CeIr₃Ge₇: A local moment antiferromagnetic metal with extremely low ordering temperature

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CeIr₃Ge₇ is an antiferromagnetic metal with a remarkably low ordering temperature $T_N = 0.63$ K, while most Ce-based magnets order between 2 and 15 K. Thermodynamic and transport properties as a function of magnetic field or pressure do not show signatures of Kondo correlations, interaction competition, or frustration, as had been observed in a few antiferromagnets with comparably low or lower T_N . The averaged Weiss temperature measured below 10 K is comparable to T_N , suggesting that the Ruderman-Kittel-Kasuya-Yosida exchange coupling is very weak in this material. The unusually low T_N in CeIr₃Ge₇ can therefore be attributed to the large Ce-Ce bond length of about 5.7 Å, which is about 1.5 Å larger than in the most Ce-based intermetallic systems.

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I. INTRODUCTION

Compounds containing Ce or Yb ions have been studied extensively due to their diverse ground states originating from the competition between several energy scales. The competition between Ruderman-Kittel-Kasuya-Yosida (RKKY) exchange interaction and Kondo coupling gives rise to intermediate valence behavior [1-4] or Kondo screening [4-9] which, in turn, often results in unconventional superconductivity, non-Fermi liquid behavior, and quantum criticality [7,10– 13]. If the hybridization between f electrons and conduction electrons is very weak, the ground states of these systems are dictated by RKKY exchange interaction and crystal electric field (CEF) effects, resulting in long-range magnetic order [14–16]. In the case of the Ce local moment metals without the Kondo effect, the ordering temperatures range from $T_{\rm C} =$ 115 K [17,18] in ferromagnetic CeRh₃B₂, to $T_{\rm C} = 0.44$ K [19,20] in ferromagnetic Ce₃Pt₂₃Si₁₁, while much lower temperatures can be expected for the Yb analogues [21]. Lower ordering temperatures in both Ce^{3+} and Yb^{3+} compounds could occur from any combination of effects including Kondo, competition between different exchange interactions, strong CEF anisotropy, or large distances between rare-earth ions (d_{R-R}) that minimize the RKKY exchange coupling J_{RKKY} . Compounds with low ordering temperatures often involve either weaker-than-RKKY exchange, as is the case in insulators, or multipolar order [22], in which case the resulting order is almost always underlined by heavy fermion (HF) behavior. Remarkably low Néel temperatures were also found in some intermetallic cage compounds having also large d_{R-R} , such as $T_N = 0.18$ K in Ce₄Pt₁₂Sn₂₅ [23] and $T_N = 0.89$ K in $CePt_4Ge_{12-x}Sb_x$ [24]. However, the low ordering temperatures have been attributed to either the onset of Kondo screening or frustration.

Here, we report on CeIr₃Ge₇, an intermetallic compound without Kondo effect and no geometric frustration, with a remarkably low antiferromagnetic (AFM) ordering temperature $T_{\rm N} = 0.63$ K. This is one of several $R = {\rm Ce}$ or Yb compounds found in the RT_3M_7 (1-3-7) class of compounds with T = transition metal and M = group 14 element [9,25], a family of rhombohedral intermetallics with the ScRh₃Si₇ structure type [26,27]. The R sublattice forms a distorted cubic structure, with nearest-neighbor d_{R-R} around 5.7 Å. Strong electron correlations in the Yb members of this family result in HF behavior and ferromagnetic or AFM ordering at temperatures as high as 7.5 K. Remarkably, CeIr₃Ge₇ is weakly correlated, and even with similar d_{R-R} , the ordering temperature is much smaller than in the Yb analogues ($T_{\rm N} =$ 7.5 K in YbRh₃Si₇ [9], $T_{\rm C} = 2.4$ K in YbIr₃Ge₇ [25], $T_{\rm N} =$ 4 K in YbIr₃Si₇ [28]). No frustration is present in CeIr₃Ge₇, as the Weiss temperature in the limit of absolute zero is close to $T_{\rm N}$. This system is a good metal, with residual resistivity values $\rho_0 \sim 20 \ \mu\Omega$ cm and a residual resistivity ratio RRR = $\rho(300 \text{ K})/\rho_0 \sim 5$. No Kondo correlations are apparent as most of the magnetic entropy is released below $T_{\rm N}$. In the absence of Kondo effect or frustration, the low T_N in CeIr₃Ge₇ is attributed to the large distance d_{R-R} . This points to the potential of the 1-3-7 family to reveal Ce or Yb compounds with low ordering temperatures, which, in turn, may be easily tuned toward absolute zero transitions and quantum critical regimes. Furthermore, an added appeal for Ce- or Yb-based metals with low magnetic exchange is their potential for demagnetization cooling below 2 K. These metallic magnets are preferable to the commonly used paramagnetic salts, which are insulators, or ³He systems, which are expensive. YbPt₂Sn [29], a metallic system with weak exchange coupling and $T_{\rm N} \sim 0.25$ K, similar to CeIr₃Ge₇, reinforces the potential of our low-temperature magnet for cooling applications.

II. EXPERIMENTAL METHODS

Temperature-dependent AC resistivity was measured using a Quantum Design (QD) physical properties measurement

TABLE I. Crystallographic parameters of CeIr₃Ge₇ single crystals at T = 298 K ($R\bar{3}c$).

<i>a</i> (Å)	7.8915(8)
<i>c</i> (Å)	20.788(6)
V (Å ³)	1121.1(4)
crystal dimensions (mm ³)	0.02 x 0.04 x 0.06
θ range (°)	5.2-30.3
extinction coefficient	0.00102(7)
absorption coefficient (mm^{-1})	86.73
measured reflections	7121
independent reflections	384
R _{int}	0.082
goodness-of-fit on F ²	1.18
$R_1(F)$ for $F_o^2 > 2\sigma (F_o^2)^a$	0.032
$wR_2(F_o^2)^{b}$	0.074

^a
$$R_1(F) = \sum || F_o | - | F_c || / \sum |F_o |$$

^b $wR_2(F_0^2) = [\sum [w(F_o^2 - F_c^2)^2] / \sum [w(F_o^2)^2]]^{1/2}$

system (PPMS) with a ³He insert using I = 2 mA and f =622.2 Hz. DC magnetic susceptibility measurements were performed on a QD magnetic properties measurement system equipped with an iHelium ³He attachment. Specific heat measurements at ambient pressure were collected using a thermal-relaxation method in QD PPMS with a ³He insert for the oriented crystal. An unoriented crystal was used for the specific heat measurement down to 0.1 K in a QD PPMS DynaCool system with a dilution refrigeration insert. Specific heat measurements under pressure were performed using a compensated heat-pulse method [30]. The sample was put into a Teflon capsule together with a piece of Pb whose superconducting transition temperature as a function of pressure served as a manometer. The capsule was mounted in a clamped-type CuBe cell using Fluorinert (3M) as a pressure transmitting medium. The background heat capacity of the empty cell was determined in separate runs and was subtracted from the raw data to obtain the sample's contribution.

III. ANALYSIS AND DISCUSSION

CeIr₃Ge₇ crystallizes in the $R\bar{3}c$ rhombohedral ScRh₃Si₇ structure type [26,27]. The very few 1-3-7 compounds known so far are RAu_3Ga_7 (R = Gd - Yb) [31,32], nonmagnetic RAu_3Al_7 (R = Ce - Sm, Gd-Lu) [33], and magnetic Eu(Rh,Ir)₃Ge₇ [34]. We recently reported on single crystal Yb Kondo systems YbRh₃Si₇ [9], YbIr₃Ge₇ [25], and YbIr₃Si₇ [28]. All the compounds were synthesized in single crystal form using a self-flux growth method [35,36], with details described elsewhere [9]. Single crystal x-ray diffraction measurements confirm the ScRh₃Si₇ structure type and verify the purity and stoichiometry of these compounds. The details of the x-ray diffraction and experimental methods are described in the Appendix and the crystallographic parameters are summarized in Table I. A nonmagnetic analog YIr₃Ge₇ polycrystalline sample was prepared by arc melting. Figure 1(a)shows a powder x-ray pattern and structural refinement for CeIr₃Ge₇ with a photo of a crystal shown in the inset. In this rhombohedral crystal structure, the R atoms form a distorted cubic sublattice [9], with the body diagonal of the cuboid



FIG. 1. (a) Room temperature (symbols) and calculated (red line) powder x-ray diffraction patterns of CeIr₃Ge₇, together with the expected peak positions (blue vertical lines) for space group $R\bar{3}c$ and lattice parameters a = 7.8915(8) Å and c = 20.788(6) Å. Violet curve is the difference between data and calculated patterns. Inset: crystal picture on millimeter-scaled paper. (b) Zero-field resistivity with current parallel to the [100] axis. (c) Inverse magnetic susceptibility H/M vs T for magnetic field $H \parallel [001]$ (blue), [100] (red), and a polycrystalline average (violet). Solid lines are high-temperature Curie-Weiss fits, with Weiss temperatures of -360 K, -20 K, and -100 K for $H \parallel [001]$, [100], and the polycrystalline average, respectively (see text). Left inset: the low-temperature H/M vs T for $H \parallel [100]$.

parallel to the *c* axis of the equivalent hexagonal unit cell. Notably, the distances $d_{R-R} \sim 5.7$ Å are larger than in many magnetic *R* intermetallics, but do not change significantly for R = Ce or Yb in the 1-3-7 structure. This observation becomes most relevant when trying to explain the low ordering temperature $T_N = 0.63$ K in CeIr₃Ge₇. Several scenarios may in principle result in low T_N in Ce compounds, such as the Kondo effect, frustration, or exchange coupling competition, weak exchange due to large $d_{\text{Ce}-\text{Ce}}$. The following discussion is based on evidence against most, if not all, of these scenarios in CeIr₃Ge₇, rendering this compound a unique non-Kondo metal with extremely low ordering temperature.

The $\mu_0 H = 0$ resistivity measurements [Fig. 1(b)] show that CeIr₃Ge₇ is a good metal, with a RRR = 5 and residual



FIG. 2. Celr₃Ge₇ M vs H isotherms for T = 0.5K (full symbols) and 1.8 K (open symbols) for $H \parallel [001]$ (blue circles) and $H \parallel [100]$ (red squares).

resistivity $\rho_0 \sim 20 \mu \Omega$ cm. However, upon cooling from room temperature, the resistivity is linear in temperature, and no signatures of Kondo correlations are apparent. The lack of Kondo effect will be further corroborated by the specific heat data shown later. For now, we turn to the magnetic susceptibility measured along $(H \parallel [001])$ and perpendicular $(H \parallel [100])$ to the c axis of the equivalent hexagonal unit cell. The inverse susceptibility H/M [Fig. 1(c)], measured up to 600 K, reveals large easy-plane CEF anisotropy. The average susceptibility is calculated as $M_{\text{ave}} = (M_{001} + 2M_{100})/3$. Fits to the Curie-Weiss law at high temperatures are shown in solid black lines. The experimental effective moment μ_{eff}^{exp} extracted from the fit of the average susceptibility [violet, Fig. 1(c)] is $\mu_{eff}^{exp} = 2.52 \ \mu_{B}/Ce^{3+}$, pointing to fully trivalent Ce ions in CeIr₃Ge₇, since the calculated Ce³⁺ effective moment $\mu_{eff}^{calc} = 2.52 \ \mu_{B}/Ce^{3+}$ 2.52 $\mu_{\rm B}/{\rm Ce^{3+}}$ is virtually identical to the experimental value. The negative Weiss temperatures indicate AFM correlations. The H/M data deviate from the Curie-Weiss law due to CEF splitting of the J = 5/2 multiplet. The deviation indicates a separation of the first excited CEF doublet of ~ 400 K, consistent with the CEF calculations, which are reported elsewhere [37].

In the $T \rightarrow 0$ limit, the inverse susceptibility intercept with the temperature axis is around -2 K for M_{ave} [left inset, Fig. 1(c)], comparable to the low ordering temperature $T_{\rm N} \sim$ 0.6 K indicated by the cusp in M/H vs T for $H \parallel [001]$ [right inset, Fig. 1(c)]. These observations can be reconciled by considering the Weiss temperatures at $T \rightarrow 0$ to reflect the exchange coupling J_{ex} , which consequently indicates that J_{ex} is inherently small in CeIr₃Ge₇. We show the M(H)isotherms in Fig. 2 for $H \parallel [100]$ (red symbols) and $H \parallel [001]$ (blue symbols), in the ordered state T = 0.5 K (full symbols) and the paramagnetic state T = 1.8 K (open symbols). The magnetization data agrees with the CEF calculations with an exchange interaction of 2.4 K [37]. The M(H) measurements confirm the out-of-plane magnetic anisotropies, and, more quantitatively, are in good agreement with the calculated moments of 0.91 μ_B /Ce and 0.33 μ_B /Ce along the easy ([100]) and hard ([001]) directions, respectively [37]. A magnetic field close to $\mu_0 H = 1.7 \text{ T}$ is required for saturation in the



FIG. 3. (a) Left axis: magnetic susceptibility M/H vs T. Right axis: d(MT)/dT vs T (solid line) for $\mu_0 H = 0.01$ T. (b) Left axis: specific heat C_p vs T (symbols) for different magnetic fields. The vertical dashed line through (a) to (b) marks T_N at zero field. (c) Temperature-dependent specific heat for different pressures. (d) Left axis: magnetic contribution to the specific heat C_{mag}/T T. Right axis: magnetic entropy S_{mag} vs T. $C_{mag}/T = C_p/T$ (CeIr₃Ge₇) - C_p/T (YIr₃Ge₇), where YIr₃Ge₇ is a nonmagnetic analog.

easy direction (squares, Fig. 2), while a linear extrapolation of $M(H \parallel [001])$ suggests a magnetic field in excess of 20 T is needed to reach saturation in the hard direction.

Because of this extremely large anisotropy, and the large magnetic field scale in the hard ([001]) direction, we focus next only on the field dependence of the ordering temperature T_N for the easy direction $H \parallel [100]$, as illustrated by the M/H and specific heat C_p data in Fig. 3. For AFM systems, a peak in C_p at T_N is expected to correspond

to a peak in d(MT)/dT[38], and this is illustrated for $\mu_0 H = 0.01 \text{ T}$ (solid line, right axis) in Fig. 3(a). Both ambient pressure M/H and C_p measurements [Figs. 3(a)-3(b)] reveal the expected suppression of T_N with increasing H, such that above $\mu_0 H = 1.7$ T, no peak can be resolved above 350 mK. Consistent with the zero-field resistivity data in Fig. 1(b), the specific heat data show an entropy release of $\sim 2/3$ R ln 2 at T_N [solid line, right axis in Fig. 3(d)], reaffirming the absence of both Kondo effect and strong correlations in CeIr₃Ge₇. To further rule out the presence of Kondo screening, one can consider the analysis by de Jongh and Miedema [39], which shows that an entropy release of 15%–40% of $R \ln 2$ above T_N can be expected in AFM systems around and above $T_{\rm N}$ due to the short-range magnetic interactions. Indeed, this is reflected in the H = 0 magnetic entropy plot of CeIr₃Ge₇ in Fig. 3(d) (black line). Furthermore, the same model indicates that the C_p contribution from Kondo is much weaker than that from classical intersite fluctuations. Within a Heisenberg model, one expects that, far above T_N , the leading term in $C_p(T)$ is proportional to $1/T^2$, i.e., $C_p/T \sim 1/T^3$. Upon applying a magnetic field [Fig. 3(b)], the dispersion of the magnons changes. In an AFM system, the energy gap at Q = 0 decreases and disappears at $\mu_0 H = \mu_0 H_c \approx 1.7$ T. This results in a large increase of the low-energy magnonlike excitations, which, in turn, shows up as a strong increase of C_p near and above T_N . For $H > H_c$ [full right triangles, Fig. 3(b)], a gap reopens in the magnon excitation spectra, and the specific heat evolves toward a broad anomaly related to the dominant Zeeman splitting.

Complementary to the field dependence, the pressure dependence of the specific heat [Fig. 3(c)] underlines the conclusion of small or negligible Kondo correlations: T_N increases linearly with pressures up to 1.6 GPa at a rate of $dT_N/dp = 3.71 \times 10^{-2}$ K/GPa. The increase of T_N under pressure, if only being ascribed to a volume effect, can be understood in the framework of the Doniach phase diagram [40]. The positive, yet very small, slope of $T_N(p)$ for CeIr₃Ge₇ suggests that this compound is located at far left of $T_N^{max}(J_{ex})$ in the Doniach diagram. The T - H phase diagram in Fig. 4(a) summarizes the T_N dependence on field at ambient pressure.

Among non-Kondo magnetic Ce compounds (see Table II), CeIr₃Ge₇ stands out [red in Fig. 4(b)] together with CeRh₃B₂ and Ce₃Pt₂₃Si₁₁. CeRh₃B₂ orders ferromagnetically with a remarkably large $T_{\rm C} \sim 115$ K due to the enhancement of the exchange interaction from the J = 7/2 multiplet, despite short $d_{\text{Ce-Ce}} = 3.096 \text{ Å}$ [18]. On the contrary, Ce₃Pt₂₃Si₁₁ orders ferromagnetically with an extraordinarily low $T_{\rm C} \sim$ 0.44 K due to large $d_{Ce-Ce} = 5.95 \text{ Å}$ [19,20]. Of note is the compound $Ce_4Pt_{12}Sn_{25}$ (Ref. [23]), which appears to have a record low $T_{\rm N} = 0.18$ K and represents a Kondo lattice in the small exchange limit of the Doniach phase diagram. In this case, however, the extremely low T_N is a result of the large $d_{\text{Ce-Ce}} \sim 6.14$ Å, weak Kondo screening just above T_{N} (marked by a tail in the magnetic specific heat peak just above the ordering), and weak geometric frustration due to the threefold point symmetry of the Ce site [60]. Except for the large $d_{\text{Ce-Ce}} \sim 5.7$ Å, none of these effects are at play in CeIr₃Ge₇: the low temperature Weiss temperatures [Fig. 1(c) inset] are comparable with $T_{\rm N}$, ruling out significant frustration effects; the specific heat peak [Fig. 3(b)] terminates abruptly at $T_{\rm N}$,



FIG. 4. (a) T - H phase diagram of CeIr₃Ge₇ at ambient pressure with $H \parallel [100]$. (b) Non-Kondo Ce compounds showing their ordering temperatures with respect to the shortest Ce-Ce bond distances [15–20,41–59]. Blue symbols represent ferromagnetic (FM) ordering temperatures, black symbols represent AFM ordering temperatures, and the red star indicates CeIr₃Ge₇. The legend for the symbols is given in the Appendix in Table II.

and $\rho(T)$ decreases linearly with temperature before it levels off at ρ_0 at the lowest temperatures [Fig. 1(b)], therefore, no Kondo screening signatures are apparent. To put CeIr₃Ge₇ into perspective, we summarize the non-Kondo Ce-based intermetallic compounds and plot their ordering temperatures vs d_{Ce-Ce} in Fig. 4(b). In these compounds, the magnetism is dictated by the RKKY interactions, where the exchange interaction J is an oscillatory function of the product of the Fermi wave vector $k_{\rm F}$ and $d_{\rm Ce-Ce}$, and the ordering temperature is proportional to J^2 . Since the $k_{\rm F}$ is not well defined for nonspherical Fermi surfaces of those compounds, we disregard it in the comparison shown in Fig. 4(b). Qualitatively, the order temperatures scale with d_{Ce-Ce} : the larger (smaller) the $d_{\text{Ce-Ce}}$, the smaller (larger) ordering temperature. Ce and Yb magnetic (trivalent) compounds are often thought as electronhole analogues. In metals, due to deeper localization of the 4 f electron and the larger strength of the spin-orbit coupling in the latter [21], smaller ordering temperatures are often expected in the latter compared to the former. What we find is that YbIr₃Ge₇ is in fact an HF ferromagnet with $T_{\rm C} \sim 2.4$ K, despite the nearly identical d_{R-R} in both the Ce and Yb analogues. [25] This may reflect that the details of the band

Compound	Symbol	$T_{ m N/C}~(m K)$	AFM/FM	Ce-Ce Bond Distance (Å)	Reference
$\mathrm{Ce_3Pt_4}$	•	2.8	AFM	3.514	41
CeMgGa	Θ	3.1	AFM	3.897	42
CeAuGe		10.9	$_{\rm FM}$	3.968	43
$\mathrm{CePd}_2\mathrm{As}_2$. ◆	14.7	AFM	4.268	16
CeAuSn	►	4.4	AFM	3.858	44, 45
CeAgSn	4	6.45	AFM	3.855	45
$\mathrm{Ce}_2\mathrm{Zn}_6\mathrm{Ge}_3$	•	7.2	AFM	4.116	46
CeCuSi		15.5	${ m FM}$	3.977	47
CeCuGe		10	${ m FM}$	3.945	48
$\mathrm{CeAu}_{2}\mathrm{Ge}_{2}$		14.5	AFM	4.367	15
$\rm Ce_2Pd_2In$	Δ	4.5	AFM	4.075	49
$\mathrm{CeRu}_{2}\mathrm{Ge}_{2}$	V	8.7	$_{\rm FM}$	4.256	50
${\rm CeNi_2As_2}$	●	4.8	AFM	4.081	51
CeCuSn	Ð	8.6	AFM	3.924	52
$CeRh_3B_2$		115	${ m FM}$	3.096	17, 18
${ m CeIr_3Si_2}$	♦	4.1	AFM	3.607	53
$\mathrm{CeRu}_{2}\mathrm{Ga}_{2}\mathrm{B}$		16.3	${ m FM}$	4.187	54
CeSbTe	A	2.75	AFM	4.37	55
CeSi	▼	5.6	AFM	3.76	56
$\mathrm{CeAg_2Ge_2}$		4.6	AFM	4.301	57
CeScGe		47	AFM	3.864	58
${\rm Ce}_3{\rm Cu}_4{\rm Si}_4$		10.4	AFM	3.922	59
$Ce_3Pt_{23}Si_{11}$	0	0.44	FM	5.95	19, 20
$\mathrm{CeIr}_3\mathrm{Ge}_7$	*	0.63	AFM	5.724	current wor

TABLE II. Non-Kondo Ce compounds. Symbols represent the legend used for Fig. 4(b).

structure near the Fermi surface of Ce and Yb analogues play an important role with regards to magnetism.

IV. CONCLUSIONS

In summary, CeIr₃Ge₇ in particular and more generally the 1-3-7 family of magnetic compounds provide a fertile ground for exploring magnetic correlations and the competition among various energy scales (RKKY, Kondo, CEF) which could result in quantum critical regimes. In addition, their rhombohedral structure allows for very weak coupling between the Ce atoms in a good metallic environment, similar to what was observed in YbPt₂Sn [29]. This is an excellent precondition for metallic magnets that can be used for adiabatic demagnetization cooling below 2 K, instead of insulating paramagnetic salts.

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APPENDIX

Room temperature powder patterns were collected using a Bruker D8 x-ray diffractometer using Cu K α radiation. The x-ray patterns were refined using TOPAS software. For single crystal x-ray refinement, fragments of CeIr₃Ge₇ were obtained by cutting larger crystals to an appropriate size. These fragments were mounted onto glass fibers using epoxy and then mounted onto a Bruker D8 Quest Kappa single crystal x-ray diffractometer equipped with an I μ S microfocus source $(\lambda = 0.71073 \text{ Å})$ operating at 50 kV and 1 mA, a HELIOS optics monochromator, and a CMOS detector. The collected data were corrected for absorption using the Bruker program SADABS (multi-scan method). The crystal structure of CeIr₃Ge₇ was solved using direct methods in SHELXS2013 [61] and all atomic sites were refined anisotropically using SHELXL2014 [62]. The orientation along the a and c axes in the hexagonal setting of CeIr₃Ge₇ single crystals were determined by the backscattering x-ray Laue method.

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