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Unusual Ordering Behavior in Single-Crystal $U_2Rh_3Si_5$

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We have grown single crystals of the ternary uranium compound $U_2Rh_3Si_5$ which crystallizes in a monoclinic $Lu_2Co_3Si_5$ structure with space group $C2/c$. $U_2Rh_3Si_5$ exhibits a single dramatic phase transition at 25.6 K. Here the specific heat peaks to more than 100 J/U-molK, and the magnetic susceptibility is strongly anisotropic and shows a steplike drop at the ordering temperature. The electrical resistivity is also anisotropic with indications for the appearance of superzone and spin-wave gaps. We relate this behavior to a (weak) first-order phase transition into a simultaneous spin-quadrupolar ordering. [S0031-9007(97)02473-3]

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Uranium intermetallic compounds are known to display a variety of exotic magnetic and (coexisting) superconducting properties as exemplified by their heavy-fermion, non-Fermi liquid and metal/insulator behaviors [1–3]. Such effects reflect the strong electron-electron correlations and large hybridization that are usually present in these materials. Especially interesting here is the magnetic ordering phenomenon which mainly depends on the crystal structure, the strength of the inter-U magnetic interactions, and the amount of Kondo screening resulting from the correlations and hybridization. Highly unusual magnetic structures of U moments have been previously observed: (i) very small moments ($\mu_{\text{eff}} \approx 0.02 \mu_B$) in a simple array of up/down ferromagnetic planes, e.g., URu_2Si_2 [4], (ii) partially frustrated and short-range order, e.g., UNi_4B [5] or UPt_3 [6], (iii) large anisotropies and incommensurate structures, e.g., UPd_2Al_3 [7] and UNi_2Al_3 [8], (iv) random freezing or spin-glass transitions, e.g., URh_2Ge_2 [9], and (v) ordering of quadrupolar and spin moments in UPd_3 [10]. Since the U ions possess an intermediate degree of localization between the mainly itinerant $3d$ systems and the fully localized $4f$ ones, their exact magnetic nature depends on the particular crystallographic and electronic structures of the given intermetallic compound. The specific crystal electric field (CEF) scheme and the coupling of the U ions to the lattice can generate additional quadrupolar and/or structural transitions. Hence new U-based materials with novel structures should lead to extraordinary behavior.

We report in this Letter the atypical magnetic behavior of a new uranium compound $U_2Rh_3Si_5$, first synthesized in 1990 by Hickey *et al.* [11]. This material forms in the monoclinic $Lu_2Co_3Si_5$ structure with space group $C2/c$. The monoclinic distortion of $U_2Rh_3Si_5$ from the orthorhombic $U_2Co_3Si_5$ structure (Ibam) is so small that we can represent our system in a quasiorthorhombic symmetry by transforming the basis \vec{a} , \vec{b} , and \vec{c} of the space group $C2/c$ (cell choice 1 with unique b axis) into $\vec{a}' = \vec{a} + \vec{c}$, while \vec{b} and \vec{c} are kept unchanged. A sketch of the crystal structure using the quasiorthorhombic

notation is given in Fig. 1. All nearest neighbor U-U bonds and other bonds shorter than 3.3 Å have been depicted. In the $\vec{b}-\vec{c}$ plane the U atoms form a corrugated planelike structure with U-U distances of 3.8 Å. Along the \vec{a}' direction the U atoms are elongated towards the rhodium atoms. The U-U planes are separated by 5.8 Å.

Previously, polycrystalline samples were reported to order antiferromagnetically below 26 K [12,13] based on susceptibility, and resistivity measurements. Our present experiments on single-crystal samples include specific heat, magnetic susceptibility and electrical resistivity in the various crystal directions. All these measurements illustrate the highly anomalous nature of the phase transition at 25.6 K. Preliminary neutron diffraction experiments on a polycrystalline sample show that in $U_2Rh_3Si_5$ the Kondo effect plays a minor role since the observed U moments are large ($1.75 \mu_B$) [14]. The moments involved in the ordering also appear to be strongly coupled to the

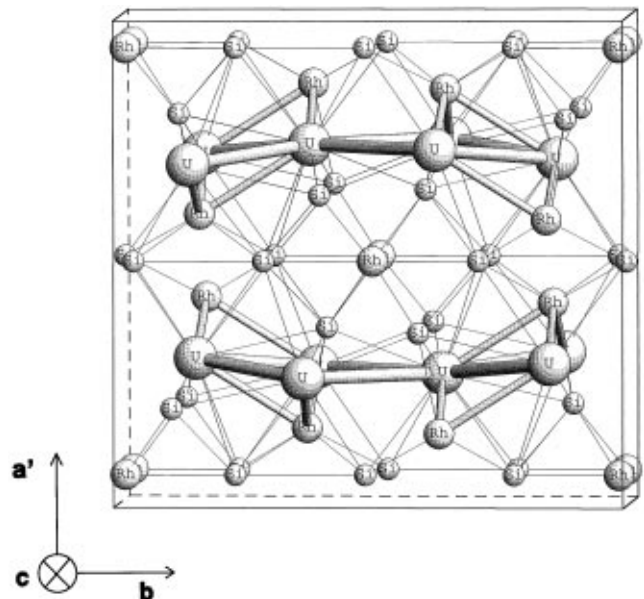


FIG. 1. Crystal structure of $U_2Rh_3Si_5$ in quasiorthorhombic coordinates.

lattice. In what follows we depict our bulk experimental results and relate these to a (weak) first-order phase transition into a simultaneous spin-quadrupolar ordering.

The single crystal has been grown in a tri-arc furnace using the Czochralski method in a high purity argon atmosphere (pulling rate: 10 mm/h; seed rotation: 20 rpm, as for the counterrotating crucible). During the growth a clear facetting has been observed. The starting materials were U 3*N*, Rh 4*N*, and Si 5*N*. The sample has been checked by Laue-x-ray diffraction, confirmed to be single crystalline and properly oriented. Electron-micron probe-analysis (EPMA) established the crystal to be single-phase material with an maximum limit of $\approx 1\%$ for impurities and second phases. Resistance bars have been cut via spark erosion along the quasiorthorhombic crystallographic axis \vec{a}' , \vec{b} , and \vec{c} . A portion of the single crystal has been annealed at 900° for seven days under high vacuum, which leads to an even higher sample quality, indicated by the sharpness of the transition.

The specific heat versus temperature is shown in Fig. 2. The measurements on the as-grown single crystal have been performed using a quasiadiabatic technique. The lattice contribution to the specific heat has been subtracted as described in Ref. [13]. At the ordering temperature T_{ord} a very sharp peak occurs with a maximum of 115 J/U-mol K. The net entropy is presented in the inset of Fig. 2. At T_{ord} the entropy reaches $R \ln 2$. The electronic linear contribution γ is determined to be 22 mJ/U-mol K², so this system is not a heavy fermion. The small contribution to the specific heat and the entropy above the transition are probably due to CEF effects.

In Fig. 3 the dc-susceptibility χ_{dc} measured on the annealed crystal is displayed. At 25.6 K the magnetic ordering is accompanied by a sharp drop in χ_{dc} for $T < T_{\text{ord}}$. The susceptibility shows a strongly anisotropic behavior: For $B \parallel \vec{b}$, χ_{dc} has the largest value at T_{ord} , four and eight times larger than for $B \parallel \vec{a}'$ and $B \parallel \vec{c}$, respectively. The inset of Fig. 3 shows the high temperature behavior of

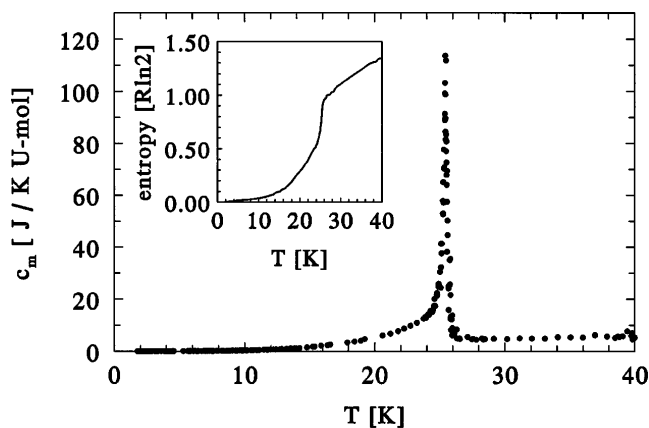


FIG. 2. Specific heat of $\text{U}_2\text{Rh}_3\text{Si}_5$ (corrected for lattice contribution) versus temperature. Inset: Net entropy gain of $\text{U}_2\text{Rh}_3\text{Si}_5$ in units of $R \ln(2)$.

$\chi_{\text{dc}}^{-1}(T)$. For a field parallel to the \vec{a}' and the \vec{b} axis a Curie-Weiss (CW) law is obeyed for $T > 100$ K as shown by the full lines. For fields parallel to the \vec{c} axis deviations from a CW behavior are found up to 300 K. From these plots we determine an average effective U moment of $3.3 \mu_B$.

The resistivity as measured on the annealed crystal is plotted versus temperature in Fig. 4. Above 30 K $\rho(T)$ shows a very weak temperature dependence irrespective of the direction of the current. The absolute values at room temperature are $\rho_{\vec{a}'} = 315 \mu\Omega \text{ cm}$, $\rho_{\vec{b}} = 214 \mu\Omega \text{ cm}$, and $\rho_{\vec{c}} = 210 \mu\Omega \text{ cm}$ (see horizontal arrows in Fig. 4). These values are large compared to other U compounds that form in the related ThCr_2Si_2 structure [15]. Around 26 K, $\rho_{\vec{a}'}$ and $\rho_{\vec{b}}$ exhibit an upward step at T_{ord} . Such “superzone-gap” features should be attributed to the changes in the Brillouin zone due to the magnetic ordering as discussed by Mackintosh [16]. In contrast to the \vec{a}' and \vec{b} directions, $\rho_{\vec{c}}$ suddenly drops upon lowering the temperature below 26 K (inset in Fig. 4). These anomalies are followed by a strong decrease of the resistivity with decreasing temperatures. The resistivity ratio between 40 and 1.3 K is $R(40 \text{ K})/R(1.3 \text{ K}) \approx 27.5$ for $i \parallel \vec{a}'$, ≈ 27 for $i \parallel \vec{b}$, and ≈ 73 for $i \parallel \vec{c}$. Below T_{ord} the resistance can be described by

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

where ρ_0 is the residual resistance, AT^2 the Fermi-liquid contribution, and Δ the gap in the spin-wave spectrum [17]. The T^2 term is quite small, which is consistent with the tiny γ value observed in the specific heat. The spin-wave gaps, as found from the temperature dependence of the resistivity, are $\Delta_{a'} = 77 \pm 2$ K, $\Delta_b = 85 \pm 2$ K, and $\Delta_c = 106 \pm 6$ K. The fits describe the resistivity remarkably well up to about

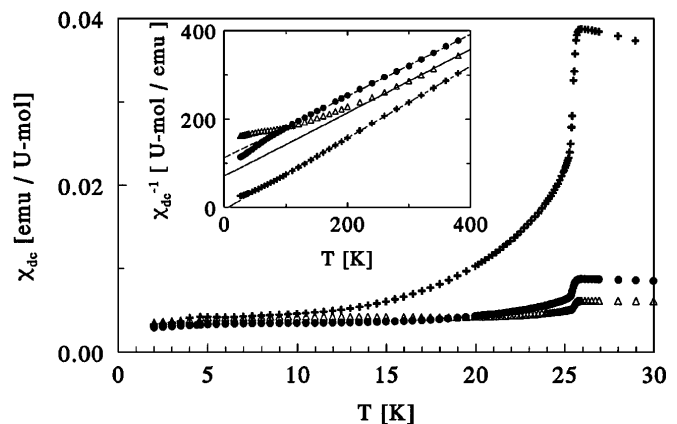


FIG. 3. Low temperature dc susceptibility χ_{dc} versus temperature of $\text{U}_2\text{Rh}_3\text{Si}_5$ with magnetic field parallel to the a' axis (\bullet), b axis ($+$), and c axis (Δ). Inset: Inverse dc-susceptibility χ_{dc}^{-1} with fits to a Curie-Weiss law as described in the text.

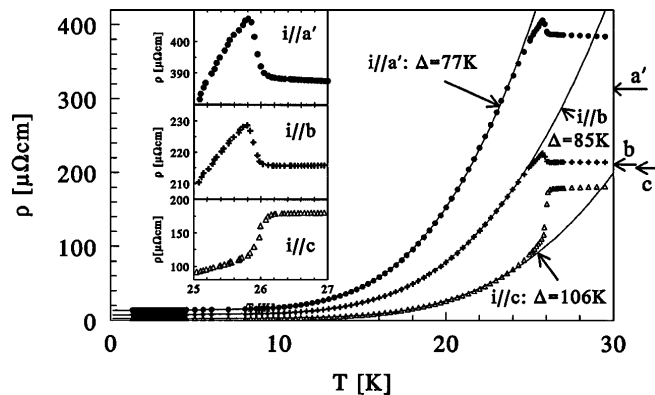


FIG. 4. Resistivity versus temperature for $U_2Rh_3Si_5$ for $i\parallel\vec{a}'$ (\bullet), $i\parallel\vec{b}$ ($+$), and $i\parallel\vec{c}$ (Δ) with fits (solid lines) to a spin-wave gap. The horizontal arrows labeled a' , b , and c indicate the resistivity at 300 K. Insets: Enlargement of the three resistivities at the phase transition.

23 K, indicating little temperature dependence of the gap up to $0.9 T_{ord}$. Within the experimental resolution of less than 50 mK no thermal hysteresis has been observed in our resistivity measurements.

The results described above show that $U_2Rh_3Si_5$ exhibits a most unusual magnetic behavior. The temperature dependence of the bulk properties reveals three distinct regions; above, around, and below T_{ord} :

(I) Above T_{ord} : free (paramagnetic) U moments and CEF effects govern the behavior. In particular, the smooth changes in $\chi_{dc}(T)$ [and in the anisotropy of $\chi_{dc}(T)$] as well as in $\rho(T)$ below 200 K should be attributed to CEF effects.

(II) A very narrow temperature regime around T_{ord} where all bulk properties of the single crystal show dramatic changes. The specific heat exhibits a large peak, and the magnetic susceptibility as well as the electrical resistivity show steps as a function of temperature. The temperature width ΔT_{ord} of these anomalies is similar for all three bulk properties: 150 mK for the as-grown sample, reduced to 80 mK after one week of annealing. Thus, $\Delta T_{ord}/T_{ord}$ is of order 10^{-3} and can be attributed to minor sample imperfections.

(III) Below T_{ord} smooth variations with temperature of the dc susceptibility and of the specific heat are observed, while the strong decrease in the resistivity with decreasing temperatures is governed by the gap in the spin-wave spectrum.

The sharp jump in the specific heat is more than 100 J/mol K, much larger than the mean field prediction for a spin $\frac{1}{2}$ doublet ground state. The sharpness and amplitude of the transition in the specific heat indicate its first order character. Indeed, neutron diffraction results [14] have shown that the antiferromagnetic ordering is accompanied by a considerable change in the lattice constants \vec{b} and \vec{c} , determined above and below T_{ord} . Since the crystallographic symmetry of $U_2Rh_3Si_5$ is

already low, no symmetry breaking is found such as occurs in cubic UPd₃ [18].

In a first attempt we model the behavior of $U_2Rh_3Si_5$ by assuming a singlet-ground state and a splitting to a first excited singlet level Δ_{CEF} with $\Delta_{CEF} > T_{ord}$ for $T < T_{ord}$, and $\Delta_{CEF} < T_{ord}$ for $T > T_{ord}$ [19,20]. This leads to a two-level model for $U_2Rh_3Si_5$ where both levels have no diagonal elements for the magnetic moment. The anomaly in the specific heat can be qualitatively understood by a rapid depopulation of the first excited state with the occurrence of antiferromagnetic ordering. The sudden changes in regime (II) could be caused by different order parameters “bootstrapping” each other into a dramatic phase transition. This suggests a strong coupling between the sublattice magnetization and the quadrupole moments which in turn greatly affects the lattice parameters and the band structure. Our preliminary thermal-expansion measurements show an anisotropic and discontinuous jump at T_{ord} [21]. The absence of fluctuations or short-range order in the specific heat and the remarkable change in the lattice constants [14,21] points to a first-order phase transition. To reveal the exact nature of the order parameters and their coupling, more detailed neutron diffraction and inelastic scattering (to determine the CEF and spin-wave gap) are required.

In summary, our investigations of the bulk properties (c_p , χ_{dc} , and ρ) on single crystal $U_2Rh_3Si_5$ have clearly shown the unusual character of the magnetic phase transition at 25.6 K which appears to be first order and strongly coupled to the lattice. A qualitative description seems possible within a singlet-singlet model where exceptionally large interactions exist among different order parameters, e.g., the sublattice magnetization and quadrupole moments. These bootstrapping interactions would significantly modify the lattice parameters and the band structure and induce superzone and spin-wave gaps. Our limited knowledge about the CEF parameters and the strengths of the magnetoelastic interactions prevents a meaningful quantitative analysis at present. Further experimentation is warranted to fully characterize $U_2Rh_3Si_5$.

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