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Giant Nernst Effect in $CeCoIn_5$

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We present a study of Nernst and Seebeck coefficients of the heavy-fermion superconductor $CeCoIn_5$. Below 18 K, concomitant with a field-dependent Seebeck coefficient, a large sublinear Nernst signal emerges with a magnitude drastically exceeding what is expected for a multiband Fermi-liquid metal. In the mixed state, in contrast with all other superconductors studied before, this signal overwhelms the one associated with the motion of superconducting vortices. The results point to a hitherto unknown source of transverse thermoelectricity in strongly interacting electrons.

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Since the discovery of superconductivity in $CeCoIn_5$ [1], this Kondo lattice has attracted much attention. Unconventional superconductivity [2,3] in this compound occurs in the vicinity of a nearly avoided antiferromagnetic order and in a metal displaying a pronounced non-Fermi-liquid behavior [4–9]. The application of pressure [6] or magnetic field [7,8] leads to the emergence of a Fermi liquid. It is now generally believed that proximity to a quantum critical point (QCP) is the origin of the non-Fermi-liquid behavior in the heavy-fermion (HF) compounds [10] and that unconventional superconductivity mediated by magnetic fluctuations may arise in such a context [11].

In this Letter, we present a study of thermoelectric coefficients in this compound. Unexpectedly, we found a large sublinear Nernst signal emerging below 18 K. The magnitude of the Nernst coefficient in the low-field regime exceeds by far what is expected in a standard metal. In this regime, enigmatically, the ratio of Nernst to Seebeck signals is diverging with decreasing temperature and the thermoelectric response of the system tends to become purely transverse. Moreover, the emergence of this anomalous Nernst signal is concomitant with a number of non-Fermi-liquid features in various transport properties of the system. Notably, below 20 K, both resistivity and the inverse of the Hall coefficient display a linear temperature dependence [9]. The latter features are archetypes of non-Fermi-liquid transport in cuprates. Our results may thus offer interesting information for the ongoing debate on the origin of the anomalous Nernst signal observed in the normal state of high-T_c cuprates [12–14]. They add a new component to the intriguing analogy drawn between the cuprates and the $CeMIn_5$ family, which has gone as far as reporting the existence of a pseudogap in the latter [6].

Single crystals of $CeCoIn_5$ were grown using a self-flux method. Thermoelectric coefficients were measured using a one-heater-two-thermometer setup. A heat current was injected into the sample with a small resistive chip. The temperature gradient created was measured with two Cernox thermometers attached to local contacts along the sample. Two N11 nanovoltmeters (EM Electronics, U.K.) were used to measure dc voltages associated with the longitudinal and transverse electric fields produced in the sample by the thermal current. In all measurements, the heat current and the thermal gradient were oriented in the basal plane of the crystal, while the magnetic field was applied along the c axis.

Figure 1 displays the temperature dependence of the Seebeck and Nernst effects in a single crystal of $CeCoIn_5$. Similar results were obtained in a second sample. As seen

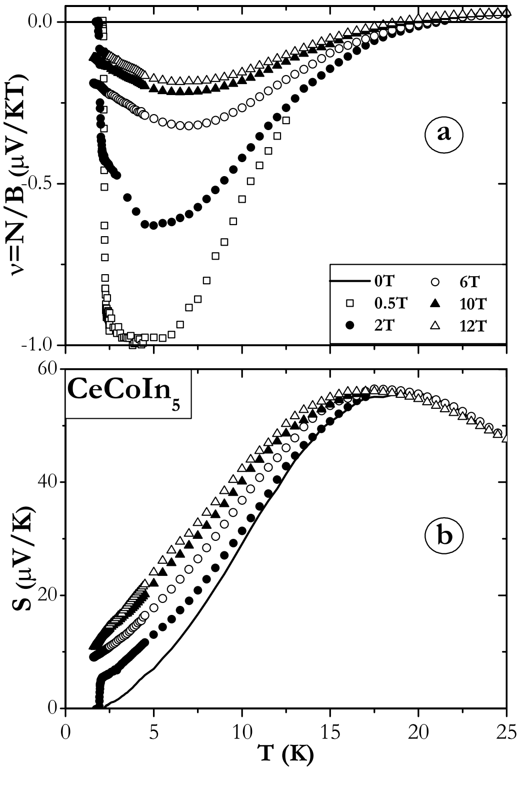

FIG. 1. (a) Temperature dependence of the Nernst coefficient for different magnetic fields. (b) Same for the Seebeck coefficient. Note also the zero-field data for the latter.

in the lower panel of the figure, the zero-field Seebeck coefficient in this sample presents a maximum at $T^* \sim$ 18 K. Generically, HF compounds present a maximum in thermopower close to the temperature which marks the onset of coherent scattering from Kondo sites [15,16]. Note that here, as in many other HF systems, the maximum in $S(T)$ occurs at a temperature which is roughly twice lower than the one associated with maximum resistivity. As seen in the figure, below T^*, the zero-field thermopower decreases rapidly before vanishing in the superconducting state. The occurrence of superconductivity apparently impedes a low-temperature sign reversal observed in many Ce-based HF systems [17]. The application of a magnetic field leads to the enhancement of the Seebeck coefficient below T^*. Previous studies of low-temperature thermopower have detected a strong field dependence in several cases, such as $CeAl_3$ [15], $CeRu_2Si_2$ [18], UBe_{13} [19], and $CeCu_{6-x}Au_x$ [20]. Interestingly, in all these cases, the magnetic field is known to strongly alter a small characteristic energy of the system. The strong variation of the Seebeck coefficient with magnetic field in $CeCoIn_5$ can be attributed to a similar effect. At zero field, superconductivity occurs before the formation of well-defined quasiparticles. Thus, T_{FL}, the temperature below which the system displays a Fermi-liquid behavior, is never attained. A field-dependent thermopower may indicate that the magnetic field significantly increases the energy scale associated with T_{FL}. Indeed, the emergence of Fermi-liquid behavior in a moderate magnetic field [7,8] points to the existence of such a tunable energy scale.

As seen in the upper panel of Fig. 1, the transverse thermoelectricity of the system displays a more striking behavior. The Nernst signal which is small, positive, and field linear above 18 K, becomes negative, large, and strongly nonlinear below this temperature. By plotting the Nernst coefficient ν, defined as the ratio of transverse electric field to longitudinal thermal gradient divided by magnetic field [$\nu = N/B = E_y/(\nabla_x T B)$], as a function of temperature, this spectacular change of regime is easily appreciated. The anomalous Nernst signal presents a broad maximum around a field-dependent temperature which is about 4 K at 0.5 T and increases to 7 K at 12 T. Note also the presence of the superconducting transition which leads to a vanishing Nernst signal in the 0.5 and 2 T data.

The maximal Nernst coefficient for $B = 0.5$ T is remarkably large ($\sim 1\ \mu$V/KT). Its magnitude is comparable with the maximum attained in the superconducting state of high-T_c superconductors at considerably higher temperatures. It is by far larger than the residual signal observed above T_c in the same compounds and attributed to vortexlike excitations [12–14]. Contrary to the case of cuprates, the negative sign of the signal observed here excludes a vortex-related interpretation. To our knowledge, with the notable exception of the giant signal recently discovered in a Bechgaard salt for fields oriented along the so-called magic angles [21], a Nernst coefficient of this size has not been reported in any other metal.

The very unusual thermoelectricity of $CeCoIn_5$ is emphasized in Fig. 2 which presents the ratio of Nernst to Seebeck signals as a function of temperature and magnetic field. This ratio represents the tangent of the angle between the thermal current and the electric field, $N/S = \tan\theta_{TE}$. As seen in the lower panel of the figure, above T^* it is a simple linear function of magnetic field. A remarkable nonmonotonous behavior emerges when the sample is cooled down below this temperature. In particular, at lower temperatures, it rapidly becomes a decreasing function of magnetic field. In other words, *the misalignment of the electric field decreases with increasing magnetic field.* As seen in the upper panel of the figure, in the low-field limit ($B = 0.5$ T), $\tan\theta_{TE}$ is a strongly diverging function of temperature and its enhancement is interrupted only by the superconducting transition. This divergence is wiped out by the application of a moderate magnetic field, and one observes a saturation of the ratio at low temperatures. Amazingly, in the zero-field limit, as the system is cooled down towards $T = 0$, *the electric field produced by a longitudinal heat current tends to become purely transverse.*

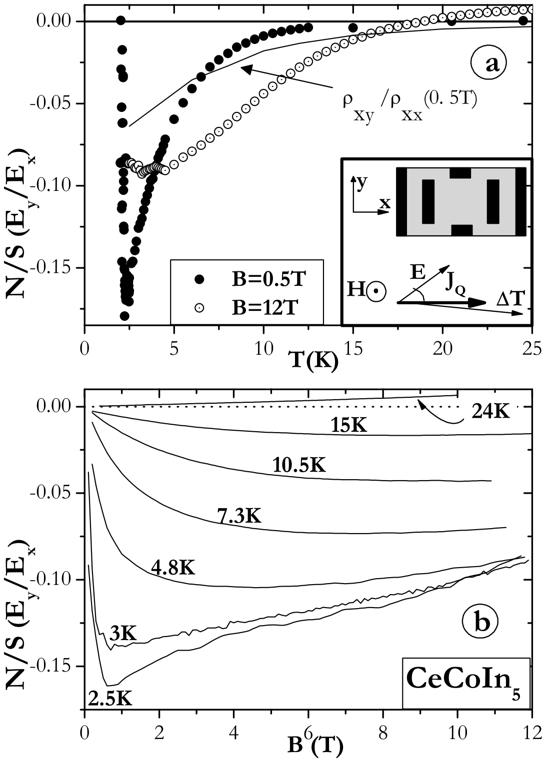

FIG. 2. The ratio of transverse to longitudinal electric fields produced by the thermal current as a function of temperature (a) and magnetic field (b). This ratio represents the tangent of the angle between the longitudinal thermal current and the induced electric field, $\tan\theta_{TE}$. The inset in (a) represents the sample geometry and miscellaneous vectors. For comparison, the temperature dependence of the electric Hall angle, ρ_{xy}/ρ_{xx}, is also plotted in (a).

Now, we turn our attention to the superconducting state. Figure 3 displays the temperature dependence of the thermoelectric coefficients in the vicinity of the superconducting transition for various magnetic fields. As seen in the figure, within the entry of the system into the mixed state, both thermoelectric coefficients of the system decrease monotonously towards zero. Since below a temperature corresponding to the solidification of the vortex lattice one would not expect any finite electric field in a superconductor, the vanishing of both Nernst and Seebeck effects is naturally expected. What is striking, on the other hand, is the absence of any easily detectable Nernst signal associated with vortex movement under the influence of the thermal gradient. In the whole range of study, the Nernst signal remains negative and displays a magnitude smaller than the Seebeck coefficient. This is in sharp contrast with any superconductor studied until now. In the mixed state, the thermoelectric response of the system has been always found to be dominated by the vortex movement along the thermal gradient due to the entropy carried by the vortex core. In CeCoIn$_5$, this signal is buried under the weight of the anomalously large signal of the normal state. Comparing the temperature dependence of Nernst and Seebeck coefficients, we found that for each field the superconducting transition leads to a faster collapse of the transverse signal. In order to resolve a Nernst signal induced by superconductivity, we subtracted a constant negative fraction of the Seebeck signal from the measured Nernst signal. This procedure allows one to detect any abrupt change in the N/S ratio in the narrow temperature range associated with vortex movement. As seen in the lower panel of the figure, which shows the case of $B = 0.5$ T, the signal thus extracted is finite and positive. Repeating the same procedure for different magnetic fields, we found a similar *positive* Nernst signal superposed on the background. Comparison with the resistive transition in the same magnetic field confirms that the extracted signal represents the vortex contribution to the overall Nernst signal. The magnitude of this contribution is comparable to what was observed in the conventional superconductor NbSe$_2$ [22].

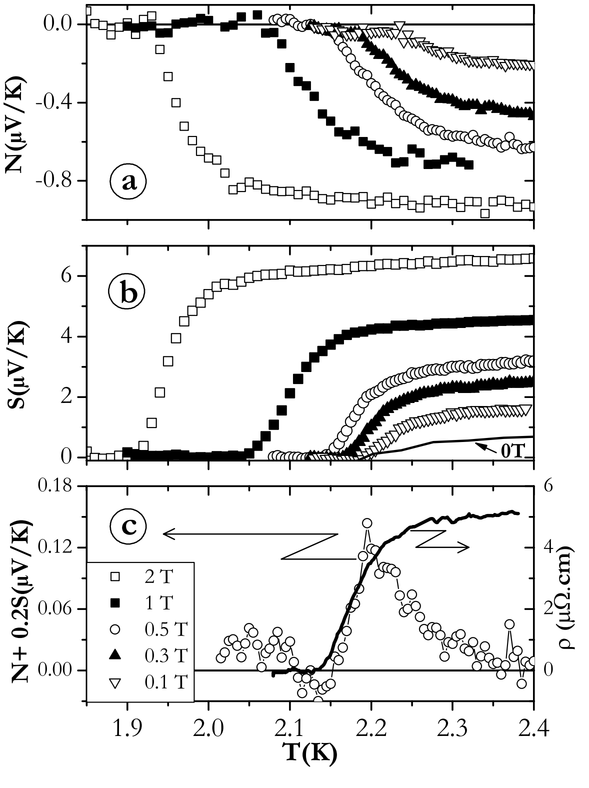

FIG. 3. Temperature dependence of the Nernst signal (a) and the Seebeck coefficient (b) for different magnetic fields in the vicinity of the superconducting transition. In panel (c), the difference between the Nernst signal and a fraction of the Seebeck signal at 0.5 T is plotted vs temperature and compared with the temperature dependence of the resistivity (solid line) measured in the same conditions.

As stated above, in CeCoIn$_5$, superconductivity occurs before the formation of well-defined quasiparticles of a Fermi liquid. It has been recently reported that, above a threshold field (~5 T) and below a field-dependent characteristic temperature, $T_{\mathrm{FL}}(H)$, two signatures of a Fermi liquid are recovered: Resistivity presents a quadratic temperature dependence [7] and the linear term of specific heat, instead of displaying a logarithmic divergence, tends to a constant value [8]. While our study of thermoelectricity is confined to an area of the (H, T) plane which has a very small overlap with the region of Fermi-liquid recovery, it presents features which support the picture of a field-induced Fermi liquid. Above a temperature-dependent threshold field, close to the $T_{\mathrm{FL}}(H)$ line, the Seebeck coefficient becomes almost field independent and temperature linear. Moreover, the magnitude of S/T yields an electronic entropy in very good agreement with the magnitude of the high-field linear term in specific heat. Indeed, multiplying S/T by the Avogadro number and the charge of electron [23], one obtains $N_{av}eS/T = 0.65$ J/mol K^2 at 12 T, surprisingly close to $C/T = 0.6$ J/mol K^2 reported at 9 T [8]. Note, however, that in this Fermi-liquid regime, the Nernst signal, while ceasing to increase as a function of magnetic field, is still sizable. It is also important to notice that the anomalous thermoelectricity reported here is a transport phenomenon. Indeed, contrary to what has been theoretically predicted [24], the zero-field Seebeck coefficient does not simply follow the temperature dependence of the specific heat C. While C/T presents a logarithmic divergence, S/T decreases rapidly with decreasing temperature. Moreover, for temperatures above $T_{\mathrm{FL}}(H)$, the application of the magnetic field leaves the specific heat virtually unchanged [8] but drastically enhances the thermopower. Finally, we notice that the anomalous transverse thermoelectricity is sharpest in the zero-field limit and not at $B = 5$ T which is supposed to host a QCP.

Explaining the large magnitude of the transverse thermoelectric response in CeCoIn$_5$ is a challenge to the theory. In a single-band metal, the Nernst coefficient is expected to be negligible [13]. The presence of carriers of different signs leads to an enhanced signal, known as the ambipolar Nernst effect [25], as recently illustrated in the case of $\mathrm{NbSe_2}$ [22]. In the latter system, at $T \sim 20$ K the contributions of holes and electrons to the Hall coefficient cancel out. Such an accidentally compensated metal is expected to yield a significant ambipolar Nernst effect and indeed the maximum Nernst signal (0.15 μV/KT) occurs at the temperature corresponding to a zero Hall coefficient. In $\mathrm{CeCoIn_5}$, the finite field-linear Nernst signal above 18 K is probably linked to the presence of several bands and carriers of both signs in the system [5]. On the other hand, the large and strongly nonlinear Nernst signal which emerges below this temperature remains enigmatic. The Hall coefficient in this compound, while displaying a nontrivial temperature and field dependence, never becomes zero [9]. As seen in Fig. 2, the temperature dependence of the electric Hall angle, $\tan\theta_H$ cannot account for the diverging $\tan\theta_{\mathrm{TE}}$ in the zero-field, zero-temperature limit. Indeed, below 6 K, the latter exceeds the former which means that the Nernst signal becomes larger than $S\tan\theta_H$ which gives an upper limit to the Nernst signal in absence of Sondheimer cancellation [13].

In the absence of systematic studies of the Nernst coefficient in other HF systems, one may speculate in several directions. We begin by noting that, beyond the relaxation time approximation, antiferromagnetic fluctuations may lead to an enhanced Nernst signal as well as other non-Fermi-liquid transport properties [26]. The possible relevance of vertex corrections to the anomalous thermoelectricity observed here remains an interesting direction of investigation.

Another possibility is that the anomalous Nernst signal is a consequence of the proximity of a QCP. At this stage, no direct connection between quantum criticality and Nernst effect has been suggested. However, the behavior of the Hall coefficient in the vicinity of a QCP has been a subject of debate [27,28]. A sudden change in the volume of the Fermi surface and, consequently, a sharp anomaly in the magnitude of the Hall coefficient may occur at the QCP [27]. Now, the Nernst coefficient is intimately related to the off-diagonal element of the Peltier conductivity tensor α_{xy} which is the energy derivative of the off-diagonal conductivity tensor, σ_{xy}, at the Fermi energy [13]. Thus, if, in the vicinity of a QCP, σ_{xy} becomes particularly sensitive to any slight modification of the volume of the Fermi surface, this will lead to an enhanced α_{xy} and a sizable Nernst signal.

An even more exotic scenario is to invoke the presence of collective modes in a Kondo lattice [29]. Very recently, the Kondo lattice problem was reconsidered in a two fluid picture with a fluid of heavy electrons (the condensate) coexisting with a normal fluid of Kondo impurities [30]. The possible association of entropy reservoirs with orbital moments in such a picture may constitute a source of Nernst signal in a manner analogous to the thermally induced motion of superconducting vortices. Future experiments on other HF compounds should tell if an enhanced Nernst effect is a generic feature of a Kondo lattice.

In summary, we measured the thermoelectric coefficients of $\mathrm{CeCoIn_5}$ and found an anomalous Nernst signal pointing to an enigmatic source of thermoelectricity in exotic metals.

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