Magnetic structure of Cd-doped CeCoIn$_5$

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The heavy-fermion superconductor CeCoIn$_5$ 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 CeCoIn$_5$, 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 $Q_{AF} = \left(\frac{1}{2}, \frac{1}{2}, \frac{1}{2}\right)$ 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 CeRhIn$_5$. Our results give insights into the interplay between superconductivity and magnetism in the family of CeTIn$_5$ (T=Co, Rh, and Ir) based compounds.

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In conventional superconductors, only small amounts of magnetic impurities destroy the superconducting state, while in the heavy-fermion systems, such as CeCu$_2$Si$_2$,[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 CeTIn$_5$ (T=Co, Rh, or Ir) compounds, forming in the tetragonal HoCoGa$_5$ structure, with CeCoIn$_5$ (Ref. 2) and CeIrIn$_5$ (Ref. 3) displaying a superconducting ground state and CeRhIn$_5$ (Ref. 4) 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 CeCoIn$_5$, pronounced non-Fermi-liquid (NFL) behavior in the normal state in thermodynamic and transport properties is observed at low temperatures.[2,5–7] Commonly, NFL behavior occurs in close proximity to a quantum critical point (QCP). The search for a magnetic phase in CeCoIn$_5$, 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 CeCoIn$_5$ 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 CeCoIn$_5$ underline its proximity to magnetism.[9,10]

Cd doping continuously suppresses the superconducting transition temperature of CeCoIn$_5$ (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 Brief 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 concentration 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) Å and c=7.5483(9) Å. Microprobe analysis revealed a uniform distribution of the Cd throughout the sample and an actual concentration of only about 1% Cd in the sample, i.e., ~10% of the nominal concentration in the flux. For an easier comparison with literature, we will refer in this Brief Report, 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 spectrom-
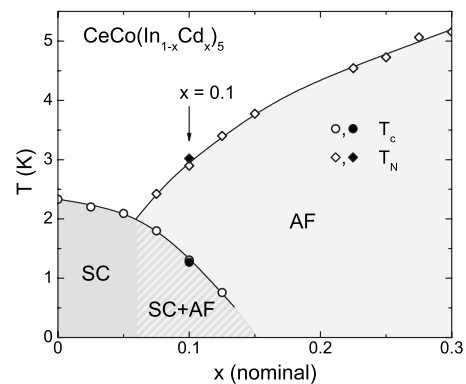

FIG. 1. Doping-temperature (x-T) phase diagram of CeCo(In$_{1-x}$Cd$_x$)$_5$, where x is the nominal Cd concentration. Diamonds indicate the Néel temperature T_N and circles the superconducting transition temperature T_c determined by specific heat measurements. The arrow indicates the concentration investigated in this work, with transition temperatures as indicated by the solid symbols. Open symbols represent data taken from Ref. 8.
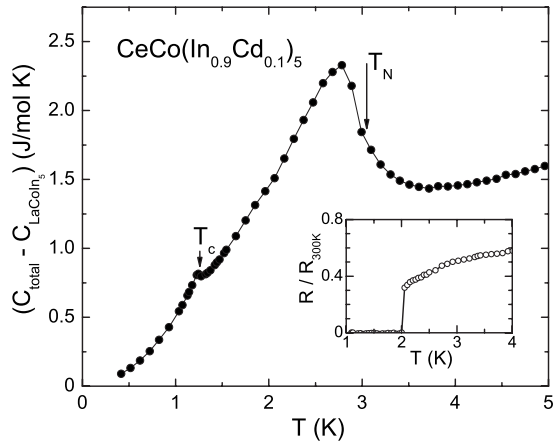
FIG. 2. Temperature dependence the specific heat, C_{total} $-C_{LaCoIn_5}$, for $CeCo(In_{0.9}Cd_{0.1})_5$. The contribution of the nonmagnetic reference compound $LaCoIn_5$ (Ref. 11) was subtracted from the specific heat of $CeCo(In_{0.9}Cd_{0.1})_5$. The Néel temperature T_N and the superconducting transition temperature T_c are indicated by arrows. The inset shows the electrical resistivity measured on the same sample.

eter V2 at the BER-II reactor of the Hahn-Meitner-Institut in Berlin, using a wavelength of the incoming neutrons of λ $=2.73$ Å, corresponding to a neutron energy $E=11$ 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 plateletlike sample of $CeCo(In_{0.9}Cd_{0.1})_5$ with a mass $m \approx 10$ mg and dimensions $2 \times 2 \times 0.3$ 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 $T=60$ mK and 3.2 K along principal and high symmetry directions in the $(hh\ell)$ 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 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 mK $\leqslant T \leqslant 10$ 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 $T_N=3.02$ K corresponding to the transition to the antiferromagnetically ordered state and the other one at $T_c=1.27$ 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 $CeIrIn_5$ (Ref. 3) as well as for Ir-rich $CeRh_{1-x}Ir_xIn_5$.[12] The specific heat exhibits a mean-field-like jump $\Delta C=1.46$ J/mol K at T_N.
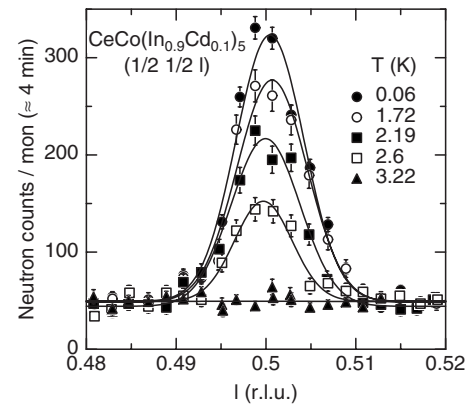

FIG. 3. Elastic scans along [001] across $Q=(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ in $CeCo(In_{0.9}Cd_{0.1})_5$ at different temperatures. No magnetic intensity can be resolved at $T=3.22$ K ($>T_N$, as determined by specific heat). The solid lines are Gaussian fits to the data.

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 $CeCo(In_{0.9}Cd_{0.1})_5$, 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 $Q=(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$, (see Fig. 3) and at symmetry equivalent positions such as $Q=(\frac{3}{2}, \frac{3}{2}, \frac{1}{2})$ 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 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=Co, Rh, or Ir) family, displays magnetic superstructure peaks.[13] Hence, the commensurate magnetic order in $CeCo(In_{0.9}Cd_{0.1})_5$ with a propagation vector $Q_{AF}=(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ 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 (see 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) instead of increasing further as anticipated, 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 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.
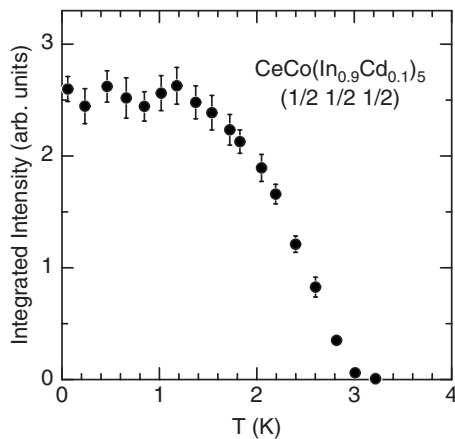
FIG. 4. Temperature dependence of the integrated magnetic intensity as obtained by Gaussian fits to the scans across $Q = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$.

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 CeCo(In$_{1-x}$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, CeCo(In$_{0.9}$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 CeCo(In$_{0.9}$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 that of CeCo(In$_{1-x}$Cd$_x$)$_5$ with CeCoIn$_5$ being situated on the purely SC side (see Fig. 1). In Rh-rich CeRh$_{1-y}$Ir$_y$In$_5$, pressure studies even reveal the same generic p-T phase diagram found for CeCo(In$_{1-x}$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 CeRh$_{1-y}$Ir$_y$In$_5$ (Ref. 21) and CeRh$_{1-z}$Co$_z$In$_5$ (Refs. 22 and 23) at low temperatures, while the same incommensurate ordering wave vector present in the pure system is still observed below T_N. 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 CeCo(In$_{0.9}$Cd$_{0.1}$)$_5$. At low temperatures, we found magnetic intensity at the commensurate wave vector $Q_{AF} = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$. 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. The magnetic intensity saturates below T_c and does not increase further with lowering temperature as expected, possibly revealing missing magnetic intensity in the superconducting state.

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