PHYSICAL REVIEW B **79**, 092415 (2009)

Commensurate magnetic structure of $\mathrm{CeRhIn_{4.85}Hg_{0.15}}$

Wei Bao,[1] Y. C. Gasparovic,[2,3] J. W. Lynn,[2] F. Ronning,[1] E. D. Bauer,[1] J. D. Thompson,[1] and Z. Fisk[4]
[1]*Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA*
[2]*NIST Center for Neutron Research, National Institute of Standards and Technology, Gaithersburg, Maryland 20899, USA*
[3]*Department of Materials Science and Engineering, University of Maryland, College Park, Maryland 20742, USA*
[4]*University of California–Irvine, Irvine, California 92697, USA*

(Received 2 October 2008; published 20 March 2009)

We show using neutron diffraction that the magnetic structure of $\mathrm{CeRhIn_{4.85}Hg_{0.15}}$ is characterized by a commensurate propagation vector (1/2,1/2,1/2). This is different from the magnetic structure in the parent compound $\mathrm{CeRhIn_5}$, which orders with an incommensurate propagation vector (1/2,1/2,0.297). The special relation between the commensurate magnetic mode and unconventional superconductivity has been shown previously for this class of heavy fermion superconductors. This work provides further evidence for the ubiquity of this antiferromagnetic mode.

DOI: 10.1103/PhysRevB.79.092415 PACS number(s): 75.25.+z, 71.27.+a, 75.20.Hr

I. INTRODUCTION

The discovery of superconductivity in $\mathrm{Ce}M\mathrm{In_5}$ (M=Rh, Ir, and Co) has reinvigorated research on heavy fermion materials.[1,2] $\mathrm{Ce}M\mathrm{In_5}$ can be viewed as built of alternate $\mathrm{CeIn_3}$ and $M\mathrm{In_2}$ layers in the tetragonal $\mathrm{HoCoGa_5}$ structure.[3,4] The layered crystal structure is reflected in the existence of a quasi-two-dimensional Fermi surface in addition to other three-dimensional Fermi surfaces.[5,6] Magnetism and transport, however, are three dimensional,[7] similar to previous heavy fermion materials.

Both $\mathrm{CeIrIn_5}$ and $\mathrm{CeCoIn_5}$ are superconductors at ambient pressure,[8,9] while $\mathrm{CeRhIn_5}$ becomes superconducting only under high pressure.[10] The phase transition of $\mathrm{CeRhIn_5}$ at ambient pressure is shown to be a magnetic one in nuclear quadrupole resonance (NQR) studies.[11] Magnetic structure was determined to be an incommensurate spiral with propagation vector (1/2,1/2,0.297) and a saturated magnetic moment $0.75\mu_B/\mathrm{Ce}$ by neutron diffraction.[12] Interesting relations between the superconductivity and antiferromagnetism are revealed in alloys with $\mathrm{CeRhIn_5}$, $\mathrm{CeIrIn_5}$, or $\mathrm{CeCoIn_5}$ as the end member.[13,14] It was discovered that an additional antiferromagnetic instability with a commensurate propagating vector (1/2,1/2,1/2) appears at the same time when superconductivity is induced by replacing Rh with Ir, suggesting an intimate relation between this magnetic mode and superconductivity.[15] Note that the coexistence of the antiferromagnetic orders with superconductivity in the alloys is not due to phase separation and the samples are highly uniform.[16,17] When Co is used to replace Rh, in addition to the (1/2,1/2,1/2) and (1/2,1/2,0.297) modes, a different incommensurate mode at $(1/2,1/2,\sim 0.4)$ also appears.[18,19] More direct evidence for the relationship between the (1/2, 1/2,1/2) mode and superconductivity is the observation of a magnetic resonance at (1/2,1/2,1/2) when $\mathrm{CeCoIn_5}$ enters the superconducting state.[20]

Before becoming a superconductor, the NQR signal from $\mathrm{CeRhIn_5}$ at the Néel temperature T_N drastically changes shape at high pressure.[21] This is likely related to an abrupt change in magnetic propagation vector to (1/2,1/2,0.4) in this pressure range.[22] Since the RKKY interaction between Ce magnetic moments is mediated by conduction electrons and the strength of the interaction in the reciprocal space that determines the magnetic ordering vector is related to the Fermi surfaces, the various magnetic orders observed so far provide important clues to the microscopic electronic processes in these heavy fermion materials. It would be interesting to establish a direct link between the propagation wave vectors and the various Fermi surfaces.[5,6,23,24]

Judging by the observed Fisher-Langer behavior in $\mathrm{CeRhIn_5}$, the complex Kondo renormalization process has reached its fixed point below $\sim$10 K at ambient pressure.[7] The same behavior can be investigated using thermal transport,[25] but one needs to keep in mind the accuracy of the Born approximation when comparing experiments to theory. The observations provide a powerful simplification for theory and justify the theoretical treatment of heavy fermion superconductivity at low temperatures without the many-body Kondo process, such as the recent one to account for the magnetic resonance in $\mathrm{CeCoIn_5}$.[26]

More recently, doping at the In site of $\mathrm{Ce}M\mathrm{In_5}$ provides another tuning parameter to explore the relation between magnetism and superconductivity.[27] The superconducting state of $\mathrm{CeCoIn_5}$ is suppressed by Cd doping, and the commensurate (1/2,1/2,1/2) antiferromagnetic state is introduced.[28] For nonsuperconducting $\mathrm{CeRhIn_5}$, T_N first decreases then rises with increasing Cd doping, suggesting a different magnetic state formation that is different from the incommensurate one in the parent $\mathrm{CeRhIn_5}$.[27] Similar behavior can be induced using Hg as the dopant.[29] Here we report neutron-diffraction studies on a Hg-doped $\mathrm{CeRhIn_5}$ sample in the present magnetic phase.

II. EXPERIMENTAL DETAILS

The single-crystal sample of nominal composition $\mathrm{CeRhIn_{4.85}Hg_{0.15}}$ was grown using a standard In self-flux technique and microprobe analysis reveals an actual Hg concentration of only 2.6%.[29] The magnetic transition temperature $T_N \approx 2.7$ K was determined by specific-heat studies (Fig. 1). An anomaly in in-plane magnetic susceptibility is also obvious at T_N. The strong anisotropy in magnetic sus-

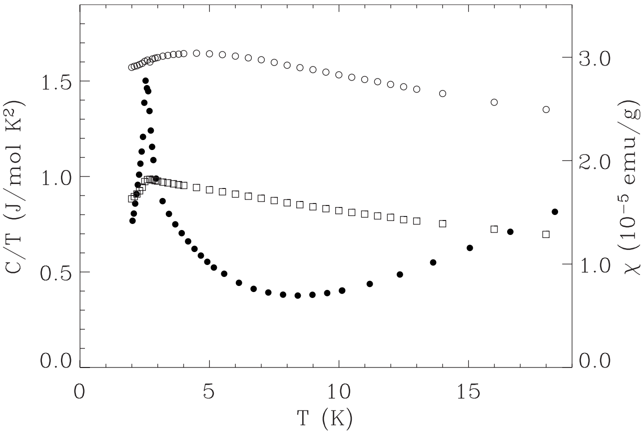

FIG. 1. Specific heat C/T (solid circle) and magnetic susceptibility χ measured with the field applied in the basal plane (square) or perpendicular to the plane (open circles).

ceptibility is a well-known effect of crystal-electric-field splitting of the incomplete Ce f-electron shell. The sample is a thin plate of approximately square shape with an edge along the (100) direction and the plate perpendicular to the (001). It was aligned with the (*hhl*) in the neutron-scattering plane of the triple-axis spectrometer BT7 at the NIST Center for Neutron Research. Both the monochromator and analyzer used the (002) reflection of pyrolytic graphite (PG) to select neutrons of energy 14.7 or 35 meV. The PG filter was used in the neutron beam path to reduce higher-order neutrons. The sample temperature was controlled by a pumped He cryostat. The lattice parameters at 1.6 K are $a=b=4.647$ and $c=7.511$ Å.

III. EXPERIMENTAL RESULTS

Temperature-dependent magnetic Bragg peaks were observed only at the (1/2,1/2,1/2) type of Bragg positions. In Fig. 2, elastic scan along the c axis at 1.6 K is shown. There are no incommensurate magnetic $(1/2,1/2,\pm\delta)$ peaks with either $-\delta \approx 0.3$ or 0.4. Thus Hg-doped CeRhIn$_5$, similar to Cd-doped CeCoIn$_5$, has only the commensurate antiferromagnetic instability. In Fig. 3, the $(1/2,1/2,-1/2)$ magnetic peak is shown at three temperatures. Consistent with a magnetic transition at $T_N \approx 2.7$ K, the intensity decreases at 2.56 K and disappears at 3.0 K.

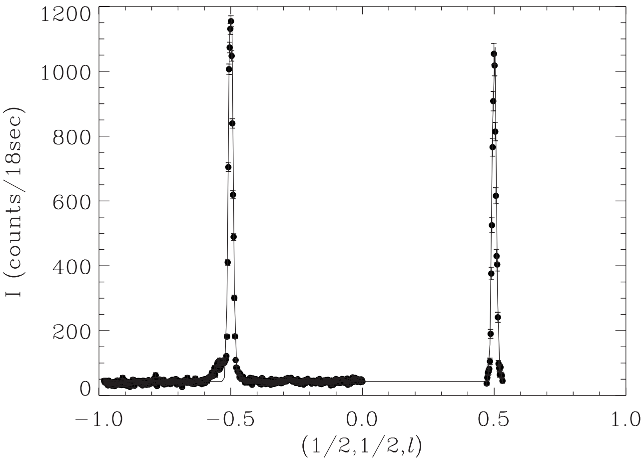

FIG. 2. Elastic scan along the c axis at 1.6 K. Only the commensurate magnetic order with propagating vector (1/2,1/2,1/2) is observed. The neutron energy was 14.7 meV.

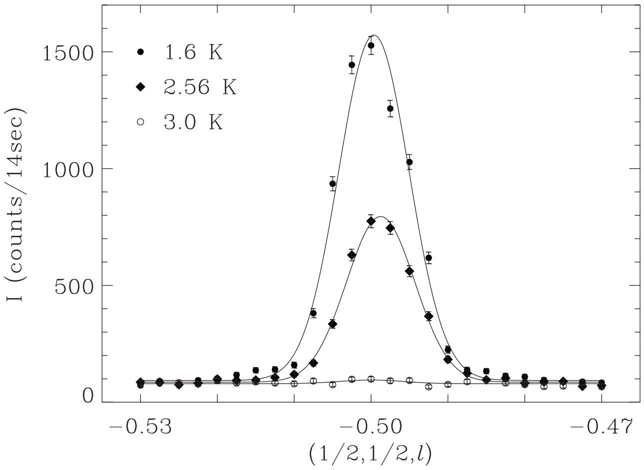

FIG. 3. The temperature dependence of the magnetic peak $(1/2,1/2,-1/2)$. It disappears above $T_N \approx 2.7$ K. The neutron energy was 35 meV.

The different intensity at (1/2,1/2,1/2) and $(1/2,1/2,-1/2)$ in Fig. 2 is attributed to the known neutron absorption effect of the platelike sample shape.[12] Even using neutrons of 35 meV, the neutron intensity absorption length for CeRhIn$_{4.85}$Hg$_{0.15}$ is shorter than the edge of the single-crystal sample. The absorption correction is further complicated by an unknown amount of In flux inclusion in the single crystal. Thus, the uncorrected magnetic Bragg intensity at 1.6 K $\sigma(1/2,1/2,1/2)=7.4(2)$ mb, $\sigma(1/2,1/2,-1/2)=8.8(2)$ mb, and $\sigma(1/2,1/2,-3/2)=7.9(2)$ mb per CeRhIn$_{4.85}$Hg$_{0.15}$ should be taken with these shortcomings in mind. The magnetic Bragg peaks were measured in the triple-axis mode using neutrons of 35 meV to minimize the absorption effect. The "Lorentz factor" is calculated using the expressions given in Refs. 30 and 31. The normalization to absolute units was performed using the (111) structural Bragg peak as the reference, assuming that CeRhIn$_{4.85}$Hg$_{0.15}$ is in the same structure as CeRhIn$_5$.[4] The error bar (one standard deviation) for σ includes only counting statistics. Assuming that the commensurate magnetic structure in CeRhIn$_{4.85}$Hg$_{0.15}$ is the same as that determined for Ce(Rh,Ir)In$_5$ and using Eq. (3) in Ref. 15, the staggered magnetic moment is estimated as $0.45\mu_B$ per CeRhIn$_{4.85}$Hg$_{0.15}$ at 1.6 K.

IV. DISCUSSIONS

So far coexistence of superconductivity and various antiferromagnetic orders has been discovered in Ce(Rh,Ir)In$_5$,[15] Ce(Rh,Co)In$_5$,[18,19] and CeCo(In,Cd)$_5$ (Ref. 28) at ambient pressure and in CeRhIn$_5$ at high pressure.[22] Both the (1/2, 1/2,1/2) and $(1/2,1/2,\sim 0.3)$ modes are present in the Ce(Rh,Ir)In$_5$ and Ce(Rh,Co)In$_5$ alloys. The additional incommensurate mode (1/2,1/2,0.4) shows up in the alloy series Ce(Rh,Co)In_5,[19] and it replaces the (1/2,1/2,0.297) mode in the superconducting phase of $CeRhIn_5$ at high pressure.[22] Spin excitation evidence so far is the resonance at (1/2,1/2,1/2) in $CeCoIn_5$.[20] The (1/2,1/2,0.4) mode in the high-pressure superconducting state of Ce$M$$In_5$ is also highly suggestive. These three magnetic propagating vectors should reflect some near nesting Fermi-surface features along the c axis, since the RKKY interaction is the responsible magnetic interaction in heavy fermion materials.

One exceptional property of the Ce$M$$In_5$ heavy fermion materials is that even in alloy form, they are clean enough to allow quantum oscillation experiments to be successfully conducted.[5,6] This greatly improves the confidence of our understanding of the electronic states at the Fermi surface. Attempts have been made to link the band structure to the magnetic instabilities[23,24] and more refined theories are called for. If for superconducting pressure and compositions the Kondo renormalization process has also reached its Fermi-liquid fixed point at low temperatures[7] and available magnetic modes have been experimentally identified, one would expect a clean explanation of superconductivity in these heavy fermion materials.

ACKNOWLEDGMENTS

Work at LANL was supported by U.S. DOE and at UC Irvine by NSF under Grant No. DMR-053360.

---

[1] J. D. Thompson *et al.*, Physica B **329-333**, 446 (2003).

[2] R. Settai, T. Takeuchi, and Y. Onuki, J. Phys. Soc. Jpn. **76**, 051003 (2007).

[3] Yu. N. Grin *et al.*, Sov. Phys. Crystallogr. **24**, 137 (1979).

[4] E. G. Moshopoulou, Z. Fisk, J. L. Sarrao, and J. D. Thompson, J. Solid State Chem. **158**, 25 (2001).

[5] U. Alver, R. G. Goodrich, N. Harrison, D. W. Hall, E. C. Palm, T. P. Murphy, S. W. Tozer, P. G. Pagliuso, N. O. Moreno, J. L. Sarrao, and Z. Fisk, Phys. Rev. B **64**, 180402(R) (2001).

[6] H. Shishido *et al.*, J. Phys. Soc. Jpn. **71**, 162 (2002).

[7] W. Bao, G. Aeppli, J. W. Lynn, P. G. Pagliuso, J. L. Sarrao, M. F. Hundley, J. D. Thompson, and Z. Fisk, Phys. Rev. B **65**, 100505(R) (2002).

[8] C. Petrovic, R. Movshovich, M. Jaime, P. G. Pagliuso, M. F. Hundley, J. L. Sarrao, Z. Fisk, and J. D. Thompson, Europhys. Lett. **53**, 354 (2001).

[9] C. Petrovic, P. G. Pagliuso, M. F. Hundley, R. Movshovich, J. L. Sarrao, J. D. Thompson, Z. Fisk, and P. Monthoux, J. Phys.: Condens. Matter **13**, L337 (2001).

[10] H. Hegger, C. Petrovic, E. G. Moshopoulou, M. F. Hundley, J. L. Sarrao, Z. Fisk, and J. D. Thompson, Phys. Rev. Lett. **84**, 4986 (2000).

[11] N. J. Curro, P. C. Hammel, P. G. Pagliuso, J. L. Sarrao, J. D. Thompson, and Z. Fisk, Phys. Rev. B **62**, R6100 (2000).

[12] W. Bao, P. G. Pagliuso, J. L. Sarrao, J. D. Thompson, Z. Fisk, J. W. Lynn, and R. W. Erwin, Phys. Rev. B **62**, R14621 (2000); **67**, 099903(E) (2003).

[13] P. G. Pagliuso, C. Petrovic, R. Movshovich, D. Hall, M. F. Hundley, J. L. Sarrao, J. D. Thompson, and Z. Fisk, Phys. Rev. B **64**, 100503(R) (2001).

[14] V. S. Zapf, E. J. Freeman, E. D. Bauer, J. Petricka, C. Sirvent, N. A. Frederick, R. P. Dickey, and M. B. Maple, Phys. Rev. B **65**, 014506 (2001).

[15] A. Llobet, A. D. Christianson, W. Bao, J. S. Gardner, I. P. Swainson, J. W. Lynn, J. M. Mignot, K. Prokes, P. G. Pagliuso, N. O. Moreno, J. L. Sarrao, J. D. Thompson, and A. H. Lacerda, Phys. Rev. Lett. **95**, 217002 (2005).

[16] G. D. Morris *et al.*, Physica B **326**, 390 (2003).

[17] M. Daniel, S. W. Han, C. H. Booth, A. L. Cornelius, P. G. Pagliuso, J. L. Sarrao, and J. D. Thompson, Phys. Rev. B **71**, 054417 (2005).

[18] S. Ohira-Kawamura, H. Shishido, A. Yoshida, R. Okazaki, H. Kawano-Furukawa, T. Shibauchi, H. Harima, and Y. Matsuda, Phys. Rev. B **76**, 132507 (2007).

[19] M. Yokoyama, N. Oyama, H. Amitsuka, S. Oinuma, I. Kawasaki, K. Tenya, M. Matsuura, K. Hirota, and T. J. Sato, Phys. Rev. B **77**, 224501 (2008).

[20] C. Stock, C. Broholm, J. Hudis, H. J. Kang, and C. Petrovic, Phys. Rev. Lett. **100**, 087001 (2008).

[21] S. Kawasaki, T. Mito, G.-q. Zheng, C. Thessieu, Y. Kawasaki, K. Ishida, Y. Kitaoka, T. Muramatsu, T. C. Kobayashi, D. Aoki, S. Araki, Y. Haga, R. Settai, and Y. Onuki, Phys. Rev. B **65**, 020504(R) (2001).

[22] S. Raymond, G. Knebel, D. Aoki, and J. Flouquet, Phys. Rev. B **77**, 172502 (2008).

[23] J. L. Wang, Z. Zeng, and H. Q. Lin, J. Appl. Phys. **99**, 08M505 (2006).

[24] S. Elgazzar, I. Opahle, R. Hayn, and P. M. Oppeneer, Phys. Rev. B **69**, 214510 (2004).

[25] J. Paglione, M. A. Tanatar, D. G. Hawthorn, R. W. Hill, F. Ronning, M. Sutherland, L. Taillefer, C. Petrovic, and P. C. Canfield, Phys. Rev. Lett. **94**, 216602 (2005).

[26] I. Eremin, G. Zwicknagl, P. Thalmeier, and P. Fulde, Phys. Rev. Lett. **101**, 187001 (2008).

[27] L. D. Pham, T. Park, S. Maquilon, J. D. Thompson, and Z. Fisk, Phys. Rev. Lett. **97**, 056404 (2006).

[28] M. Nicklas, O. Stockert, T. Park, K. Habicht, K. Kiefer, L. D. Pham, J. D. Thompson, Z. Fisk, and F. Steglich, Phys. Rev. B **76**, 052401 (2007).

[29] E. D. Bauer, F. Ronning, S. Maquilon, L. D. Pham, J. D. Thompson, and Z. Fisk, Physica B **403**, 1135 (2008).

[30] M. J. Cooper and R. Nathans, Acta Crystallogr., Sect. A: Cryst. Phys., Diffr., Theor. Gen. Crystallogr. **24**, 619 (1968).

[31] S. A. Werner and R. Pynn, J. Appl. Phys. **42**, 4736 (1971).