

## Magnetism and superconductivity in $\text{Ce}_2\text{RhIn}_8$

M. Nicklas, V. A. Sidorov,\* H. A. Borges,<sup>†</sup> P. G. Pagliuso, C. Petrovic,<sup>‡</sup> Z. Fisk,<sup>§</sup> J. L. Sarrao, and J. D. Thompson  
 Los Alamos National Laboratory, Los Alamos, New Mexico 87545

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We report the discovery of pressure-induced superconductivity, with  $T_c=2$  K, in the heavy-fermion antiferromagnet  $\text{Ce}_2\text{RhIn}_8$ , where superconductivity and magnetic order coexist over an extended pressure interval. A  $T$ -linear resistivity in the normal state, accessed by an applied magnetic field, does not appear to derive from the existence of a two-dimensional quantum-critical spin-density wave.

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Discoveries of pressure-induced superconductivity in several cerium-based heavy-fermion antiferromagnets have provided a qualitative perspective on the complex relationship between magnetism and superconductivity in these highly correlated systems.<sup>1</sup>  $\text{CeIn}_3$  is a typical example. Application of pressure suppresses its Néel temperature from  $T_N \sim 10$  K at atmospheric pressure to zero temperature at a critical pressure  $P_c \sim 2.5$  GPa.<sup>2</sup> Neutron-diffraction studies<sup>3</sup> show that the ordered  $4f$  moment decreases with  $T_N(P)$ , behavior also reflected in a monotonic depression of a specific-heat anomaly at  $T_N$  that disappears as  $P$  approaches  $P_c$ .<sup>4</sup> This evolution in magnetic properties arises from a pressure-induced increase in hybridization between Ce's  $f$  electron and conduction-band electrons. Near  $P_c$ , the electrical resistivity assumes a quasilinear temperature dependence, in contrast to  $\rho \propto T^2$  expected of a Landau-Fermi liquid, and is consistent with quasiparticle scattering from a quantum-critical spin-density wave.<sup>1</sup> Damped spin fluctuations in this critical region mediate Cooper pairing in the unconventional superconducting state that emerges in a narrow pressure range centered around  $P_c$ .<sup>1</sup> Like  $\text{CeIn}_3$ , which has a maximum  $T_c \approx 0.25$  K, other examples in this class have  $T_c$ 's well below 1 K and superconductivity appears only in the "cleanest" samples with a long electronic mean-free path.

A counter example to this view is  $\text{UPd}_2\text{Al}_3$  in which local-moment antiferromagnetism and unconventional superconductivity coexist from atmospheric to high pressures.<sup>5</sup> In this case, the three  $5f$  electrons in U assume dual characters: two localized  $f$ 's are responsible for antiferromagnetism at  $T_N = 14.5$  K and the other hybridizes with conduction states to form a liquid of heavy quasiparticles that becomes unstable below 2 K with respect to a pairing interaction derived from dispersive excitations of the ordered moments.<sup>6</sup>

In both examples, unconventional superconductivity is mediated by a magnetic interaction, but the bosonic excitations are distinctly different. In the Ce superconductors there is only a single  $f$  electron participating in both magnetism and superconductivity through its hybridization with itinerant electrons; whereas, there is a functional separation of  $f$  electrons in  $\text{UPd}_2\text{Al}_3$ . In the following, we present pressure-dependent measurements of the heavy-fermion antiferromagnet  $\text{Ce}_2\text{RhIn}_8$  in which magnetic order and superconductivity appear to coexist over a rather broad pressure range and are accompanied by an unexpected  $T$ -linear variation in electrical

resistivity. These results suggest that  $\text{Ce}_2\text{RhIn}_8$  and perhaps the structurally related compound  $\text{CeRhIn}_5$  present a different example of the interplay between magnetism and superconductivity in strongly correlated matter.

$\text{Ce}_2\text{RhIn}_8$  is a member of the family of heavy-fermion antiferromagnets  $\text{Ce}_n\text{RhIn}_{3n+2}$  composed of  $n$  layers of  $\text{CeIn}_3$  separated by a single layer of  $\text{RhIn}_2$ , a sequence repeated along the tetragonal  $c$  axis.<sup>7</sup> The  $n=1$  member,  $\text{CeRhIn}_5$ , becomes superconducting at pressures above 1.6 GPa with a  $T_c$  exceeding 2 K,<sup>8</sup> nearly an order of magnitude higher than the infinite layer member  $\text{CeIn}_3$ . Inserting a second layer of  $\text{CeIn}_3$  into  $\text{CeRhIn}_5$  gives  $\text{Ce}_2\text{RhIn}_8$ , which orders in a commensurate antiferromagnetic structure at 2.8 K with an ordered moment of  $0.55 \mu_B$ ,<sup>9</sup> slightly reduced by Kondo-spin compensation from the moment expected in the ground-state crystal-field doublet. It undergoes a second transition to an incommensurate magnetic structure at  $T_{LN} = 1.65$  K,<sup>10</sup> which, as will be shown, is irrelevant to the superconductivity that appears with applied pressure.

$\text{Ce}_2\text{RhIn}_8$  single crystals were grown out of excess In flux. X-ray diffraction on powdered crystals revealed single-phase material in the primitive tetragonal  $\text{Ho}_2\text{CoGa}_8$  structure with lattice parameters  $a = 0.44665$  nm and  $c = 1.2244$  nm at room temperature. There was no evidence for intergrowth of  $\text{CeRhIn}_5$ . Four-probe ac-resistance measurements were made with current flow in the  $(a,b)$  plane. Clamp-type cells generated hydrostatic pressures to 0.6 GPa for dc magnetization and 2.3 GPa for resistivity measurements. Flourinert-75 served as the pressure medium. Hydrostatic pressures to 5.0 GPa were produced in a toroidal anvil cell using a glycerol-water mixture.<sup>11</sup> In both cases, the superconducting transition of Pb (Sn), which served as a pressure gauge, remained sharp at all pressures, indicating a pressure gradient of less than 1–2% of the applied pressure.

The overall behavior of the resistivity is shown in Fig. 1. The high-temperature resistivity increases with increasing pressure over the temperature range between about 25 K and room temperature. There is well-defined maximum in the resistivity at  $T_{\text{max}} = 5$  K that initially decreases with  $P$  before increasing at a rate of  $\sim 20$  K/GPa for  $P \geq 2.0$  GPa. This initial negative  $\partial T_{\text{max}} / \partial P$  is unexpected for a Ce-based compound but is found in  $\text{CeRhIn}_5$ .<sup>8,12</sup> In that case,  $T_{\text{max}}(P)$  followed the pressure dependence of a maximum in the static susceptibility that is produced by the development of anisotropic antiferromagnetic correlations above  $T_N$ .<sup>13</sup> Presum-

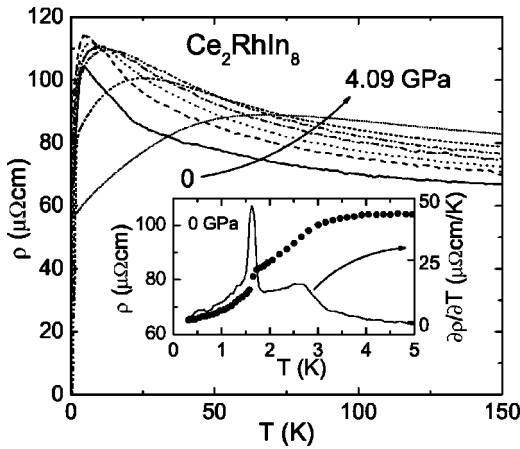


FIG. 1. Temperature dependence of the in-plane resistivity of  $\text{Ce}_2\text{RhIn}_8$  at various fixed pressures from ambient pressure to 4.09 GPa. The inset shows the  $P=0$  low-temperature resistivity and its derivative. The magnetic transitions are clearly indicated at  $T_N=2.8$  K and  $T_{LN}=1.65$  K, respectively.

ably, the initial decrease in  $T_{\text{max}}(P)$  in  $\text{Ce}_2\text{RhIn}_8$  has the same origin as in  $\text{CeRhIn}_5$ . The increase in  $T_{\text{max}}(P)$  at higher pressures is due to a shift of the characteristic spin-fluctuation temperature to progressively higher energies.

The inset of Fig. 1 shows the low-temperature resistivity and its derivative at atmospheric pressure. Compared to the typically small residual resistivity  $\rho_0 \approx 1 \mu\Omega \text{ cm}$  of  $\text{CeIn}_3$  and  $\text{CeRhIn}_5$ ,<sup>1,8</sup>  $\rho_0$  for  $\text{Ce}_2\text{RhIn}_8$  is one to two orders of magnitude higher. A low residual resistivity ratio and high  $\rho_0$  are reproduced in all of many high-quality crystals of  $\text{Ce}_2\text{RhIn}_8$  we have studied and, therefore, appears to be an intrinsic property of this compound. In spite of the high resistivity,  $\rho(T)$  and  $\partial\rho/\partial T$  clearly reveal the commensurate and incommensurate antiferromagnetic transitions at  $T_N=2.8$  K and  $T_{LN}=1.65$  K, respectively. Using the data in Fig. 1 and the maxima in  $\partial\rho/\partial T$  to track the pressure evolution of these phase transitions, we find that  $T_N(P)$  decreases linearly at a rate  $\partial T_N/\partial P \approx -0.76$  K/GPa. This slope is confirmed by dc-susceptibility measurements to 0.5 GPa.

Figure 2 gives a detailed view of the low-temperature resistivity at intermediate pressures. We see that the incommensurate transition is very sensitive to pressure.  $T_{LN}$  shifts from 1.65 K at ambient pressure to 0.95 K at 0.02 GPa. This gives an estimate of a critical pressure  $P_{c, LN} \approx (0.04 \pm 0.01)$  GPa for suppressing  $T_{LN}$  and a corresponding slope of  $\partial T_{LN}/\partial P \approx -(43 \pm 15)$  K/GPa that is consistent with  $\partial T_{LN}/\partial P$  derived from Ehrenfest's relation and measurements of the low-temperature specific heat and volume thermal expansion.<sup>10</sup> Therefore, only the commensurate phase survives for  $P \geq 0.04$  GPa. At 0.55 GPa a weak decrease in the resistivity appears at  $T_7=420$  mK. Nearly the same temperature dependence of the resistivity and same resistivity feature at  $T_7$  are found for 0.69 and 0.89 GPa (not shown). We do not know the origin of this feature. At 1.10 GPa the data develop a steeper slope below  $\sim 1$  K followed by a kink near  $T_c=380$  mK. The kink shifts continuously to higher temperatures with increasing pressure and evolves smoothly into a zero-resistance state below 600 mK at 1.63 GPa. Mea-

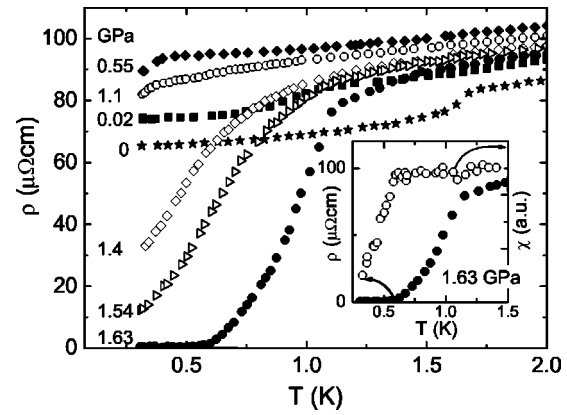


FIG. 2. Low-temperature resistivity of the in-plane resistivity of  $\text{Ce}_2\text{RhIn}_8$  at various fixed pressures. See text for details. With increasing pressure, a zero resistivity and diamagnetic state evolves below 600 mK at 1.63 GPa as shown in the inset.

surements of the ac susceptibility, plotted in the inset of Fig. 2, show the onset of a diamagnetic response at the same temperature where the resistance goes to zero. Although perfect diamagnetism could not be observed in the experimentally accessible temperature range, it is clear from the size of the signal change that the diamagnetic response is due to bulk superconductivity. Reproducibility of a zero-resistance state for  $P \geq 1.6$  GPa was confirmed on another crystal.

To provide additional confirmation of bulk superconductivity, we determined the upper critical field  $H_{c2}(T)$  at 1.63 GPa using data plotted in Fig. 3. The resistive onset defines  $H_{c2}(T)$ , which is shown in the inset. A fit of  $H_{c2} \propto [H_{c2}(T=0) - H_{c2}(T)]^2$  describes the data reasonably well with  $H_{c2}(0)=53.6$  kOe and an initial slope  $-dH_{c2}/dT|_{T=T_c}=91.8$  kOe/K.<sup>14</sup> The Ginzburg-Landau coherence length in the  $c$ -axis direction  $\xi_{GL} = [\Phi_0/2\pi H_{c2}(0)]^{0.5} \approx 7.7$  nm, which is comparable to the volume-averaged electronic mean-free path,  $l \approx 6.5$  nm.<sup>15</sup> The dirty-limit relationship  $-dH_{c2}/dT|_{T=T_c} \propto \rho_0 \gamma$  gives  $\gamma \approx 0.20$  J/mol Ce K<sup>2</sup> at 1.63 GPa, which is one-half the value measured directly at atmo-

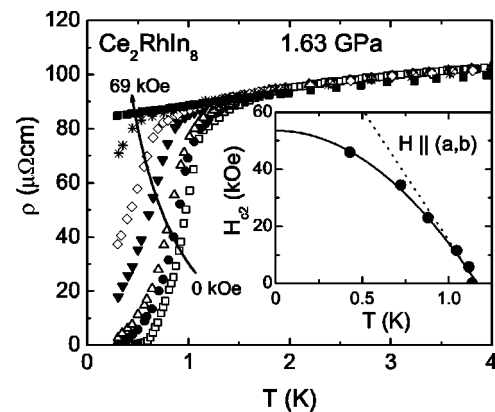


FIG. 3. Effect of a magnetic field on the resistivity of  $\text{Ce}_2\text{RhIn}_8$  at  $P=1.63$  GPa and in various magnetic fields up to 69 kOe applied parallel to the basal plane. The inset shows  $H_{c2}$  as a function of the temperature. The solid curve is a fit described in the text. The initial slope of  $H_{c2}(T)$  is indicated by the dotted line.

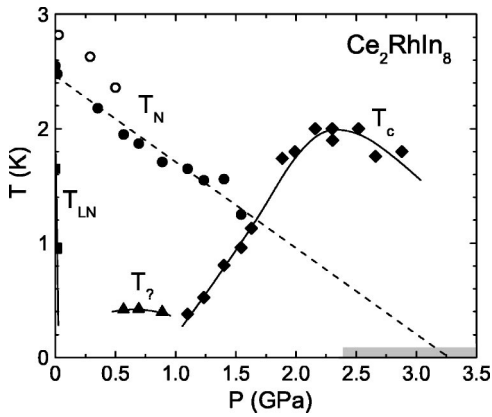


FIG. 4. The temperature-pressure phase diagram for  $\text{Ce}_2\text{RhIn}_8$  determined by  $\rho(T)$  (solid symbols) and dc-magnetization (open circles). The lines are guides to the eyes.

spheric pressure just above  $T_N$ . Halving of the Sommerfeld coefficient at 1.63 GPa is expected from the relationship  $\gamma(P) \propto 1/T_{\max}(P)$  obeyed by several heavy-fermion systems and our observation that  $T_{\max}(1.63)/T_{\max}(0) = 2.5$  in  $\text{Ce}_2\text{RhIn}_8$ . Furthermore, bulk superconductivity evolves out of a distinctly non-Fermi-liquid-like state. A fit to  $\rho(T)$  at 1.63 GPa and 69 kOe, solid squares in Fig. 3, gives  $\rho = \rho_0 + A'T^n$  with  $n = 0.95 \pm 0.05$  for  $0.3 \text{ K} \leq T \leq 1.8 \text{ K}$ . An approximately  $T$ -linear resistivity also is found above  $T_c$  in  $\text{CeRhIn}_5$ <sup>12</sup> and is qualitatively different from the  $T^{-1.6}$  dependence observed in  $\text{CeIn}_3$  near its critical pressure.<sup>1</sup>

Measurements to 5.0 GPa, but for  $T \geq 1 \text{ K}$ , show the onset of superconductivity reaching a maximum of 2.0 K near 2.3 GPa before decreasing below 1 K above 3.5 GPa. See Fig. 4.  $T_c$  reaches a maximum close to the pressure at which  $T_N$  extrapolates to zero. Unless  $T_N$  drops precipitously above  $\sim 1.5 \text{ GPa}$ , superconductivity and local moment, commensurate antiferromagnetism coexist over a substantial range of pressures. A critical pressure of  $\sim 2.5 \text{ GPa}$  and a “dome” of superconductivity with a maximum  $T_c$  centered near the extrapolated critical pressure also are found in  $\text{CeIn}_3$ ;<sup>1</sup> however, the maximum  $T_c$  of  $\text{Ce}_2\text{RhIn}_8$  reaches a value comparable to  $T_N(P=0)$ , as it does in  $\text{CeRhIn}_5$ ,<sup>8,12</sup> and is an order of magnitude higher than in  $\text{CeIn}_3$ , where  $T_c/T_N \approx 0.025$ . Interestingly, the dome of superconductivity exists over a rather broad pressure range, at least 2.5 GPa, in  $\text{Ce}_2\text{RhIn}_8$  but is very narrow,  $\sim 0.4 \text{ GPa}$ , in  $\text{CeIn}_3$ ; the pressure range over which superconductivity exists scales roughly with  $T_c$ .

To interpret these observations, we first consider the perspective developed from other examples of  $P$ -induced superconductivity in Ce-based systems. For magnetically mediated superconductivity,  $T_c \propto T_{sf}$ , where  $T_{sf}$  is the characteristic spin-fluctuation temperature that is inversely proportional to the specific-heat Sommerfeld coefficient  $\gamma$ .<sup>16</sup> For  $\text{CeRhIn}_5$ ,<sup>17</sup>  $\text{Ce}_2\text{RhIn}_8$  and  $\text{CeIn}_3$ ,<sup>4</sup>  $\gamma \sim 0.4, 0.2,$  and  $0.37 \text{ J/mol Ce K}^2$ , respectively, at  $P \leq P_c$ . With all other factors equal,  $T_c$ 's, then, should be approximately ( $\pm 50\%$ ) the same within this family of materials; instead,  $T_c$ 's of the layered compounds are much higher than in  $\text{CeIn}_3$ . Though  $T_{sf}$  sets the overall scale for the magnitude of  $T_c$ ,  $T_c$  also depends on the effective dimensionality of the spin fluctua-

tions and the electronic structure: reduced dimensionality favors a higher  $T_c$ .<sup>18</sup>

Support for this scenario comes from conventional models of antiferromagnetic quantum criticality.<sup>19</sup> These models predict that, near  $P_c$ ,  $T_N \propto (P_c - P)^{z/d}$  and  $\rho(T) \propto T^{d/z}$ , where the dynamical exponent  $z=2$  and  $d$  is the effective dimensionality of the spin-fluctuation spectrum. Experimental observations on  $\text{CeIn}_3$  are consistent with theoretical predictions for  $d=3$  and with  $d=2$  in  $\text{Ce}_2\text{RhIn}_8$  and provide a plausible explanation for the unexpectedly high  $T_c$  of  $\text{Ce}_2\text{RhIn}_8$ . In this picture, the  $d$ -wave superconductivity<sup>17</sup> and even somewhat higher  $T_c$  of  $\text{CeRhIn}_5$  would be attributable to more nearly optimal matching of the momentum dependence of the dynamic spin susceptibility  $\chi(\mathbf{q}, \omega)$  to its quasi-two-dimensional (2D) electronic structure.<sup>20</sup> Further, this interpretation supports superconductivity existing over a much wider range of pressures in  $\text{Ce}_2\text{RhIn}_8$  than in  $\text{CeIn}_3$  because the effective pairing interaction is expected to be stronger in quasi-2D than in 3D.<sup>18</sup>

Though providing a qualitative account of our observations, the interpretation outlined above relies on a model<sup>19</sup> in which the non-Fermi-liquid temperature dependence of  $\rho(T)$  arises from Bragg diffraction of heavy quasiparticles off a quantum-critical spin-density wave (SDW). In this case, the scattering is critical only on “hot” portions of the Fermi surface spanned by the antiferromagnetic ordering wave vector  $\mathbf{Q}$ ; whereas, other parts of the Fermi surface are unaffected.<sup>21</sup> Unless all of the Fermi surface is hot, the resistivity should vary as  $T^{1+\epsilon}$ , where  $0 < \epsilon < 1$ , in contradiction to our observations and those<sup>12</sup> on  $\text{CeRhIn}_5$ . Neutron-diffraction studies of  $\text{CeRhIn}_5$ <sup>22</sup> show that  $\mathbf{Q}$  and the local moment remain well defined and weakly changing as  $P \rightarrow P_c$ , supporting speculation of similar behavior in  $\text{Ce}_2\text{RhIn}_8$ . We cannot rule out the possibility that most of the Fermi surface is exactly spanned by  $\mathbf{Q}$  in both compounds, but this seems very unlikely.

Alternatively, we speculate that the entire Fermi surface is hot and  $\rho(T)$  takes a non-Fermi-liquid form because the quantum criticality is local.<sup>21,23</sup> At a local quantum-critical point,  $\chi(\mathbf{q}, \omega)$  has an anomalous frequency dependence throughout the entire Brillouin zone and not just at  $\mathbf{Q}$ . Unlike the weak-coupling SDW limit, local criticality, which is facilitated by two dimensionality, requires the physics of a Kondo lattice, i.e., a local-moment coupled antiferromagnetically to a bath of itinerant spins and a fluctuating field produced by surrounding *local* moments.<sup>23</sup> The nature of superconductivity that might develop near a local quantum-critical point remains to be investigated, but, because the basic interactions are antiferromagnetic, as in the SDW limit, superconductivity of  $d$ -wave symmetry would be expected.<sup>24</sup> Unconventional superconductivity near a quantum-critical SDW is favored when  $\xi_{GL} \ll l$ ,<sup>1</sup> which is satisfied in  $\text{CeIn}_3$ ; however,  $l/\xi_{GL} \sim 1$  in  $\text{Ce}_2\text{RhIn}_8$  and  $\sim 3$  in  $\text{CeRhIn}_5$ . This, the much higher  $T_c$ 's, quasi-two dimensionality and unexpected  $\rho \propto T$  suggest that unconventional superconductivity in the latter two compounds may be mediated by qualitatively different spin fluctuations than in  $\text{CeIn}_3$ . Finally, we note that a

theory<sup>25</sup> unifying the order parameters of antiferromagnetism and *d*-wave superconductivity predicts a *T*–*P* phase diagram like that shown in Fig. 4 and allows  $T_c/T_N \sim 1$  as found in Ce<sub>2</sub>RhIn<sub>8</sub>.

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- \*Permanent address: Institute for High Pressure Physics, Russian Academy of Sciences, Troitsk, Russia.
- †Permanent address: Departamento de Física, Pontifícia Universidade Católica do Rio de Janeiro, Brazil.
- ‡Permanent address: Brookhaven National Laboratory, Upton, NY.
- §Permanent address: NHMFL, Florida State University, Tallahassee, FL.
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