Non-Fermi Liquid behavior in CeIrIn$_5$ near a metamagnetic transition.

C. Capan, A. Bianchi, F. Ronning, A. Lacerda, J. D. Thompson, P. G. Pagliuso, J. L. Sarrao, and R. Movshovich

1 Los Alamos National Laboratory, Los Alamos, New Mexico 87545
2 National High Magnetic Field Laboratory, Los Alamos, New Mexico 87545
3 Instituto de Física Gleb Wataghin, UNICAMP, 13083-970, Campinas, Brazil

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We present specific heat and resistivity study of CeIrIn$_5$ in magnetic fields up to 17 T and temperature down to 50 mK. Both quantities were measured with the magnetic field parallel to the c-axis ($H \parallel [001]$) and within the a-b plane ($H \perp [001]$). Non-Fermi-liquid (NFL) behavior develops above 12 T for $H \parallel [001]$. The Fermi liquid state is much more robust for $H \perp [001]$ and is suppressed only moderately at the highest applied field. Based on the observed trends and the proximity to a metamagnetic phase transition, which exists at fields above 25 T for $H \parallel [001]$, we suggest that the observed NFL behavior in CeIrIn$_5$ is a consequence of a metamagnetic quantum critical point.

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Investigations of the material properties near a zero-temperature phase transition (Quantum Critical Point or QCP) is at present a very active area of research, attracting both experimental and theoretical attention. It is common for metallic compounds in the vicinity of a QCP to display a variety of physical properties at odds with those expected for a Fermi Liquid (FL), a concept that forms the basis for our understanding of the physics of a vast majority of metals. Characteristic of a FL are such properties as a linear-in-temperature specific heat $C$ and T-squared resistivity $\rho$. In contrast, materials near QCPs often display a diverging Sommerfeld coefficient $\gamma = C/T$ and a power law temperature dependent resistivity $\rho = \rho_0 + AT^\alpha$, with $\alpha$ significantly different from 2 [1]. The theoretical picture of a system near a QCP is not complete at the moment, and the origins of the behavior described above are the subject of intense theoretical investigations.

For a large number of the materials studied, the two competing phases at the QCPs are antiferromagnetically (AF) ordered and paramagnetic ones. It was demonstrated that for this class of compounds pressure was an effective parameter to tune a system through an AF QCP, particularly for the Ce-based heavy-fermion compounds. This group of materials include, for example, CeCu$_2$Ge$_2$, CeRh$_2$Si$_2$, CePd$_2$Si$_2$, and CeIn$_3$ [2]. Alternatively, in an increasing number of materials, i.e. CeCu$_5$Ag$_0.1$, CeCoIn$_5$ [3, 4], and YbRh$_2$Si$_2$ [5, 6], it was found to be possible to tune the system through a QCP by varying the magnetic field.

A novel route to quantum criticality was proposed based on Sr$_3$Ru$_2$O$_7$, when its resistivity in magnetic field close to the metamagnetic field $H_M = 8$ T displayed pronounced NFL behavior [10, 11]. On general grounds a first order $T = 0$ phase transition does not lead to a QCP. Nevertheless, it was suggested that quantum critical behavior can be associated with a first order phase transition when the transition’s critical end point is driven to zero temperature [10, 12]. In Sr$_3$Ru$_2$O$_7$ the critical end point of the metamagnetic phase transition temperature $T_{Xc}$ can be tuned by varying the direction of the magnetic field with respect to the tetragonal crystal lattice. When magnetic field is close to $H \parallel [001]$, $T_{Xc}$ is suppressed close to zero, leading to the quantum critical behavior observed in Sr$_3$Ru$_2$O$_7$ [10]. Recent analysis indicate that CeRu$_2$Si$_2$ may also be close to a metamagnetic QCP [13].

Related phenomena may be in play in CeIrIn$_5$. For most of the compounds with AF QCPs magnetic field suppresses the AF state, with the Fermi Liquid behavior recovered in the paramagnetic state above the critical field of the QCP. In this Letter we present results of the specific heat and resistivity measurements in CeIrIn$_5$ which show the reverse trend, with magnetic field suppressing rather than enhancing the Fermi Liquid state, pointing perhaps to a different route to Quantum Criticality. Based on our results we suggest that the NFL behavior in CeIrIn$_5$ is due to the proximity to a magnetic phase transition and a metamagnetic QCP, perhaps similar to the recently discussed case of Sr$_3$Ru$_2$O$_7$ [10].

The details of sample growth and characterization are described in Ref. [14]. Large plate-like single crystals, up to 1 cm long, are grown from an excess In flux. CeIrIn$_5$ is a layered tetragonal heavy fermion compound from the 1-1-5 family, with no long-range magnetic order but a superconducting ground state [14]. The presence of the cylindrical Fermi surface sheet, inferred from de Haas-van Alphen studies [15], and a ratio of 4.8 of the effective masses between the c-axis and the CeIn$_3$ planes [16] makes CeIrIn$_5$ a moderately anisotropic system. Moreover, the crystal electric field effects result in an anisotropic susceptibility, with a step-like feature around 50 K along the c-axis [17]. The anisotropy of the spin fluctuations is evidenced in the temperature dependence of $T_1$ derived from the NQR data [18]. This anisotropy is reflected in both the specific heat and resistivity data shown below.

We measured the specific heat of a CeIrIn$_5$ single crystal with the quasi-adiabatic heat pulse method in a dilu-
magnetic fields up to 17 T. Fig. 1 shows the specific heat as a function of temperature in the normal state, for magnetic fields up to 17 T applied parallel and perpendicular to the c-axis. At low temperature (below 200 mK) the specific heat is dominated by the nuclear Schottky anomaly, which is mainly due to In nuclear levels split by the magnetic field. The Schottky anomaly can be well approximated with an $\alpha / T^2$ dependence in the whole field range, with $\alpha \propto H^2$ for both field orientations, as expected. The 17 T field induces a significant shift between the in-plane and out-of-plane specific heat, for temperatures ranging from 0.2 K to 3 K.

Figure 2 shows the electronic specific heat as a function of temperature on a semi-log scale, for magnetic fields oriented (a) in-plane and (b) out-of-plane, with field values ranging between 1 T and 17 T. The electronic specific heat is obtained after subtracting the lattice and the nuclear Schottky contribution from the measured specific heat. The lattice contribution is small in the temperature range of interest (only 2.8% of the total specific heat at 3K) and has been calculated from the LaIrIn$_5$ specific heat in the Debye approximation.

The Sommerfeld coefficient $\gamma = C/T$ rises as the temperature is reduced below 3 K, consistent with earlier reports on non-Fermi liquid behavior in this compound, both of specific heat and thermal expansion, and reaches a plateau below about 1 K in low magnetic fields. The saturation of $\gamma$ marks the onset of the Fermi liquid regime below the temperature $T_{FL}$ for both field orientations. However, $\gamma$ has a remarkably different evolution for the two field orientations studied when the magnetic field is increased. Namely, the field in-plane does not have a strong effect on the overall shape of $\gamma$, and just slightly suppresses $T_{FL}$ to lower temperatures and makes the slope above 1 K steeper. In contrast, for $H \parallel c$ the knee in $\gamma$ gradually disappears, and the overall slope becomes more flat with increasing field, leading to the gap between the bare specific heat curves described earlier (see Fig. 1). Note that the difference in the slopes above 1 K cannot be due to the error in subtraction of the Schottky contribution, which drops to $\approx 50\%$ of the total specific heat at 300 mK for 17 T.

The difference in the evolution of the specific heat with the field in different orientations is emphasized in the inset of Fig. 1 where we plot the difference between the in-plane and out-of-plane specific heat divided by temperature, $(C_{H \perp c} - C_{H \parallel c})/T$. A broad maximum is resolved above the field of 3 T, reflecting the suppression of the specific heat in the $H \parallel [001]$ orientation. This maximum increases in magnitude and shifts to lower temperatures as the field is increased, reaching 0.23 J/molK$^2$ at 17 T around 0.6 K, or about 27% of the total specific heat.

There is a clear anisotropy in the evolution of $T_{FL}$ as well. At 1 T the Fermi liquid behavior survives up to $\sim 0.9$ K independent of the field orientation. However, as the magnetic field is increased, the Fermi temperature is depressed much faster when the field is along the c-axis, in contrast to a very gradual decrease observed when the field is in-plane. The knee eventually vanishes completely and $\gamma$ becomes divergent down to the lowest temperatures measured for fields above 12 T with $H \parallel c$. This NFL behavior suggests that CeIrIn$_5$ may be approaching a QCP for fields $H \parallel c$ above 12 T.

Figure 3 shows resistivity data for magnetic fields of 12 T, 15 T, and 17 T applied both within and out of the a-b plane of CeIrIn$_5$. Fermi liquid $\rho = \rho_0 + AT^2$ behavior
is clearly obeyed by the resistivity for all fields \( H \perp [001] \) (Fig. 3(a)) below a well defined temperature, marked by the arrow for 17 T data as an example. This behavior of the resistivity is consistent with the FL behavior displayed at low temperature by the specific heat for \( H \perp [001] \) (see Fig. 2(a)). To analyze the data further we plot coefficient \( A \) of the \( T^2 \) term in resistivity versus square of the Sommerfeld coefficient of \( \gamma \propto C/T \) for \( H \perp [001] \), testing the Kadowaki-Woods relation (see the text). (b): \( H \perp [001] \) orientation. Inset: resistivity for \( H \parallel [001] \) plotted vs. \( T^o \) for best fit of the lowest temperature data. \( \alpha \neq 2 \) for these fields.

Fig. 3: Resistivity of CeIrIn\(_5\) at (○) 12 T, (⋆) 15 T, and (□) 17 T. (a): \( H \parallel [001] \) orientation. Arrow indicate the onset of the deviation of the 17 T data from the Fermi Liquid \( \rho \propto T^2 \) behavior (straight line) at \( T_{FL} \). Inset: coefficient \( A \) of the \( T^2 \) term in resistivity versus square of the Sommerfeld coefficient of \( \gamma \propto C/T \) for \( H \perp [001] \), testing the Kadowaki-Woods relation (see the text).

The NFL behavior in CeIrIn\(_5\) is qualitatively different from that commonly observed in many heavy fermion compounds in the vicinity of the QCP, where application of the magnetic field suppresses the magnetically ordered state and stabilizes the FL behavior for \( H > H_{QCP} \). In contrast, in CeIrIn\(_5\) increasing magnetic field suppresses the FL state. Magnetic field in the range investigated here, therefore, drives the system closer to a QCP. Such behavior is likely related to the properties discovered in the very high field investigations of CeIrIn\(_5\). Magnetization studies of CeIrIn\(_5\) revealed a metamagnetic anomaly at a field of 42 T [22] at 1.3 K. Subsequent specific heat measurements of CeIrIn\(_5\) uncovered a phase transition into a magnetic state between 35 T at 1.8 K and 45 T at 4.2 K, extrapolating to \( H_M \approx 26 \) T at zero temperature [19]. More recent data from cantelever magnetometry investigation of CeIrIn\(_5\) show an anomaly in magnetic response at 30 T at 45 mK [23]. Despite the quantitative differences, these measurements are most likely studying the same phenomenon of metamagnetism in CeIrIn\(_5\).

It is instructive to compare behavior of CeIrIn\(_5\) with that of Sr\(_3\)Ru\(_2\)O\(_7\), a material whose NFL behavior close to a metamagnetic phase transition have recently received both experimental and theoretical attention [10, 12]. The Sommerfeld coefficient \( \gamma \) of Sr\(_3\)Ru\(_2\)O\(_7\) diverges in magnetic field close to the metamagnetic field \( H_M = 8T \) for \( H \parallel [001] \), close to the orientation when the first order critical end point is suppressed to \( T = 0 \) [24].
It is interesting to note that even when the critical end point temperature $T^*_M$ is finite (i.e. $H \perp [001]$, where $H_M \approx 6$ T) the phase diagram showing NFL behavior, based on the resistivity data, is remarkably similar to the $H \parallel [001]$ case with $T^*_M = 0$ [11]. In addition, $\gamma$ in Sr$_3$Ru$_2$O$_7$ is close to being logarithmically divergent with temperature for $H \perp [001]$ at 6 T as well [22]. Finally, we note that MnSi provides evidence that a first order phase transition does not preclude NFL behavior [26].

The above comparison leads us to suggest that the NFL behavior in CeIrIn$_5$ is due to the proximity to a metamagnetic phase transition. Figure 4 shows the magnetic field - temperature phase diagram. The open and solid circles represent the Fermi Liquid temperature $T_{FL}$ obtained from the analysis of the specific heat data for $H \perp c$ and $H \parallel c$, respectively. The data for $H \parallel c$ extrapolates to a value close to 25 T. Stars, representing the metamagnetic transition temperature $T_M$ from Ref. [19], extrapolate to about 26 T. The two values are very close and hint at the possibility that the NFL behavior in CeIrIn$_5$ for $H \parallel c$ between 12 and 17 T may be related to the metamagnetic phase transition observed at higher fields. To test this hypothesis further we compared the entropy associated with the metamagnetic transition at 45 T with the entropy of CeIrIn$_5$ at 17 T. We estimated the specific heat of CeCoIn$_5$ at 42 T and 45 T below 1.2 K by linearly extrapolating $\gamma$ from Ref. [10] from 1.2 K to $T = 0$. The extrapolated curves were integrated and the resulting entropy curves are displayed in the inset of Fig. 4. The entropy values are very close at high temperature above the metamagnetic anomaly in specific heat. This indicates that the metamagnetic anomaly is built out of the spin fluctuations that lead to the NFL behavior of specific heat of CeIrIn$_5$ at 17 T, and presents a strong argument in favor of the metamagnetic quantum critical point in CeIrIn$_5$ being the origin of the NFL properties of CeIrIn$_5$ we observed.

For $H \perp c$, the metamagnetic transition does not occur below 52 T [22]. This is reflected in our data, with FL behavior being much more robust for this orientation. In fact, from the field dependence of $T_{FL}$ for $H \perp c$ displayed in Fig. 4 we can roughly estimate the metamagnetic transition field $H_M$ to be between 70 T and 90 T for $H \perp c$.

In conclusion, specific heat and resistivity measurements of CeIrIn$_5$ in magnetic field up to 17 T revealed NFL behavior in both of these properties for the field out of the plane ($H \parallel c$, easy axis) orientation. This behavior develops above 12 T. Fermi liquid is robust for $H \perp c$ in this field range. On the basis of the phase diagram and the entropy analysis we suggest that the NFL behavior in CeIrIn$_5$ is due to the metamagnetic QCP point.

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