First-order magnetization process as a tool of magnetic-anisotropy determination: Application to the uranium-based intermetallic U₃Cu₄Ge₄

D. I. Gorbunov,^{1,2,*} M. S. Henriques,^{2,3} A. V. Andreev,² Y. Skourski,¹ M. Richter,⁴ L. Havela,⁵ and J. Wosnitza^{1,6}

¹Dresden High Magnetic Field Laboratory (HLD-EMFL), Helmholtz-Zentrum, Dresden-Rossendorf, D-01314 Dresden, Germany

²Institute of Physics, Academy of Sciences of the Czech Republic, Na Slovance 2, 182 21 Prague, Czech Republic

³CCTN, Instituto Superior Técnico, Universidade de Lisboa, Estrada Nacional 10, 2695-066 Bobadela LRS, Portugal

⁴IFW Dresden e.V., P.O. Box 270116, D-01171 Dresden, Germany

⁵Department of Condensed Matter Physics, Faculty of Mathematics and Physics, Charles University in Prague, Ke Karlovu 5, 121 16 Prague, Czech Republic

⁶Institut für Festkörperphysik, Technische Universität Dresden, D-01062 Dresden, Germany

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Uranium-based intermetallic compounds often display very strong magnetic anisotropies, the energy of which is usually not directly accessible by common experimental methods. Here, we report on static- and pulsed-field studies of $U_3Cu_4Ge_4$. This material orders ferromagnetically at $T_C = 73$ K with the easy magnetization direction along the *a* axis and a strong *bc*-plane anisotropy. The magnetization measured for fields along the hard *b* direction displays a first-order magnetization process that can be described well by use of a phenomenological theory yielding anisotropy constants up to the sixth order. This phenomenological description, working excellently for $U_3Cu_4Ge_4$, may also be applied for other uranium-based compounds.

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I. INTRODUCTION

The 5 *f* states in uranium intermetallic compounds typically form narrow bands with dispersions comparable to those of the late 3*d* metals. Thus, the 5 *f* bandwidth, the screened intraatomic $f \cdot f$ Coulomb interaction, the spin-orbit interaction, and the exchange interaction are all on a similar energy scale, which results in a rich variety of electronic ground states. Noteworthy examples of this variety include the heavyfermion superconductivity in UPt₃ [1–3] and UBe₁₃ [4–6], pressure-induced superconductivity coexisting with ferromagnetic order in UGe₂ [7–9], their coexistence at ambient pressure, reentrant superconductivity in URhGe [10–12], and hidden order in URu₂Si₂ [13–15]. A crucial role in the formation of such complex phases is played by the hybridization of the uranium 5 *f* electrons with *s*, *p*, and *d* electrons of ligand atoms.

Hence, the strength of the 5f-3d hybridization is crucial for the magnetism of uranium-iron intermetallic compounds. The 5 f bands are broadened by hybridization, which reduces the U or Fe magnetic moments or even prevents magnetic order. For instance, strong 5 f-3d hybridization is the reason for the Pauli paramagnetism without long-range magnetic order observed in UFeAl [16] and UFeGe [17] despite the large moments of free U and Fe atoms or ions. In contrast to these extreme examples, there exist compounds with ordered magnetic moments within only one of the sublattices. A material that is representative of this behavior and motivated the present work is U₃Fe₄Ge₄, crystallizing in the orthorhombic crystal structure of Gd₃Cu₄Ge₄ type (space group *Immm*) [18]. U₃Fe₄Ge₄ is an itinerant ferromagnet below $T_{\rm C} = 18$ K [19,20]. The magnetic order originates entirely from the U sublattice, while the Fe atoms do not carry any ordered magnetic moment observable by ⁵⁷Fe Mössbauer spectroscopy. The average moment per U atom amounts to $0.4 \mu_B$ at 2 K. U₃Fe₄Ge₄ displays a strong uniaxial anisotropy with the *a* axis being the easy magnetization direction and an anisotropy field between the *a* axis and *bc* plane of about 60 T. However, a strong anisotropy is also present in the *bc* plane.

The prominent role of the 5f-3d hybridization provides a versatile tool to tune the magnetism in the $U_3T_4G_4$ compounds (*T* is a transition metal). The strength of such hybridization can be affected by changing the transition metal. In our case, Fe with an unfilled 3d shell may be replaced by Cu with the 3d shell filled up. 3d bands, located away from the Fermi level, lead to a reduced 5f-3d hybridization (the 5fstates remain pinned at the Fermi level), which should support the formation of 5f moments and may also enhance the magnetic-ordering temperature. In this sense, $U_3Fe_4Ge_4$ and $U_3Cu_4Ge_4$ are particularly interesting since in both systems ordered magnetic moments are found only on the U sublattice. This should make it possible to study the effect of the *T* element on the 5f states without dealing with an additional magnetic sublattice.

The electronic and magnetic properties of $U_3Cu_4Ge_4$ have previously been examined on polycrystals. A magnetic phase transition has been found at about 70 K, as follows from magnetization and resistivity [21,22] and from neutrondiffraction measurements [23]. Below this T_C , $U_3Cu_4Ge_4$ is a collinear ferromagnet with the U magnetic moments lying along the *a* axis as in the case of $U_3Fe_4Ge_4$. The moment per uranium atom, $1.71 \mu_B$, exceeds the value of $0.4 \mu_B$ of $U_3Fe_4Ge_4$ substantially and points to considerably less-hybridized 5f states in $U_3Cu_4Ge_4$. It should be noted that the related compound UCuGe with a stoichiometry close to $U_3Cu_4Ge_4$ crystallizes in a hexagonal structure and has an antiferromagnetic ground state [24].

While general information on the structural, electronic, and magnetic properties of $U_3Cu_4Ge_4$ is available, we are aware of no detailed data on the magnetic anisotropy, which requires single-crystal studies. Particularly important are single crystals

^{*}Corresponding author: d.gorbunov@hzdr.de

of uranium compounds due to their very strong anisotropy, assumed to be due to the directional bonding of the 5fstates (two-ion hybridization-induced anisotropy [25,26]). Furthermore, investigations of strongly anisotropic materials require high magnetic fields. For these reasons, we studied the properties of U₃Cu₄Ge₄ on a single crystal in static and pulsed magnetic fields up to 60 T by use of electricalresistivity, specific-heat, and magnetization measurements. The most intriguing finding is the observation of a firstorder magnetization process that makes it possible to deduce anisotropy constants by means of a phenomenological model.

II. EXPERIMENTAL AND NUMERICAL METHODS

A U₃Cu₄Ge₄ single crystal was grown by a modified Czochralski method in a triarc furnace from a stoichiometric mixture of the pure elements (99.9% U, 99.99% Cu, and 99.99% Ge) on a water-cooled copper hearth under protective argon atmosphere. A tungsten rod was used as a seed, and the pulling speed was varied between 10 and 20 mm/h. The diameter of the obtained single crystal was 4 mm, the length 30 mm. The crystal structure was determined by standard x-ray diffraction using Cu radiation on a part of the single crystal crushed into fine powder. The refined lattice parameters of the orthorhombic unit cell are a = 4.277(2) Å, b = 6.576(3) Å, and c = 13.934(6) Å. Backscattered Laue patterns were used to verify the quality of the crystal and to orient it along the [100] (*a*), [010] (*b*), and [001] (*c*) axes for magnetization measurements.

Single-crystal x-ray diffraction was measured at 300 K in order to check the structural model. A crystal of dimensions $0.06 \times 0.05 \times 0.02 \text{ mm}^3$ was glued to the top of a glass needle, mounted on a goniometer head, and placed on a four-circle diffractometer (Agilent Xcalibur Gemini Ultra) equipped with a Mo anode, Mo-enhanced collimator, graphite monochromator, and CCD detector. The software CRYSALIS [27] was used to collect and reduce the data. A Gaussian integration method was applied for the absorption correction, using the crystal shape and dimensions optimized by the program X-SHAPE [28]. The software SUPERFLIP [29] was employed for structure solution and the JANA2006 package [30] for structure refinement. The refinement confirmed the correctness of the structural model with an *R* factor converging to $R_{obs} = 4.3\%$. The lattice parameters obtained are a = 4.270(4) Å, b = 6.565(6) Å, and c = 13.921(9) Å, in good agreement with those determined from powder diffraction. Refined atomic positions and atomic displacement parameters (ADPs) are presented in Table I.

Temperature and field dependences of the magnetization along the principal crystallographic directions of a 30 mg single-crystal sample were measured using a commercial extraction magnetometer [physical properties measurement system (PPMS), Quantum Design] in fields up to 14 T.

The PPMS was also used to measure the specific heat by use of the relaxation method and the electrical resistivity using a four-point method as functions of temperature. The geometry of the electrical contacts did not allow us to determine the precise orientation of the current with respect to crystallographic directions.

The high-field magnetization was measured at fixed temperatures between 2 and 60 K in pulsed magnetic fields (pulse

TABLE I. Atomic coordinates, equivalent isotropic displacement parameters, and their estimated standard deviations from singlecrystal refinement for $U_3Cu_4Ge_4$. U_{eq} is defined as one-third of the trace of the orthogonalized U_{ij} tensor.

Atom	Wyckoff position	x	у	z	$U_{\rm eq}({\rm \AA}^2)$
U1	2a	0	0	0	0.0019(4)
U2	4 <i>j</i>	0.5	0	0.3701(7)	0.0031(4)
Cu	81	0	0.3117(4)	0.3321(7)	0.0062(1)
Ge1	4h	0	0.1933(4)	0.5	0.0027(8)
Ge2	4i	0	0	0.2145(2)	0.0030(6)

duration 20 ms) up to 60 T at the Dresden High Magnetic Field Laboratory. The magnetization was measured by the induction method using a coaxial pickup coil system. A detailed description of the high-field magnetometer is given in Ref. [31].

III. RESULTS AND DISCUSSION

A. Low-field magnetization, specific heat, and resistivity

Figure 1(a) shows the temperature dependences of the magnetization, M, measured along the principal crystallographic directions of U₃Cu₄Ge₄ in a field of 5 T. The anisotropy is clearly evident. In particular, at low temperatures, the magnetization for field along the *a* axis is significantly larger than for fields in the *bc* plane. The onset of magnetic order is signaled by the pronounced increase of M for H along the *a* axis when cooling below about 100 K and from the maxima of M for H along the *b* and *c* axes at about 75 K. The magnetization measured in a lower field, 0.02 T, allows us to locate the phase transition at 74 K [inset in Fig. 1(a)]. Although the transition looks rather abrupt and could be classified as being first order, the specific heat displays a typical λ -type anomaly [Fig. 1(b)] which suggests that it is of second order.

The specific heat, *C*, and the electrical resistivity, ρ , provide additional information on the onset of magnetic order [Figs. 1(b) and 1(c)]. The pronounced anomaly in *C* reflects the transition into the ordered state and can be used to determine the Curie temperature $T_{\rm C} = 73 (\pm 0.3)$ K. This value should be regarded as correct since it was obtained in zero magnetic field. This is in good agreement with earlier reported values, 67 and 71 K, from magnetization and electrical-resistivity measurements [21,22], and 69 K, from neutron-diffraction data [23].

The specific heat can be analyzed using three terms:

$$C = C_{\rm el} + C_{\rm ph} + C_{\rm magn},\tag{1}$$

where $C_{\rm el} = \gamma T$ is the electronic, $C_{\rm ph} = \beta T^3$ the phonon, and $C_{\rm magn}$ the magnetic contribution. From the low-temperature linear fit of C/T vs T^2 [orange line in the inset in Fig. 1(b)] $\gamma = 92 \text{ mJ mol}^{-1} \text{ K}^{-2}$, i.e., $\gamma = 31 \text{ mJ (mol U)}^{-1} \text{ K}^{-2}$, and $\beta = 0.00125 \text{ J mol}^{-1} \text{ K}^{-4}$. The latter corresponds to a Debye temperature of $\Theta_{\rm D} = 258 \text{ K}$. Between 2 and 25 K the specific heat can be well described using the formula

$$C(T) = \gamma T + \beta T^3 + \delta T^{1/2} \exp\left(-\frac{\Delta_1}{T}\right), \qquad (2)$$



FIG. 1. Temperature dependence of (a) the magnetization, M, for a field of 5 T applied along the principal crystallographic directions; (b) the specific heat, C, and (c) the electrical resistivity, ρ , of U₃Cu₄Ge₄. In (b) and (c), the black symbols represent the experimental data, and the orange lines are fits (see text for details). The insets in (a), (b), and (c) show the low-temperature dependences of M in a field of 0.02 T applied along the a axis, C/T, and the temperature derivative of ρ , $d\rho/dT$, respectively.

where the last term takes into account the contribution of magnons with an energy gap, Δ_1 . When using the γ and β values as extracted above, we obtain $\Delta_1 = 41$ K and $\delta =$ $10 \,\mathrm{J}\,\mathrm{mol}^{-1}\,\mathrm{K}^{-3/2}$. In order to estimate the magnetic entropy connected with the phase transition, the nonmagnetic part of the specific heat, $C_{\rm el} + C_{\rm ph}$, was estimated up to 300 K. For $C_{\rm el}$ the above-mentioned γ value was used, while $C_{\rm ph}$ was approximated by the Debye integral with $\Theta_{\rm D}$ as above. The specific heat determined in this way is shown by the orange line in Fig. 1(b). It should be noted that even such a simple model provides good agreement with the experimental data. The magnetic entropy estimated as the difference between the experimental data and calculated curve practically saturates just above $T_{\rm C}$ and amounts to S = $5.3 \,\mathrm{J}\,\mathrm{mol}^{-1}\,\mathrm{K}^{-1} \approx R\ln(2)$ per mol U. This implies that the U moments in U₃Cu₄Ge₄ can be taken as local moments, in contrast to the isostructural U₃Fe₄Ge₄, which has a magnetic entropy of only about 0.17Rln(2) per mol U [20].

The electrical resistivity of $U_3Cu_4Ge_4$ increases with temperature in the magnetically ordered state [Fig. 1(c)].

Between 2 and 40 K, the data follow the dependence

$$\rho = \rho_0 + AT^2 + bT \left(1 + \frac{2T}{\Delta_2} \right) \exp\left(-\frac{\Delta_2}{T} \right), \quad (3)$$

where the first term is the residual resistivity, the second term describes the electron-electron scattering, and the third term describes the electron scattering on magnetic excitations with a gap Δ_2 (the fit is shown by the orange line in the lower part of the figure) [32]. This expression neglects the electron-phonon scattering, which has been assumed to be small in the given temperature range. The fit to the experimental data yields $\rho_0 = 84 \,\mu\Omega \,\mathrm{cm}$, $A = 0.022(2) \,\mu\Omega \,\mathrm{cm} \,\mathrm{K}^{-2}$, $b = 3.77 \,\mu\Omega \,\mathrm{cm} \,\mathrm{K}^{-1}$, and $\Delta_2 = 50 \,\mathrm{K}$. Δ_2 represents the energy gap in the spin-wave excitation spectrum, corresponding to the magnetic anisotropy between the two easiest-magnetization directions. The obtained value is in reasonable agreement with the energy gap found from the specific heat, $\Delta_1 = 41 \,\mathrm{K}$.

At high temperatures, $\rho(T)$ exhibits a weak but noticeable negative temperature coefficient. Such behavior has been found for a number of compounds (typically U-based materials), which exhibit large U moments and contribute to the resistivity by spin-disorder scattering. In case this scattering is very strong, the resistivity is like that for metals with strong crystallographic disorder. For those, quantum interference phenomena (weak localization, Ref. [33]) can increase the resistivity towards low temperatures. Such a situation was discussed for the case of UGa₂ [34], in which large magnetic moments $(3 \mu_B/U)$ in the ferromagnetic state and a very low $\gamma = 10 \text{ mJ mol}^{-1} \text{ K}^{-2}$ do not allow speculations about the Kondo effect as the source of $d\rho/dT < 0$. U₃Cu₄Ge₄ can be seen as an analog; the resistivity exceeds 300 $\mu\Omega$ cm (similar to that of UGa₂), which indeed should lead to negative $d\rho/dT$ by applying the Mooij criterion [35] which predicts the gradual development of a negative $d\rho/dT$ derivative for resistivities exceeding 100-150 $\mu\Omega$ cm. The conspicuous similarity with UGa₂ suggests a similar nature of the magnetic excitations in both materials. The common feature is that both compounds exhibit one hard-magnetization direction whereas a moderate anisotropy between the other two directions allows excitations of magnons.

B. High-field magnetization

In order to further study the strong magnetic anisotropy of $U_3Cu_4Ge_4$ reflected in the magnetization shown in Fig. 1(a), we performed a comprehensive investigation in static (up to 14 T) and pulsed (up to 60 T) magnetic fields. Magnetization data measured at 2 and 60 K in fields up to 14 T applied along the principal crystallographic directions are shown in Fig. 2. These data confirm that the *a* axis is the easy magnetization direction. We find a spontaneous magnetic moment, $M_s =$ $5.0 \mu_B/f.u.$ (formula unit) at 2 K, corresponding to 1.67 μ_B per uranium atom. In a powder neutron-diffraction study, the uranium magnetic moments were indeed found to be oriented along the *a* axis in U₃Cu₄Ge₄ and a value of $1.71 \mu_B$ was reported for both crystallographically inequivalent sites of the uranium atoms [23]. The inset in Fig. 2 shows the temperature dependence of M_s , as determined by means of Arrott plots. The spontaneous moment decreases monotonically towards zero at $T_{\rm C}$.



FIG. 2. Magnetization as function of magnetic field applied along the *a*, *b*, and *c* axes of a $U_3Cu_4Ge_4$ single crystal at 2 and 60 K. The inset shows the temperature dependence of the spontaneous magnetic moment determined by the Arrott-plot method.

As is typical for uranium intermetallics, the magnetic anisotropy of $U_3Cu_4Ge_4$ is very strong. This is true not only for the anisotropy between the *a* axis and the *bc* plane, but also within the *bc* plane, where the *b* axis is the hardest magnetization direction. At 60 K, the *M*(*H*) curve for field along the *b* axis displays a field-induced magnetic transition around 12 T. Since the transition occurs along the hard magnetization direction, it is a first-order magnetization process (FOMP) [36]. It is reasonable to assume that at lower temperatures the FOMP occurs at higher magnetic fields. Additionally, the magnetization curve along the *c* axis at 2 K displays a small positive curvature, which might be a precursor of a transition in a higher field.

Figure 3(a) shows magnetization of U₃Cu₄Ge₄ measured at 2 K in pulsed magnetic fields up to 60 T. The data obtained in static fields are shown as well (closed symbols). A hysteretic FOMP occurs along the b axis at a critical field of $\mu_0 H_{cr} = 25$ T. This is a so-called type-II FOMP since immediately above the transition the magnetization does not reach the easy axis value [36]. Rather, it grows steadily up to the highest available field. The magnetization jump at the transition amounts to $\Delta M = 2.5 \,\mu_B/f.u.$ Further, the magnetization for fields aligned along the c axis exhibits a noticeable curvature change with inflection in the range of 15-20 T, which might indicate a gradual rotation of the magnetic moments. The magnetization along the easy a axis grows monotonically with increasing field, most likely due to an additional splitting of spin-up and spin-down bands of itinerant electrons in the magnetic field.

The magnetization dependence of $U_3Cu_4Ge_4$ in applied magnetic fields strongly contrasts with that of the antiferromagnets $UCu_{0.95}Ge$ and UCuGe [37–39]. These materials



FIG. 3. Magnetization vs magnetic field applied along (a) the a, b, and c axes at 2 K; (b) the b axis and (c) the c axis of U₃Cu₄Ge₄ between 2 and 60 K. The symbols represent static-field measurements, the solid lines pulsed-field measurements. The dashed line in (a) represents a description of the b-axis magnetization curve by use of Eq. (4). The inset shows the temperature dependence of the FOMP critical field and of the magnetization jump at the FOMP.

also display field-induced magnetic transitions but they are observed at much higher fields and correspond to a change of the magnetic structure from antiferromagnetic to ferromagnetic, whereas the compound studied in the present work is ferromagnetic already below the transition.

The magnetization for different fixed temperatures and fields applied along the *b* and *c* axes is shown in Figs. 3(b) and 3(c). The FOMP critical field decreases with increasing temperature. Simultaneously, the transition becomes less pronounced and finally disappears above 60 K. The temperature dependences of H_{cr} and ΔM are shown in the inset of Fig. 3. For fields applied along the *c* axis the curvature change is no longer observed above 20 K; instead the magnetization at 40 and 60 K shows first a linear field dependence and a decreasing slope at higher fields.

Since the 5f electronic states of uranium intermetallic compounds are itinerant, they also participate in bonding. This

leads to occupancy of those orbitals which are oriented towards the nearest actinide neighbor, providing the best bonding conditions. The magnetic moments are then perpendicular to the bonding direction. The bonding anisotropy of the 5f electronic shell in combination with strong spin-orbit coupling leads to a very strong two-ion anisotropy (see, e.g., Refs. [24,26,40,41]). Some exceptions are UFe₂ [42] and U_2Fe_3Ge [43]. The reason for the low anisotropy observed in UFe₂ might be a mutual cancellation of intrinsic and magnetoelastic contributions to the anisotropy. The anisotropy constants and anisotropy fields of most uranium intermetallics cannot be measured directly. They are typically estimated by high-field extrapolations of the magnetization measured in fields applied along the easy and hard directions. For U₃Cu₄Ge₄, the unusual finding of a FOMP for fields applied along the hardest direction provides a way to describe the magnetic anisotropy quantitatively. This anisotropy can be evaluated with much higher precision than would be possible by the usual extrapolation.

A proper description of a type-II FOMP requires anisotropy constants at least up to sixth order. The equilibrium equation for the field-dependent magnetization containing a FOMP for materials with uniaxial magnetic symmetry, as appropriate for $U_3Cu_4Ge_4$, is given by

$$HM_s = 2K_1 \frac{M}{M_s} + 4K_2 \left(\frac{M}{M_s}\right)^3 + 6K_3 \left(\frac{M}{M_s}\right)^5,$$
 (4)

where K_1 , K_2 , and K_3 are the second-, fourth-, and sixth-order anisotropy constants, respectively [36]. Figure 3(a) shows a fit using Eq. (4) describing the magnetization data for fields applied along the *b* axis at 2 K. The initial almost linear part of the fit is followed by an S-like shape. The irreversible rotation of the magnetization occurs in this field interval as a first-order transition between inequivalent magnetization states. The fit was done in such a way that the areas enclosed between the experimental and calculated curves on both sides of the transition are equal. This procedure was performed for all temperatures where the FOMP is observed. The obtained temperature dependences of the anisotropy constants are plotted in Fig. 4.

The rather large anisotropy constants, $K_1 = 17.6 \text{ MJ/m}^3$, $K_2 = -33 \text{ MJ/m}^3$, and $K_3 = 26 \text{ MJ/m}^3$, are found at 2 K, which characterizes U₃Cu₄Ge₄ as a system with strong magnetic anisotropy. K_1 remains positive, as expected for a uniaxial magnetic anisotropy. On the other hand, K_2 is negative, giving the magnetization curve a positive curvature upon approaching the FOMP. Finally, K_3 is positive which produces a negative curvature above the transition. At low temperatures, the relation $|K_2| > K_3 > K_1$ holds. All three constants decrease gradually with temperature and around 40 K the relation changes to $|K_2| > K_1 > K_3$. This is in accordance with the common behavior that higher-order anisotropy constants fall off more rapidly with increasing temperature than lower-order constants (see also the inset in Fig. 4) [44,45].

The large uniaxial magnetic anisotropy of U₃Cu₄Ge₄ leads to strong magnetic hysteresis, as shown in Fig. 5. The coercive field, $\mu_0 H_c$, attains more than 0.4 T at 2 K and rapidly falls off with temperature (inset in Fig. 5). The virgin curve at



FIG. 4. Second-, fourth-, and sixth-order anisotropy constants of $U_3Cu_4Ge_4$ as functions of temperature. The inset shows the temperature dependence of the anisotropy constants normalized at 2 K.

2 K indicates that the sample reaches a single-domain state abruptly at about 0.2 T when domain-wall motion is activated. However, $\mu_0 H_c = 0.4$ T exceeds this value after a complete hysteresis cycle. Therefore, the dominant mechanism of the magnetic hysteresis of U₃Cu₄Ge₄ is the domain nucleation



FIG. 5. Hysteresis loops for fields applied along the *a* axis of $U_3Cu_4Ge_4$ between 2 and 20 K. The inset shows the temperature dependence of the coercive field.

lag. The practically instantaneous remagnetization at $H = H_c$ supports this suggestion.

IV. SUMMARY AND CONCLUSIONS

We have carried out a comprehensive study of the magnetic properties of single-crystalline $U_3Cu_4Ge_4$ in conjunction with phenomenological modeling of the magnetic anisotropy. A first-order magnetization process was observed in large applied magnetic fields which allowed us to determine the magnetocrystalline anisotropy constants. For the case of uranium intermetallic compounds, this is a rare and favorable circumstance, as normally the very high anisotropy of such compounds is not accessible with most experimental methods.

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