

Complex magnetic behavior of the Ce sawtooth chains in CeRhSn₂

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(Received 16 January 2026; revised 24 March 2026; accepted 7 May 2026; published 17 July 2026)

Conflicting reports exist on the ground state of the intermetallic compound CeRhSn₂. This can be rooted in the sawtoothlike arrangement of two inequivalent Ce sites in the unit cell, which suggests potential geometric magnetic frustration. To resolve these conflicting reports, we conducted a comprehensive study on high-quality single crystals of CeRhSn₂ by means of magnetization (M), specific heat (C_p/T), and resistivity (ρ). The system exhibits strong magnetic anisotropy, confirming the b axis as the easy magnetic axis. We establish three successive transitions: an antiferromagnetic order at $T_N = 3.65$ K, a first-order ferromagnetic (FM) order at $T_C = 1.7$ K and final transition, at $T = 1.5$ K. The transition temperatures are highly field-directional dependent: in a magnetic field, the lowest transition is immediately suppressed while $\mathbf{H} \parallel b$ rapidly merges T_C and T_N into a single second-order transition. Conversely, $\mathbf{H} \parallel c$ suppresses the FM order and reduces T_N . Additional *ab initio* calculations affirm the FM ground state of CeRhSn₂. The observation of an enhancement of the Sommerfeld coefficient ($\gamma = 76.5$ mJ/mol K²) may arise from geometric frustration, but it is most consistently attributed to weak Kondo hybridization as frustration cannot be conclusively established through our data.

DOI: [10.1103/physrevb.114.045118](https://doi.org/10.1103/physrevb.114.045118)

I. INTRODUCTION

Over the past decades, rare-earth intermetallic compounds containing cerium have commanded considerable scientific interest. These materials exhibit a range of complex phenomena, including valence fluctuations, heavy-fermion behavior, unconventional superconductivity, and various types of magnetic ordering. The origin of these diverse ground states arises from the interaction of $4f$ electrons with the crystal electric field (CEF), as well as the subtle competition between intersite Ruderman-Kittel-Kasuya-Yosida exchange interactions and the on-site Kondo effect. When Ce atoms occupy multiple nonequivalent crystallographic sites, the ground state properties become significantly more complex due to the distinct local environments experienced by the f electrons and the presence of additional sublattice interactions. An earlier example is Ce₂Sn₅ and Ce₃Sn₇ [1]. They are both superstructures of CeSn₃ exhibiting two different cerium sites: Ce1 is

coordinated by 12 Sn atoms, whereas Ce2 is surrounded by 2 Ce and 10 Sn atoms. Despite similar local environments of the Ce atoms, the magnetic ground state of the two compounds differs markedly: Ce₂Sn₅ experiences strong Kondo interaction evidenced by an enhanced value of γ of about 350 mJ/mol K², and orders in a modulated magnetic structure with moments of $1.3\mu_B$ aligned along the a axis. In contrast, Ce₃Sn₇ is an antiferromagnet with reduced moments of only $0.36\mu_B$ parallel to c and a γ close to zero. More recent investigated examples are Ce₃Pd₂₀Si₆ [2,3], CeRuSn [4–7], Ce₅Rh₄Sn₁₀ [8,9], Ce₃Pd₆Sb₅ [10], and, for instance, Ce₃(Pd, Pt)In₁₁ [11–13]. The latter two compounds exhibit antiferromagnetic order coexisting with heavy-fermion superconductivity, and have been discussed within a framework in which the two distinct Ce sublattices play separate roles—one driving magnetic ordering and the other facilitating superconductivity.

Against this backdrop, CeRhSn₂ emerges as an intriguing case. The compound forms in an orthorhombic structure with space group $Cmcm$ (No. 63) and harbors two crystallographically independent cerium sites [14]. Initial experiments classified the material as nonmagnetic, exhibiting only weak Kondo interactions [15]. A later study identified a ferro- or ferrimagnetic transition near 4 K [14]. Subsequent work by Hossain *et al.* [16] uncovered a second magnetic transition around 3 K and, based on low-temperature specific heat fits

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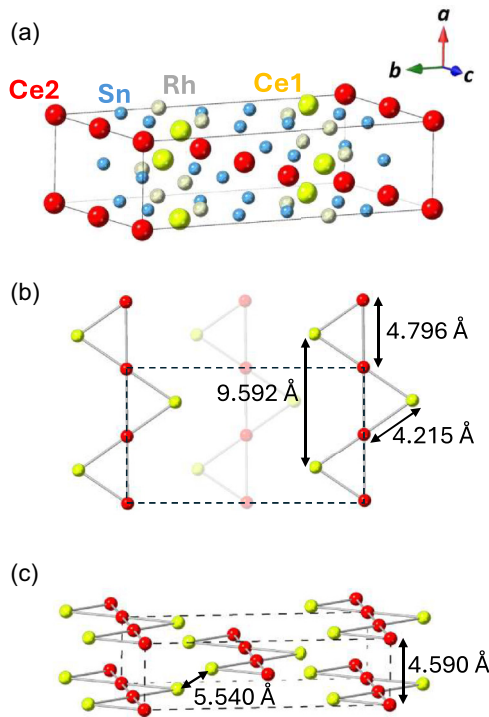


FIG. 1. Picture of the crystal structure of CeRhSn_2 emphasizing (a) the two inequivalent Ce sites. The view from above of the bc plane (b) shows the magnetic lattice of CeRhSn_2 . Ce2 (Wyckoff position $4a$) sites align in linear chains along the c axis and link with Ce1 (Wyckoff position $4c$) sites to create a pattern of alternating sawtooth-shaped isosceles triangles. The shortest Ce2-Ce2 distance (c) is along the a axis.

and magnetoresistance behavior, proposed that the ground state is ferrimagnetic or exhibits canted antiferromagnetic order. The specific heat analysis also yielded a relatively large Sommerfeld coefficient of approximately 300 mJ/mol K^2 , suggesting enhanced quasiparticle masses, characteristic of heavy-fermion systems, or (in part due to) the existence of low-energy magnetic excitations. X-ray photoemission spectroscopy, in conjunction with local (spin) density approximation [L(S)DA] calculations, revealed that both Ce sites adopt a stable Ce^{3+} configuration, each carrying a moment of $\sim 1\mu_B$. Moreover, the LDA calculations yield an expected Sommerfeld coefficient of only 15.6 mJ/mol K^2 [17], significantly lower than the experimentally obtained value.

The peculiar arrangement of the Ce ions in CeRhSn_2 could be responsible for the observed enhancement in γ . As illustrated in Fig. 1, the Ce ions form an equilateral triangular pattern, closely resembling the sawtooth geometry found in, for instance, Mn_2SiX_4 ($X = \text{S, Se}$) [18]. Such a geometry is prone to magnetic frustration, which may contribute to the enhanced γ coefficient. Alternatively, strong Kondo interactions could underlie the observed heavy-fermion-like behavior, placing CeRhSn_2 in proximity to a ferromagnetic quantum critical point (FM QCP). To date, only a limited number of Ce- and Yb-based compounds are known to host a FM QCP [19]. Moreover, it has been proposed that in systems with multiple magnetic sublattices, each sublattice may be characterized by a distinct Kondo scale, potentially giving

rise to a complex phase diagram with multiple quantum phase transitions [20].

Together, these considerations identify CeRhSn_2 as a particularly promising system for further investigation. As a first step, it is essential to resolve the discrepancies in the reported magnetic ordering and to establish a precise characterization of its magnetic properties. In this work, we report on the synthesis of single crystals of CeRhSn_2 and a comprehensive study of their low-temperature properties. The magnetic response was examined using dc magnetization, specific heat, and electrical resistivity measurements. Complementary *ab initio* calculations were also performed to independently determine the ground state.

II. METHODOLOGY

A. Experimental details

Single crystals of CeRhSn_2 were synthesized in India (sample #1) and subsequently reproduced in Prague (crystal cut in samples #2–#5) using the same growth procedure, in order to verify or extend selected experiments. Appropriate amounts of high-purity elements (Ce: 99.95% and solid state electrotransport purified [21,22]; Rh: 99.95%; and Sn: 99.995%) were melted together in an arc-melting furnace under argon atmosphere. The obtained precursor was turned over and remelted several times to achieve a better homogeneity. Following this, single crystals were grown from the molten precursor using the Czochralski method under an Ar atmosphere. Crystal #1 was prepared in a tetra-arc furnace, while the second crystal (samples #2–#5) was grown using a tri-arc furnace. In both cases, a tungsten rod was used as a seed rod, and a pulling rate of 12 mm/h was deployed.

Subsequently, the crystal structure and chemical composition for both crystals were verified by the X-ray Laue method (Laue diffractometer of Photonic Science), powder X-ray diffraction (Bruker AXS D8 Advance X-ray diffractometer using the $\text{Cu K}\alpha$ line), and energy dispersive spectrometer analysis (scanning electron microscope Tescan Mira I LMH). A small piece of size $49 \times 25 \times 16 \mu\text{m}^3$ from crystal #1 and of size $62 \times 45 \times 22 \mu\text{m}^3$ from sample #2 was cut off for further investigation by means of X-ray diffraction experiments. On both, single crystal diffraction data were collected at 293 K using an automatic 4-circle Bruker Kappa-Apex II diffractometer equipped with an Apex II CCD detector and a Mo x-ray source with a graphite monochromator. The collection strategy was performed employing Apex II software [23] and the data were analyzed using crysalispro software [24]. The cell parameters were defined, and the study of the reciprocal $(hkl)^*$ planes, reconstructed using the unwarp option, was done to evidence systematic absences. Integration of the data was performed with crysalispro and an empirical absorption correction using spherical harmonics (ABSPACK scaling algorithm) was applied. The structural resolution and refinement were done using superflip [25] and JANA2006 [26] programs. The analysis revealed that the CeRhSn_2 single crystals crystallize in an orthorhombic lattice with space group $Cmcm$. The refined unit-cell parameters are $a = 4.5905(10)$, $b = 16.9758(5)$, and $c = 9.5924(3) \text{ \AA}$ for crystal #1, and $a = 4.5913(9)$, $b = 16.9824(6)$, and $c = 9.5915(3) \text{ \AA}$ for sample #2. These values are in good agreement with previously

reported data [14,16]. Inspection of the reconstructed reciprocal planes (hkl)* reveals a systematic absence condition $h + k = 2n + 1$, consistent with C centering. The refinement of crystal #1 was performed using 11114 measured reflections, of which 899 were independent with $I > 3\sigma(I)$. A total of 32 parameters were refined. Twinning by a 180° rotation about the (100) axis was taken into account, with the minor twin component refined to a volume fraction of 0.10. For sample #2, 10591 reflections were measured, yielding 734 independent reflections. In this case, 30 parameters were refined, and no evidence of twinning was observed. The final agreement factors are $R = 2.49\%$ for crystal #1 and $R = 2.18\%$ for crystal #2. These values demonstrate the good quality of the resolved structure. In addition, it is supported by the theoretical calculations obtaining $a = 4.5971$, $b = 17.2251$, and $c = 9.6186$ Å (see Sec. III C for more details).

Magnetization measurements were conducted on a superconducting quantum interference device (SQUID) magnetometer (MPMS-7, Quantum Design) in the temperature range from 1.8 to 300 K and in external fields up to 7 T. Subsequent measurements down to 0.4 K were performed using a Quantum Design MPMS-3 SQUID magnetometer with the ^3He option in Košice and Augsburg. Heat capacity measurements using the relaxation method and resistivity measurements utilizing the standard low-frequency ac four-probe technique were carried out on a commercial physical property measurement system (PPMS-9, Quantum Design) at a T interval of $0.35 < T < 10$ K.

B. Computational method

Quantum-mechanical (*ab initio*) calculations were performed within density functional theory (DFT) [27,28] by using the generalized gradient approximation parametrized by Perdew-Burke-Ernzerhof [29] for electron exchange and correlation effects as implemented in the Vienna *ab initio* simulation package (VASP) code [30]. Atoms were represented by the projector-augmented wave pseudopotentials provided by vasp with the following electron configurations $5s^25p^64f^16s^25d^1$, $5s^25p^2$, and $5s^24d^8$ for Ce, Sn, and Rh atoms, respectively. A plane-wave expansion of 500 eV, combined with conjugate gradient energy minimization and quasi-Newton force minimization, was used to optimize the geometry and atomic positions of the primitive cell (8 f.u.). Atomic positions were relaxed until the forces were smaller than 10^{-3} eV/Å, while the total energy was converged down to 10^{-6} eV. The Brillouin zone was sampled using the k -point mesh generated by the Γ -centered Monkhorst-Pack scheme with equidistant length vectors of 100 [30], equivalent to a grid of $22 \times 6 \times 10$.

III. RESULTS

A. Magnetic properties of CeRhSn₂

The inset of Fig. 2 displays the inverse magnetic susceptibility $\chi^{-1}(T) = H/M$ versus the temperature between 1.8 and 300 K measured in a field of 1 T applied along the three main axes. The inverse susceptibility is strongly anisotropic in the entire temperature range. Above 200 K, the Curie-Weiss (CW) expression $\chi = \frac{N\mu_B^2\mu_{\text{eff}}^2}{3k_B(T-\theta_p)}$, where the various symbols have their usual meaning [31], provides a good

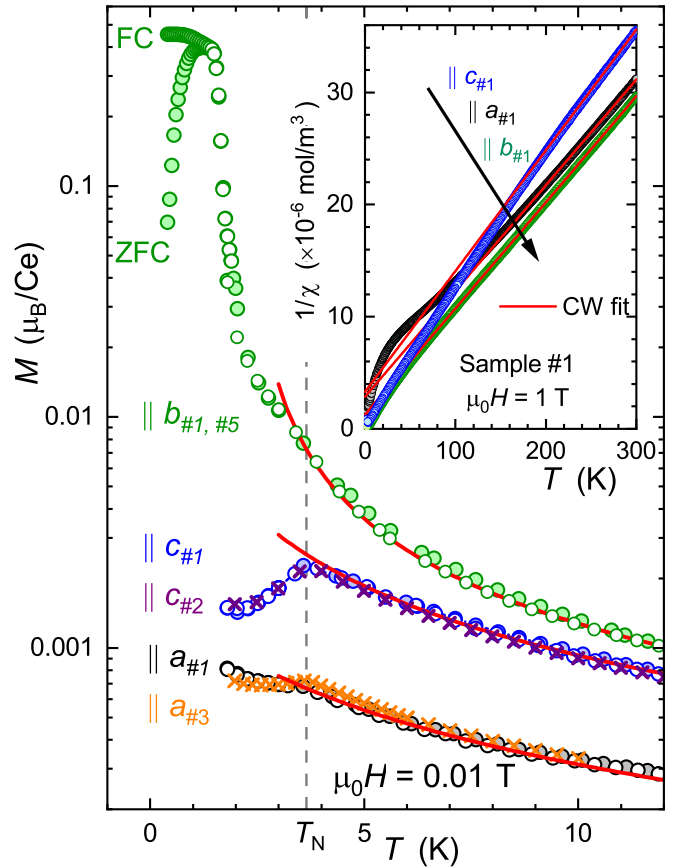


FIG. 2. Evolution of the magnetization M at low temperature and in an applied field of $\mu_0 H = 0.01$ T. Closed and open symbols denote zero-field-cooled (ZFC) and field-cooled (FC) measurements, respectively, with subscripts indicating sample numbers. The red lines are low-temperature Curie-Weiss fits ($4 < T < 10$ K). Measurements $T < 2$ K ($T \geq 1.8$ K) in the orientation $\mathbf{H} \parallel b$ were carried out on sample #5 (#1). The inset plots the inverse of magnetic susceptibility $\chi^{-1} = H/M$ vs temperature T in a field of $\mu_0 H = 1$ T along the a axis (black circle), b axis (green circle), and c axis (blue circle). The red lines represent Curie-Weiss fits applied to the data within the temperature range $200 < T < 300$ K.

fit to the data giving $\mu_{\text{eff}} = 2.59\mu_B/\text{Ce}$, $\mu_{\text{eff}} = 2.58\mu_B/\text{Ce}$, and $\mu_{\text{eff}} = 2.44\mu_B/\text{Ce}$ for applied field along the a , b , and c directions, respectively, which are in good agreement with the theoretical value of $\mu_{\text{eff}} = 2.54\mu_B$ for a free Ce^{3+} ion. Previous reports on polycrystalline specimens [14,16] showed that the zero-field-cooled (ZFC) and field-cooled (FC) $M(T)$ curves bifurcate below around $T_{M1} \approx 4$ K, indicative of a weak ferromagneticlike transition. Toward lower temperatures, a second transition manifesting through a small cusp in the ZFC cycle emerges at 3 K [16]. In the main panel of Fig. 1 we present the low-temperature ZFC and FC magnetization data ($M^{\parallel a}$ and $M^{\parallel c}$) down to 1.8 K, and $M^{\parallel b}$ down to 0.4 K in $\mathbf{H} = 0.01$ T for various samples. At $T_N = 3.6$ K, a pronounced cusp in $M^{\parallel c}$ signals the onset of antiferromagnetic ordering. Interestingly, the corresponding feature appears only as a faint kink along the a axis and is entirely absent along the b axis, reflecting strong directional dependence in the magnetic response. This is also reflected in the low- T CW

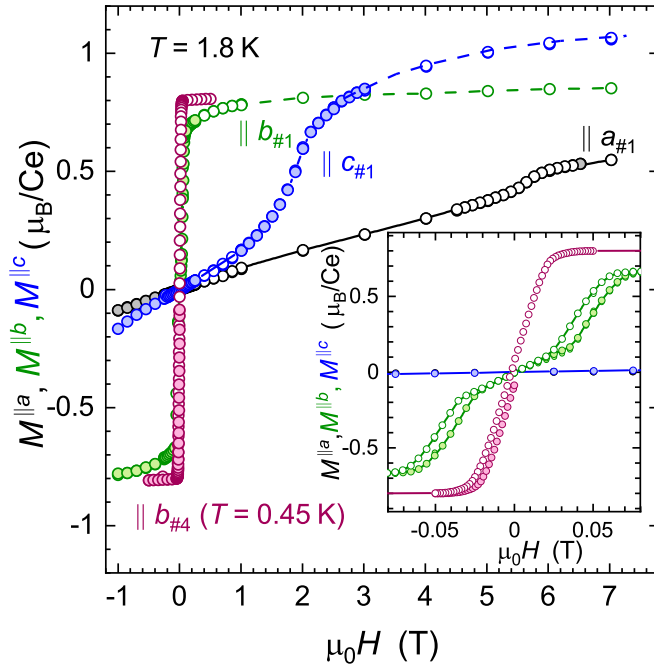


FIG. 3. Field dependence of the magnetization for field applied a , b , and c axes measured at 1.8 K and at $T = 0.45$ K ($\parallel b$). The inset shows a zoom-in of the low-field region of M vs H . Closed (open) symbols show the data in increasing (decreasing) field. The data $\parallel a$ and $\parallel c$ overlap in low fields. A finite M at zero field persists along b in the $T = 0.45$ K measurement.

behavior. The fitting of the low-temperature χ ($4 < T < 10$ K) yields $\mu_{\text{eff}} = 1.29\mu_B/\text{Ce}$ ($\theta_p = -1.94$ K), $\mu_{\text{eff}} = 2.09\mu_B/\text{Ce}$ ($\theta_p = 2.29$ K), and $\mu_{\text{eff}} = 2.04\mu_B/\text{Ce}$ ($\theta_p = -0.006$ K) for the a , b , and c axes, respectively, showing that FM interaction dominates along the b axis. Note that the previously mentioned bifurcation between the ZFC and FC data below T_N is not observed. Instead, for $T < T_N$ the magnetization along this direction increases rapidly and a second transition into a FM order takes place at $T_C = 1.7$ K, marked by a sudden steep increase, reaching a maximum value of $M^{\parallel b} = 0.45\mu_B/\text{Ce}$ (FC). The splitting of the ZFC and FC curves below T_C ($T_{\text{bf}} \approx 1.5$ K) is in agreement with the expected behavior for a ferromagnet with domain-wall pinning, which is enforced by the high magnetocrystalline anisotropy.

To further contrast our data with previous results, we calculate the average susceptibility, defined as $\chi_{\text{avg}} = \frac{1}{3}(\chi^{\parallel a} + \chi^{\parallel b} + \chi^{\parallel c})$, where $\chi^{\parallel a} = M^{\parallel a}/H$, etc., from Fig. 2 at 5 K yielding a value of $1.47 \times 10^{-6} \text{ m}^3/\text{molCe}$. In comparison, the susceptibility of the polycrystalline sample equals $\chi_{\text{pol}}(5 \text{ K}) = 1.75 \times 10^{-6} \text{ m}^3/\text{molCe}$ [16]. The discrepancy can be explained due to the preferred orientation of the investigated polycrystalline sample. This is corroborated by isothermal magnetization experiments at 1.8 K (see Fig. 3). The average magnetic moment M_{avg} at the field of 5 T yields $0.74\mu_B/\text{Ce}$, which is in excellent agreement with the value of $0.75\mu_B/\text{Ce}$ reported in [14] but lower than the moment obtained by Hossain *et al.* [16] being $M = 0.9\mu_B/\text{Ce}$. From our data we conclude that their polycrystalline specimen had a preferred orientation along the c direction.

More importantly, the isothermal magnetization at $T = 0.45$ K along $\mathbf{H} \parallel b$ displays clear hysteresis, with a remanent magnetization of $0.03\mu_B/\text{Ce}$ at the zero applied field crossing, providing additional evidence for the ferromagnetic nature of the lower transition. The figure also illustrates the strongly anisotropic nature of the intermediate AFM state in CeRhSn_2 . At $T = 1.8$ K, hence slightly above T_C , $M^{\parallel b}(H)$ initially displays a linear field dependence. When the field is increased above 0.035 T a sharp metamagnetic transition (MT) occurs, resulting in a magnetization step of $0.6\mu_B$. In fields $\mathbf{H} > 0.07$ T, $M^{\parallel b}(H)$ saturates reaching a value of $0.85\mu_B$ at 7 T. This is smaller than the $2.14\mu_B$ value for the Ce^{3+} due to the presence of crystal field effects. A narrow hysteresis occurs when the field is swept down, which closes at 0.015 T (see the inset in Fig. 3). In the case of $\mathbf{H} \parallel a$, $M(H)$ increases monotonically in field up to 5.3 T where an MT emerges. Unlike for $\mathbf{H} \parallel b$, the MT is broad, more “S” shapeliike, and the magnetization step is small from $M^{\parallel a} = 0.4\mu_B/\text{Ce}$ at 5.3 T to $M^{\parallel a} = 0.5\mu_B/\text{Ce}$ at 6.0 T. In higher fields $M^{\parallel a}$ continues to increase to a value of $0.55\mu_B/\text{Ce}$ at 7 T. The $M^{\parallel c}$ behavior resembles the former. In low fields the curve initially shows a linear in H dependence. At ≈ 0.7 T, an upturn in field sets is followed by a gradual saturation for fields > 2 T to a value of $1.06\mu_B/\text{Ce}$ at 7 T. In comparison to the others, the MT is much broader. However, its increase is significant and for $\mu_0H > 2.7$ T the magnetization $M^{\parallel c}$ exceeds $M^{\parallel b}$. In neither of the two field directions, $\parallel a$ and $\parallel c$, was a hysteresis in $M(H)$ found.

To characterize the magnetic behavior more comprehensively, we conducted several $M(H)$ measurements at various temperatures below and above T_N . These are summarized in Fig. 4. The evolution of $M(H)$ for fields $\parallel a$ and $\parallel c$ [Figs. 4(a) and 4(c), respectively] are very similar. For $T < T_N$, which is below the antiferromagnetic ordering temperature, the magnetization initially increases linearly with the increase of magnetic field. As noted earlier, a weak-field-induced metamagnetic transition from AFM to a field-induced polarized state occurs at higher fields, above which magnetization saturates slowly. The transition decreases with increasing temperature. Above 3 K, the MT is no longer observed and $M(H)$ recovers Brillouin-like behavior [Figs. 4(a) and 4(c)], consistent with the observation that the system undergoes its initial magnetic ordering near $T_N = 3.6$ K. In contrast, as the temperature increases, the sharp MT in $M^{\parallel b}(H)$ subsequently broadens and reduces in size, and shifts to higher fields until it disappears for $T \geq 3.5$ K. This behavior is characteristic of FM ordering, and accordingly, we associate the peak position of dM/dH in increasing field (not shown) with the transition from the AFM state into the ferromagnetically ordered state.

B. Specific heat and electrical resistivity of CeRhSn_2

Low-temperature heat capacity measurements C_p/T for CeRhSn_2 are shown in Figs. 5 and 6 for fields applied parallel to the b axis and c axis, respectively. The data is presented without subtraction of a phonon contribution due to the lack of the nonmagnetic counterpart LaRhSn_2 . However, in this temperature range, the phonon contribution is often negligible and the contribution of the s , p , and d electrons is small compared to that of the $4f$ electrons. To elucidate, based on

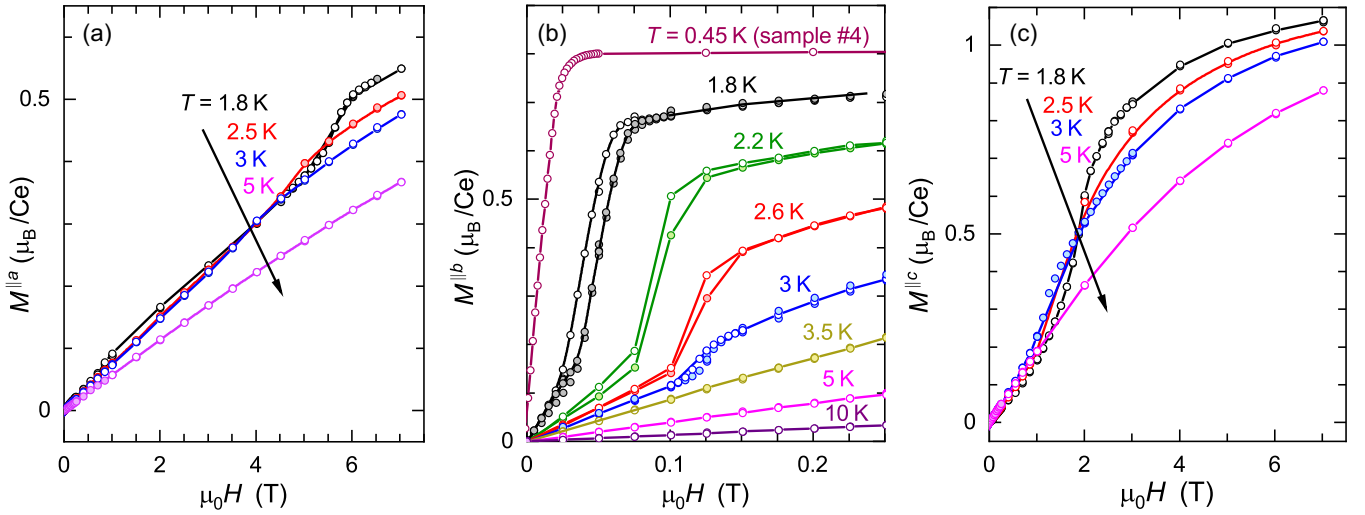


FIG. 4. Magnetic field isotherms of CeRhSn₂ at various temperatures for $\mathbf{H}$ parallel (a) a axis, (b) b axis, and (c) c axis, respectively. Open (closed) symbols depict ZFC (FC) results.

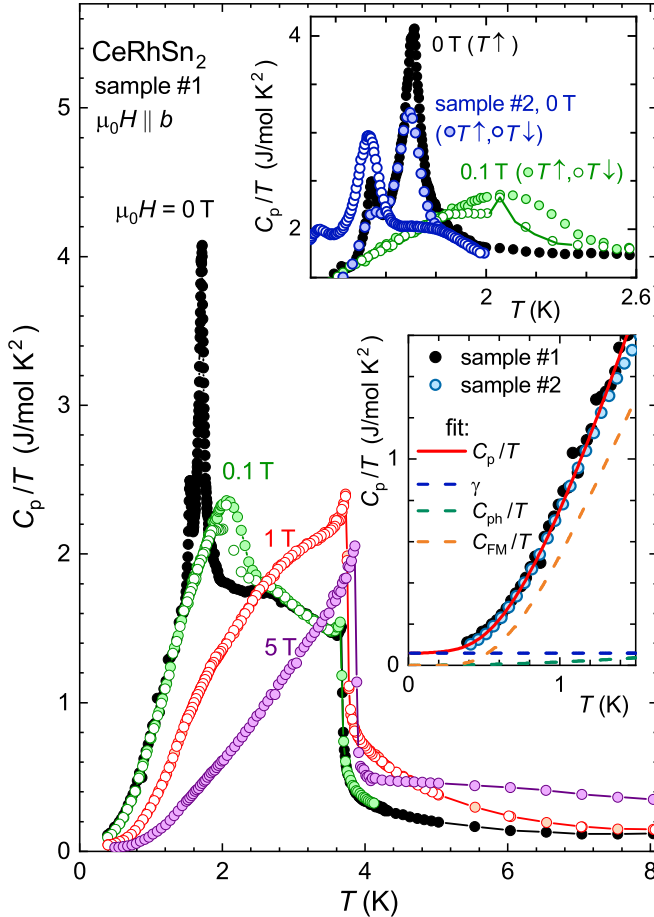


FIG. 5. The heat capacity C_p divided by temperature T vs T in various fields applied parallel to the b axis. Open (closed) symbols are in the cooling down (warming up) cycle. The upper inset enlarges the region around the double transition in zero field and from a second sample (#2). The lower inset presents the fit to the data using Eq. (1).

LDA calculations, the Sommerfeld coefficient γ of LaRhSn₂ is estimated to be of the order of 5 mJ/mol K² [17]. In zero field, C_p/T exhibits a well-defined anomaly at $T_N \approx 3.6$ K, consistent with the onset of AFM order inferred from the $M(T)$ data and in agreement with the report by Hossain *et al.* [16]. However, the low-temperature behavior below T_N differs markedly from previous results. Rather than a single broadened maximum, $C_p(T)$ increases gradually upon cooling, indicative of residual spin fluctuations within the ordered state. At lower temperatures, two closely spaced anomalies are resolved. A pronounced, λ -like peak at $T_C \approx 1.7$ K signals

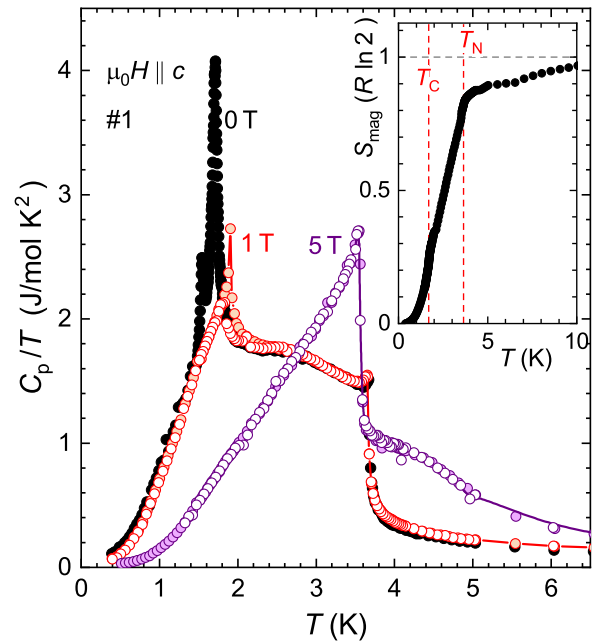


FIG. 6. The ratio C_p/T as a function of temperature T of sample #1, under varying magnetic fields aligned parallel to the c axis, with open (closed) symbols representing the cooling down (warming up) cycle. The inset shows the zero-field magnetic entropy S_{mag} vs T .

the onset of FM order, followed by a second, smaller anomaly at $T \approx 1.5$ K. Both features exhibit characteristics consistent with weakly first-order behavior and are observed across different samples, indicating their intrinsic nature. Upon further cooling, the heat capacity is rapidly suppressed, reflecting the freezing out of magnetic excitations in the low-temperature ground state. To resolve the discontinuous character of these transitions, we employed a long heat-pulse technique [32] on sample #1 in the range $1.4 < T < 2$ K (Fig. 5, black symbols), while all other data were collected using the standard relaxation method. Clear hysteresis is observed upon temperature cycling, as highlighted in the upper-right inset of Fig. 5, corroborating the first-order nature of both transitions. Under applied magnetic field, the anomaly at 1.7 K broadens, shifts to higher temperatures, and evolves toward a more second-order-like character, consistent with the stabilization of FM order. In contrast, the lower-temperature feature at 1.5 K is progressively suppressed for $\mathbf{H} \parallel b$ and c . Its microscopic origin remains unclear but is suggestive of an abrupt spin reconfiguration.

Our data further contradict earlier reports [16,17] which suggested a monotonic suppression of T_N under applied magnetic field. Instead, we observe a pronounced anisotropic response: T_N decreases with increasing field for $\mathbf{H} \parallel a$ and c , whereas it increases for $\mathbf{H} \parallel b$. At low temperatures ($T < 1.5$ K), the specific heat is well described by (see inset of Fig. 5)

$$C_p/T = \gamma + \beta T^2 + A\sqrt{T} \exp^{-\Delta/T}. \quad (1)$$

Here, γ denotes the electronic (Sommerfeld) contribution, βT^2 represents the phonon contribution, and the last term accounts for the spin-wave contribution of a strongly anisotropic ferromagnet with a gap Δ in the magnon spectrum [33]. The best fit yields $\gamma = 76.5(3)$ mJ/mol K², $\beta = 0.67(4)$ mJ/mol K⁴, corresponding to a Debye temperature $\Theta_D \approx 226$ K, $A = 4.54(7)$ J/mol K^{3/2}, and a magnon gap $\Delta = 1.92(5)$ K, the latter being comparable to T_C . The extracted Sommerfeld coefficient is significantly smaller than the previously reported value of ~ 300 mJ/mol K², which was obtained from a fit to the rising flank of a broad anomaly [16]. Nevertheless, γ remains enhanced compared to other Ce-based ferromagnets such as CeRu₂Ge₂ ($\gamma \approx 20$ mJ/mol K²) [34,35] and CePd₂Ga₃ ($\gamma \approx 9$ mJ/mol K²) [36]. Similarly, the previous study using local spin density approximation indicates that the local electronic and exchange effects leads also to low γ value ($\gamma \approx 15$ mJ/mol K²) [17]. This is even lower if localization by the Hubbard model is taken into account for Ce f states [17], indicating that in reality many-body electronic effects enhance the states near Fermi level. While the obtained value is below that typically observed in strongly correlated Ce-based heavy-fermion systems ($\gamma \gtrsim 100$ mJ/mol K²), it nevertheless indicates moderate mass enhancement. The inset of Fig. 6 displays the magnetic entropy $S_{\text{mag}}(T)$ obtained by subtracting the phonon contribution derived from Eq. (1) and integrating the resulting C_p/T over temperature. We observe that at the ordering temperature $T_N \approx 3.6$ K, the entropy amounts to $S_{\text{mag}} \approx 0.84R \ln 2$. The value agrees well with previous results on polycrystalline samples [16] but is below the value expected for a well-

isolated doublet ground state. The full $R \ln 2$ is recovered only at higher temperatures, $T \gtrsim 10$ K, consistent with a CEF scheme in which the Ce³⁺ ions possess a well-separated Kramers doublet ground state arising from the splitting of the $J = 5/2$ multiplet. The substantial fraction of entropy released at T_N indicates predominantly localized f -electron moments. To put this in perspective, known frustrated systems show a significant portion of $S_{\text{mag}}(T)$ being shifted to higher temperatures. For example, in the olivine compound Mn₂SiS₄, where the Mn ions form a sawtooth lattice [37], the entropy released at the magnetic transition is exceedingly small, amounting to only $3.6 \times 10^{-5} R \ln 6$ [38]. Similarly, in the heavy-fermion metal CePdAl, which crystallizes in the ZrNiAl-type structure and hosts geometrically frustrated Ce moments on a distorted Kagome lattice, the entropy recovered at T_N remains below $0.5R \ln 2$ [39]. In addition, CePdAl exhibits a pronounced maximum in the magnetization above T_N that is not accompanied by a corresponding anomaly in C_p/T [40]. The absence of these features in CeRhSn₂ indicates that magnetic frustration plays at most a minor role. In this context, the slightly reduced value of S_{mag} at T_N , together with the moderately enhanced γ is more naturally attributed to weak but finite Kondo screening.

Figure 7(a) displays the temperature dependence of the electrical resistivity $\rho(T)$, measured in zero magnetic field with the current $\mathbf{j}$ applied along the principal axes. The data reveal a pronounced anisotropy in the resistivity values with $\rho(T)$ being highest for $\mathbf{j} \parallel b$ (sample #2), and roughly five times lower when $\mathbf{j} \parallel a$ (sample #2). Despite this magnitude difference, the temperature evolution of $\rho(T)$ is qualitatively similar across all directions. Upon cooling, the resistivity monotonically decreases, reaching a minimum near 17 K for $\mathbf{j} \parallel b$ and c , and around 22 K for $\mathbf{j} \parallel a$. This temperature behavior mimics the previously reported data on polycrystalline samples, which also shows a resistance minimum around 20 K [15,16]. At lower temperatures, $\rho(T)$ exhibits an upturn followed by a sharp drop, signaling the onset of magnetic ordering at T_N . The low- T resistivity regime is shown in more detail in Figs. 7(b)–7(d). Below T_N , a second drop in resistivity appears, marking the onset of the FM ordering. This transition displays a clear hysteresis, corroborating the first-order character as previously identified in the C_p/T measurements. However, the third transition, which appears as a subtle peak in C_p/T , seemingly lacks a distinct counterpart in the resistivity data. In Figs. 8(a) and 8(b), we directly compare the experimental data sets near the ferromagnetic transition. The closed (open) symbols denote the data collected during increasing (decreasing) temperature sweeps. Clearly, the FM anomaly in C_p/T coincides with the drop in $\rho(T)$ in increasing and decreasing temperature runs, while the subsequent small peak in C_p/T may be associated with a faint kinklike irregularity in $\rho(T)$.

The resistivity response under applied magnetic field is consistent with the behavior observed in the specific heat data. For $\mathbf{H} \parallel b$ [Figs. 7(b) and 7(d)], a small field of $\mu_0 H = 0.1$ T strongly enhances the FM transition, shifting T_C from 1.55 K (zero field, decreasing temperature) up to 2.3 K. Concurrently, the kinklike structure in $\rho(T)$, which correlated with the lowest transition observed in C_p/T , vanishes. In contrast, the AFM ordering temperature at $T_N = 3.65$ K remains robust

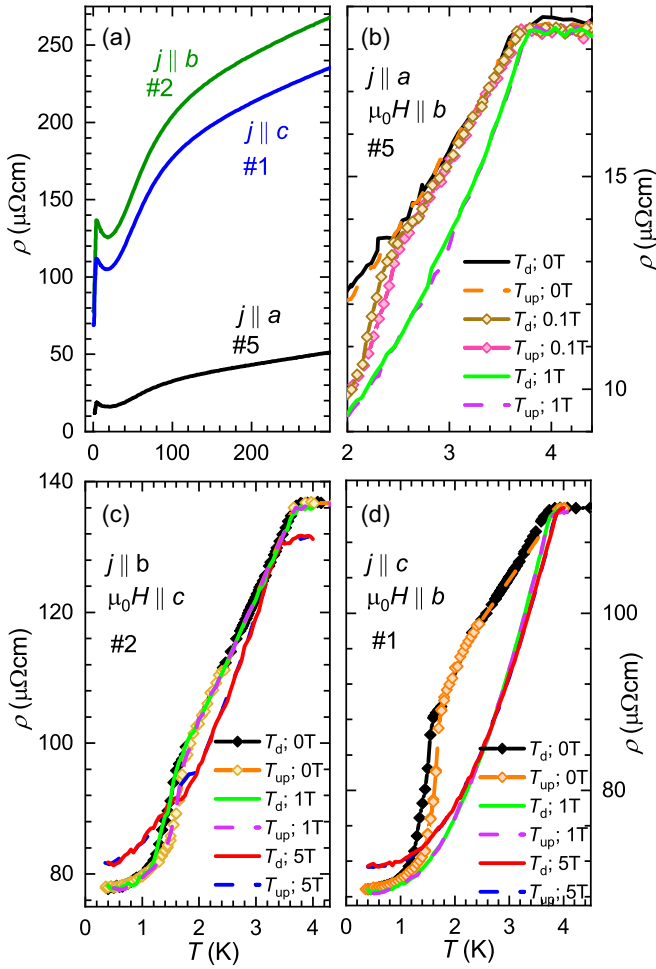


FIG. 7. Temperature-dependent electrical resistivity $\rho(T)$. (a) Zero-field data with current density $\mathbf{j}$ applied along the principal axis; measurements were performed on different samples. (b)–(d) Low-temperature $\rho(T)$ in applied magnetic fields: (b) $\mathbf{H} \parallel \mathbf{b}$ with $\mathbf{j} \parallel \mathbf{a}$, (c) $\mathbf{H} \parallel \mathbf{c}$ with $\mathbf{j} \parallel \mathbf{b}$, and (d) $\mathbf{H} \parallel \mathbf{b}$ with $\mathbf{j} \parallel \mathbf{c}$. Data taken upon cooling and warming are denoted by T_d and T_{up} , respectively.

and largely unaffected by 0.1 T. In a field of 1 T the FM and AFM transitions are merged, as clearly seen in Figs. 7(b) and 7(d), and the thermal hysteresis in the resistivity sweep has disappeared. Furthermore, the upper AFM order is slightly pushed to a higher temperature. Conversely, the transitions are much more robust against a field applied along the c axis. For $\mathbf{H} \parallel \mathbf{c}$ [Fig. 7(c)], a field of 1 T does not significantly affect the resistivity, with the 1 T data collapsing on top of the zero-field data within the measurement resolution. No kinklike anomaly below T_C is observed along this current direction. Even at a high field of $\mu_0 H = 5$ T, we still detect a small thermal hysteresis in $\rho(T)$ around 1.9 K. This hysteresis suggests that the FM transition has not fully merged with T_N , even though no corresponding anomaly is detected in the C_p/T data at this field. Moreover, the AFM ordering temperature is noticeably reduced at 5 T. The field dependence of the magnetic transitions is further detailed in Figs. 8(c) and 8(d), where we plot the zero-field and $\mu_0 H = 5$ T data for C_p/T , the temperature derivative of the magnetic specific heat proxy $d(\chi T)/dT$ ($\mathbf{H} \parallel \mathbf{c}$ in 0.01 T), and the second derivative of

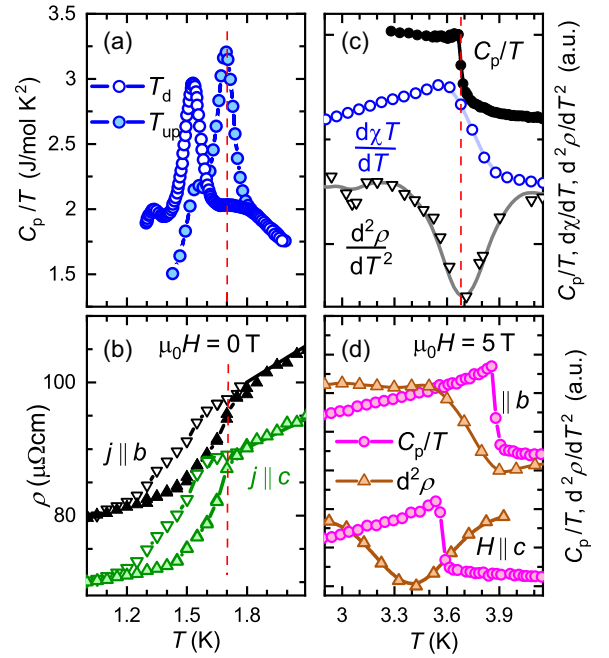


FIG. 8. Overview of magnetic transitions: open symbols denote data acquired in the cooling cycle (T_d), and closed symbols represent measurements from the warming cycle (T_{up}). (a) C_p/T vs T in zero field measured using a heat-pulse technique and (b) the respective low- T resistivity for $\mathbf{j} \parallel \mathbf{b}$ (black) and $\mathbf{j} \parallel \mathbf{c}$ (green). The red dashed line marks T_C obtained from $\chi(T)$ (see Fig. 2). (c) Shows the definition of the AFM transition temperature T_N from C_p/T (zero field), $d\chi T/dT$ (0.01 T), and $d^2\rho/dT^2$ (zero field, $\mathbf{j} \parallel \mathbf{b}$) and (d) in a field of $\mu_0 H = 5$ T (no susceptibility data available).

resistivity $d^2\rho/dT^2$. While the function $d(\chi T)/dT$ is generally expected to resemble the C_p/T data near the critical region for simple systems [41], the observed deviation here suggests that we are not dealing with a simple antiferromagnetic transition, highlighting the complexity of the ordering. Crucially, Fig. 7(d) clearly illustrates that the AFM ordering temperature shifts in opposite directions depending on whether $\mathbf{H}$ is applied along b or c as discussed previously. Furthermore, the discrepancy observed at $\mu_0 H = 5$ T ($\mathbf{H} \parallel \mathbf{c}$), where the C_p/T anomaly appears at a higher temperature than the minimum in $d^2\rho/dT^2$, suggests a potential influence from sample misalignment. This mismatch could be attributed to a slight tilt of the measured platform (e.g., the heat capacity sample) toward the highly responsive b direction, leading to an apparent shift due to the large magnetic anisotropy.

C. Theoretical results

The initial spin-polarized calculations were performed, leading to a ferromagnetic ground state. The lattice parameters and internal degrees of freedom of certain atoms are summarized in Table I and compared with experimental data. In general, we obtain very good agreement regarding the lattice parameters (the volume is only 2% larger than the experimental one) as well as the atomic positions of the optimized internal degrees of freedom (see Table I). The ground state ferromagnetic order exhibits two different spin moments on Ce atoms according to their Wyckoff positions, namely, the

TABLE I. The lattice parameters in angstrom as well as the internal degrees of freedom of atomic positions for various Wyckoff positions of constituent elements. Two numbers (y/z) denote the internal degree of freedom along the y and z directions.

method	lattice			atomic position				
	a	b	c	Ce 4c	Sn 8f	Sn1 4c	Sn2 4c	Rh 8f
expt.	4.5905	16.9758	9.5924	0.20418	0.69789/0.58981	0.57350	0.40848	0.64684/0.49519
theory	4.5971	17.2251	9.6186	0.20446	0.698915/0.58935	0.57435	0.40995	0.646401/0.49418

4c and 4a amount to $0.4\mu_B$ and to $0.9\mu_B$, respectively. The results coincide with the magnetization presented in Fig. 3 in an interesting way; if the field is along the b axis (c axis), it saturates close to the value of the spin moment of Ce at the 4a (4c) Wyckoff position, that is, Ce2 (Ce1). However, these would suggest that the external magnetic field will induce antiferromagnetic order on one or the other Ce sublattice. Even if the starting magnetic ordering was set up to be antiferromagnetic, the ferrimagnetic order prevails, i.e., the Ce atoms carrying significant magnetic moments do not compensate each other; rather, different magnitudes were found, and such ordering possesses higher energy than the pure ferromagnetic order. Hence, we conclude that the FM state is preferred by means of at least 50 meV/f.u. with respect to any antiferromagnetic or ferrimagnetic possible arrangements based on the simplest and standard DFT calculations, whereas more sophisticated approaches, such as DMFT [42] or multiplet calculations, might be necessary to fully address the anisotropic behavior in the external magnetic field along the a , b , and c axes.

IV. CONCLUSION

We have presented a comprehensive study of single-crystalline CeRhSn₂ that resolves long-standing ambiguities regarding its low-temperature properties and magnetic ground state [14–17]. Thermodynamic and transport measurements performed on multiple single crystals reproduce the behavior reported for polycrystalline samples, demonstrating that the observed features are intrinsic. Our results establish that CeRhSn₂ undergoes three magnetic transitions: an antiferromagnetic transition at $T_N = 3.65$ K, followed by ferromagnetic ordering below $T_C = 1.7$ K, and a third anomaly at $T = 1.5$ K. The pronounced hysteresis in specific heat and resistivity at the two lower transitions, together with the analysis of thermodynamic data and magnetization measurements, provides clear evidence for a ferromagnetic ground state, thereby resolving the long-standing controversy. This conclusion is further supported by our *ab initio* calculations.

We further show that previously reported enhanced values of the Sommerfeld coefficient γ arose from measurements that did not fully capture the completion of ferromagnetic order. The revised value, $\gamma = 76.5$ mJ/mol K², is only moderately enhanced compared to local-density approximation calculation. We attribute this to weak Kondo hybridization.

In addition, we demonstrate that the magnetic behavior of CeRhSn₂ is highly anisotropic. In particular, the anti-

ferromagnetic transition at $T_N = 3.65$ K is not observed in magnetization for fields applied along the b axis. Furthermore, magnetic fields along specific crystallographic directions appear to selectively stabilize antiferromagnetic correlations on distinct Ce sublattices, which may account for discrepancies in earlier studies on polycrystalline samples. Although the crystal structure, with its sawtoothlike arrangement of Ce ions, suggests possible geometric frustration, our results indicate that frustration does not play a dominant role.

Hydrostatic pressure measurements are currently underway to explore the possibility of tuning CeRhSn₂ toward a ferromagnetic quantum critical point and to examine whether multiple quantum critical points, as proposed in Ref. [20], are realized. In addition, neutron-scattering experiments would be highly desirable to resolve the magnetic structures of the ordered phases and to follow their evolution under applied magnetic fields and pressure. Such studies would provide deeper insight into the interplay of competing magnetic interactions in systems with multiple magnetic sublattices.

ACKNOWLEDGMENTS

J.C. thanks J. Schnack for fruitful discussion. This work was supported by the INTEREXCELLENCE-INTERCOST Project no. LUC24139 of Czech Ministry of Education, Youth and Sports (MŠMT) within the framework of the COST Action SUPERQUMAP (CA21144). The experiments were carried out at the Materials Growth and Measurement Laboratory (MGML [43]), which is supported by the Czech Research Infrastructure program (Project No. LM2023065). Z.H. acknowledges financial support from the Polish National Agency for Academic Exchange under the Ulam fellowship. D.L. acknowledges the Czech Science Foundation Grant No. 25-16339S and the project e-INFRA CZ (ID 90254) for access to computational resources by the Ministry of Education, Youth and Sports of the Czech Republic. Work at the University of Augsburg was supported by the Bavarian-Czech Academic Agency (Project no. BTHA-JC-2024-15) and by the German Research Foundation (DFG) through TRR360 (Project no. 492547816). The work acknowledges Project QM4ST No. CZ.02.01.01/00/22_008/0004572 funded by the MŠMT.

DATA AVAILABILITY

The data of the figures have been made available at [44].

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