

UC Irvine

UC Irvine Previously Published Works

Title

Magnetic structure of Cd-doped CeCoIn₅

Permalink

<https://escholarship.org/uc/item/2x87g3dn>

Journal

Physical Review B, 76(5)

ISSN

2469-9950

Authors

Nicklas, M
Stockert, O
Park, Tuson
[et al.](#)

Publication Date

2007-08-01

DOI

10.1103/physrevb.76.052401

Copyright Information

This work is made available under the terms of a Creative Commons Attribution License, available at <https://creativecommons.org/licenses/by/4.0/>

Peer reviewed

Magnetic structure of Cd-doped CeCoIn₅

M. Nicklas,^{1,*} O. Stockert,¹ Tuson Park,² K. Habicht,³ K. Kiefer,³
L. D. Pham,⁴ J. D. Thompson,² Z. Fisk,⁵ and F. Steglich¹

¹Max Planck Institute for Chemical Physics of Solids, Nöthnitzer Str. 40, 01187 Dresden, Germany

²Los Alamos National Laboratory, Los Alamos, NM 87545, USA

³Hahn-Meitner-Institut, Glienicke Str. 100, 14109 Berlin, Germany

⁴University of California, Davis, CA 95616, USA

⁵University of California, Irvine, CA 92697, USA

(Dated: February 6, 2008)

The heavy fermion superconductor CeCoIn₅ is believed to be close to a magnetic instability, but no static magnetic order has been found. Cadmium doping on the In-site shifts the balance between superconductivity and antiferromagnetism to the latter with an extended concentration range where both types of order coexist at low temperatures. We investigated the magnetic structure of nominally 10% Cd-doped CeCoIn₅, being antiferromagnetically ordered below $T_N \approx 3$ K and superconducting below $T_c \approx 1.3$ K, by elastic neutron scattering. Magnetic intensity was observed only at the ordering wave vector $Q_{AF} = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ commensurate with the crystal lattice. Upon entering the superconducting state the magnetic intensity seems to change only little. The commensurate magnetic ordering in CeCo(In_{1-x}Cd_x)₅ is in contrast to the incommensurate antiferromagnetic ordering observed in the closely related compound CeRhIn₅. Our results give new insights in the interplay between superconductivity and magnetism in the family of CeTIn₅ ($T = \text{Co, Rh, and Ir}$) based compounds.

PACS numbers: 74.70.Tx, 75.25.+z, 71.27.+a, 75.30.Mb

In conventional superconductors only small amounts of magnetic impurities destroy the superconducting state, while in the heavy-fermion systems, like CeCu₂Si₂,¹ the presence of a dense lattice of magnetic rare-earth atoms is needed to generate unconventional superconductivity (SC). The discovery of SC and antiferromagnetism in the family of CeTIn₅ ($T = \text{Co, Rh, or Ir}$) compounds, forming in the tetragonal HoCoGa₅ structure, with CeCoIn₅² and CeIrIn₅³ displaying a superconducting ground state and CeRhIn₅⁴ being antiferromagnetically ordered, offers an ideal opportunity to study the peculiar interplay of these two ground states. Though no sign of static magnetic order has been found in CeCoIn₅, pronounced non-Fermi-liquid (NFL) behavior in the normal state in thermodynamic and transport properties is observed at low temperatures.^{2,5,6,7} Commonly, NFL behavior occurs in the close proximity to a quantum critical point (QCP). The search for a magnetic phase in CeCoIn₅, which could give rise to such a QCP by a continuous suppression of the transition temperature via, e.g., applying external pressure or chemical doping has not been successful until recently. Studies of Cd doping on the In-site in CeCoIn₅ revealed that only a few percent of Cd doping indeed lead to the development of antiferromagnetic (AF) order.⁸ Also, reports on the existence of field-induced magnetism in the Abrikosov vortex state in CeCoIn₅ underline its proximity to magnetism.^{9,10}

Cd-doping continuously suppresses the superconducting transition temperature of CeCoIn₅ ($T_c = 2.3$ K), as shown in the phase diagram depicted in Fig. 1. For a nominal Cd-concentration in excess of 7.5%, AF order has been observed, coexisting with SC at low temperatures.⁸ With further increasing Cd-concentration

the Néel temperature, T_N , increases monotonically and no SC is found above 12.5% Cd anymore. In this report we present neutron scattering data on a CeCo(In_{1-x}Cd_x)₅ sample with $x = 0.1$, where x represents the nominal concentration, situated in the concentration range where SC and antiferromagnetism coexist at low temperatures.

Single crystals of CeCo(In_{0.9}Cd_{0.1})₅ were grown using a standard In-flux technique with a nominal concentra-

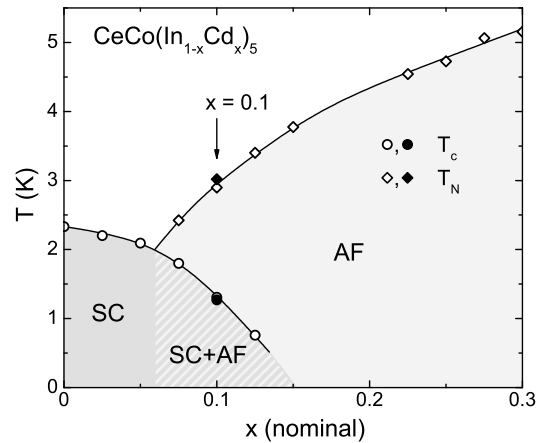


FIG. 1: Doping - temperature ($x - T$) phase diagram of CeCo(In_{1-x}Cd_x)₅, where x is the nominal Cd concentration. Diamonds indicate the Néel temperature, T_N , and circles the superconducting transition temperature, T_c , determined by specific heat measurements. The arrow indicates the concentration investigated in this work, with transition temperatures as indicated by the solid symbols. Open symbols represent data taken from Ref. 8.

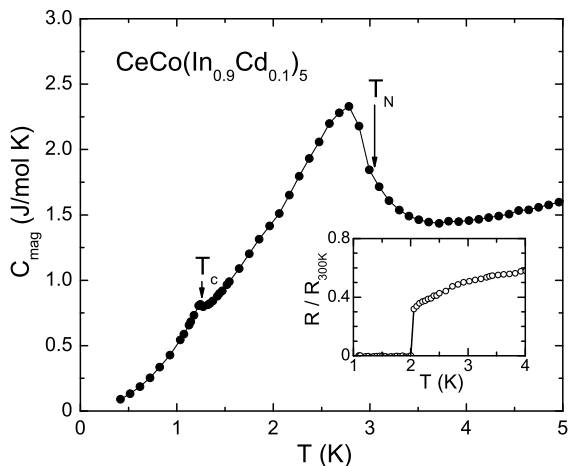


FIG. 2: Temperature dependence of the magnetic contribution to the specific heat, C_{mag} , for $\text{CeCo}(\text{In}_{0.9}\text{Cd}_{0.1})_5$. To obtain C_{mag} , the contribution of the non-magnetic reference compound LaCoIn_5 ¹¹ was subtracted from the specific heat of $\text{CeCo}(\text{In}_{0.9}\text{Cd}_{0.1})_5$. The Néel temperature, T_N , and the superconducting transition temperature, T_c , are indicated by arrows. Inset shows the electrical resistivity measured on the same sample.

tion of 10% Cd in the indium flux. X-ray diffraction confirmed that the samples crystallize in the tetragonal HoCoGa_5 type of crystal structure with lattice parameters $a = 4.6122(4) \text{ \AA}$ and $c = 7.5483(9) \text{ \AA}$. Microprobe analysis revealed a uniform distribution of the Cd throughout the sample and an actual concentration of only about one percent Cd in the sample, i.e. $\sim 10\%$ of the nominal concentration in the flux. For an easier comparison with literature, we will refer in this paper, too, to the nominal concentration as used in Ref.⁸. The magnetic order was investigated by elastic neutron scattering. The experiments were performed on the cold triple-axis spectrometer V2 at the BER-II reactor of the Hahn-Meitner-Institut in Berlin, using a wavelength of the incoming neutrons of $\lambda = 2.73 \text{ \AA}$, corresponding to a neutron energy $E = 11 \text{ meV}$. Pyrolytic graphite, PG(002), was used as monochromator and analyzer. The horizontal collimation before the monochromator was given by the ^{58}Ni guide and was $60'$ before the sample, before the analyzer and in front of the detector. A tunable PG-filter in the scattered beam reduced the contamination of second-order neutrons. The platelet-like sample of $\text{CeCo}(\text{In}_{0.9}\text{Cd}_{0.1})_5$ with a mass $m \approx 10 \text{ mg}$ and dimensions $2 \times 2 \times 0.3 \text{ mm}^3$, 0.3 mm being the thickness along the tetragonal c -axis, was mounted on a copper pin attached to the mixing chamber of a dilution refrigerator. Data were recorded at temperatures between $T = 60 \text{ mK}$ and 3.2 K along principal and high symmetry directions in the $(h h \ell)$ scattering plane. Analyzing the scattered neutrons, i.e. performing elastic scattering, considerably improves the signal-to-background ratio in comparison to (standard) diffraction. This holds especially true in our case since the sample and thus

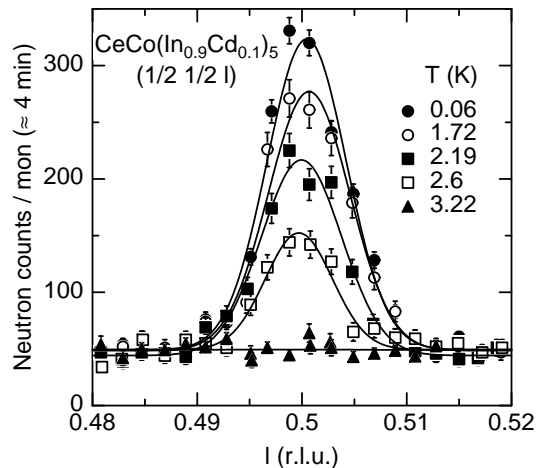


FIG. 3: Elastic scans along $[001]$ across $Q = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ in $\text{CeCo}(\text{In}_{0.9}\text{Cd}_{0.1})_5$ at different temperatures. No magnetic intensity can be resolved at $T = 3.22 \text{ K}$ ($> T_N$, as determined by specific heat). The solid lines are Gaussian fits to the data.

the signal was quite small. In addition to these neutron scattering studies, we conducted heat capacity and electrical resistivity measurements in a Physical Properties Measurement System (Quantum Design) in the temperature range $350 \text{ mK} \leq T \leq 10 \text{ K}$. In order to correlate the results of the microscopic and the macroscopic studies, all experiments were performed on the same single-crystalline sample.

The specific heat data taken on this sample (see Fig. 2) show two anomalies, one at $T_N = 3.02 \text{ K}$ corresponding to the transition to the antiferromagnetically ordered state and the second one at $T_c = 1.27 \text{ K}$ indicating the phase transition to the superconducting state, in good agreement with literature.⁸ Despite this T_c value, zero resistance is already observed below 2 K . Similar deviations between thermodynamic and electrical transport results are also known for CeIrIn_5 ³ as well as for Ir-rich $\text{CeRh}_{1-x}\text{Ir}_x\text{In}_5$.¹² The specific heat exhibits a mean-field like jump $\Delta C = 1.46 \text{ J/mol K}$ at T_N . Below the AF transition only 30% of the magnetic entropy ($R \ln 2$) is released suggesting a substantially Kondo-compensated ordered moment.

To determine the magnetic structure of $\text{CeCo}(\text{In}_{0.9}\text{Cd}_{0.1})_5$ we carried out elastic neutron scattering experiments and performed elastic scans along $(\frac{1}{2}, \frac{1}{2}, \ell)$ at 60 mK . Magnetic intensity was detected at $Q = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$, cf. Fig. 3, and at symmetry equivalent positions like $Q = (\frac{3}{2}, \frac{3}{2}, \frac{1}{2})$ and $Q = (\frac{3}{2}, \frac{3}{2}, \frac{3}{2})$. Due to the small sample size long counting times (several minutes per point) were required. The magnetic peak at $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ monotonically decreases upon heating the sample and vanishes at T_N . Scans along other high symmetry directions revealed no additional intensity, e.g. no intensity was found at $(1, 1, \frac{1}{2})$, $(0, 0, \frac{1}{2})$, $(\frac{3}{2}, \frac{3}{2}, 0)$, or $(\frac{3}{2}, \frac{3}{2}, 1)$. In particular, no magnetic intensity could

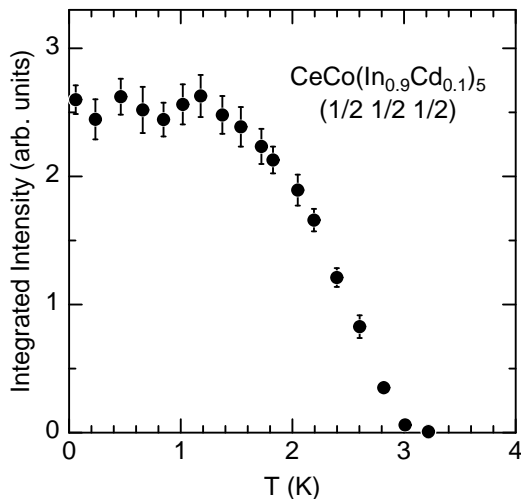


FIG. 4: Temperature dependence of the integrated magnetic intensity as obtained by Gaussian fits to the scans across $Q = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$.

be detected at $(\frac{1}{2}, \frac{1}{2}, \delta)$, $\delta \approx 0.3$, an incommensurate position where CeRhIn_5 , a closely related member of the CeTIn_5 ($T = \text{Co, Rh, or Ir}$) family, displays magnetic superstructure peaks.¹³ Hence, the commensurate magnetic order in $\text{CeCo}(\text{In}_{0.9}\text{Cd}_{0.1})_5$ with a propagation vector $Q_{\text{AF}} = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ is in marked contrast to the incommensurate order observed in other compounds of the CeTIn_5 family as well as in CeCu_2Si_2 .¹⁴ It is speculated that rather small changes in the Fermi surface are responsible for this behavior.

The integrated magnetic intensity obtained from Gaussian fits to the data (cf. Fig. 3) is depicted in Fig. 4. The magnetic intensity starts to build up below $T_N \approx 3$ K, in agreement with T_N determined from specific heat, increases continuously and eventually saturates below $\approx T_c$ ($= 1.27$ K), potentially indicating missing magnetic intensity in the superconducting state. No distinct anomaly is resolved at the superconducting transition, in particular, the magnetic intensity does not vanish below T_c . Our neutron scattering results clearly demonstrate the existence of AF order well below T_c , down to lowest temperatures ($T = 60$ mK). Nuclear magnetic quadrupole resonance (NQR) experiments give supplementary evidence for the microscopic coexistence of SC and antiferromagnetism, and the estimated moment of approximately $0.7\mu_B$ is in line with the observed magnetic intensity,¹⁵ however, we cannot calculate the magnetic moment precisely.

As shown by Pham et al.,⁸ applying pressure reverses the effect of Cd doping. A generalized pressure-temperature ($p-T$) phase diagram for CeRhIn_5 and the doping series $\text{CeCo}(\text{In}_{1-x}\text{Cd}_x)_5$ (including pure CeCoIn_5)^{16,17} describing the pressure dependence of

$T_N(p)$ and $T_c(p)$ suggests the same underlying physics. According to this $p-T$ phase diagram, $\text{CeCo}(\text{In}_{0.9}\text{Cd}_{0.1})_5$ can be considered to correspond to CeRhIn_5 at $p = 1.6$ GPa. As already mentioned, at atmospheric pressure CeRhIn_5 orders magnetically below $T_N = 3.8$ K with an incommensurate ordering wave vector $(\frac{1}{2}, \frac{1}{2}, \delta)$, $\delta = 0.297$. Pressure dependent neutron scattering experiments on CeRhIn_5 at 1.8 K show that the incommensurability, δ , and the ordered moment are changing only slightly.¹⁸ In marked contrast to the simple expectation inferred from our present results, *no* magnetic intensity at 1.6 GPa is reported at $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ in CeRhIn_5 corresponding to $\text{CeCo}(\text{In}_{0.9}\text{Cd}_{0.1})_5$ at ambient pressure.

In CeRhIn_5 AF order can be suppressed by either Co or Ir substitution on the Rh-site. With increasing substitution level a superconducting phase, first coexisting with antiferromagnetism, develops until antiferromagnetism becomes suppressed and only SC survives.^{12,19} The corresponding phase diagram is similar to the one of $\text{CeCo}(\text{In}_{1-x}\text{Cd}_x)_5$ with CeCoIn_5 being situated on the purely SC side, cf. Fig. 1. In Rh-rich $\text{CeRh}_{1-y}\text{Ir}_y\text{In}_5$ pressure studies even reveal the same generic $p-T$ phase diagram found for $\text{CeCo}(\text{In}_{1-x}\text{Cd}_x)_5$ ⁸ suggesting a close relationship.²⁰ In striking contrast to the neutron scattering results obtained for CeRhIn_5 under pressure, a commensurate magnetic structure with ordering wave vector $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ develops in the doping series $\text{CeRh}_{1-y}\text{Ir}_y\text{In}_5$ and $\text{CeRh}_{1-z}\text{Co}_z\text{In}_5$ at low temperatures, while the same incommensurate ordering wave vector present in the pure system is still observed below T_N .^{21,22} We speculate that the appearance of the commensurate magnetic ordering wave vector $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ is related to SC. Perhaps this commensurate magnetic ordering has so far been overlooked in superconducting CeRhIn_5 under pressure. Improved experiments with a better signal-to-background ratio are called for to answer this question.

In summary, we carried out elastic neutron scattering experiments on $\text{CeCo}(\text{In}_{0.9}\text{Cd}_{0.1})_5$. At low temperatures we found magnetic intensity at the commensurate wave vector $Q_{\text{AF}} = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$. The magnetic intensity is building up below T_N , with T_N being in good agreement with specific heat data. No indication for additional intensity was observed at incommensurate positions where CeRhIn_5 , the related AF member in the CeTIn_5 family, orders. At low temperatures magnetic order is coexisting with SC. A saturation of the magnetic intensity below T_c possibly reveals missing magnetic intensity in the superconducting state.

We acknowledge useful discussions with P. Thalmeier and G. Knebel. We would like to thank A. D. Bianchi for the X-ray diffraction and M. Meissner for his assistance with the low temperature equipment at HMI. Work at Los Alamos was performed under the auspices of the U.S. DOE/Office of Science. Work at UC Davis and UC Irvine has been supported by NSF Grant No. DMR 053360.

-
- * Electronic address: nicklas@cpfs.mpg.de
- ¹ F. Steglich, J. Aarts, C. D. Bredl, W. Lieke, D. Meschede, W. Franz, and H. Schäfer, *Phys. Rev. Lett.* **43**, 1892 (1979).
 - ² C. Petrovic, P. G. Pagliuso, M. F. Hundley, R. Movshovich, J. L. Sarrao, J. D. Thompson, Z. Fisk, and P. Monthoux, *J. Phys. Cond. Mat.* **13**, L337 (2001).
 - ³ 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).
 - ⁴ 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).
 - ⁵ Y. Kohori, Y. Yamato, Y. Iwamoto, T. Kohara, E. D. Bauer, M. B. Maple, and J. L. Sarrao, *Phys. Rev. B* **64**, 134526 (2001).
 - ⁶ A. Bianchi, R. Movshovich, I. Vekhter, P.G. Pagliuso, and J. L. Sarrao, *Phys. Rev. Lett.* **91**, 257001 (2003).
 - ⁷ J. Paglione, M. A. Tanatar, D.G. Hawthorn, E. Boaknin, R.W. Hill, F. Ronning, M. Sutherland, and L. Taillefer, *Phys. Rev. Lett.* **91**, 246405 (2003).
 - ⁸ L. D. Pham, Tuson Park, S. Maquilon, J. D. Thompson, and Z. Fisk, *Phys. Rev. Lett.* **97**, 056404 (2006).
 - ⁹ V. F. Mitrović, M. Horvatić, C. Berthier, G. Knebel, G. Lapertot, and J. Flouquet, *Phys. Rev. Lett.* **97**, 117002 (2006).
 - ¹⁰ B.-L. Young, R. R. Urbano, N. J. Curro, J. D. Thompson, J. L. Sarrao, A. B. Vorontsov, and M. J. Graf, *Phys. Rev. Lett.* **98**, 036402 (2007).
 - ¹¹ M. F. Hundley, private communication.
 - ¹² 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).
 - ¹³ 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); W. Bao, P. G. Pagliuso, J. L. Sarrao, J. D. Thompson, Z. Fisk, J. W. Lynn, and R. W. Erwin, *Phys. Rev. B* **63**, 219901(E) (2001).
 - ¹⁴ O. Stockert, E. Faulhaber, G. Zwicknagl, N. Stüßer, H. S. Jeevan, M. Deppe, R. Borth, R. Küchler, M. Loewenhaupt, C. Geibel, and F. Steglich, *Phys. Rev. Lett.* **92**, 136401 (2004).
 - ¹⁵ R. R. Urbano, B.-L. Young, N. J. Curro, J. D. Thompson, L. D. Pham, and Z. Fisk, *cond-mat/0702008* (2007).
 - ¹⁶ M. Nicklas, R. Borth, E. Lengyel, P. G. Pagliuso, J. L. Sarrao, V. A. Sidorov, G. Sparn, F. Steglich, and J. D. Thompson, *J. Phys. Condens. Matter* **13**, L905 (2001).
 - ¹⁷ V. A. Sidorov, M. Nicklas, P.G. Pagliuso, J. L. Sarrao, Y. Bang, A.V. Balatsky, and J. D. Thompson, *Phys. Rev. Lett.* **89**, 157004 (2002).
 - ¹⁸ A. Llobet, J. S. Gardner, E. G. Moshopoulou, J.-M. Mignot, M. Nicklas, W. Bao, N. O. Moreno, P. G. Pagliuso, I. N. Goncharenko, J. L. Sarrao, and J. D. Thompson, *Phys. Rev. B* **69**, 024403 (2004).
 - ¹⁹ 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).
 - ²⁰ M. Nicklas, V. A. Sidorov, H. A. Borges, P. G. Pagliuso, J. L. Sarrao, and J. D. Thompson, *Phys. Rev. B*, **70**, 020505(R) (2004).
 - ²¹ A. D. Christianson, A. Llobet, Wei 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).
 - ²² M. Yokoyama, H. Amitsuka, K. Matsuda, A. Gawase, N. Oyama, I. Kawasaki, K. Tenya, and H. Yoshizawa, *J. Phys. Soc. Jpn.* **75**, 103703 (2006).