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

C. Capan,$^1$ A. Bianchi,$^1$ F. Ronning,$^1$ A. Lacerda,$^2$ J. D. Thompson,$^1$ M. F. Hundley,$^1$ P. G. Pagliuso,$^3$ J. L. Sarrao,$^1$ and R. Movshovich$^1$

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

(Received 13 July 2004; published 22 November 2004)

We present a 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 ($H||[001]$) and within the $a$-$b$ plane ($H \perp [001]$). Non-Fermi-liquid (NFL) behavior develops above 12 T for $H||[001]$. The Fermi-liquid state is much more robust for $H||[001]$ 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 $H||[001]$, we suggest that the observed NFL behavior in CeIrIn$_5$ is a consequence of a metamagnetic quantum critical point.

DOI: 10.1103/PhysRevB.70.180502

PACS number(s): 74.70.Tx, 71.27.+a, 74.25.Fy, 75.40.Cx

Investigations of the material properties near a zero-temperature phase transition [quantum critical point (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 material 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 driving the tetragonal crystal lattice. When the magnetic field is close to the metamagnetic phase transition temperature $T_M$, 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.11,12

In Sr$_2$Ru$_2$O$_7$, the critical end point of the metamagnetic phase transition temperature $T'_M$ can be tuned by varying the direction of the magnetic field with respect to the tetragonal crystal lattice. When the magnetic field is close to $H||[001]$, $T'_M$ is suppressed close to zero, leading to the quantum critical behavior observed in Sr$_3$Ru$_2$O$_7$.11 Recent analysis indicates that CeRu$_2$Si$_2$ may also be close to a metamagnetic QCP.

Related phenomena may be in play in CeIrIn$_5$. For most of the compounds with AF QCPs the 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 Rapid Communication, we present results of the specific heat and resistivity measurements in CeIrIn$_5$ which show the reverse trend, with the 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 metamagnetic phase transition and a metamagnetic QCP, perhaps similar to the recently discussed case of Sr$_3$Ru$_2$O$_7$.11

The details of sample growth and characterization are described in Ref. 14. Large platelike 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 makes CeIrIn$_5$ a moderately anisotropic system. Moreover, the crystal electric field effects result in an anisotropic susceptibility, with a steplike 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 quasiadiabatic heat pulse method in a dilution refrigerator between 100 mK and 3 K and in magnetic fields up to 17 T. Figure 1 shows the specific heat as a function of temperature in the normal state, for the magnetic field of 17 T applied perpendicular and parallel to the $c$ axis. At low temperature (below 200 mK) the specific heat is dominated by $T^4$ as expected for the Fermi-liquid state. As the field is increased, the specific heat develops a power-law divergence, characteristic of a FL.

The field dependence of the specific heat in the vicinity of the QCP is shown in Fig. 2. The temperature for $C(T=0)=0$, i.e., $T_M$, is suppressed close to zero, leading to the quantum critical behavior observed in Sr$_3$Ru$_2$O$_7$.11 Recent analysis indicates that CeRu$_2$Si$_2$ may also be close to a metamagnetic QCP.13

Recently, we suggested that CeIrIn$_5$ is a Fermi liquid near the quantum critical point, with the Fermi-liquid behavior recovered in the paramagnetic state above the critical field of the QCP. In this Rapid Communication, we present results of the specific heat and resistivity measurements in CeIrIn$_5$ which show the reverse trend, with the 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 metamagnetic phase transition and a metamagnetic QCP, perhaps similar to the recently discussed case of Sr$_3$Ru$_2$O$_7$.11

The details of sample growth and characterization are described in Ref. 14. Large platelike 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 makes CeIrIn$_5$ a moderately anisotropic system. Moreover, the crystal electric field effects result in an anisotropic susceptibility, with a steplike 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 quasiadiabatic heat pulse method in a dilution refrigerator between 100 mK and 3 K and in magnetic fields up to 17 T. Figure 1 shows the specific heat as a function of temperature in the normal state, for the magnetic field of 17 T applied perpendicular and parallel to the $c$ axis. At low temperature (below 200 mK) the specific heat is dominated by $T^4$ as expected for the Fermi-liquid state. As the field is increased, the specific heat develops a power-law divergence, characteristic of a FL.
FIG. 1. Specific heat as a function of temperature at 17 T for field oriented (●) parallel to the c axis and (○) in the plane. Inset: Anisotropy of specific heat (difference of in-plane and out-of-plane specific heat) as a function of temperature at various magnetic fields up to 17 T: (□) 1 T, (●) 3 T, (△) 6 T, (×) 9 T, (○) 12 T, and (□) 17 T.

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 $a/H^2$ dependence in the whole field range, with $a\propto H^2$ 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 to 3 K.

Figure 2 shows the electronic specific heat as a function of temperature on a semilogarithmic scale, for magnetic fields ranging between 1 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 3 K) 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 NFL 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 $H||c$ 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, $(C_{H\parallel c} - C_{H\perp c})/T$. A broad maximum is resolved above the field of 3 T, reflecting the suppression of the specific heat in the $H||[001]$ orientation. This maximum increases in magnitude and shifts to lower temperatures as the field is increased, reaching 0.23 J/mol K$^2$ 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 $H||c$. This NFL behavior suggests that CeIrIn$_5$ may be approaching a QCP for fields $H|c$ above 12 T. Using the expression $C/T = -0.25R/T_0 \ln(T/0.41T_0)$ from Ref. 1, we obtain the Kondo temperature for CeIrIn$_5$ of 10.6 and 26.4 K for 1 and 17 T, respectively. This difference of the characteristic energy scales is responsible for the apparent decrease of the specific heat between 1 and 17 T in the temperature range below roughly 3 K. We emphasize here that in spite this decrease, the specific heat for fields above 12 T does not saturate to the lowest temperatures measured.

Figure 3 shows resitivity data for magnetic fields of 12, 15, 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 $\gamma^2$ in the inset of Fig. 3(a). For many heavy fermion compounds $A$ vs $\gamma^2$ points fall close to a single straight line.\textsuperscript{21} Similar behavior is observed for CeIrIn$_5$, with $A \approx \gamma^2$, as is seen in the inset. The coefficient of proportionality (Kadowaki-Woods ratio) is about a factor of 5 smaller than the average value for other heavy fermions. This, however, is within the scatter displayed by this class of materials. Therefore, both resistivity and specific heat of CeIrIn$_5$ have a well-developed heavy-fermion FL ground state for $H \perp c$ below 17 T.

Figure 3(b) shows resistivity data for $H \parallel [001]$, which we plot versus $T^4$. For all three fields the data fall on straight lines below $\approx 0.5$ K. Such NFL behavior of resistivity with the power law exponent different from two was observed in other compounds near a QCP? The NFL resistivity behavior is consistent with the NFL behavior displayed by the specific heat of CeIrIn$_5$ for $H \parallel [001]$ [see Fig. 2(b)]. Together, resistivity and specific heat data point to an approaching QCP with magnetic field $H\parallel[001]$ increasing above 12 T.

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 (Ref. 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.\textsuperscript{23} More recent data from can-telever magnetometry investigation of CeIrIn$_5$ show an anomaly in magnetic response at 30 T at 45 mK.\textsuperscript{24} Despite

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 FL temperature $T_{FL}$ derived from the electronic specific heat for $H \parallel [001]$ and $H \perp [001]$, respectively. The data for $H \parallel [001]$ extrapolates to a value close to 25 T. Stars, representing the magnetic transition temperature $T_M$ from Ref. 23, 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 CeIrIn$_5$ at 42 and 45 T below 1.2 K by linearly extrapolating $\gamma$ from Ref. 23 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 (Ref. 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 and 90 T for $H \perp c$.

In conclusion, specific heat and resistivity measurements of CeIrIn$_5$ in magnetic fields up to 17 T revealed NFL behavior in both of these properties for the field out of the plane ($H||c$, 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.

We thank R. G. Goodrich, C. Varma, and J. Lawrence for stimulating discussions. Work at Los Alamos National Laboratory was performed under the auspices of the U.S. Department of Energy. F.R. thanks Reines Postdoctoral Fellowship (DOE-LANL) for support. Work at the NHMFL was performed under the auspices of the National Science Foundation, the State of Florida, and the U.S. Department of Energy.