Magnetooelastic phenomena in antiferromagnetic uranium intermetallics: the UAu₂Si₂ case

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Thermal expansion, magnetostriction and magnetization measurements under magnetic field and hydrostatic pressure were performed on a UAu₂Si₂ single crystal. They revealed a large anisotropy of magnetoelastic properties manifested by prominent length changes leading to a collapse of the unit-cell volume accompanied by breaking the fourfold symmetry (similar to that in URu₂Si₂ in the hidden-order state) in the antiferromagnetic state as consequences of strong magnetooelastic coupling. The magnetostriction curves measured at higher temperatures confirm a bulk character of the 50 K weak ferromagnetic phase. The large positive pressure change of the ordering temperature predicted from Ehrenfest relation contradicts the more than an order of magnitude smaller pressure dependence observed by the magnetization and thermal-expansion measured under hydrostatic pressure. A comprehensive magnetic phase diagram of UₐAu₂Sn₂ in magnetic field applied along the c axis is presented. The ground-state antiferromagnetic phase is suppressed by a field-induced metamagnetic transition that changes its character from the second to the first order at the tricritical point.

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I. INTRODUCTION

The anharmonic lattice vibrations due to the asymmetric bonding potential lead to increasing equilibrium interatomic distances with rising temperature in solids. The corresponding thermal expansion is a monotonously increasing function of temperature. The anisotropy of bonding within the crystal lattice causes the anisotropy of thermal expansion which is manifested by different temperature dependences of the linear thermal expansion (\(\Delta l/l_i\)) along the different crystallographic axes, \(i\). The thermal expansion of metals includes also a conduction-electron contribution. This plays a considerable role at low temperatures where the phonon term almost vanishes.

The magneto-structural coupling, reflecting the interplay between the spin and lattice degrees of freedom, brings additional contributions to the thermal expansion in magnetic compounds. The magnetocrystalline anisotropy leads to anisotropic magnetic contributions to the thermal expansion. Magnetic materials then exhibit unusual thermal-expansion behavior especially in a magnetically ordered state.

The thermal expansion, similar to the specific heat, is thus a useful probe for investigations of thermodynamic phenomena in magnetic materials (without applying magnetic field). The specific heat has only bulk character, whereas the thermal expansion enables us to study also the anisotropy of thermodynamic properties.

The spatially extended uranium 5f-electron wave functions in solids considerably interact with the overlapping 5f orbitals of the nearest-neighbor U ions and the 5f-electron states hybridize with valence-electron states of non-uranium ligands (5f-ligand hybridization) and the 5f electrons even participate in bonding. The exchange interactions that are coupling the uranium 5f-electron magnetic moments in U antiferromagnets are strongly anisotropic. The direct exchange interactions are due to the 5f–5f orbitals overlap. The anisotropy of these, typically ferromagnetic (FM), interactions and as well as the magnetocrystalline anisotropy are determined by the arrangement of the nearest-neighbor U ions in the lattice. The antiferromagnetic (AFM) interactions in U compounds are usually mediated by the anisotropic 5f-ligand hybridization. The magnetoelastic coupling then produces highly anisotropic magnetic contributions to the thermal expansion and magnetostriction, especially in U antiferromagnets.

The anisotropy of magnetoelastic phenomena is a subject of numerous papers on U magnetics. Most frequently they have been dedicated to the intriguing properties of URu₂Si₂, the most thoroughly studied uranium compound in the last three decades. The volume of this compound reduces considerably below the hidden-order transition temperature \(T_{\text{HO}}\) as manifested by a sharp positive peak in the thermal-expansion coefficient at \(T_{\text{HO}}\). The volume reduction of the tetragonal structure is due to the basal-plane shrinkage. The simultaneous lattice
expansion along the c axis is too small to compensate the negative basal-plane effect.\textsuperscript{4–7} The possibility of a slight orthorhombic distortion of the tetragonal lattice at temperatures below $T_{\text{HTO}}$ plays an important role in the physics of URT$_2$Si$_2$. We will come back to this point in the Discussion section.

The influence of anisotropic exchange interactions on the anisotropy of thermal expansion in U antiferromagnets is manifested by the magnetoelastic behavior of two other UT$_2$X$_2$ ($\text{T} - \text{T} - \text{T}$ transition metal, $\text{X} - \text{p}$-electron metal) compounds with the tetragonal ThCr$_2$Si$_2$ structure, UC$_2$Si$_2$\textsuperscript{19} and UN$_2$Si$_2$\textsuperscript{10} and several antiferromagnets from the family of hexagonal UTX compounds crystallizing in the ZrNiAl structure. In the both structures the nearest U-U neighbors are located in the basal plane where the U magnetic moments are coupled ferromagnetically. All these compounds exhibit the strong uniaxial anisotropy fixing the U moments to the c axis, which is the easy magnetization direction. The AFM structures in these materials are built of the FM basal-plane layers antiferromagnetically coupled along the c axis. The thermal expansion below the Neel temperature ($T_N$) in these antiferromagnets (similar to URT$_2$Si$_2$ below $T_{\text{HTO}}$) is strongly anisotropic as well as the magnetostriction accompanying field-induced metamagnetic transitions from the AFM to paramagnetic state. The corresponding $a$- and $c$-axis linear thermal expansions ($\Delta l/l|_a$ and ($\Delta l/l|_c$, respectively have in all cases opposite signs. The volume thermal expansion calculated according to

$$\Delta V/V = 2 \cdot (\Delta l/l|_a + (\Delta l/l|_c)$$

for the UT$_2$X$_2$ compounds is small as a result of compensation of the opposite-sign linear expansions. The linear magnetostrictions $\lambda_a$ and $\lambda_c$ accompanying a metamagnetic transition are also of opposite signs leading to small volume magnetostriction. However, they have opposite polarities with respect to the corresponding-direction of thermal expansions. In fact, the magnetic contributions to thermal expansion of an antiferromagnet below $T_N$ are suppressed by the opposite polarity corresponding to magnetostrictions caused by the metamagnetic transition.

URT$_2$Si$_2$ belongs to the family of UT$_2$Si$_2$ compounds which adopt the tetragonal ThCr$_2$Si$_2$ structure (URT$_2$Si$_2$ and UPT$_2$Si$_2$ crystallize in the CaBe$_2$Ge$_2$ structure\textsuperscript{11}). These compounds exhibit a spectrum of physical properties ranging from Pauli paramagnets (UF$_2$Si$_2$,\textsuperscript{12} URE$_2$Si$_2$, and UO$_2$Si$_2$\textsuperscript{13}) to magnetically ordered systems which are mostly complex and either AFM (UCr$_2$Si$_2$,\textsuperscript{14} UC$_2$Si$_2$,\textsuperscript{15} UN$_2$Si$_2$,\textsuperscript{16} UR$_2$Si$_2$,\textsuperscript{17} UP$_2$Si$_2$,\textsuperscript{17,18} UR$_2$Si$_2$,\textsuperscript{19,13} and UPT$_2$Si$_2$\textsuperscript{13}), or FM (UMn$_2$Si$_2$\textsuperscript{12}). UC$_2$Si$_2$\textsuperscript{15,20–22} exhibits a FM ground state with an additional AFM phase at higher temperatures. An exceptional case among them is the well-known URT$_2$Si$_2$\textsuperscript{23} exhibiting the hidden-order transition. The magnetism of URT$_2$Si$_2$ was for many years left unclear mainly due to metallurgical difficulties.\textsuperscript{11}

Quite recently we have successfully prepared a URT$_2$Si$_2$ single crystal and commenced systematic investigations of its intrinsic properties. The results obtained by measurements of magnetization, specific heat and electrical resistivity\textsuperscript{24} followed by neutron diffraction\textsuperscript{25} and Si-NMR\textsuperscript{26} experiments corroborate the conclusion about the ground state of URT$_2$Si$_2$ as an uncompensated antiferromagnet, contrary to previous reports on polycrystals.\textsuperscript{13,27–30} URT$_2$Si$_2$ undergoes a FM-like transition at 50 K (referred to as $T_2$) followed by another magnetic phase transition ($T_m$) around 20 K. All our previous measurements show a large magnetocrysalline anisotropy with the direction of magnetic moments along the c axis. The propagation vector is $(2/3, 0, 0)$ and the magnetic structure can be described as a stacking sequence $(+ + -)$ of the FM ac plane sheets along the c axis.\textsuperscript{25} The specific-heat measurements point to an enhanced value of the Sommerfeld coefficient $\gamma \sim 180 \text{ mJ K}^{-2} \text{ mol}^{-1}$. Now, we investigated the URT$_2$Si$_2$ single crystal by use of thermal-expansion, magnetostriction and magnetization measurements up to high magnetic fields and under hydrostatic pressure. The results of the present paper, are complementary to our previous neutron-diffraction work, confirming the AFM ground state of the compound\textsuperscript{25} and bring evidence for the intrinsic nature of the FM component below $\sim 50 \text{ K}$ previously reported as parasitic in our single-crystal study.\textsuperscript{24} Magnetization measurements in pulsed high magnetic fields helped us to complete the phase diagram of URT$_2$Si$_2$ and revealed signs of the presence of a tricritical point (TCP).

II. EXPERIMENTAL DETAILS

The URT$_2$Si$_2$ single crystal used in this study was prepared using the float-in-zone method in an optical furnace (Crystal Systems Co.) in a similar way as in our previous work.\textsuperscript{24} Nevertheless, to obtain a higher-quality and larger single crystals we have optimized the entire growth process. The initial polycrystalline rod with diameter of 6 mm and length of $\sim 100 \text{ mm}$ was prepared from the starting elements of U (initially 99.9% and consequently purified by the Solid State Electrotransport Method under ultra high vacuum\textsuperscript{31}), Au (99.99%) and Si (99.999%). The rod was subsequently annealed at 1000°C for three days, cut in two parts and placed in the optical furnace, where the shorter bottom part served as a polycrystalline seed and the main larger rod hung from the top as feed material for the growth. The chamber of the optical furnace was evacuated to $\sim 10^{-6} \text{ mbar}$ and the growth itself was done under the protective Ar atmosphere with a flow of 1.5 l min$^{-1}$ in an overpressure of $\sim 0.2 \text{ MPa}$. The power of the lamps in the furnace was adjusted to keep the temperature of the hot zone slightly above the melting point. Both, the seed and feed rod were slowly pulled through the hot zone with a speed of 1 mm h$^{-1}$ and with-
out rotation. The quality of the grown single crystal was checked by the x-ray Laue method and Energy-dispersive x-ray (EDX) analysis.

Length changes were measured using a miniature capacitance dilatometer mounted in PPMS 9 T and PPMS 14 T (Quantum Design Co.) between 2 K and 100 K in magnetic fields up to 14 T. Magnetization measurements in static fields were done using the VSM option (Vibrating Sample Magnetometer) implemented in the PPMS 14 T.

The magnetization in pulsed magnetic fields up to $\sim 58$ T was measured at the Dresden High Magnetic Field Laboratory using a coaxial pick-up coil system. The high-field magnetometer is described in Ref. 33. Absolute values of the magnetization were calibrated using static-field measurements.

The magnetization measurements under hydrostatic pressure were performed in the MPMS SQUID (Quantum Design Co.) magnetometer using a CuBe pressure cell with a liquid pressure medium and a piece of lead as manometer. The linear thermal expansion under hydrostatic pressure was measured in a double-layered CuBe/NiCrAl piston cylinder cell under pressures up to $\sim 2$ GPa, mounted in PPMS 14 T. Manganin manometer and a Daphne 7373 pressure medium were used. Miniature low-temperature strain gauges (Kyowa Co., KFL series) were used to measure the expansion of sample along both principal axes and simultaneously also a reference copper sample in the single loading.

III. RESULTS

A. Thermal-expansion measurements

The $a$- and $c$-axis linear thermal expansions together with the volume expansion calculated according to Eq. (1) are plotted in Fig. 1. The individual data are vertically shifted to set them equal to 0 at $T_m = 20$ K. Below this magnetic phase transition we observe a significant change in the temperature dependence of the thermal expansion in line with our previous reports. 24,25

There obviously is a large anisotropy of the thermal expansion over the entire temperature range. The linear thermal expansion along the $a$ axis shows a continuous decrease from 100 K down to the ordering temperature $T_m = 20$ K, where it bends down rapidly pointing at a large contraction of the unit cell along the $a$ axis ($-1.5 \times 10^{-4}$ between $T_m$ and 2 K). On the other hand, the thermal expansion along the $c$ axis shows a broad minimum around 75 K followed by an increase at lower temperatures. The ordering temperature appears as an inflection point and the $c$-axis expansion below 20 K is $6.6 \times 10^{-5}$. The volume thermal expansion is calculated using Eq. (1). The obtained relative volume change shows a large reduction below $T_m (-2.3 \times 10^{-4}$ between $T_m$ and 2 K) as shown in Fig. 1. The continuous character of the linear thermal expansion at $T_m$ points to a second-order transition.

The linear thermal-expansion coefficients $\alpha_i$ are defined as temperature derivatives of the linear thermal expansion $(\Delta l/l)_i$, i.e., $\alpha_i = d(\Delta l/l)_i/dT$. The calculated linear thermal-expansion coefficients are plotted in Fig. 2 together with the volume thermal-expansion coefficient defined as $\alpha_v = 2\alpha_a + \alpha_c$. It is also useful to determine the temperature dependence of the $c/a$ ratio. We, therefore, define the following temperature coefficient $\alpha_{c/a} = \alpha_c - \alpha_a$. Both quantities are plotted in Fig. 2.

The linear thermal-expansion coefficient for the $c$ axis becomes negative below 75 K where the minimum of its...
relative length change occurs. There are sharp peaks at $T_m$ for all measured curves and a small change of the slope above 50 K visible in $\alpha_a$ coefficient that is projected in the remaining computed quantities. Integration of the $\alpha_{c/a}$ coefficient along the whole temperature range results in the relative temperature dependence of the $c/a$ ratio, see inset in Fig. 1.

The temperature dependence of this ratio is monotonous and decreasing nearly linear above $T_m$. The slope increases below the $T_m$, emphasizing again the prominent contraction of the $a$ axis.

As will be discussed below, ultra-pure samples of the isostructural compound $\text{URu}_2\text{Si}_2$ studied by synchrotron x-ray diffraction show a small orthorhombic distortion when entering the hidden-order state. The size of the distortion and/or sample quality are possible reasons why it was not observed by the thermal-expansion measurements.  

In order to test the presence of a lattice distortion in $\text{UAu}_2\text{Si}_2$ we measured the thermal expansion also along the $a$ axis and [110] direction. The corresponding thermal expansions and the linear thermal-expansion coefficients are plotted in Fig. 3.

We clearly observe an anisotropy of the thermal expansion in the basal plane. An undistorted tetragonal plane would show the same relative length changes for the $a$ axis and for the [110] direction (face diagonal).

The effective Grüneisen parameter is defined as

$$\Gamma_{\text{eff}} (T) = \frac{V_m \alpha_a (T)}{\kappa C (T)},$$

where $V_m$ is the molar volume (5.45 × 10$^{-5}$ m$^3$ mol$^{-1}$ for $\text{UAu}_2\text{Si}_2$ using the unit-cell parameters from our neutron-diffraction work) and $\kappa$ is the compressibility of the material. Calculating $\Gamma_{\text{eff}}$ requires the knowledge of the specific heat $C (T)$. We use data from our previous study.  

$\Gamma_{\text{eff}}$ is expected to be about 2 at higher temperatures where the phonon contribution dominates the volume dependence. The phonon contribution can be neglected at sufficiently low temperatures and the so-called electronic Grüneisen parameter $\Gamma_e$ evolves. The specific heat can be approximated as $C = \gamma T$ at the lowest temperature giving the extrapolated value of $C/T$ to 0K as $\sim 150 \text{ mJ mol}^{-1} \text{ K}^{-2}$ (Ref. 24). A similar linear trend appears in the low-temperature part of the volume thermal-expansion coefficient described by $\alpha_a = a T$. For that purpose, we plot $\alpha_a/T$ vs. $T^2$ in the inset of Fig. 4.

The linear extrapolation to 0K gives $a \approx 2.93 \times 10^{-7} \text{ K}^{-2}$. The electronic Grüneisen parameter can be calculated as $\Gamma_e = (V_m a)/ (\langle \kappa \gamma \rangle)$. We can estimate the pressure dependence of the specific-heat coefficient $\gamma$, which is defined as

$$\frac{d (\ln \gamma)}{dp} = -\kappa \Gamma_e .$$

This results in the negative relative pressure dependence of $d (\ln \gamma)/dp \approx -1.1 \times 10^{-4} \text{ GPa}^{-1}$. Further calculations of the electronic Grüneisen parameter requires the compressibility data. For that reason we measured the compressibility of $\text{UAu}_2\text{Si}_2$ using the strain gauges as part of our hydrostatic pressure study (see Sec. III C). The approximate linear slopes of the length changes of the $a$ and $c$ axis measured up to 1.65 GPa were comparable and resulted in the compressibility of $\kappa = 1.11(6) \times 10^{-11} \text{ m}^2 \text{ N}^{-1}$ at room temperature. This value is two times larger than the value of $0.52 \times 10^{-13} \text{ m}^2 \text{ N}^{-1}$.
Table I. The jumps of the thermal-expansion coefficients $\Delta \alpha_i$ at $T_m$ and the corresponding hydrostatic-pressure and uniaxial-pressure derivatives of $T_m$ predicted using Ehrenfest relation.

|       | $\Delta \alpha_i$ (K$^{-1}$) | $\frac{dT_m}{dP}$ (K GPa$^{-1}$) |
|-------|-----------------------------|----------------------------------|
| $a$   | $2.69 \times 10^{-5}$       | 5.9                              |
| $c$   | $-4.36 \times 10^{-6}$      | -0.9                             |
| Volume| $5.84 \times 10^{-5}$       | 11.1                             |

(Ref. 20) for the isostructural URu$_2$Si$_2$. The electronic Grüneisen parameter obtained under these conditions is 9.6(6). We can consequently compute the temperature dependence of the effective Grüneisen parameter using above value for $\kappa$, see Fig. 4.

The calculated effective Grüneisen parameter $\Gamma_{\text{eff}}$ shows an almost constant temperature dependence at higher temperatures with a value close to 2 and with a shallow minimum around 50 K. $\Gamma_{\text{eff}}$ exhibits a very sharp peak at $T_m$, reaching about 17 and finally $\Gamma_{\text{eff}}$ drops down in the ordered state and steadily increases to the extrapolated value $\Gamma_c \approx 9.6(6)$.

The thermodynamic relation for second-order phase transitions known as Ehrenfest relation allows us to estimate the pressure dependence of the ordering temperature based on the jumps in the specific heat $\Delta C$ and the thermal-expansion coefficient $\Delta \alpha$ at $T_m$. It is defined as

$$\frac{dT_m}{dP} = \frac{\Delta \alpha V_m}{\Delta C/T}, \quad (4)$$

and it can serve as an estimation for the hydrostatic-pressure dependence using the volume thermal-expansion coefficient $\alpha_V$, or for the uniaxial pressure dependences when the linear thermal-expansion coefficients $\alpha_a$ or $\alpha_c$ are used. The jumps of the thermal-expansion coefficients at $T_m$ and the corresponding hydrostatic-pressure and uniaxial-pressure derivatives of $T_m$ predicted using Eq. (4) are shown in Table I.

### B. Magnetization and magnetostriction measurements

In a first stage we measured the magnetization at various temperatures from 2 to 50 K as function of magnetic field up to 14 T applied along the $c$ axis. This was followed by high-field measurements in pulsed magnetic fields up to $\sim 58$ T. The results are shown in Fig. 5. The magnetization data up to 13 K show a low-field inflection point in the hysteric part of the curve labeled as $\mu_0 H_1$. Another step-like feature is present at higher field and is labeled as $\mu_0 H_2$. Unlike the low-field transition, the $\mu_0 H_2$ anomaly can be traced to temperatures above $T_m$ and is clearly distinguishable even at 40 K. The magnetization isotherms in the vicinity of $T_m$ show another field-induced step-like transition at higher fields. Similar transitions were observed in our previous work on a different single crystal.54 There, the anomalies labeled as $\mu_0 H_m$ were, however, much less pronounced probably due to lower crystal quality. The values of characteristic fields at different temperatures have been determined using plots of $d\mu/d\mu_0 H$ vs. $\mu_0 H$ shown in Fig. 6 for results at 2 and 19 K.

Further magnetization measurements in pulsed fields were performed to track the metamagnetic transition $\mu_0 H_m$. The measured magnetization was scaled using the static-field data obtained at 2 K and a small linear background was subtracted to give the correct absolute values. Results of the high-field measurement are plotted in Fig. 7. A clear metamagnetic transition at $\mu_0 H_m$ is visible on all measured isotherms up to 20.1 K. The metamagnetic transition is most probably of spin-flip type40 as one would expect for the system with large magnetocrystalline anisotropy. The low-temperature curves also show significant hystereses ($\Delta \mu_0 H_m$) between the up and down sweeps. The hysteresis narrows with increasing temperature and disappears around 16 K when $\mu_0 H_m \sim 16$ T. The temperature dependence of $\Delta \mu_0 H_m$ is shown in Fig. 12(b).

The longitudinal magnetostriction was measured at various temperatures with magnetic fields up to 9 T ap-
Figure 6. Field derivatives of the magnetization data at 2 and 19 K depicting the determination of $\mu_0 H_1$, $\mu_0 H_2$, and $\mu_0 H_m$. The solid lines correspond to field-up and dotted lines to field-down sweeps.

Figure 7. Magnetization measured in pulsed fields applied along the $c$ axis up to $\approx 58$ T. The curves are consecutively vertically shifted by $0.5 \mu_B$/f.u. The solid lines correspond to field-up and dash-dotted lines to field-down sweeps. The arrows mark the $\mu_0 H_m$ transitions. The gray curve overlapping data at 1.8 K are the static-field data at 2 K up to 14 T shown in Fig. 5.

Figure 8. Longitudinal magnetostriction measured along the $c$ axis. The solid lines correspond to field-up sweeps and dash-dotted lines to field-down sweeps. The curves are vertically shifted for better clarity.

Figure 9. How the fields were determined.

As in the magnetization data, we can clearly observe anomalies at $\mu_0 H_1$ and $\mu_0 H_2$. Figure 9 shows how these fields were determined.

There is a significant change of the shape of the magnetostriction curves when UAl$_2$Si$_2$ crosses $T_m = 20$ K, going from a concave to a convex curvature. We have consequently conducted further measurements of the longitudinal magnetostriction along the $c$ axis up to 14 T at selected temperatures close to $T_m$. These isotherms show the high-field anomaly as a pronounced sharp drop at $\mu_0 H_m$ (see Fig. 10), as determined from the magnetization measurements.

We have also measured the temperature dependence of the magnetization in various fields up to 14 T applied along the $c$ axis. These data agree with our previous results obtained on a different single crystal. We can clearly see the anomaly labeled as $T_m$, determined from the upturn of the magnetization for curves below 5 T and from the peak at higher fields. There is also another transition marked as $T_2$ that can be distinguished only in the low-field data at 0.1 T (see Fig. 11).

The whole set of anomalies observed in the magnetization, magnetostriction and thermal-expansion measurements allows us to construct the magnetic phase diagram as plotted in Fig. 12(a). Fig. 12(b) shows the tempera-
Figure 9. Field derivatives of the magnetostriction data at 2
and 24 K showing the determination of $\mu_0 H_1$ and $\mu_0 H_2$. The
solid lines correspond to field-up scans and dash-dotted lines
to field-down sweeps.

Figure 10. Longitudinal magnetostriction measured with the
fields up to the 14 T applied along the c axis.

ture dependence of the hysteresis of the $\mu_0 H_m$ transition.

C. Hydrostatic pressure study

As our calculations using the Ehrenfest relation pre-
dict a rather dramatic positive effect of hydrostatic pres-
sure on the ordering temperature ($\approx 11.1$ K GPa$^{-1}$) we
wanted to verify this hypothesis. For that purpose,
we measured the magnetization in a field of 0.1 T ap-
pved along the c axis under hydrostatic pressures up to
$\sim 1$ GPa (see Fig. 13). The measured data were cor-
rected for the diamagnetic contribution of the pressure
cell. The shape of the ambient pressure curve differs
from those obtained under pressure. This can be an ef-
effect of a slightly different orientation of the sample in
the pressure cell. Contrary to our prediction, we ob-
serve only a small shift of the transition temperature $T_m$
with applied pressure (see inset of Fig. 13). The tran-
sition temperature $T_m$ again is defined by the upturn of
the magnetization curve. The resulting small ratio of the
pressure change of the ordering temperature is
$\frac{dT_m}{dp} \approx 0.6(1)$ K GPa$^{-1}$. A larger effect is visible in the reduc-
tion of the spontaneous magnetic moment $\mu_{spont}$ with the slope $\frac{d\mu_{spont}}{dp} \approx -0.019(6) \mu_B / (f.u. \text{ GPa})$.
due to magnetic contributions. The ordered state below the ordering temperature of temperatures up to the highest magnetic field. At the inset of Fig. 14. Compared to the data at the ambient temperature dependence of the strain gauge itself. Large upturn at low temperatures is a consequence of the features connected with phase transitions even without the proper calibration. The strain-gauge was glued to the sample to be sensitive for the length change along the axis and the magnetic field was applied in the perpendicular direction along the axis. The relative change of the strain gauge resistance measured at 1.65 GPa in various magnetic fields is plotted in Fig. 14, together with a sketch of the geometry of the experiment. The large upturn at low temperatures is a consequence of the temperature dependence of the strain gauge itself. The exact position of the field dependence of the function of the strain gauges is known to be smooth. Nevertheless, the correction to the data at the ambient temperature dependence up to 100 K is not visible for the strain gauge resistance measured at the ordering temperature whereas the axis consequently exhibits a sharp positive anomaly at the ordering temperature, accenting the contraction in the basal plane, and entering the hidden-order state at 17.5 K. The linear thermal-expansion coefficient for the axis consequently exhibits a sharp positive anomaly at the ordering temperature whereas the c-axis anomaly is negative and less pronounced (Fig. 2). The volume thermal-expansion coefficient of URu$_2$Si$_2$ also exhibits a sharp and positive peak at 17.5 K that can be translated to a volume decrease in the ground state. A very similar behavior for both compounds can be found in the temperature dependence of the c/a ratio. Both materials show an upturn below the ordering temperature, accenting the contraction in the basal plane, and nearly linear temperature dependence at higher temperatures up to 40 K. Nevertheless, the c/a ratio of URu$_2$Si$_2$ has a pronounced minimum around 60 K, that is not visible for UAu$_2$Si$_2$ which shows a linear temperature dependence up to 100 K. Even though the overall character of the thermal expansion of URu$_2$Si$_2$ and UAu$_2$Si$_2$ is qualitatively similar, it does differ quantitatively. The step in the volume thermal-expansion coefficient $\alpha^*_v = (\alpha_a + \alpha_b + \alpha_c)/3$ is $\sim 2.5 \times 10^{-6}$ K$^{-1}$ for URu$_2$Si$_2$ and almost an order of magnitude larger ($\sim 2 \times 10^{-5}$ K$^{-1}$) for UAu$_2$Si$_2$. It is believed, that anomalies in the thermal-expansion coefficient of the order of $10^{-4}$, $10^{-5}$ can be connected with a structural transitions as.

**IV. DISCUSSION**

Our thermal-expansion measurements on a UAu$_2$Si$_2$ single crystal revealed a large anisotropy which is mainly due to magnetic contributions. The ordered state below $T_m$ is connected with a dramatic lattice contraction in the basal plane (a axis). This together with the relatively small expansion of the c axis leads to the ground state volume collapse (Fig. 1). This behavior strongly resembles the case of the isostructural heavy-fermion compound URu$_2$Si$_2$ entering the hidden-order state at 17.5 K. The linear thermal-expansion coefficient for the a axis consequently exhibits a sharp positive anomaly at the ordering temperature whereas the c-axis anomaly is negative and less pronounced (Fig. 2). The volume thermal-expansion coefficient of URu$_2$Si$_2$ also exhibits a sharp and positive peak at 17.5 K that can be translated to a volume decrease in the ground state. A very similar behavior for both compounds can be found in the temperature dependence of the c/a ratio. Both materials show an upturn below the ordering temperature, accenting the contraction in the basal plane, and nearly linear temperature dependence at higher temperatures up to 40 K. Nevertheless, the c/a ratio of URu$_2$Si$_2$ has a pronounced minimum around 60 K, that is not visible for UAu$_2$Si$_2$ which shows a linear temperature dependence up to 100 K. Even though the overall character of the thermal expansion of URu$_2$Si$_2$ and UAu$_2$Si$_2$ is qualitatively similar, it does differ quantitatively. The step in the volume thermal-expansion coefficient $\alpha^*_v = (\alpha_a + \alpha_b + \alpha_c)/3$ is $\sim 2.5 \times 10^{-6}$ K$^{-1}$ for URu$_2$Si$_2$ and almost an order of magnitude larger ($\sim 2 \times 10^{-5}$ K$^{-1}$) for UAu$_2$Si$_2$. It is believed, that anomalies in the thermal-expansion coefficient of the order of $10^{-4}$, $10^{-5}$ can be connected with a structural transitions as.
in the case of UPd$_3$. In that sense, URu$_2$Si$_2$ does not evidence a structural change in the hidden-order state. Nevertheless, there is a list of studies which suggest the breaking of the fourfold rotational symmetry of the tetragonal c axis, whereas the high-resolution x-ray backscattering and thermal–expansion data do not confirm this. However, lattice-symmetry breaking from the fourfold tetragonal to twofold orthorhombic structure was unambiguously observed by high-resolution synchrotron x-ray diffraction measurements in zero field. The fact that this distortion is observed only in ultrapure samples may explain the long list of more or less unsuccessful attempts to observe this.

As the thermal-expansion coefficients of UAu$_2$Si$_2$ are even one order of magnitude larger (i.e., $\sim 10^{-5}$) the possibility of some kind of lattice distortion should be seriously considered. Our thermal-expansion measurements show an anisotropic expansion in the basal plane breaking the fourfold symmetry along the c axis. The body-centered room-temperature tetragonal structure of UAu$_2$Si$_2$ belongs to the $I4/mmm$ space group. It has 15 maximal non-isomorphic subgroups and only two of them have no fourfold symmetry along the c axis. These are the orthorhombic $Fmmm$ and $Immm$ space groups. The same space groups were also considered in the synchrotron x-ray diffraction study of URu$_2$Si$_2$. The $Fmmm$ space group was found to describe the system in the hidden-order state.

The measurement of thermal expansion, as a macroscopic quantity, is not sufficient to properly describe the space group of the distorted structure, even though it is more sensitive to detect distortions than diffraction studies. In that sense, high-resolution x-ray diffraction experiments are needed to resolve the structure of UAu$_2$Si$_2$ in the ordered state. Our results from an ultrasonic study show a Curie-type softening in the transverse $(C_{11} - C_{12})/2$ mode toward $T_m$, that could also point to orthorhombic distortion at $T_m$. In the analogy with the URu$_2$Si$_2$, there was also observed softening of the same mode suggesting that the $\Gamma_{3g}(B_{gg})$-type lattice instability is innate in these systems.

The large ground-state volume collapse of UAu$_2$Si$_2$ indicates initial positive pressure dependence of the ordering temperature of $dT_m/dp \approx 11.1 \text{ K GPa}^{-1}$, according to the Ehrenfest relation. Uniaxial pressure applied along the a axis should have a positive effect as well ($\approx 5.9 \text{ K GPa}^{-1}$). On the other hand, uniaxial pressure along the tetragonal c axis should lower $T_m$ at a rate of $\approx -0.9 \text{ K GPa}^{-1}$. These findings qualitatively agree with the experimentally confirmed behavior of URu$_2$Si$_2$ where the predicted pressure dependences are approximately eight times smaller. Ehrenfest-relation estimates give a pressure dependence of the hidden-order transition of $1.4 \text{ K GPa}^{-1}$ (Ref.4) and high-pressure resistivity measurements show an experimental initial slope of $1.01 \text{ K GPa}^{-1}$ (Ref.52). The estimated pressure changes of the ordering temperature of the hidden order of URu$_2$Si$_2$ and the AFM state of UAu$_2$Si$_2$ are largely different. A similar dramatic change of the $dT/dp$ values was observed for the U (Ru, Fe)$_2$Si$_2$ system, where doping of Fe leads to a change of the hidden order to “large-moment antiferromagnetism”. However, our measurement of the magnetization under hydrostatic pressure up to 1.0 GPa show only a weak pressure dependence of $T_m$ (Fig. 13). The estimated slope is $dT_m/dp \approx 0.6(1) \text{ K GPa}^{-1}$. We also observed the lowering of the spontaneous magnetization with increasing pressure $d\mu_{\text{spont}}/dp \approx -0.019(6) \mu_B$ (f.u. GPa). The thermal-expansion measurement of the a axis under hydrostatic pressure up to 1.65 GPa revealed a small increase of $T_m$ at zero magnetic field to $T_m,1.65 \text{GPa} = 20.6\text{K}$. Applying magnetic fields along the c axis results in a similar shape of the phase boundary as for the ambient-pressure state, but shifted to slightly higher temperatures. The obtained slope of the pressure dependence of the ordering temperature is $dT_m/dp \approx 0.36 \text{ K GPa}^{-1}$, even smaller than the value obtained from the magnetization measurements. The observed inconsistency with the expected trend from the Ehrenfest relation is rather unexpected. It may be caused by a structural distortion that takes place at $T_m$. In that case Eq. (1) is not valid and the real volume change can be different, i.e., possibly smaller. Another question is the applicability of the Ehrenfest relation itself. Although it is widely and successfully used to characterize the pressure dependence (both positive and negative) of AFM and FM second-order phase transitions, it may strictly be applied only for the superconducting transitions. And even for some superconductors the predicted pressure dependence determined by use of the Ehrenfest relation differs from the experimental findings, such as in the case of PuCoGa$_5$ by an order of magnitude or even by sign in the layered iron-based superconductors of the Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ series. Both of these systems exhibit very anisotropic thermal-expansion coefficients, similar as for UAu$_2$Si$_2$.

The calculated effective Grüneisen parameter exhibits an unusually sharp peak ($\Gamma_{\text{eff}} \sim 17$) at $T_m$ followed by a drop and an almost constant value in the ordered state, that reaches the electronic Grüneisen parameter $\Gamma_e = 9.6(6)$ in the 0 K limit (Fig. 4). The latter value is an order of magnitude larger than the free-electron value, but lower than 25 found for URu$_2$Si$_2$.

The magnetization isotherms (Fig. 5 and 7) clearly and reproducibly show transitions at $\mu_0 H_1$, $\mu_0 H_2$, and $\mu_0 H_m$ in line with our previous study on a different single crystal. However, we now found a much sharper character of the step-like transition at $\mu_0 H_m$. We suggest, that the deviation from the linear dependence of the magnetization, that was marked as $\mu_0 H_m$ in our previous work, is a low-field sign of the step-like transition which takes place at higher fields. We have previously not observed this transition, possibly due to a lower crystal quality or slightly improper orientation of the c axis with respect to the applied field.

Our magnetostriction measurements (Fig. 8 and 10)
with field applied along the c axis reproduce the transitions observed in the magnetization data. The transition at $T_m$ is reflected in the thermal-expansion data only by a small slope change in $\alpha_a$, $\alpha_c$, and $\alpha_{c/a}$ around 50 K (Fig. 2). The size of the relative length change of the c axis at the $\mu_0H_2$ transition, is of the order of $10^{-6}$. This provides an evidence of bulk character of the transition, which can be traced up to $\sim 40$ K as in the magnetization data.

A larger relative length contraction ($\sim 10^{-5}$) takes place at $\mu_0H_m$. It resembles the c-axis contraction at the field-induced phase transition of URu$_2$Si$_2$.60

We now discuss the most important regions in the revised magnetic phase diagram of UAu$_2$Si$_2$ (Fig. 12). Phase I is the low-field section defined by the $\mu_0H_1$ boundary and lies below the ordering temperature. Previous neutron-diffraction experiments revealed that the magnetic structure of UAu$_2$Si$_2$ at 2 K and in zero magnetic field, thus strictly in this phase, is a squared-up AFM structure with the + + - stacking of the FM ac-plane sheets along the a axis.25 The critical field applied along the c axis is $\sim 0.8$ T, the same field where the phase boundaries of the FM-like phase II and the major AFM phase cross. At the critical magnetic field for this phase ($\sim 5$ T) the ordering temperature $T_m$ reaches its maximal value $T_{m,\text{max}} \approx 20.8$ K. In that sense, the vanishing of the phase II leads to a slight strengthening of the major AFM phase. Our high-field magnetization measurements show that the temperature dependence of the hysteresis of the $\mu_0H_m$ transition vanishes around 16 K (and 16 T) where the transition changes its character from step like to continuous. This is attributed to the change of the order of the phase transition from first order (in higher fields) to second order (in lower fields). Such a point is usually referred to as a tricritical point (TCP).40 We marked this point by a star in the phase diagram in Fig. 12(a). Similar tricritical points have recently been reported in the uranium-based antiferromagnets USB$_2$61 and UN.92 The critical field, where the transition temperature $T_m$ is suppressed to 0 K in UAu$_2$Si$_2$ is extrapolated to be about 22 T.

V. CONCLUSIONS

Our thermal expansion, magnetostriction and magnetization study allowed us to complete a comprehensive magnetic phase diagram for UAu$_2$Si$_2$.

The magnetostriction curves measured at higher temperatures confirm bulk character of the 50 K weak FM phase. The large volume contraction in the AFM ordered state suggests a large positive pressure dependence of $T_m$. The linear thermal-expansion data point on the opposite effect for uniaxial pressure applied along the tetragonal c axis and within the basal plane. Magnetization measurements in a hydrostatic pressure cell, however, revealed a negligible hydrostatic pressure effect on $T_m$, namely $dT_m/dp \approx 0.6(1)$ K GPa$^{-1}$ in pressures up to 1.0 GPa. An even smaller pressure dependence of the ordering temperature, $dT_m/dp \approx 0.36$ K GPa$^{-1}$, was obtained from the measurement of the thermal expansion of the a axis under a hydrostatic pressure of 1.65 GPa. These values are much smaller than the prediction from the Ehrenfest relations ($dT_m/dp \approx 11.1$ K GPa$^{-1}$). Further complex studies involving hydrostatic and uniaxial pressure would be desired to shed more light on the nature of this controversy.

As the order of all the relative length changes is $\sim 10^{-5}$, we can expect some structural changes or distortions of the UAu$_2$Si$_2$ unit cell in the ground state. Our comparative dilatometry measurements of the linear thermal expansion along the a axis and along the [110] direction clearly show the fourfold symmetry breaking in the basal plane. This may also affect the real low-pressure dependence of the ordering temperature. High-resolution diffraction measurements are needed to find the ground-state space group. Possible candidates are the orthorhombic non-isomorphic subgroups Fmmm and Immm, where the first one was found to describe the structure of the high-quality samples of URu$_2$Si$_2$ in the hidden-order state. Our high-field magnetization measurements revealed a critical field of $\approx 22$ T where the ordering temperature $T_m$ is suppressed to 0 K. The hysteresis of this transition emerges at a tricritical point given by $T_m \approx 16$ K and $\mu_0H_m \approx 16$ T as a sign of the change of the transition from second to first order.

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