Superconductivity and Quantum Criticality in CeCoIn$_5$

V. A. Sidorov, M. Nicklas, P. G. Pagliuso, J. L. Sarrao, Y. Bang, A. V. Balatsky, and J. D. Thompson
Los Alamos National Laboratory, Los Alamos, NM 87545
(Dated: November 10, 2021)

Electrical resistivity measurements on a single crystal of the heavy-fermion superconductor CeCoIn$_5$ at pressures to 4.2 GPa reveal a strong crossover in transport properties near $P^* \approx 1.6$ GPa, where $T_c$ is a maximum. The temperature-pressure phase diagram constructed from these data provides a natural connection to cuprate physics, including the possible existence of a pseudogap.

PACS numbers: 74.70.Tx, 74.62.Fj, 75.30.Mb, 75.40.-s

The relationship between unconventional superconductivity and quantum criticality is emerging as an important issue in strongly correlated materials, including cuprates, organics and heavy-fermion intermetallics. In each of these, either Nature or an externally imposed parameter, such as pressure or chemical substitutions, tunes some magnetic order to a $T = 0$ transition where quantum fluctuations introduce new excitations that control thermodynamic and transport properties over a broad range of temperature-parameter phase space. A frequent, but controversial assumption is that these same excitations may mediate Cooper pairing in the anisotropic superconductivity that appears in the vicinity of a quantum-critical point (QCP). Indeed, the effective potential generated by proximity to a quantum-critical spin-density wave can also lead to anisotropic superconductivity.

Though the issue of quantum criticality and superconductivity came to prominence in the context of the cuprates (see, eg), there have been several recent examples of this interplay in heavy-fermion compounds. Like the cuprates, unconventional superconductivity in these Ce-based heavy-fermion materials develops out of a distinctly non-Fermi-liquid normal state that evolves in proximity to a continuous $T = 0$ antiferromagnetic transition; but unlike the cuprates, this state can be accessed cleanly by applied hydrostatic pressure and without introducing extrinsic disorder associated with chemical substitutions. In addition to chemical inhomogeneity in the cuprates, a further ‘complication’ is the existence of a pseudogap state above $T_c$ for dopings less than optimal. Whether evolution of the pseudogap with doping produces another QCP near the optimal $T_c$ in the cuprates is an open question as is the origin of the pseudogap. Thus far, a pseudogap has not been detected in canonical quantum-critical heavy-fermion systems, such as CeIn$_3$ or CePd$_2$Si$_2$ under pressure, but this may not be too surprising. In these cases, superconductivity appears only at very low temperatures, less than 0.4 K, and over a narrow window of pressures ($\leq 0.8$ GPa) centered around the pressure where $T_N$ extrapolates to zero. In analogy to the cuprates, we would expect a pseudogap in these Ce compounds to exist only over a narrow temperature window, 0.1 – 0.2 K above $T_c$, and in a fraction of the narrow pressure window, which certainly would make detection of a pseudogap difficult.

CeCoIn$_5$ offers the possibility of making a connection between other Ce-based heavy-fermion materials and the cuprates. Bulk, unconventional superconductivity is present in CeCoIn$_5$ at atmospheric pressure and develops out of a heavy-fermion normal state in which the resistivity is approximately linear in temperature. Application of a magnetic field sufficient to quench superconductivity reveals a specific heat diverging as $-T \ln T$ and associated low-temperature entropy consistent with its huge zero-field specific heat jump at $T_c$ ($\Delta C/\gamma T_c = 4.5$). Together, these properties suggest that CeCoIn$_5$ may be near an antiferromagnetic quantum-critical point at $P = 0$. Its isostructural, antiferromagnetic relative CeRhIn$_5$ becomes a pressure-induced unconventional superconductor near 1.6 GPa, with a $T_c = 2.1$ K close to that of CeCoIn$_5$ at $P = 0$. This and other similarities between CeCoIn$_5$ at $P = 0$ and CeRhIn$_5$ at $P = 1.6$ GPa reinforce speculation that the nearby antiferromagnetic QCP in CeCoIn$_5$ may be at an inaccessible slightly negative pressure. The 1.7% smaller cell volume of CeCoIn$_5$ compared to that of CeRhIn$_5$ is consistent with this view. We have studied the effect of pressure on the electrical resistivity and superconductivity of CeCoIn$_5$ and find striking correlations between them that are reminiscent of cuprate behaviors.

Four-probe AC resistivity measurements, with current flowing in the tetragonal basal plane, were made on a single crystal of CeCoIn$_5$ grown from excess In flux. Hydrostatic pressures to 5 GPa were generated in a toroidal anvil cell in which a boron-epoxy gasket surrounds a teflon capsule filled with a glycerol-water mixture (3:2 volume ratio) that served as the pressure transmitting fluid. Pressure inside the capsule was determined at room temperature and at low temperatures from the pressure-dependent electrical resistivity and $T_c$ of Pb, respectively. The width of superconducting transition of Pb did not exceed 15 mK, indicating good hydrostatic conditions and providing an estimate of the pressure-measurement uncertainty, $\pm 0.04$ GPa.

The response of $\rho(T)$ to pressure over a broad temperature scale is typical of many Ce-based heavy-
fermion compounds and reflects a systematic increase in the energy scale of spin fluctuations. Most interesting is the low-temperature response (Fig. 1) where \( T_c \) and the resistivity just above \( T_c \) \( (\rho(T_c^+)) \) decrease rapidly with pressures \( P > 1.35 \) GPa. We have analyzed the low-temperature resistivity, plotted over a broader temperature range chosen for fitting. The more sensitive and revealing approach is the latter in which we adjust \( \rho_0 \) and \( n \) fitting parameters, or from plots of \( (\rho(T) - \rho_0)/T^n \). A drawback to the former approach is that parameters can be sensitive to the temperature range chosen for fitting. The more sensitive and revealing approach is the latter in which \( \rho_0 \) and \( n \) are adjusted to give the best horizontal line. This procedure requires one less parameter, \( A \), which is provided directly by the magnitude of \( (\rho(T) - \rho_0)/T^n \). Representative examples of this approach are plotted in Fig. 2. Parameters obtained from these plots are well defined and agree, typically to 10% or better, with parameter values extracted from straightforward fits. As indicated by arrows on the lowest pressure curve in Fig. 2, \( (\rho(T) - \rho_0)/T^n \) deviates from the horizontal trend at a temperature \( T_{pg} \) well above the onset of superconductivity. This departure implies either a decrease in the scattering rate or increase in carrier density. Resistivity measurements at \( P = 0 \) and in a magnetic field (6 T) greater than \( H_{c2}(0) \) find \( a \) similar departure of \( (\rho(T) - \rho_0)/T^n \) from a \( T \)-independent curve at \( T_{pg} = 3 \) K. Thus, \( T_{pg} \) does not originate from superconducting fluctuations or from trace amounts of free indium. We also note that this behavior was present in previous measurements on other single crystals CeCoIn\(_5\) but was missed in inspections of \( \rho \) versus \( T \) curves \cite{13}. We will return to a discussion of \( T_{pg} \) later.

Values of \( \rho_0 \), \( n \) and \( A \) obtained from these plots are summarized in Figs. 3a and b. There is an unmistakable crossover in the pressure dependence of \( \rho_0 \) and \( n \) at \( P^* \approx 1.6 \) GPa. Below \( P^* \), \( n = 1.0 \pm 0.1 \) for \( T_{pg} \leq T \leq 10 \) K. This value of \( n \) is expected for a 2-dimensional, antiferromagnetic quantum-critical system \cite{21} and is not too surprising given the layered crystal structure \cite{22} and anisotropy in electronic states of CeCoIn\(_5\) \cite{10, 23}. At pressures greater than \( P^* \), \( n \) rapidly approaches the Fermi-liquid value of 2.0, which holds from just above \( T_c \) to \( T_{FL} \) (see Fig. 2). Though not shown in either Fig. 3 or 4, at these higher pressures \( n \) assumes a value of \( 1.3 \pm 0.1 \), characteristic of a 3-D antiferromagnetic QCP, in a temperature interval from slightly greater than \( T_{FL} \) to as high as \( \sim 60 \) K. For \( P \geq P^* \), \( \rho_0 \) decreases reversibly by an order of magnitude to a very small value of about \( 0.2 \mu\Omega \text{cm} \). Clearly, pressure does not remove impurities from the sample; the large decrease in \( \rho_0 \) must be due to a pressure-induced change in inelastic scattering processes. Theories of electronic transport in quantum-critical systems show \cite{24, 23} that impurity scattering can be strongly enhanced by quantum-critical fluctuations. This sensitivity provides a natural explanation for a decrease in scattering with increasing pressure, if there is a crossover near \( P^* \) from quantum-critical \( (P < P^*) \) to a Fermi-liquid-like state for \( (P > P^*) \). Such a crossover is suggested by the pressure variation of \( n \).

Experimental values of the \( T^n \)-coefficient of resistivity, plotted in Fig. 3a, also decrease strongly with increasing pressure for \( P \lesssim P^* \). For comparison, we show
the variation of the electronic specific heat divided by temperature \( (C/T \equiv \gamma) \) at 3 K for pressures less than 1.5 GPa \(^{26}\) and, at higher pressures, we plot values of \( \gamma \) inferred from the \( T^2 \)-coefficient of resistivity, followed by several heavy-fermion compounds \(^{23}\). Though a \( T^2 \) coefficient proportional to \( \gamma^2 \) is expected for a Landau Fermi-liquid \(^{3}\), it is not obvious \( a \ priori \) why \( A(P) \) approximately tracks the directly measured \( \gamma \) at low pressures, but this seems to be the case. In any event, both \( A(P) \) and \( \gamma(P) \) exhibit qualitatively different magnitudes and functional dependencies above and below \( P^* \). The dramatic differences in \( \gamma(P) \) at low and high pressures suggest that either the empirical relationship is invalid and underestimates \( \gamma \) in the high-pressure regime by at least two orders of magnitude or there is a very rapid crossover in the density of low-energy excitations in the vicinity of \( P^* \). In the Fermi-liquid-like state above \( P^* \), we would expect the temperature scale \( T_{FL} \) (see Fig. 2) to be proportional to \( A^{-0.5} \) instead of the approximate \( T_{FL} \propto \sqrt{A} \) found experimentally. We do not understand this discrepancy. Direct measurements of \( C/T \) at pressures above \( P^* \) would be valuable.

Finally, we note that the resistive transition to the superconducting state is equally sensitive to the crossover at \( P^* \). As shown in Fig. 3c, the relative transition width \( \Delta T_c/T_c \) passes through a pronounced minimum near \( P^* \). To some extent this behavior is accentuated by the pressure variation of \( T_c \), but \( \Delta T_c \) itself is a minimum near \( P^* \). It is as if the optimally homogeneous state is singular in the vicinity of \( P^* \).

From the data presented above, we construct a temperature-pressure phase diagram for \( \text{CeCoIn}_5 \) given in Fig. 4a. This phase diagram is more like that of the cuprates around optimal doping than the canonical behavior of heavy-fermion systems exemplified, for example, by \( \text{CeIn}_3 \) or \( \text{CePd}_2 \text{Si}_2 \). There is no obvious long-range ordered state in \( \text{CeCoIn}_5 \) at atmospheric or higher pressures. Rather, in view of its relationship to \( \text{CeRhIn}_5 \), a reasonable speculation is that the antiferromagnetic QCP of \( \text{CeCoIn}_5 \) is at a slightly negative pressure. The resistivity exponent \( n \) and specific heat (ref.
are those of a non-Fermi-liquid up to $P^*$ where $T_c$ is a maximum. Beyond $P^*$, $T_c$ drops rapidly and parameters characterizing electronic transport are qualitatively different, Fermi-liquid-like at temperatures just above $T_c$ and those of a 3-D quantum-critical antiferromagnetic state at higher temperatures.

The generic $T$–$P$ phase diagram in Fig. 1 provides a broader perspective of our experimental observations and their relationship to CeRhIn$_5$. In the spirit of this diagram, the non-Fermi-liquid transport and thermodynamic properties of CeCoIn$_5$ below $P^*$ are controlled by an antiferromagnetic QCP at the inaccessible negative pressure denoted by $P_c$ in Fig. 1. We have chosen $P_c$ to correspond approximately to the critical pressure at which superconductivity appears in CeRhIn$_5$. Spin-lattice relaxation measurements on CeRhIn$_5$ identify a pseudogap whose signature first appears at pressures slightly less than $P_c$ and at a temperature of $\sim 5$ K and then decreases toward $T_c$ with increasing pressure until its signature is lost at $P \approx P_c$. If we associate the temperature $T_{pg}(P)$ in Fig. 1 with a resistive signature for a pseudogap and extrapolate $T_{pg}(P)$ in CeCoIn$_5$ to slightly negative pressures, there is a natural connection between our observations and properties of CeRhIn$_5$. Neither the $1/T_1T$ measurements on CeRhIn$_5$ nor our transport measurements on CeCoIn$_5$ permit definitive statements about the origin or nature of the pseudogap. We note, however, that 4-fold anisotropy of the thermal conductivity in the basal plane of CeCoIn$_5$ persists to 3.2 K $\approx T_{pg}$ at atmospheric pressure. If this in-plane modulation of the normal-state thermal conductivity is due to the presence of a pseudogap, it implies d-wave symmetry.

The schematic phase diagram, Fig. 1b, reflects the relationship among various phases of CeCoIn$_5$ and CeRhIn$_5$ and is similar to the $T$-doping phase diagram of the cuprates. This diagram and experimental data from which it is inferred indicate that the physics of heavy-fermion systems may be more closely related to that of the cuprates than previously appreciated. Aside from the structural layering in CeCoIn$_5$ and CeRhIn$_5$ and their relatively high $T_c$’s, they are similar in many respects to other Ce-based heavy-fermion materials in which superconductivity develops near a QCP. Further study of CeCoIn$_5$, in parallel with the cuprates and other pressure-induced heavy-fermion superconductors, holds promise for bridging our understanding of the inter-relationship between unconventional superconductivity and quantum criticality across these interesting classes of correlated matter.

We thank R. Movshovich for helpful discussions. VS acknowledges S. M. Stishov for his support during the initial stage of this research. Work at Los Alamos was performed under the auspices of the US Department of Energy.

---

* Permanent address: Institute for High Pressure Physics, Russian Academy of Sciences, Troitsk, Russia.

[1] J. Orenstein and A. J. Millis, Science 288, 468 (2000); S. Sachdev, ibid. 288, 475 (2000).
[2] A. Y. Chubukov, D. Pines, and J. Schmalian, cond-mat/0201140.
[3] F. W. Anderson, Physica B 318, 28 (2002).
[4] P. Coleman and C. Pepin, Physica B 312-313, 383 (2002).
[5] P. Menthoux, A. V. Balatsky, and D. Pines, Phys. Rev. Lett. 67, 3448 (1991).
[6] C. M. Varma, Z. Nussinov, and W. van Saarloo, Phys. Rep. 361, 267 (2002).
[7] N. D. Mathur et al., Nature 394, 39 (1998).
[8] T. Timusk and B. Statt, Rep. Prog. Phys. 62, 61 (1999).
[9] See, for example, C. M. Varma, Phys. Rev. Lett. 83, 3538 (1999); A. Sokol and D. Pines, Phys. Rev. Lett. 71, 2813 (1993); S. Chakravarty et al., Phys. Rev. B 63, 094503 (2001); Ar. Abanov, A.V. Chubukov, and J. Schmalian, Europhys. Lett. 55, 369 (2001); and references therein.
[10] C. Petrovici et al., J. Phys.: Condens. Matter 13, L337 (2001).
[11] R. Movshovich et al., Phys. Rev. Lett. 86, 5152 (2001).
[12] K. Izawa et al., Phys. Rev. Lett. 87, 057002 (2001).
[13] Y. Kohori et al., Phys. Rev. B 64, 134526 (2001).
[14] H. Hegger et al., Phys. Rev. Lett. 84, 4986 (2000).
[15] R. A. Fisher et al., Phys. Rev. B 65, 224509 (2002).
[16] M. Nicklas et al., J. Phys.: Condens. Matter 13, L905 (2001).
[17] H. Shishido et al., J. Phys. Soc. Jpn. 71, 162 (2002).
[18] L. G. Khvostantsev, V. A. Sidorenko, and O. B. Tsiok, in: Properties of Earth and Planetary Materials at High Pressures and Temperatures, Ed. by M.H. Manghnani and T. Yagi, Geophysical Monograph 101, American Geophysical Union, 1998, p.89.
[19] A. Eliing and J. S. Schilling, J. Phys. F: Metal Phys. 11, 623 (1981).
[20] R. Movshovich, private communication.
[21] S. Sachdev, Quantum Phase Transitions (Cambridge University Press, Cambridge, 1999).
[22] E. G. Moshopoulou et al., J. Solid State Chem. 158, 25 (2001).
[23] R. Settai et al., J. Phys.: Condens. Matter 13, L627 (2001).
[24] A. Rosch, Phys. Rev. Lett. 82, 4280 (1999).
[25] K. Miyake and O. Narikiyo, J. Phys. Soc. Jpn. 71, 867 (2002).
[26] G. Sparn, et al. Physica B 312-313, 138 (2002); E. Lengyel et al. High Pressure Res. 22, 185 (2002). In these references, for $H \geq H_c(0)$ $C/T = -\ln T$ persists to $P = 1.48$ GPa, but the rate of divergence decreases with increasing $P$.
[27] K. Kadowaki and S. B. Woods, Solid State Comm. 58, 507 (1986).
[28] S. Kawasaki et al., Phys. Rev. B 65, 020504 (2001).
[29] There is no obvious evidence for a pseudogap at $P = 0$ in specific heat or magnetic susceptibility data for CeCoIn$_5$, possibly because the signature is expected to be weak and the temperature range between $T_{pg}$ and $T_c$ is limited to no more than 1.1 K.