the anomalous term $\alpha_{xy}$ arises from phase-slip events caused by the passage of individual vortices, the Nernst signal probes the existence of vorticity in a condensate with strong phase fluctuations. Thus it does not rely on having regions with long-range phase coherence, and can survive to large fields. This scenario is compatible with the rapid changes in $\alpha$ in weak $H$, as well as the relative robustness of $\alpha_{xy}$ to moderately large fields.

The vortex liquid above $T_c$ should exhibit a sizeable diamagnetic signal, as has been confirmed in the case of cuprates [17]. However, our measurements of magnetization $M$ have not uncovered such diamagnetism in CeCoIn$_5$ (fig. 6a). Above $T_c$, the magnetization is dominated by a $T$-dependent, paramagnetic susceptibility that is large ($\chi \sim 10^{-3}$) [9]. Within our resolution, $M$ is observed to be strictly linear in $H$ from 0 to 5 T at 3.5 K, with no trace of a diamagnetic fluctuation contribution (fig. 6b). Moreover, below $T_c$, the upper critical field $H_{c2}$ is sharply defined and given by the mean-field form $H_{c2} \sim (T_c - T)$, very unlike the situation in cuprates. Clarification of the magnetization may require experiments that can resolve a fluctuating $M$ at the level of a $1-10$ A/m [13].

Summary. – The thermoelectric current response in a field has revealed several unusual characteristics of the precursor state in CeCoIn$_5$ which appear below 15 K. In zero $H$, the systematic decrease of $S$ and $\alpha$ towards $\sim 0$ near $T_c$ implies a sharp loss of the entropy current carried by quasiparticles. The suppression of the entropy current is a new feature of the precursor state of CeCoIn$_5$. The striking field sensitivity of $\alpha$ implies that this zero-entropy feature occurs in regions with long-range phase coherence that are readily decohered by field. Concurrently, the off-diagonal current $\alpha_{xy}(-\nabla T)$ gains an anomalous positive contribution, which is readily distinguished from the negative $qp$ contribution. In contrast to these very prominent anomalies in the thermoelectric current, the modification to the resistivity is quite modest [8]. The contribution to the magnetization is currently below our resolution. Careful measurements of the electronic heat capacity above $T_c$ may be rewarding.

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