Basal plane anisotropy in Ce_2RhIn_8

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We report on the measurements of the magnetization and heat capacity of the tetragonal heavy-fermion compound Ce_2RhIn_8, dependent on the direction of the magnetic field applied perpendicular to the *c* axis. A notable anisotropic magnetization component having a fourfold symmetry has been observed in the antiferromagnetic state in low fields. The angular dependence of the heat capacity does not indicate any additional phase which implies that the observed magnetization anisotropy is an intrinsic property of Ce_2RhIn_8. A clear angular dependence of magnetic phase boundaries on the applied magnetic field has been also revealed. A possible interpretation of these results is discussed in the context of previously reported studies of magnetic structures in Ce_2RhIn_8.

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I. INTRODUCTION

The family of compounds of the general chemical formula $Ce_nT_mIn_{3n+2m}$ ($n = 1, 2$; $m = 1, 2$; T = a transition *d* metal) have been of interest of the condensed matter physics community since the discovery of the pressure induced superconductivity (SC) in $CeRhIn_5$ [1] and the ambient pressure SC in $CeCoIn_5$ [2] and $CeIrIn_5$ [3]. The antiferromagnetic (AF) $Ce_nT_mIn_{3n+2m}$ compounds exhibit a plethora of interesting phenomena in the vicinity of the magnetic quantum critical point, including non-Fermi liquid behavior, coexistence of magnetism and superconductivity, and unconventional superconductivity [4,5].

The $Ce_nT_mIn_{3n+2m}$ compounds adopt the corresponding tetragonal $Ho_nCo_mGa_{3n+2m}$-type crystal structures (space group $P4/mmm$, No. 123). This crystallographic arrangement can be viewed as *n* layers of $CeIn_3$ and *m* layers of TIn_2 alternating along the *c* axis. The freedom of changing the magnetic dimensionality by varying the *m* and *n* parameters together with the possibility of selecting the transition metal species provide a vast pitch for tuning the ground-state properties.

One of them, Ce_2RhIn_8, is a heavy-fermion antiferromagnet [4] with Néel temperature $T_N = 2.8$ K and the electronic specific heat coefficient [6] $\gamma = 400$ mJ mol^{-1} K^{-2}. At temperatures below T_N a commensurate AF structure with the propagation vector $k = (0.5, 0.5, 0)$ and an ordered Ce magnetic moments of 0.55 μ_B pointing 38° out from the tetragonal *c* axis has been observed. This intermediate phase exists at temperatures down [7] to $T^* = 1.65$ K where it transforms to an incommensurate AF ground-state structure [8]. An *H*-*T* magnetic phase diagram of Ce_2RhIn_8 in the magnetic field applied along the *a* axis was reported by Cornelius *et al.* [9]. The proposed [9] magnetic structures in region I (low field, low temperatures) and region II (intermediate phase at finite magnetic field in the vicinity of the ordering temperatures) are spin-density waves (incommensurate with the lattice) where region II has a larger magnetic moment on each Ce atom. Region III (low temperatures, high field) corresponds to a spin-density wave that is commensurate with the lattice. However, no details of the ground-state magnetic structure have been reported so far.

In this paper we present an experimental study of angular dependencies of the magnetization and heat capacity of Ce_2RhIn_8 single crystals on the direction of the magnetic field applied perpendicular the *c* axis. A magnetization anisotropy of a fourfold magnetic symmetry within the tetragonal basal plane has been observed in a limited region of the *H*-*T* phase space.

II. EXPERIMENT

The Ce_2RhIn_8 single crystals have been grown from indium flux using high purity constituent elements Ce (99.9%,

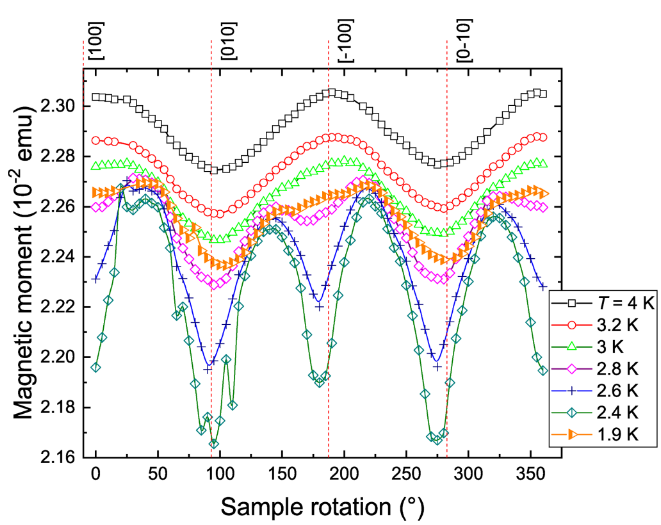

FIG. 1. Rotation scans of the Ce_2RhIn_8 magnetization measured at several temperatures between 4 and 1.9 K in the field of 4 T. The transition between the twofold and fourfold rotation symmetries commences at $T \sim 3$ K. The vertical dashed lines note the respective orientations in the tetragonal lattice.

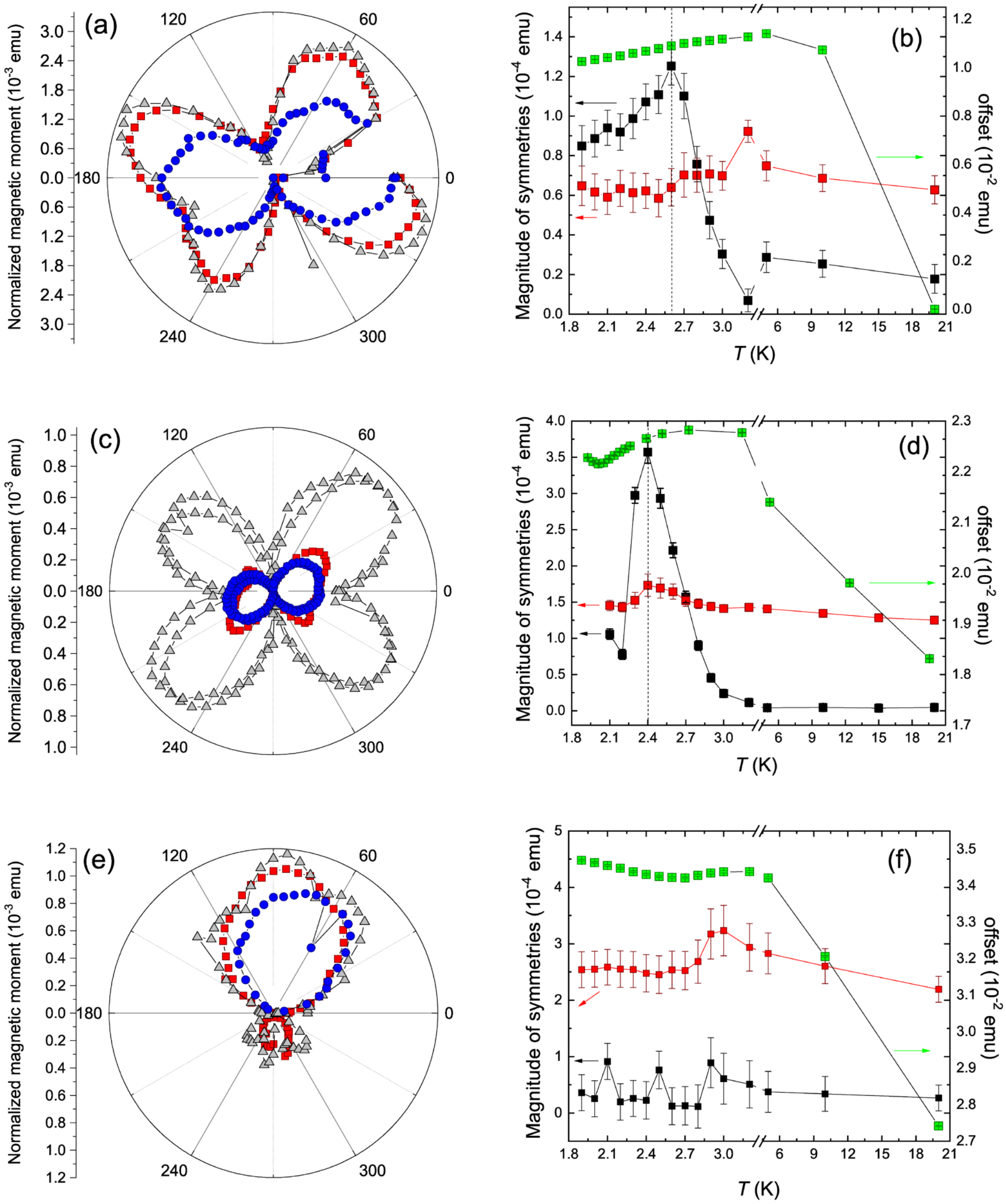

FIG. 2. Polar plots of the magnetic moment of Ce_2RhIn_8 measured at temperatures T = 2 K (red square), 2.5 K (gray triangle), and 3 K (blue circle), and in external magnetic fields $\mu_0 H = 2$ T (a) and (b), 4 T (c) and (d), and 6 T (e) and (f), applied perpendicular to the c axis (left-hand panels). Evolution of the corresponding fourfold (full black square) B, twofold (full red square) C, and isotropic component of magnetization (full green square) A parameters in Eq. (1) (right-hand panels).

additionally purified by solid state electrostransport method), Rh (99.95%), and In (99.9999%) in the ratio Ce: Rh : In = 2 : 1 : 27. The mixture was heated up to a maximum temperature of 1000 °C which was then kept stable for 10 h to homogenize the melt. Subsequently, it was cooled down at a rate of 8 °C h^{-1} to a temperature $T_{decant} = 350$ °C at which the remaining In flux was decanted in a centrifuge. Platelike single crystals of the typical dimension of ~2 × 2 × 1 mm^3 (directions $a \times a \times c$ of the tetragonal lattice) and mass ~30–50 mg were obtained. The single crystals were oriented using the Laue backscattering method. The homogeneity and chemical composition of selected single crystals were studied using a scanning electron microscope (Tescan MIRA I LMH SEM) equipped with an energy dispersive x-ray analyzer (Bruker AXS). The magnetization and heat capacity measurements were performed in the temperature range $1.7 \leqslant T \leqslant 20$ K using MPMS7 and PPMS9 (Quantum Design Inc.), respectively. The magnetic field has been applied in various directions within the basal plane of the tetragonal crystal structure. The heat capacity data were analyzed using the dual-slope method [10]. The angular dependent of heat capacity data were measured using a modified Quantum Design rotator option.

III. RESULTS

The isothermal rotation scans of magnetization have been taken at an external magnetic field of 2, 4, and 6 T, always

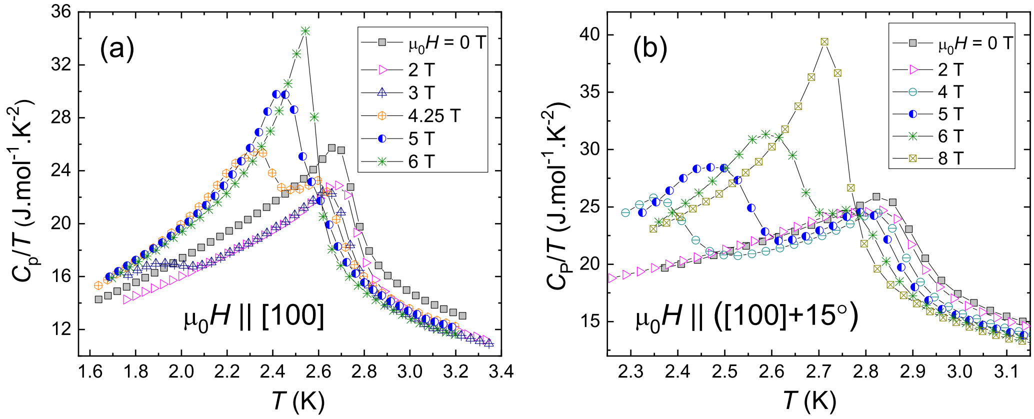

FIG. 3. Temperature dependencies of the specific heat of Ce_2RhIn_8 (C_p/T vs T plots) in selected external magnetic fields applied ∥ a axis (a) and oriented 15° (b) away from the a axis.

after cooling in the field of measurement. The zero (0°) rotator position was adjusted to coincide with the [100] direction. The magnetization was measured starting either at the 0° or 100° rotator position, respectively, providing no observable difference of corresponding magnetization data. As an example we show in Fig. 1 the angular dependencies of magnetic moments of Ce_2RhIn_8 measured at selected temperatures between 1.9 and 4 K in the magnetic field of 4 T.

Two components show up and develop with decreasing temperature. A twofold rotational symmetry is seen at temperatures between 3 and 4 K while the onset of a fourfold symmetry is clearly visible in a temperature interval below 3 K. The fourfold symmetry component is partially suppressed at lower temperatures but it remains traceable down to the lowest temperature of measurements.

In order to capture the symmetry evolution the measured data were fitted using a phenomenological function

$$f(\theta) = A + B\cos[4(\theta - D)] + C\cos[2(\theta - E)], \quad (1)$$

where A represents the isotropic component of magnetization, and B and C are the magnitudes of fourfold and twofold symmetry components, respectively. The parameters D and E represent the temperature and field independent phase factors for a given symmetry. Figure 2 represents a comprehensive summary of the angular measurements performed in fields $\mu_0 H = 2$, 4, and 6 T, respectively, applied within the basal plane and rotated around the [001] axis. It is seen that the 6 T scans do not show any significant fourfold contribution, contrary to an angular dependence observed in 4 T. On the other hand, the 2 T rotational scans show the fourfold symmetry component emerging in coincidence with the onset of magnetic ordering and varying only slightly with decreasing temperature.

In order to exclude the possibility of magnetic phase boundary crossing and to shed light on this peculiar behavior we decided to further map the basal plane magnetic phase diagram of Ce_2RhIn_8 by means of measurements of angular dependencies of the heat capacity on the direction of the external magnetic field ($\mu_0 H$ ranging from 0 to 9 T) rotating in the basal plane from $\theta = 0°$ to 45°. The $\theta = 0°$ setup corresponds to a field parallel to the [100] direction in the tetragonal lattice. For clear visual representation we discuss $\theta = 0°$, 15°, 30°, and 45° rotation steps corresponding to the span from the [100] to the [110] direction. The representative C_p/T vs T data sets for $\theta = 0°$ and $\theta = 15°$ are shown in Fig. 3. A clear λ-type anomaly at $T_N = 2.75$ K is seen in the zero-field heat capacity data and corresponds to the transition from the paramagnetic (PM) to the AF phase. T_N slightly decreases with increasing the magnetic field applied along the a axis and reaches the value of 2.6 K at $\mu_0 H = 5$ T. In the field of 3 T the low-temperature phase transition at $T_M = 1.97$ K has been observed in agreement with previous reports [9]. This phase transition T_M increases with further increasing magnetic field and reaches the value of 2.55 K in $\mu_0 H = 5$ T. The T_N and T_M transitions merge at 6 T. Data measured with the field applied 15° away from the a axis show a much slower field evolution of both T_M and T_N. Consequently, the corresponding phase boundaries merge at a higher magnetic field (7.5 T).

In order to summarize the temperature, magnetic field, and angular dependencies of heat capacity, a three-dimensional magnetic phase diagram has been constructed (Fig. 4). The phase diagram tracks the evolution of the T_N (AF phase), as well as the low temperature phase transition, denoted by T_M.

Starting from $\theta = 0°$ one can see the convergence of the two phases below the critical field of $\mu_0 H = 6$ T. The value of this critical magnetic field is strongly dependent on the orientation within the basal plane. By increasing the degree of rotation one see a strong unfolding of the λ-shaped structure in the phase diagram observed in the $\theta = 0°$ setup. The magnetic field needed for merging T_M and T_N transitions rapidly increases with increasing deflection of the field vector from the [100] direction. It reaches the maximum field used in this study (9 T) at the deflection of ∼20°. Both $T_N(H)$ and $T_M(H)$ become steeper functions of the applied magnetic field $\mu_0 H$, with a further increase of the θ angle. An additional transition line between T_N and T_M becomes more prominent and clearly visible—at approximately 1.5–2.5 T we see a reflection of the additional transition in the temperature dependence of heat capacity (slightly decreasing in temperature while increasing the applied magnetic field). In contrast to the angular dependence of T_M and T_N, this transition remains almost unaffected by the change of the field. We link it to the phase boundary between phases I and II reported in Ref. [9],

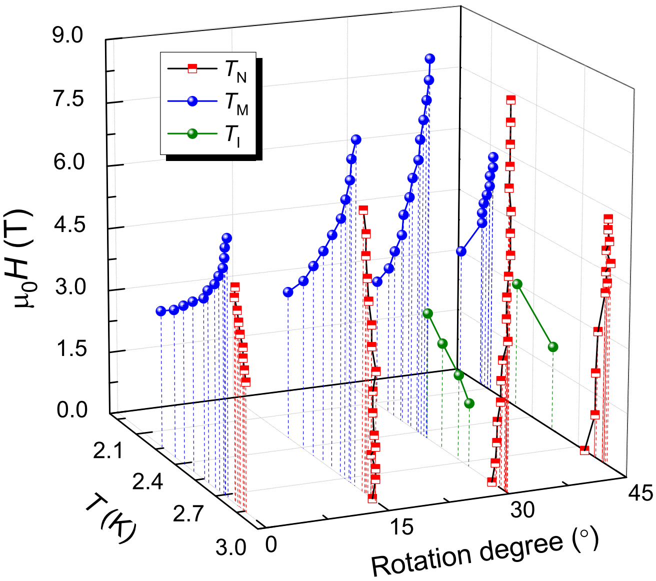

FIG. 4. A three-dimensional magnetic field-temperature-rotation phase diagram of the tetragonal basal plane for Ce_2RhIn_8. The $\theta = 0° \parallel [100]$.

which was indistinguishable for lower deflection angles due to the immediate vicinity of the T_M and T_N transitions and become apparent with their unfolding with increasing θ.

IV. DISCUSSION AND CONCLUSIONS

Low-temperature measurements of the magnetization of Ce_2RhIn_8 were performed in magnetic fields applied in various directions within the basal plane of tetragonal crystal structure. The experimental results document the presence of a low-field fourfold symmetric phase in the AF ordered state.

Our data indicate a complex interplay of exchange interactions in Ce_2RhIn_8. In agreement with the previous studies we have observed predominantly AF ordering, however peculiar features could be identified when the magnetic field is applied in various directions within the basal plane. The major part of magnetization is isotropic and proportional to the applied magnetic field as can be seen from rotational scans (Fig. 1) and directly in magnetization isotherms (not shown). However a notable (up to 4%) anisotropic fourfold-symmetry component of the measured magnetic moment within the basal plane has been revealed. The anisotropic component emerges in coincidence with the onset of the long range AF order and remains to the lowest investigated temperatures. In small fields (2 T) it is almost temperature independent. For any direction of magnetic field applied within the basal plane it is related to the ground-state properties as shown by the heat capacity measurements. In a higher field (4 T) the fourfold symmetry of magnetization is observed in a narrower temperature interval. At high fields (6 T) the anisotropic part of the magnetization is dominated by the polarization and only a remainder of the fourfold symmetry is visible upon inspection of polar diagrams [Fig. 2(c)] in the narrow intermediate temperatures $T_M < T < T_N$.

As the experimental microscopic evidence is limited we can only speculate about the origin of the anisotropy. The best description of the measured neutron diffraction data provided [8] a collinear AF structure of Ce moments tilted away from the c axis (Rh magnetic moments were not considered). The AF domains formed due to the tetragonal symmetry prevented determination of the direction of the Ce magnetic moment projection within the basal plane. The muon spectroscopy [11] indicated the presence of Rh magnetic moments of the order of 0.1 μ_{Ce} forming a cone around the c axis. One may speculate that the observed fourfold symmetry of magnetization within the basal plane is due to the Rh moment component within the basal plane. The magnitude of this component is determined by the competition of applied magnetic field and magnetocrystalline anisotropy associated with tetragonal symmetry of the crystal structure. The Rh moment vanishes when crossing the phase boundaries: Either T_N by increasing temperature in low fields; or T_M by increasing the magnetic field at low temperatures. Consequently, the fourfold symmetry of magnetization disappears. A detailed polarized neutron diffraction study of Ce_2RhIn_8 single crystals in magnetic fields is highly desirable to test our tentative scenario.

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