Downloaded from UvA-DARE, the institutional repository of the University of Amsterdam (UvA)
http://hdl.handle.net/11245/2.2626

File ID uvapub:2626
Filename 25446y.pdf
Version unknown

SOURCE (OR PART OF THE FOLLOWING SOURCE):
Type article
Title Coupled spin-lattice transition in U2Rh3Si5: Thermal expansion and magnetization
Author(s) T. Takeuchi, T. Yamada, Y. Miyako, K. Oda, K. Kindo, B. Becker, A.A. Menovsky, G.J. Nieuwenhuys, J.A. Mydosh
Faculty FNWI: Van der Waals-Zeeman Institute (WZI)
Year 1997

FULL BIBLIOGRAPHIC DETAILS:
http://hdl.handle.net/11245/1.132883

Copyright

It is not permitted to download or to forward/distribute the text or part of it without the consent of the author(s) and/or copyright holder(s), other than for strictly personal, individual use, unless the work is under an open content licence (like Creative Commons).

UvA-DARE is a service provided by the library of the University of Amsterdam (http://dare.uva.nl)
(pagedate: 2016-08-01)

Coupled spin-lattice transition in $U_2Rh_3Si_5$: Thermal expansions and magnetization

T. Takeuchi, T. Yamada, and Y. Miyako
Low Temperature Center and Department of Earth and Space Science, Osaka University, Toyonaka 560, Osaka, Japan

K. Oda and K. Kindo
Research Center for Materials Science at Extreme Conditions, Osaka University, Toyonaka 560, Osaka, Japan

B. Becker, S. Ramakrishnan,* A. A. Menovsky,† G. J. Nieuwenhuys, and J. A. Mydosh
Kamerlingh Onnes Laboratory, Leiden University, 2300 RA Leiden, The Netherlands
(Received 28 July 1997)

We have measured the thermal expansion, $\Delta l_i/l_i$, and high-field magnetization, M_i, along the three quasiorthorhombic axes of $U_2Rh_3Si_5$. There are discontinuities in $\Delta l_i(T)/l_i$ at $T_0 = 25.7$ K of opposite sign in the a' and b directions while the c axis changes steplike, which tracks a large volume change. The magnetization along the b axis, $M_b(H)$, exhibits a dramatic temperature-dependent metamagnetic transition where the induced U moments switch from 0.1 to $1.7\mu_B$, while the a' and c axes remain unperturbed. We relate these anomalies to a first-order phase transition of strongly coupled spin and quadrupole orderings. [S0163-1829(97)01242-3]

The search for unusual behavior in uranium-based intermetallic compounds has been underway for many years[1] and has had a varied and fruitful history.[2] During the past decade the main focus was on the strongly correlated materials where a vigorous hybridization occurs between the conduction electrons and the nearly localized $5f$ electrons of the U ion. Here a variety of unconventional magnetic and (coexisting) superconducting ground states are observed[3] and nicely described via a heavy-mass Fermi-liquid model.[4] More modern interest has shifted to non-Fermi-liquid behavior,[5] usually caused by disorder, and now an unordered ground state is under study.

A somewhat neglected phenomenon (from the modern standpoint) is the magnetoelastic interaction between the U ion with its spin and orbital moments and the underlying lattice. Yet past experience[6,7] has taught us that some U compounds do exhibit magnetic transitions which are strongly coupled to the lattice and thus affect the crystal structure. This, in a few cases, e.g., (insulating) fcc-UO_2 (Ref. 8) and (metallic) dhcp-UPd_3,[9] results in a change of the crystal symmetry via lattice distortions, usually denominated a quadrupolar ordering or cooperative Jahn-Teller effect. The above two systems represent well-known examples of such spin-lattice transitions: at a single first-order transition temperature for UO_2: $T_N = 30.8$ K, or at two different critical temperatures for UPd_3: $T_1 = 6.5$ K, quadrupolar and $T_2 = 4.5$ K, also magnetic. These distinct compounds (one insulating, the other metallic) seem to possess similar U^{4+} ($5f^2$) valencies whose crystal electric field (CEF) splitting allows quadrupolar active states, e.g., Γ_5 (triplets) or Γ_3 (doublets), to interact and also to couple with the usual spin antiferromagnetism, thereby creating the quadrupole-dipole transitions. Nevertheless, in the vast literature of the U-based materials reports on these types of transitions are surprisingly limited to compounds with well-defined (nonhybridized) U moments.

Recently Becker *et al.*[10] have grown well-characterized single crystals of a new uranium compound, $U_2Rh_3Si_5$, and investigated its bulk physical properties, i.e., specific heat, magnetic susceptibility and electrical resistivity. $U_2Rh_3Si_5$ forms in the monoclinic $Lu_2Co_3Si_5$ structure with space group $C2/c$.[11] The monoclinic distortion is so small that we can represent our system in a quasiorthorhombic symmetry ($U_2Co_3Si_5$ structure, *Ibam*) with orthogonal axes $\vec{a'}$, $\vec{b}$, and $\vec{c}$.[12] Dramatic effects have been observed in all of the above bulk properties at a *single* critical temperature $T_0 = 25.7$ K (Ref. 10) that suggest a first-order phase transition into a simultaneous spin-quadrupolar ordering.

In order to directly probe the lattice behavior and to further determine the field dependence of this unusual transition, we have measured the thermal expansion, $\Delta l_i/l_i$, and high-field magnetization, M_i, along the three main quasiorthorhombic axes of $U_2Rh_3Si_5$. Discontinuities are found in $\Delta l_i/l_i$ at T_0 where there is also a step in the crystal volume. In contrast, only along the b axis does $M_b(H)$ exhibit a dramatic, T-dependent metamagnetic transition where the U moments switch within a tiny field interval from 0.05 to 1.7 μ_B. The a' and c axes remain unperturbed up to 30 T with small induced moments. Our thermal expansion results illustrate that the lattice is strongly involved in the first-order phase transition at T_0 and the crystal structure distorts along the c axis. The metamagnetic transition (also of first order) along the b axis indicates the easy direction of spin alignment[12] and its sensitivity to a ferromagnetic reorientation in sufficiently large fields.

The sample of $U_2Rh_3Si_5$ has been grown in a triarc furnace using the Czochralski method in high-purity argon atmosphere and is taken from the same single crystal as in Ref. 10. An electron-probe microanalyzer (EPMA) established the sample to be single-phase material with a maximum limit of $\approx 1\%$ for impurities and second phases. Laue–x-ray diffraction verified the single crystallite and proper orientation.

 © 1997 The American Physical Society

A cube of 1.4 mm × 1.4 mm × 1 mm aligned along the quasiorthorhombic axes $\vec{a'}$, $\vec{b}$, and $\vec{c}$ [with $\vec{a'} \perp (\vec{b}, \vec{c})$] has been formed by spark erosion. The accuracy of the alignment of the sample is of the same order as the monoclinic distortion, viz., less than 1°.

Measurements of thermal expansion have been performed by using a sensitive three-terminal capacitance method and the temperature varied from 4.3 K to 50 K. The sample was mounted in a parallel-plate capacitance cell made of oxygen-free high conductivity copper. The high-field magnetization measurements were performed in pulsed magnetic fields up to 30 T at the Research Center for Materials Science at Extreme Conditions, Osaka University.

The thermal expansion $\Delta l_i/l_i \equiv [l_i(T) - l_i(4.5\,\mathrm{K})]/l_i(4.5\,\mathrm{K})$ versus temperature curves are shown in Fig. 1 (upper panel) along the $\vec{a'}$, $\vec{b}$, and $\vec{c}$ directions. The volume expansion $\Delta V/V \equiv [V(T) - V(4.5\,\mathrm{K})]/V(4.5\,\mathrm{K})$ as a function of temperature has been calculated from the length changes along the three perpendicular axes and is also presented in Fig. 1. Note that the same scale is used in Fig. 1 but the volume change has been shifted downward by one unit for clarity.

Between 50 K and 27 K the length change in the $\vec{a'}$, $\vec{b}$, and $\vec{c}$ directions are linear in temperature, increasing along the a' and c axes, decreasing along the b axis, with decreasing temperature. Below 25 K the a' and b axes hardly expand in contrast to the c direction that continuously contracts as the temperature increases. At the ordering temperature all directions show very steep changes in the thermal expansion. To illustrate this the center panel of Fig. 1 presents an expanded view around $T_0 = 25.7$ K. The measurements along $\vec{a'}$ and $\vec{b}$ are fitted to linear dependences above and below T_0. The resulting ''ideal'' jumps are shown by the solid lines. A temperature smearing of the transition about 120 mK is visible. We attribute this broadening to tiny imperfections and inhomogeneities of the ternary single crystal (also observed by Becker *et al.*[10]). The thermal expansion along the a' and b axes exhibit a jump of $\delta_{a'} = +2\times10^{-5}$ and $\delta_b = -4\times10^{-5}$ at T_0. The c-axis thermal expansion shows a steep, steplike change that merges into a weaker temperature dependence for $T < T_0$. An upper estimate of δ_c is $\approx -1.6\times10^{-5}$.

Figure 1 (lower panel) displays the thermal expansion coefficient $\alpha_i(T) = d(\Delta l_i/l_i)/dT$ along the a' and c axes, respectively. The curves have been calculated by numerically differentiating the thermal expansion data. The above-mentioned temperature smearing prevents the α_i from diverging, nevertheless rather large values of $6\times10^{-5}\mathrm{K}^{-1}$ are reached at $T_0 = 25.7$ K. $\alpha_{a'}(T)$ and $\alpha_b(T)$ (not shown) change sign at T_0 while $\alpha_c(T)$ remains negative in the whole measured temperature range.

In Fig. 2 the magnetization versus field, $M_i(H)$, up to 30 T is plotted as measured along the different crystallographic axis: $\vec{a'}$, $\vec{b}$, and $\vec{c}$ at 4.5 K. $M_{a'}(H)$ and $M_c(H)$ increase linearly with field up to 30 T. At 29 T induced moments of $0.13\mu_B$/U atom (a' axis) and $0.155\mu_B$/U atom (c axis) are created. For the b axis $M_b(H)$ increases linearly in field up to a small induced moment of $0.056\mu_B$/U atom. However, at 14.1 T a sharp jump in $M_b(H)$ occurs within

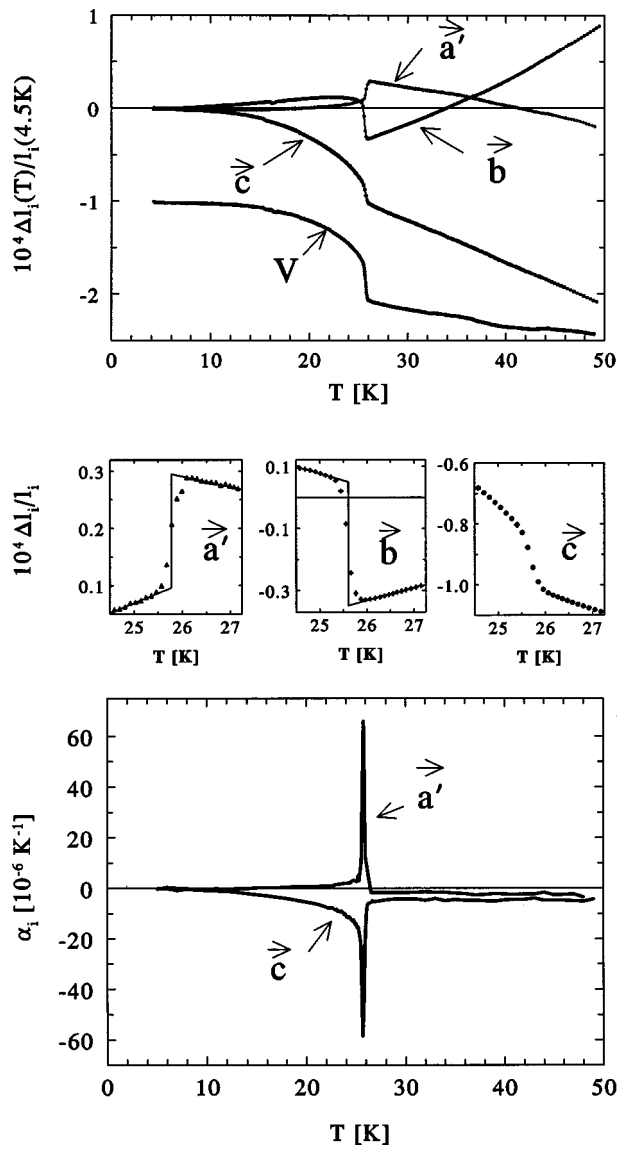

FIG. 1. (Upper panel) Thermal expansion $\Delta l_i/l_i$ vs temperature for the $\vec{a'}$, $\vec{b}$, and $\vec{c}$ directions of $U_2Rh_3Si_5$. The net volume change $\Delta V/V$ given by the sum of the three $\Delta l_i/l_i$ is also shown. (Middle panel) Expanded view of the measured $\Delta l_i/l_i$ near T_0 (data points), including modeled ''ideal'' jumps (solid lines). (Lower panel) Coefficient of thermal expansion $\alpha_i(T) \equiv d(\Delta l_i/l_i)/dT$ vs temperature for the $\vec{a'}$ and $\vec{c}$ direction.

90 mT, and the moment switches to a value of $1.65\mu_B$/U atom. By further increasing the field up to 30 T only a slight increase in $M_b(H)$ to $1.78\mu_B$/U atom is observed. The inset in Fig. 2 shows the derivative of the magnetization with respect to the applied field along the b axis. The jump in the magnetization is clearly reflected in the peak. In addition, a tiny hysteretic behavior with a width of ≈ 50 mT is observed by sweeping the field up or down through the metamagnetic transition using a slowly varying dc field up to 16 T.

Figure 3 presents the magnetic phase diagram obtained from magnetization and resistivity measurements in longitudinal fields. At 4.5 K the high-field result of Fig. 2 has been included. The low-field data have been obtained in a Quantum Design SQUID (to 5 T), a vibrating sample magnetome-

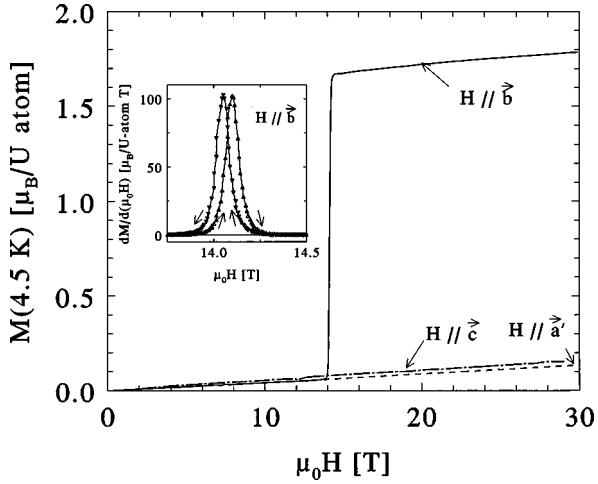

FIG. 2. Magnetization, $M_i(H)$, vs pulsed magnetic field ($\vec{H}\|\vec{a'}$, dashed line; $\vec{H}\|\vec{b}$, solid line; and $\vec{H}\|\vec{c}$, dash-dotted line). The inset shows the field derivative $dM_b/d(\mu_0 H)$ close to the metamagnetic transition (field sweep up, △ and field sweep down, ▽).

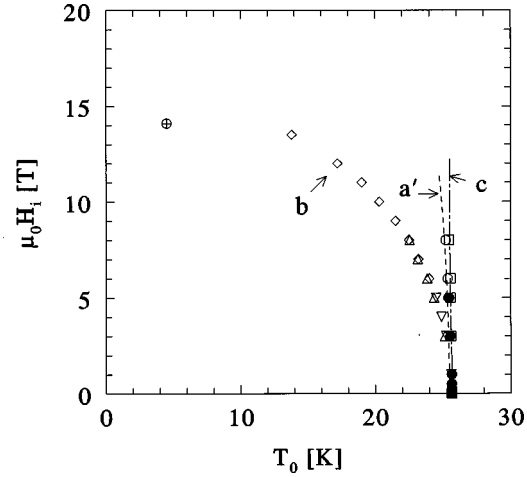

FIG. 3. The magnetic phase diagram: magnetic field H_i applied along $\vec{a'}$, $\vec{b}$, and $\vec{c}$ vs transition temperature T_0. The data were obtained by pulsed field magnetization ($m_{\vec{b}}$: ⊕), vibrating sample magnetometer ($m_{\vec{b}}$: ◇), dc-SQUID susceptibility ($\chi_{dc,\vec{a'}}$: ●, $\chi_{dc,\vec{b}}$: ▽ and $\chi_{dc,\vec{c}}$: +), and ac resistivity ($\rho_{i\|\vec{a'},\vec{H}}$: ○, $\rho_{i\|\vec{b},\vec{H}}$: △ and $\rho_{i\|\vec{c},\vec{H}}$: □).

ter (to 13.5 T) and via ac four-probe resistance measurements (to 8 T). Note that no field transitions are found along the a' and c axes up to 30 T. The dashed (guide to the eye) lines indicate the large anisotropies in $U_2Rh_3Si_5$ between the b axis and the a'/c axes. The latter two are essentially similar (see also Fig. 2).

The thermal expansion measurements with their distinct steps at T_0 offer strong evidence for a first-order phase transition. The experimental fact that there is only one critical temperature, $T_0 = 25.7$ K, as determined by both magnetic and structural probes, demonstrates the strong spin-lattice coupling. With respect to the single, first-order transition $U_2Rh_3Si_5$ can be compared to UO_2.[8] However, in $U_2Rh_3Si_5$, a metal, the Fermi surface is involved in the transition. Due to the magnetic ordering, the Fermi surface reconstructs[13] as is indicated by the observed steps in the electrical conductivity.[10] Since the density of states at E_F directly influences the electronic contribution to $\alpha_i(T)$, this reconstruction could lead to the large c-axis expansion which tracks the volume change.

Remarkably, the thermal expansion coefficients of the a', c direction and of the volume are negative even in the paramagnetic state ($T > T_0$). As phonons generally lead to positive values for $\alpha_i(T)$, the thermal expansion in $U_2Rh_3Si_5$ must be governed by crystalline electric field (CEF) effects. In fact, smooth changes in susceptibility and resistivity measurements have also been attributed to CEF.[10] The influence of the CEF on the thermal expansion has been recently studied in orthorhombic $ErCu_2$. A knowledge of the CEF parameters and the elastic constants enables Gratz *et al.*[14] to calculate the thermal expansion in the paramagnetic state. Unfortunately the above quantities are not yet known for $U_2Rh_3Si_5$.

Although in the paramagnetic state the thermal expansion behavior of $U_2Rh_3Si_5$ is governed by the CEF, at T_0 additional ''bootstrapping'' interactions come into play and lead to the single first-order transition. The lattice deformations are the result of a delicate balance between the magnetic, electronic, and elastic energies that are controlled by the exchange, magnetoelastic, quadrupolar,[7] and electron-local moment interactions.[13] The experimentally observed scaling of the sublattice magnetization [$\mu_{a'} = 0.89\mu_B$ and $\mu_b = 1.5\mu_B$ (Ref. 12)] with the jumps in the thermal expansion ($\delta_{a'} = +2\times10^{-5}$ and $\delta_b = -4\times10^{-5}$) suggests the importance of magnetoelastic effects, which could be established via magnetostriction experiments.[7] Measurements of the elastic constants would enable one to understand the quadrupolar interactions present in $U_2Rh_3Si_5$.[15]

Our field-dependent studies demonstrate the sensitivity of the ordered magnetic structure to a field of singular alignment. The magnetization measurements show the b direction to be the easy magnetic axis. This agrees with the magnetic structure as determined by neutron diffraction.[12] The metamagnetic transitions, illustrated in the magnetic phase diagram of Fig. 3, are of first order with a small hysteresis and large induced moment change. The $M_{a'}$ and the M_c are hardly affected by fields up to 30 T. This indicates the strong magnetic anisotropy that is different from the crystallographic one found in $\Delta l_i/l_i$.

In summary we have detected a first-order phase transition in the lattice parameters and the total volume of $U_2Rh_3Si_5$ at $T_0 = 25.7$ K that is similar to the magnetic transition observed previously in the specific heat, susceptibility, and resistivity. A first-order metamagnetic transition occurs only in the b-axis magnetization for $T < T_0$ where there is a large moment change. These results evince the strong coupling of the lattice to the spin system. The Fermi surface reconstruction at T_0 adds a facet to the behavior of $U_2Rh_3Si_5$ that warrants further investigations.

We wish to acknowledge the metallurgical assistance of R. Hendrikx and T. J. Grotenmulder. This work was partially supported by the Nederlandse Stichting Fundamental Onderzoek der Materie (FOM) and by the Grant-in-Aid from the Ministry of Education, Science and Culture.

*Permanent address: Tata Institute of Fundamental Research, Bombay, India.

†Also at Van der Waals–Zeeman Laboratory, University of Amsterdam, 1018 XE Amsterdam, The Netherlands.

[1] See, for example, *The Actinides, Electronic Structure and Related Properties*, edited by A.J. Freemann and J.B. Darly, Jr. (Academic Press, New York, 1964), Vols. I and II; M.B. Brodsky, Rep. Prog. Phys. **41**, 1547 (1978).

[2] For more recent reviews, see V. Sechovsky and L. Havela, in *Ferromagnetic Materials*, edited by E.P. Wohlfarth and K.H.J. Buschow (Elsevier, Amsterdam, 1988), Vol. 4, p. 309; G.J. Nieuwenhuys, in *Handbook of Magnetic Materials*, edited by K.H.J. Buschow (Elsevier, Amsterdam, 1995), Vol. 9, p. 1.

[3] N. Grewe and F. Steglich, in *Handbook on the Physics and Chemistry of Rare Earth*, edited by K.A. Gschneider and L. Eyring (Elsevier, Amsterdam, 1991), Vol. 14, p. 343.

[4] A.C. Hewson, *The Kondo Problem to Heavy Fermions* (Cambridge University Press, Cambridge, England, 1993).

[5] D. Finsterbusch, H. Willig, B. Wolf, G. Bruls, B. Lüthi, M. Waffenschmidt, O. Stockert, A. Schröder, and H. v. Löhneysen, Ann. Phys. (Leipzig) **5**, 184 (1996); *Proceedings of the Conference on Non-Fermi-Liquid Physics in Metals, 1996* [J. Phys. Condens. Matter **8** (1996)].

[6] G.A. Gehring and K.A. Gehring, Rep. Prog. Phys. **38**, 1 (1975).

[7] P. Morin and D. Schmitt, in *Ferromagnetic Materials*, edited by K.H.J. Buschow and E.P. Wohlfarth (Elsevier, Amsterdam, 1990), Vol. 5, p. 1.

[8] See, for example, P. Giannozzi and P. Erdös, J. Magn. Magn. Mater. **67**, 75 (1987).

[9] See, for example, K.A. McEwen, U. Steigenberger, and J.L. Martinez, Physica B **186–188**, 600 (1993).

[10] B. Becker, S. Ramakrishnan, A.A. Menovsky, G.J. Nieuwenhuys, and J.A. Mydosh, Phys. Rev. Lett. **78**, 1347 (1997).

[11] E. Hickey, B. Chevalier, P. Gravereau, and J. Etourneau, J. Magn. Magn. Mater. **90&91**, 501 (1990).

[12] Preliminary neutron diffraction on polycrystalline $U_2Rh_3Si_5$ show a canted antiferromagnetic structure with large spin component along the *b* axis [R. Feyerherm, B. Becker, M.F. Collins, J.A. Mydosh, G.J. Nieuwenhuys, and S. Ramakrishnan, Physica B **234-236**, 891 (1997)].

[13] See, for example, V.N. Antonov, A.Ya. Perlov, P.M. Oppeneer, A.N. Yaresko, and S.V. Hallieov, Phys. Rev. Lett. **77**, 5253 (1996).

[14] E. Gratz, M. Rotter, A. Lindbaum, H. Müller, E. Bauer, and H. Kirchmayr, J. Phys.: Condens. Matter **5**, 567 (1993).

[15] P. Thalmeier and B. Lüthi, in *Handbook on the Physics and Chemistry of Rare Earth* (Ref. 3), p. 225.