Available online at www.sciencedirect.com

ScienceDirect

Journal of Magnetism and Magnetic Materials 310 (2007) 532–535

Journal of magnetism and magnetic materials

www.elsevier.com/locate/jmmm

Magnetism and unconventional superconductivity in isostructural cerium and plutonium compounds

J.D. Thompson*, Tuson Park, N.J. Curro, F. Ronning, R. Movshovich, E.D. Bauer, J.L. Sarrao

Los Alamos National Laboratory, MS K764, Los Alamos, NM 87545, USA

Available online 10 November 2006

Abstract

The heavy-fermion antiferromagnet $CeRhIn_5$ exhibits a complex interplay between magnetism and unconventional superconductivity (SC) as a function of applied pressure and magnetic field. This interplay leads to a line of magnetic quantum-critical points within the superconducting state. A comparison of nuclear spin-relaxation measurements on $CeRhIn_5$ to those made on $CeCoIn_5$, $PuCoGa_5$, and $PuRhGa_5$ suggests that SC and magnetism also may be related closely in these isostructural superconductors.

PACS: 75.20.Hr; 74.70.Tx; 74.20.Mn; 75.30.Mb

Keywords: Superconducting mechanisms; $CeRhIn_5$; $PuCoGa_5$; Pressure; Heavy fermion; NQR

1. Introduction

Magnetism and unconventional superconductivity (SC) are closely coupled broken symmetries in heavy-fermion materials. The connection between these two states is most apparent in Ce- and U-based compounds in which magnetism and SC are invariably nearby each other as a function of some tuning parameter and often coexist [1]. $CeRhIn_5$ provides one of several examples of this interplay that suggests magnetic fluctuations play an essential role in creating unconventional SC [2]. On the other hand, Pu-based superconductors are rare, with only two examples $PuCoGa_5$ [3] and $PuRhGa_5$ [4]. These materials form in the same tetragonal, $HoCoGa_5$ (1 1 5) structure type as $CeRhIn_5$ and have modestly enhanced electronic specific heats of order 100 mJ/mol K^2 [3,4]. So far, no magnetically ordered Pu-member of the 115 family has been found, which makes it less clear that their SC arises in proximity to magnetism. Nevertheless, relationships between the Ce- and Pu115s, discussed below, allow the reasonable plausibility that magnetic fluctuations are involved as well in Cooper pairing in the Pu superconductors.

2. Results and discussion

$CeRhIn_5$ is an instructive starting point for considering the relationship between magnetism and SC in the 115 family of materials. At atmospheric pressure, neutron diffraction [5] finds an ordered moment ($0.8\,\mu_B$) below a Néel temperature of $T_N = 3.8$ K that is close to that expected for a localized 4f electron in a crystal-field doublet ground state. The Fermi-surface topology of $CeRhIn_5$ is nearly identical to its La-analog, additionally implying that the 4f electron is essentially localized [6].

The response of $CeRhIn_5$ to applied pressure is shown by data in the vertical plane in Fig. 1 [7]. T_N decreases for $P > 1$ GPa and extrapolates to $T = 0$ at $P_2 \approx 2.4$ GPa; however, the pressure-induced superconducting transition temperature $T_c = T_N$ for $P_1 \approx 1.7$ GPa, which hides evidence for coexistence of SC and magnetic order (MO) above P_1 and prevents a reliable extrapolation of $T_N(P)$ to $T = 0$. Qualitatively, this T–P relationship between magnetism and SC is typical of other Ce-based heavy-fermion systems [8]. Recent specific heat measurements have

*Corresponding author. Tel.: +1 505 667 6416; fax: +1 505 665 7652.
E-mail address: jdt@lanl.gov (J.D. Thompson).

0304-8853/$ - see front matter © 2006 Elsevier B.V. All rights reserved.
doi:10.1016/j.jmmm.2006.10.138

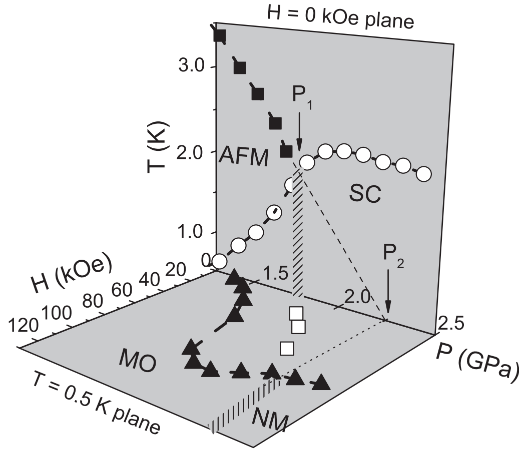

Fig. 1. Temperature–pressure–magnetic field phase diagram of $CeRhIn_5$ [7]. Points in the vertical T–P plane are determined from specific heat measurements for $H = 0$. Solid squares denote $T_N(P)$ and open circles show $T_c(P)$. For $P \leqslant P_1$, antiferromagnetism and SC coexist below the $T_c(P)$ boundary, but magnetism disappears for $P \geqslant P_1$. The horizontal H–P plane shows $H_{c2}(P, T = 0.5\,K)$, solid triangles, and field values at which a magnetic transition is induced in the superconducting state, open squares. MO: magnetically ordered normal state; NM: paramagnetic normal state.

established that magnetism, hidden by SC above P_1, is revealed at low temperatures by an applied magnetic field [7,9], which is illustrated by data points in the horizontal plane of Fig. 1. A linear extrapolation of the line of field-induced magnetic transitions intersects the upper critical field boundary ($H_{c2}(P,T)$) at a pressure very near P_2, where there is a magnetic to non-magnetic transition. P_2 appears to be a bone fide quantum-critical point, as evidenced by deHaas–van Alphen experiments that find the cyclotron mass diverging at P_2 [10]. At P_2, dHvA frequencies increase abruptly and above P_2 correspond closely to those of isostructrural $CeCoIn_5$, which is superconducting at atmospheric pressure with nearly the same T_c as $CeRhIn_5$ at P_2 and whose dHvA frequencies are consistent with electronic structure calculations assuming an itinerant 4f electron [10]. Consequently, these experiments also are consistent with a 4f-localized to 4f-delocalized, or at least partially delocalized, transition in $CeRhIn_5$ at P_2.

Nuclear spin-relaxation ($1/T_1$) experiments are especially useful for probing magnetism and SC. For $CeRhIn_5$ at $P \geqslant P_1$, $1/T_1$ is proportional to T^3 for $T < T_c$, which, together with the absence of a Hebel–Slichter coherence peak near T_c, is consistent with gap nodes in an unconventional superconductor; whereas, above T_c, $1/T_1$ increases as $T^{1/2}$, as expected for relaxation dominated by fluctuations in a 3-dimensional metal near an antiferromagnetic instability [11,12]. Very near a magnetic quantum-phase transition, $1/T_1$ is predicted [13] to exhibit a weaker temperature dependence ($1/T_1 \propto T^{1/4}$), which is observed in $CeCoIn_5$ at $P = 0$ and $T \geqslant T_c$ [14], but NQR experiments have not been performed on $CeRhIn_5$ at P_2. Nevertheless, Fig. 2 shows that, once zero-field magnetism disappears in $CeRhIn_5$ above P_1, there is a strong similarity

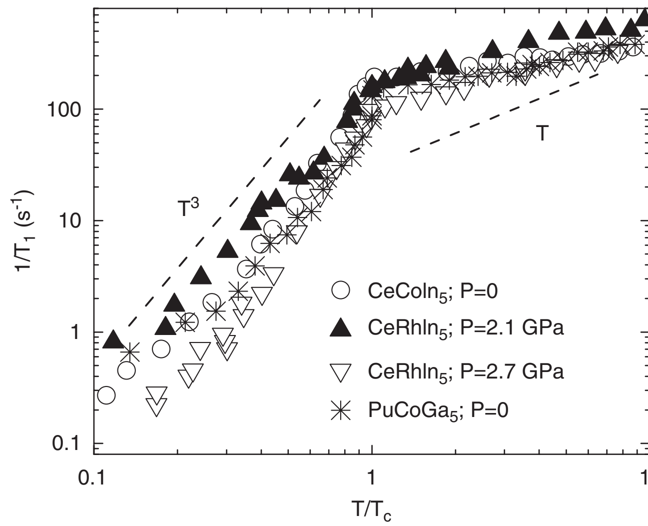

Fig. 2. Nuclear spin-relaxation rate $1/T_1$ as a function of reduced temperature T/T_c, both on logarithmic scales. Values of T_c for $CeCoIn_5$ and $CeRhIn_5$ under pressure are similar and range from 2.05 to 2.3 K, but T_c for $PuCoGa_5$ is 18.5 K. In each e.g., $1/T_1$ is approximately proportional to T^3 below T_c and increase as T^n, where $n \approx -1/4 - 1/2$, over an extended temperature range above T_c. At the lowest temperatures, $1/T_1$ begins to deviate from a T^3 dependence, which may be due to low-lying excitations in the gap nodes. $CeCoIn_5$ (Ref. [14]); $CeRhIn_5$ at 2.1 GPa (Ref. [12]); $CeRhIn_5$ at 2.7 GPa (Ref. [15]); $PuCoGa_5$ (Ref. [16]).

in the evolution of $1/T_1$ above and below T_c in both $CeRhIn_5$ and $CeCoIn_5$.

If antiferromagnetic fluctuations are essential for SC in these Ce-115 compounds, SC should weaken as magnetic fluctuations are suppressed by moving away from a magnetic boundary. The $H = 0$ plane in Fig. 1 shows that T_c begins to decrease slowly above P_2, and measurements to higher pressures establish that T_c in $CeRhIn_5$ becomes zero or very low at a pressure near 6 GPa[17]. This trend is expected for spin-mediated SC, but a predictive understanding of the relationship between T_c and complex spin dynamics reflected in $1/T_1$ experiments is lacking.

As seen in Fig. 2, the relaxation rate in $PuCoGa_5$ also varies as T^3 below its T_c of 18.5 K and increases as a weak power law above T_c [16], similar to what is found in $CeCoIn_5$ and in $CeRhIn_5$ under pressure. These results are the strongest evidence for unconventional SC in $PuCoGa_5$ and also suggest that SC develops out of a normal state in which magnetic fluctuations are present. However, experiments sensitive to the phase of the superconducting order parameter are required to prove that SC in these 115 materials is unconventional. Establishing that magnetic fluctuations are responsible, at least in part, for SC is even more challenging and likely will come only from a body of indirect evidence and comparison to theoretical models.

The role of magnetic fluctuations in $PuRhGa_5$ is less clear. Though $1/T_1$ follows at T^3 dependence below T_c, $1/T_1T$ is a constant from T_c to about 30 K [18]. This Korringa-like normal state relaxation also is approached in $CeCoIn_5$ at pressures near 3 GPa [15], well away from its quantum-critical regime, and by analogy suggests that

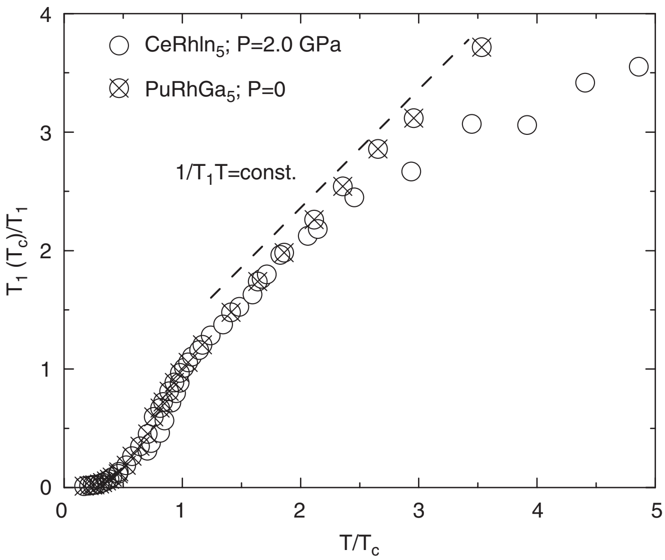

Fig. 3. Relaxation rate $1/T_1$, normalized by its value at T_c, as a function of reduced temperature for $PuRhGa_5$ (Ref. [18]) and $CeRhIn_5$ at 2.0 GPa (Ref. [21]). There is a small difference in the absolute value of pressure, at which P_1 occurs in Ref. [21] and in data plotted in Fig. 1. The dashed line corresponds to a Korringa-like temperature dependence.

$PuRhGa_5$ may not be so close to a magnetic instability. In the case of $CeCoIn_5$, the emergence of a Korringa-like relaxation rate corresponds with the development of a Fermi-liquid-like T^2 variation in electrical resistivity above T_c; [19] this correspondence also may be found in $PuRhGa_5$, but a non-Fermi-liquid temperature dependence ($\rho \propto T^{1.35}$) appears to describe the resistivity equally well [4].

An alternative to a Fermi-liquid explanation for the Korringa-like dynamics is that $PuRhGa_5$ is near an antiferromagnetic instability, but the competition between contributions to $1/T_1$ from magnetic fluctuations and from the presence of a pseudogap results in a temperature-independent $1/T_1T$ above T_c [20]. With presently available data, it is not possible to distinguish, which of these two interpretations is correct; however, it is interesting that $1/T_1 \propto T$ behavior also is observed in $CeRhIn_5$ at pressures just below P_1, where the resistivity is not proportional to T^2, and is interpreted as being due to pseudogap formation [21]. Fig. 3 compares the temperature dependences found in $PuRhGa_5$ and in $CeRhIn_5$ very close to P_1.

As seen in results of Fig. 2, the $1/T_1 \propto T$ dependence in $CeRhIn_5$ evolves away from Korringa-like behavior with increasing pressure. It would be interesting to determine how the relaxation rate of $PuRhGa_5$ changes with applied pressure, especially because its $T_c(P)$ also exhibits a maximum [22] similar to that found in $CeRhIn_5$. Likewise, inelastic neutron scattering studies of the Ce- and Pu-based 115 would be a valued complement to nuclear relaxation and other existing measurements for understanding more thoroughly the relationship between magnetism and SC in these systems.

3. Summary

In the absence of more definitive experiments and of a microscopic theory against which experiment can be compared, we cannot state unequivocally that superconductivity in the 115 compounds is magnetically mediated. There is, however, a growing body of evidence that consistently points to an intimate relationship between magnetism and SC in the isostructural and nominally isoelectronic 115 heavy-fermion materials [16,23]. One of many outstanding questions yet to be answered, especially in regard to $CeRhIn_5$, is how does magnetic order affect SC, and conversely, how does SC affect magnetism. With similarities between Ce115 and Pu115s, it seems plausible that magnetism could be hidden by SC in the Pu superconductors as it is in $CeRhIn_5$. This possibility becomes more plausible, given recent studies of $CeCoIn_5$, that suggest some form of magnetism may appear in its superconducting state below the upper critical field boundary at low temperatures [24,25].

Unfortunately, the Pu-bearing 115 s can be studied experimentally only at a few institutions and even there with some difficulty and with the complication of self-damage due to radioactive decay of the Pu nucleus. As a result, progress will be slow. These limitations do not apply to the Ce115s, which, with their relative high T_c's and interesting responses to modest pressures and accessible magnetic fields, offer an exceptional opportunity to study and understand the interplay between magnetism and SC in strongly correlated electron systems.

Acknowledgments

Work at Los Alamos was performed under the auspices of the US Department of Energy, Office of Science. T.P., F.R., and E. D. B also acknowledge fellowships supported by the Los Alamos Laboratory Directed Research and Development program.

References

[1] M. Sigrist, K. Ueda, Rev. Mod. Phys. 63 (1991) 239.
[2] H. Hegger, C. Petrovic, E.G. Moshopolou, M.F. Hundley, J.L. Sarrao, Z. Fisk, J.D. Thompson, et al., Phys. Rev. Lett. 84 (2000) 4986.
[3] J.L. Sarrao, L.A. Morales, J.D. Thompson, B.L. Scott, G.R. Stewart, F. Wastin, J. Rebizant, P. Boulet, E. Colineau, G.H. Lander, Nature (London) 420 (2002) 297.
[4] F. Wastin, P. Boulet, J. Rebizant, E. Colineau, G.H. Lander, J. Phys.: Condens. Matter 15 (2003) S2279.
[5] A. Llobet, J.S. Gardner, E.G. Moshopolou, J.M. Mignot, M. Nicklas, W. Bao, N.O. Moreno, P.G. Pagliuso, I.N. Goncharenko, J.L. Sarrao, J.D. Thompson, Phys. Rev. B 69 (2004) 024403.
[6] D. Hall, E.C. Palm, T.P. Murphy, S.W. Tozer, C. Petrovic, E. Miller-Ricci, L. Peabody, C.Q.H. Li, U. Alver, R.G. Goodrich, J.L. Sarrao, P.G. Pagliuso, J.M. Wills, Z. Fisk, Phys. Rev. B 64 (2001) 064506.
[7] T. Park, F. Ronning, H.Q. Yuan, M.B. Salamon, R. Movshovich, J.L. Sarrao, J.D. Thompson, Nature (London) 440 (2006) 65.
[8] J. Flouquet, in: W. Halperin (Ed.), Progress in Low Temperature Physics, vol. XV, Elsevier, Amsterdam, 2005, p. 3.

[9] G. Knebel, D. Aoki, D. Braithwaite, B. Salce, J. Flouquet, Phys. Rev. B 74 (2006) 020501.
[10] H. Shishido, R. Settai, H. Harima, Y. Onuki, J. Phys. Soc. Japan 74 (2005) 1103.
[11] Y. Koroi, Y. Yamato, Y. Iwamoto, T. Kohara, Eur. Phys. J. B 18 (2000) 601.
[12] T. Mito, S. Kawasaki, G.Q. Zheng, Y. Kawasaki, K. Ishida, Y. Kitaoka, D. Aoki, Y. Haga, Y. Onuki, Phys. Rev. B 63 (2001) 220507.
[13] A. Ishigaki, T. Moriya, J. Phys. Soc. Japan 65 (1996) 3402;
H. Kondo, J. Phys. Soc. Japan 71 (2002) 3011.
[14] Y. Kohori, Y. Yamato, Y. Iwamoto, T. Kohara, E.D. Bauer, M.B. Maple, Phys. Rev. B 64 (2001) 134526;
Y. Kawasaki, S. Kawasaki, M. Yashima, T. Mito, G.-Q. Zheng, Y. Kitaoka, H. Shishido, R. Settai, Y. Haga, Y. Onuki, J. Phys. Soc. Japan 72 (2003) 2308.
[15] Y. Kohori, H. Taira, H. Fukazawa, T. Kohara, Y. Iwamoto, T. Matsumoto, M.B. Maple, J. Alloys Compd. 51 (2006) 408–412.
[16] N.J. Curro, T. Caldwell, E.D. Bauer, L.A. Morales, M.J. Graf, Y. Bang, A.V. Balatsky, J.D. Thompson, J.L. Sarrao, Nature (London) 434 (2005) 622.
[17] T. Muramatsu, T.C. Kobayashi, K. Shimizu, K. Amaya, D. Aoki, Y. Haga, Y. Onuki, Physica C 539 (2003) 388–389.
[18] H. Sakai, Y. Tokunaga, T. Fujimoto, S. Kambe, R.E. Walstedt, H. Yasuoka, D. Aoki, Y. Homma, E. Yamamoto, A. Nakamura, Y. Shiokawa, K. Nakajima, Y. Arai, T. Matsuda, Y. Haga, Y. Onuki, J. Phys. Soc. Japan 74 (2005) 1710.
[19] V.A. Sidorov, M. Nicklas, P.G. Pagliuso, J.L. Sarrao, Y. Bang, A.V. Balatsky, J.D. Thompson, Phys. Rev. Lett. 89 (2002) 157004.
[20] Y. Bang, M.J. Graf, N.J. Curro, A.V. Balatsky, cond-mat/0603042.
[21] S. Kawasaki, M. Yashima, T. Mito, Y. Kawasaki, G.-q. Zheng, Y. Kitaoka, D. Aoki, Y. Haga, Y. Onuki, J. Phys: Condens. Matter 17 (2005) S889.
[22] J.-C. Griveau, P. Boulet, E. Colineau, F. Wastin, J. Rebizant, Physica B 1093 (2005) 359–361.
[23] E.D. Bauer, J.D. Thompson, J.L. Sarrao, L.A. Morales, F. Wastin, J. Rebizant, J.-C. Griveau, P. Javorsky, P. Boulet, E. Colineau, G.H. Lander, G.R. Stewart, Phys. Rev. Lett. 93 (2004) 147005.
[24] V.F. Mitrovic, M. Horvatic, C. Berthier, G. Knebel, G. Lapertot, J. Flouquet, cond-mat/0605305.
[25] B.L. Young, R.R. Urbano, N.J. Curro, J.D. Thompson, J.L. Sarrao, A.B. Vorontsov, M.J. Graf, cond-mat/0608040.