The Lorenz number in CeCoIn$_5$ inferred from the thermal and charge Hall currents

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Abstract – The thermal Hall conductivity $\kappa_{xy}$ and Hall conductivity $\sigma_{xy}$ in CeCoIn$_5$ are used to determine the Lorenz number $L_H$ at low temperature $T$. This enables the separation of the observed thermal conductivity into its electronic and non-electronic parts. We uncover evidence for a charge-neutral, field-dependent thermal conductivity, which we identify with spin excitations. At low $T$, these excitations dominate the scattering of charge carriers. We show that suppression of the spin excitations in high fields leads to a steep enhancement of the electron mean-free-path, which leads to an interesting scaling relation between the magnetoresistance, thermal conductivity and $\sigma_{xy}$.

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Introduction. – The heavy-electron system CeCoIn$_5$ exhibits a host of unusual electronic properties of current interest. In the superconducting state, strong evidence for $d$-wave pairing symmetry has been reported [1–5]. The FFLO state involving pairing with unequal spin populations in an in-plane magnetic field ($\mathbf{H} \perp \mathbf{c}$) has been proposed [6]. The phase diagram in a perpendicular field ($\mathbf{H} | \mathbf{c}$) has also received wide attention [7,8]. In an extended region in $T$-$H$ plane surrounding the superconducting (SC) state (labelled I in fig. 1a), the resistivity $\rho$ and heat capacity exhibit distinctive “non-Fermi liquid” characteristics: $\rho \sim T$ [7], while the Sommerfeld parameter $\gamma(T) \sim \log T$ [8]. When $H$ exceeds the boundary $H_s(T)$, $\rho$ recovers the Fermi-liquid form $\rho = \rho_0 + AT^2$ and the unconventional features of $\gamma(T)$ are suppressed. The high-field region (labelled II in fig. 1a) is called the Fermi-liquid region. The boundary field $H_s(T)$ and the upper critical field $H_{c2}$ terminate at a quantum critical point (QCP) as $T \rightarrow 0$ (the field scale $H_Z$ is discussed below). Several parameters characterizing resistivity display divergent behavior as the QCP is approached [7]. In addition, a large Nernst signal is observed in I [9,10].

To clarify the electronic state in the region I, we have measured extensively the in-plane thermal conductivity $\kappa \equiv \kappa_{xx}$ with $\mathbf{H} | \mathbf{c} | \mathbf{z}$, and the thermal Hall conductivity $\kappa_{xy}$ (the Righi-Leduc effect) in crystals with very long

Fig. 1: (a) The phase diagram of CeCoIn$_5$ adapted from refs. [7,8], showing some features inferred from our experiment. The dashed line is the boundary $H_s(T)$ between the Fermi liquid (II) and non-Fermi Liquid (I) regions reported in refs. [7,8]. Solid triangles are our values for $H_s(T)$. Open circles indicate the field scale $H_Z$ (this work) above which the current ratio $R_e = Z$ (see eq. (3)). (b) The $T$ dependence of $\kappa$ in zero field (solid circles) showing a large $q_p$ peak below $T_c$. The background term $\kappa_b$ measured at 6 T is shown as solid triangles. Below $\sim 8$ K, $\kappa_b$ is largely comprised of a term $\kappa_s$ that is very field dependent (and identified with spin excitations).
electron mean-free-path $\ell$. In addition, we measured the electrical conductivity $\sigma \equiv \sigma_{xx}$ and Hall conductivity $\sigma_{xy}$. The crystals, grown from metallic flux [1], are plate-like with the $c$-axis normal to the broadest faces and the $a$-axis along one edge (for structure, see ref. [11]). The zero-field $\kappa$ displays a prominent peak below $T_c$ (solid circles in fig. 1b), which arises from the steep increase in $\ell$ of Bogolyubov excitations in the superconducting state. Using the Hall conductivities $\kappa_{xy}$ and $\sigma_{xy}$, we demonstrate the validity of the Wiedemann-Franz (WF) law, and then use the Lorenz number to separate the total $\kappa$ into its electronic and non-electronic components $\kappa_e$ and $\kappa_b$, respectively. From the strong field dependence observed in $\kappa_e$, we infer that spin excitations provide the dominant scattering mechanism for the charge carriers in the region I. The application of an intense field leads to suppression of this scattering channel and a sharp increase in $\ell$. This insight sheds light on the large magnetoresistance and the unusual features of the Hall effect. We discuss the implications for Cooper pairing in the SC region.

**Thermal and charge conductivity tensors.** The thermal resistivity tensor $W_{ij}$ is measured by applying a weak gradient ($\delta T \sim 10 \text{ mK}$ along length of the crystal at 0.5 K). Below 2 K, the strong variation of $\ell$ with $T$ and $H$ is potentially the largest source of error in comparing $W_{ij}$ (measured in finite $\delta T$) with $\rho_{ij}$ ($\delta T = 0$). We minimized the uncertainties by extensive calibration of the RuO$_2$ thermometers (glued to the crystal), and using very slow field scans (0.1–0.2 T/min at 0.5 K).

We emphasize that, because $\ell$ attains very large values below 10 K, it is necessary to use the full matrix inversion to reliably convert the measured tensors $W_{ij}$ and $\rho_{ij}$ into their reciprocal conductivity tensor, e.g. $\kappa_{xx} = W_{xx}/(W_{xx}^2 + W_{xy}^2)$. Experimentally, this means that $\kappa$ and $\sigma$ in strong fields cannot be obtained without measuring simultaneously the diagonal and off-diagonal elements of $W_{ij}$ and $\rho_{ij}$ (leaving out the Hall tensor elements leads to errors in $\kappa$ and $\sigma$ of 30% or more at low $T$).

Figure 2 compares the field dependences of $\kappa$ (panel a) and the in-plane resistivity $\rho$ (panel b) at temperatures from 5 K to 0.5 K with $H||c$. Above 5 K, an increasing field decreases slightly the observed $\kappa$. With decreasing $T$, however, this trend changes. At 2 K, $\kappa$ rises gradually with $H$. At even lower $T$ (0.5–1.5 K), this rising trend becomes firmly established in the normal state when $H$ exceeds $H_{c2}$ (step in $\kappa$). In the SC region below $H_{c2}$, a prominent feature in $\kappa$ is the sharp, narrow spike which represents the rapid field suppression of the zero-field peak caused by scattering of Bogolyubov excitations from vortices [12–14]. The spike in $\kappa$ vs. $H$ below $T_c$ is much larger than previously reported [3]. This reflects a much longer $\ell$ in the present samples.

The complicated behavior of $\kappa(T,H)$ arises because it is the sum of the electronic term $\kappa_e$ and a “background” term $\kappa_b$ carried by charge-neutral excitations (spin excitations and phonons), viz.

$$\kappa(T,H) = \kappa_e(T,H) + \kappa_b(T,H). \quad (1)$$

Our main finding is that, in CeCoIn$_5$, the charge-neutral term $\kappa_b(T,H)$ displays an unexpectedly strong $H$ dependence. Its $T$-profile at $H = 6 \text{ T}$ is shown as solid triangles in fig. 1b.

As previously found [7,15], CeCoIn$_5$ exhibits a large magnetoresistance (MR) (fig. 2b). The initial positive MR ($H < 3 \text{ T}$) is caused by suppression of superconducting fluctuations which we discuss elsewhere [10]. Our focus is on the negative MR that prevails for $H > 4 \text{ T}$ at all $T$ below $\sim 30 \text{ K}$. As $T$ decreases towards $T_c$, the negative MR becomes pronounced. At 2 K, $\rho$ decreases by $\sim 2.5$...
when $H$ reaches 14T. Both the sign and magnitude preclude classical MR associated with the Lorentz force. As shown below, the MR results from a steep enhancement of $\ell$ with increasing field.

**Lorenz number from scaling of $\kappa_{xy}/T$ to $\sigma_{xy}$.**

The field enhancement of $\ell$ strongly influences the field profiles of the heat and charge currents. To disentangle these effects, we exploit the WF law, which states that the ratio $\kappa_e/T\sigma$ is close to the Lorenz factor $L_0 = \frac{1}{4}\pi^2 (k_B/e)^2$. In the elemental metals, the WF law is nearly universally obeyed at 300 K as well as in the impurity-scattering regime below 4K, while deviations are common in between. However, in many interesting metals with low carrier densities, $\kappa_e$ cannot be measured directly because the charge-neutral term $\kappa_b$ (usually from phonons) is comparable in size or larger.

Recently, a way to separate $\kappa_e$ from $\kappa$ using the Righi-Leduc effect was introduced. Zhang et al. [16] have shown that the Lorenz ratio may be determined from the ratio $L_H \equiv \kappa_{xy}/T\sigma_{xy}$. (Essentially, the Righi-Leduc effect senses only the electronic entropy current, while filtering out the charge-neutral components which do not have a Hall response. Since the latter also do not contribute to $\sigma_{xy}$, the ratio of the 2 Hall currents yields the WF ratio. The WF-Hall method was tested on high-purity Cu and applied to cuprates [16].) CeCoIn$_5$ is well-suited for this method because the 2 Hall conductivities are large.

The Hall resistivity in CeCoIn$_5$ was previously reported [15], but it is the Hall conductivity that is of interest here. At each $T$, we find that the profile of $\sigma_{xy}$ vs. $H$ matches that of $\kappa_{xy}$ even when the two curves display strong curvature vs. $H$. The curves of $\kappa_{xy}/T$ and $L_H\sigma_{xy}$ are plotted together in fig. 3 for $T \leq 3$K (panel a), and $T > 3$K (b). Let us first note that the curves share 2 characteristics rarely seen in Hall experiments. In weak $H$, the curves rise from zero with strong negative curvature to produce a knee-like feature. In addition, the curvature changes its sign to positive in higher fields; both Hall conductivities increase more rapidly than the first power in $H$ in strong fields. Further, we note the peak anomaly in weak $H$ shown by $\kappa_{xy}$ below 1.5K. We return to these unusual features later.

At each $T$, $\kappa_{xy}/T$ and $L_H\sigma_{xy}$ may be scaled together over the entire field range by adjusting $L_H$. We emphasize that $L_H$ is an $H$-independent scaling parameter (otherwise, it does not make sense to discuss scaling between $\kappa_{xy}/T$ and $\sigma_{xy}$). In view of the pronounced nonlinearity, the close match between the 2 field profiles is strong evidence that the WF law is valid with a field-independent Lorenz number. The inferred values of $L_H$ are plotted in fig. 4. Between 2 and 10 K, $L_H/(k_B/e)^2$ is close to the Sommerfeld value $\pi^2/3$, but seems to deviate slightly downwards below 2K.

**Separation of electronic and non-electronic heat currents.**

We next determine $\kappa_e(T,H)$ and $\kappa_b(T,H)$ in eq. (1). Using the values of $L_H$ in fig. 4, we convert the measured $\sigma$ into $\kappa_e(T,H)$ via

$$\kappa_e(T,H) = T\sigma(T,H)\mathcal{L}_H(T).$$

Subtracting the curve of $\kappa_e(T,H)$ from $\kappa(T,H)$ at each $T$, we finally determine $\kappa_b(T,H)$, which is plotted in fig. 5a.

At low $T$, $\kappa_b$ is found to be strongly $H$-dependent. In general, $\kappa_b$ is the sum of the spin-excitation conductivity $\kappa_s$ and the phonon conductivity $\kappa_{ph}$, viz. $\kappa_b(T,H) = \kappa_s(T,H) + \kappa_{ph}(T)$. When spin-disorder scattering of phonons is important, an applied field generally leads to an increase in $\kappa_{ph}$ because $H$ suppresses spin disorder, which is opposite to what is observed. Consequently, we identify all the field dependence with
the spin-excitation term $\kappa_s(T, H)$. Figure 5a shows that $\kappa_s$ accounts for a large fraction of $\kappa_b$ between 5K and 1K. At the lowest $T$ (0.5–1.5K), the curve of $\kappa_b$ falls to a floor value at the field scale $H_s(T)$, which is observable as a break-in-slope. In the phase diagram in fig. 1a, $H_s(T)$ is seen to lie close to the I/II boundary (solid triangles). With the present data, we cannot determine $H_s$ above 1.5K. Nonetheless, the trends of the curves in fig. 1a suggest that, throughout the region I up to 5K, $\kappa_b$ is greatly reduced from its zero-field values when $H$ reaches the I/II boundary in the phase diagram (fig. 1a).

Spin degrees and charge transport. – In a conventional magnet, an external $H$ raises the magnon dispersion energy which reduces the spin-wave population and their thermal conductivity. In the region I of CeCoIn$_5$, the uniform susceptibility $\chi$ is strongly enhanced, but conventional long-range magnetic order seems to be absent. However, in heavy fermions, spin-ordered states involving the local moments in the $f$ bands are widely postulated. Incipient spin ordering may exist above $T_c$ in CeCoIn$_5$ (see Broholm [20]). Although our analysis is guided by the known properties of conventional spin waves, a more exotic kind of spin ordering is not precluded, and $\kappa_s$ may derive from spin-excitations in unconventional spin-ordered states. Because of hybridization between the $f$ and $s$-$p$-$d$ states and large spin-orbit coupling, the spin excitations will strongly scatter the charge carriers. We write $\kappa_s = \frac{1}{3} c_s v \lambda$, where $c_s$ is the heat capacity of the spin excitations, $v$ the average velocity and $\lambda$ their mean free path. As the local moments align with $H$, the spin excitation population $n_s \sim c_s$ decreases steeply in field. We interpret the curves of $\kappa_b$ in fig. 5a as the sharp field-suppression of $n_s$ at low $T$. While evidence for heat currents carried by spin excitations have been reported

Fig. 4: The Lorenz number $L_H$ obtained by scaling $\kappa_{xy}/T$ to $\sigma_{xy}$ in fig. 3. $L_H$ is plotted in units of $(k_B/e)^2$. The dashed line is the Sommerfeld value $\pi^2/3$.

Fig. 5: (a) The curves of $\kappa_b = \kappa_s + \kappa_{ph}$ vs. $H$ obtained by subtracting $\kappa_e$ from the observed $\kappa$ at each $T$ ($\kappa_b$ is not obtained below $H_{c2}$ because $\rho = 0$). At 0.5, 0.75 and 1K, the $H$-dependence shows a kink at $H = H_s(T)$ at the boundary between I and II (fig. 1a). The curve of $\kappa_b$ vs. $T$ at 6T is shown in fig. 1b. (b) Comparison of $\sigma_{xy}$ (bold curves) with the quantity $\sigma^2 B Z$ (thin curves). Above the field $H_Z(T)$, the 2 quantities match over nearly 2 decades with one scaling constant $Z = 1 \times 10^{-7}$ cm$^3$/C. Below $H_Z$, however, the scaling is spoilt by an “excess Hall current”.
for low-dimensional oxides [21], a distinguishing feature in CeCoIn$_5$ is that changes in $\kappa_b$ strongly affect the charge currents, which we describe next.

The curves in fig. 5a reveal that the charge-neutral conductivity $\kappa_b$ decreases strongly with increasing $H$. This trend is opposite to that in the electronic conductivity $\sigma$. As the former decreases, the latter rises in almost direct proportion. (The ratio of $\kappa_b(H)$ evaluated at $H = 0$ and 14 T, $\kappa_b(0)/\kappa_b(14) \approx 1.5$ and 2.7 at 5K and 2K, respectively. These ratios match the corresponding ratios $\rho(0)/\rho(14)$ at the same $T$ in fig. 2b.) Panel b of fig. 5 shows the steep increase of $\sigma^2$ and $\sigma_{xy}$ with $H$. As discussed above, the sharp decrease in $\kappa_b$ with $H$ reflects a decrease in the density of spin excitations $n_s$. Hence the correlated increase in $\sigma$ implies that spin excitations are the dominant scattering mechanism of the carriers at these temperatures. The steep increase in $\ell$ in high field is caused by field-suppression of the spin excitations. Full suppression of this scattering channel, attained when $H$ reaches $H_s$, leads to the unusually large $\ell$ in the region II.

Magnetoresistance and current ratio $R_{\sigma}$. – We have also found that the strong enhancement of $\ell$ forges a link between the unusual MR and Hall effect. The observed $\sigma_{xy}$ is the sum of contributions $\sigma_{xy}^i$ from each FS sheet $i$. Assuming that the lifetime $\tau_i$ on each sheet is dominated by spin-disorder scattering, all $\tau_i$ follow the same monotonically rising function of field $g(B)$. As a result, we have $\sigma_{xy} \sim B g(B)^2$, while the conductivity $\sigma \sim g(B)$: The Hall current grows in direct proportion to the square of the longitudinal current multiplied by $B$.

To test this assumption, we compare the curves of $\sigma_{xy}$ with $\sigma^2 B$ at low $T$ (fig. 5b). In high fields, the quantity $\sigma^2 B Z$ (thin curves) can be made to match $\sigma_{xy}$ (bold curves) by setting the $T$-independent constant $Z = 1 \times 10^{-9} \text{cm}^2/\text{C}$. The match is excellent for fields above a cross-over field $H_Z(T)$. As $H$ decreases below $H_Z$, however, $\sigma_{xy}$ increasingly exceeds $\sigma^2 B Z$. The negative curvature (knee) feature described earlier now appears as a small “excess” Hall current below $H_Z$.

This high-field scaling is made more apparent if we plot the quantity

$$R_{\sigma}(T, B) = \frac{\sigma_{xy}}{\sigma^2 B},$$

which measures the ratio of the Hall current and the longitudinal current squared (see fig. 6). We note that $R_{\sigma} = R_H[1 + (\tan \theta_H)^2]$ deviates from the ordinary Hall coefficient $R_H$ when the Hall angle $\theta_H$ is large. Remarkably, fig. 6 shows that, below 1K, $R_{\sigma}$ is just a constant equal to $Z$, even though both $\sigma$ and $\sigma_{xy}$ are increasing with strong curvature. Above 1K, $R_{\sigma}$ deviates significantly from $Z$ as the excess Hall current grows, but only for fields $H < H_Z$. Above $H_Z$ (arrow), we see that $R_{\sigma}$ again settles down to the value $Z$. The constancy of $R_{\sigma}$ is direct evidence that both the anomalous MR and $\sigma_{xy}$ reflect the enhancement in $\ell$. As seen in the phase diagram fig. 1a, $H_Z$ (open circles) lies significantly below $H_s$, the simple Hall response determines the high-field Hall behavior in the regions I and II. Hence the complicated Hall response in CeCoIn$_5$ arises solely from the excess Hall current which is responsible for the weak-field “knee”, but is suppressed above $H_Z$.

Quasiparticles in superconducting state. – These findings have interesting implications for the superconducting state. In heavy-electron systems, pairing mediated by the exchange of spin fluctuations has been proposed as the likely mechanism for the SC state. However, the evidence to date for spin exchange has been indirect. Here, we have exploited the unusually large strong $H$ dependence of the tensors $\kappa_{ij}$ and $\sigma_{ij}$ to show that spin excitations play a dominant role. The high-field scaling is made more apparent if we plot the quantity

$$R_{\sigma}(T, B) = \frac{\sigma_{xy}}{\sigma^2 B},$$

(3)

which measures the ratio of the Hall current and the longitudinal current squared (see fig. 6). We note that $R_{\sigma} = R_H[1 + (\tan \theta_H)^2]$ deviates from the ordinary Hall coefficient $R_H$ when the Hall angle $\theta_H$ is large. Remarkably, fig. 6 shows that, below 1K, $R_{\sigma}$ is just a constant equal to $Z$, even though both $\sigma$ and $\sigma_{xy}$ are increasing with strong curvature. Above 1K, $R_{\sigma}$ deviates significantly from $Z$ as the excess Hall current grows, but only for fields $H < H_Z$. Above $H_Z$ (arrow), we see that $R_{\sigma}$ again settles down to the value $Z$. The constancy of $R_{\sigma}$ is direct evidence that both the anomalous MR and $\sigma_{xy}$ reflect the enhancement in $\ell$. As seen in the phase diagram fig. 1a, $H_Z$ (open circles) lies significantly below $H_s$. The steep variation in weak $H$ reflects the excess Hall current discussed in fig. 5b.

Finally, we comment on the extraordinary peak in $\kappa$ vs. $H$ that appears below $T_c$ (fig. 7). The sharp reduction of the peak amplitude in $H$ is very similar to the $\kappa$ vs. $H$ curves below $T_c$ in unwinned YBa$_2$Cu$_3$O$_7$ (YBCO) [13]. The extreme sensitivity to $H$ is interpreted as caused by scattering of nodal qp by vortices [12–14]. In YBCO, the observation of a large anomaly in $\kappa_{xy}$ at peaks at finite field provided early key evidence that the peak in $\kappa$ arises from enhancement of $\ell$ of nodal excitations in a $d$-wave superconductor. Similarly, the low-field peak reported here in $\kappa_{xy}$ at 0.5 and 0.75 K (fig. 3a) confirms that the cusp-anomaly in $\kappa_{xy}$ is electronic in origin. The steep increase in $\ell$ below $T_c$ implies that the nodal qp does not experience the intense scattering from spin excitations.
The close similarity between CeCoIn$_5$ and YBCO suggests that a steep enhancement of $\ell$ below $T_c$ associated with nodal quasiparticles may be generic to electronic-mediated pairing with $d$-wave symmetry.

**Discussion.** We have exploited the unusually large Righi-Leduc effect in CeCoIn$_5$ to determine the Wiedemann-Franz ratio of its charge carriers at low $T$. As shown in fig. 3, the 2 Hall conductivities $\kappa_{xy}$ and $\sigma_{xy}$ are strongly non-linear, with a change-in-sign of the curvature occurring as $H$ increases from 0 to 12 T. Remarkably, over a broad interval of $T$, the 2 quantities may be scaled together using an $H$-independent Lorenz parameter $L(H)$. We find that $L(H)$ is weakly $T$ dependent and close to the Sommerfeld value ($\pi^2/3)(k_B/e)^2$. The strict insensitivity of $L(H)$ to field allows the electronic heat conductivity $\kappa_e$ to be determined unambiguously. On subtracting $\kappa_e$ from the observed $\kappa_{xx}$, we uncover a large background charge-neutral term $\kappa_b$ that is field sensitive. As $H$ increases, $\kappa_b$ falls while $\sigma$ rises in proportion. This implies that spin excitations are the dominant scatterers of the electrons.

The transport picture that emerges is that, throughout region I in zero $H$ (fig. 1a), the electrons are strongly scattered by spin excitations. Moreover, the spin excitations contribute the dominant share of the charge neutral thermal conductivity $\kappa_b$, which accounts for $\sim 45\%$ of the observed $\kappa_{xx}$ at $T_c$. In a finite $H|\mathbf{c}$, the density of spin excitations is strongly suppressed. This leads to 2 correlated effects. The neutral heat term $\kappa_b$ is suppressed, while the 3 electronic currents $\sigma$, $\sigma_{xy}$ and $\kappa_e$ grow in proportion, as a result of strong enhancement of $\ell$. The trend in $\kappa_b$ suggests that the full suppression of spin scattering is attained when $H \rightarrow H_s$. An interesting scaling relationship between $\sigma^2$ and $\sigma_{xy}$ is found. (Two recent findings related to this work are refs. [22,23].

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Fig. 7: Expanded view of $\kappa_{xx} \equiv \kappa$ vs. $H$ in the vortex state of CeCoIn$_5$ ($T < T_c$). The sharp peaks in weak $H$ arise from the field suppression of the broad peak in $\kappa(0,T)$ below $T_c$ (see fig. 1b).