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Non-collinear magnetic ordering in $U_2Rh_3Si_5$

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Abstract

Neutron powder diffraction reveals a non-collinear antiferromagnetic arrangement of the eight uranium magnetic moments within the unit cell of $U_2Rh_3Si_5$ below $T_N = 26$ K. The μ_x component of the moments orders in ferromagnetic wavy sheets stacked antiferromagnetically along x, while μ_y exhibits a two-up, two-down configuration along y, else being stacked like μ_x. The magnetic structure is associated with the ordered moment $\mu = 1.81(5)\mu_B$ at $T = 8.5$ K.

Keywords: $U_2Rh_3Si_5$; Antiferromagnetism; Non-collinear structures; Actinides

$U_2Rh_3Si_5$, whose crystal structure is related to that of the heavy-fermion compounds URh_2Si_2 and URu_2Si_2, orders antiferromagnetically at $T_N = 26$ K [1,2]. Its heat capacity, magnetic susceptibility and electrical resistivity exhibit unusually large anomalies at T_N, e.g., a resistivity drop reaching a factor of 65 at $T \ll T_N$. This extraordinary behaviour was discussed in terms of overcritical-exchange-induced magnetic ordering within a two-level system [3]. In this work we present the determination of the magnetic structure of $U_2Rh_3Si_5$ by neutron powder diffraction.

A single-phase polycrystalline sample of mass 5 g has been prepared and characterized by techniques described previously [3]. The neutron diffraction studies in the temperature range 8.5–40 K have been carried out on the high-resolution DUALSPEC C2 diffractometer at the AECL Chalk River Laboratories. The Rietveld-analysis of the data was carried out using GSAS [4].

The crystal structure of $U_2Rh_3Si_5$ is monoclinic of type $Lu_2Co_3Si_5$ with space-group C2/c [1]. This structure can be also represented in the non-standard quasi-orthorhombic spacegroup I12/c1 [5]. In the following, we will use the I12/c1 notation. At $T = 40$ K, we find the lattice constants $a' = 9.8096(7)$ Å, $b = 11.3857(8)$ Å, $c = 5.8415(4)$ Å, $\beta' = 90.045(10)°$ ($a = 11.4132(8)$ Å, $\beta = 120.74(1)°$ in C2/c). The corresponding atom coordinates are close to the ones observed in $U_2(Ru_{0.65}Rh_{0.35})_3Si_5$ [6].

Fig. 1 shows the difference of the diffraction patterns recorded at $T = 8.5\ \text{K} < T_N$ and $T = 40\ \text{K} > T_N$. Several Bragg peaks of magnetic origin are observed, which can be indexed with integer h, k, l with $h + k + l =$ odd. This shows that the magnetic and the crystallographic unit cells are identical and that the body-centering translation $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ involves a 180° rotation of the magnetic moments.

It is not possible to reproduce the observed diffraction pattern assuming a collinear arrangement of the ordered magnetic moments. However, if the absolute value of the ordered moment is taken to be equal at all crystallographically identical uranium sites, one finds two (and only two) possible non-collinear arrangements. These structures are shown as insets in Fig. 1.

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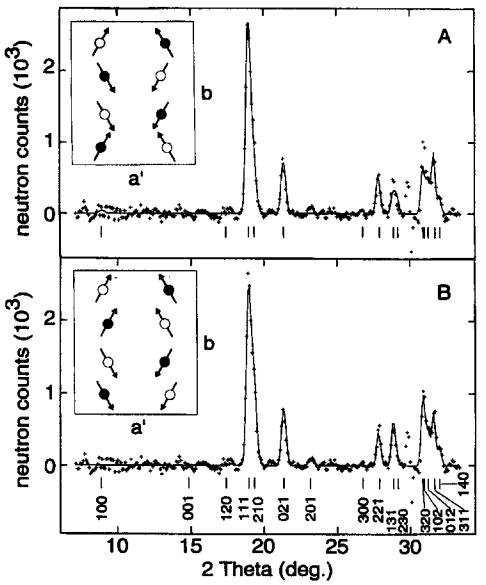

Fig. 1. Difference of the neutron diffraction patterns at $T = 8.5$ K and $T = 40$ K after correction for a small smooth paramagnetic contribution. The neutron wavelength is $\lambda = 1.505$ Å. Full lines are fits to the experimental data (+) of two possible magnetic structures shown in the inserts. U atom positions: $z = 0 \pm \delta$ (open cirles), $z = \frac{1}{2} \pm \delta$ (closed circles) with δ small. Structure B is more probable, see text.

Note that they are closely related, the overall magnetic pattern being shifted along b by one $a'c$ layer. The full lines in Fig. 1 are fits of these structures to the observed diffraction patterns, the only free parameters being the components μ_x and μ_y ($x \| a'$, $y \| b$) of the ordered moments. All other parameters were taken from the fit to the nuclear Bragg peaks. The component μ_z was set to zero in the final fits, because it proved to be below the detection limit of about 0.1–0.2μ_B. Structure B gives a better fit to the data ($R_p = 1.85\%$) than structure A ($R_p = 1.92\%$) and therefore is more probable (see also the 201 peak at $\approx 23°$). However, structure A cannot be definitely excluded.

We obtain ordered moments of $\mu^A = (\pm 0.87(4), \pm 1.52(3), 0)\mu_B$ for structure A and $\mu^B = (\pm 0.89(4), \pm 1.58(3), 0)\mu_B$ for structure B at $T = 8.5$ K, i.e., the moments are oriented within the $a'b$ plane and its angles are at $\pm 60(2)°$ or $\pm 120(2)°$ to the a' axis. These angles are temperature-independent below T_N. The absolute value of the ordered moments are $\mu^A = 1.75(5)\mu_B$ and $\mu^B = 1.81(5)\mu_B$. The temperature dependence of μ is consistent with a continuous magnetic transition at $T_N = (26.0 \pm 0.4)$ K.

The overall non-collinear arrangement of the ordered moments results from a different stacking of the μ_x and μ_y components. While μ_x orders in ferromagnetic 'wavy' bc-planes stacked antiferromagnetically along a', μ_y exhibits a two-up-two-down structure along b, else being stacked like μ_x. The different stacking of μ_x and μ_y may point to an anisotropic exchange interaction between the U ions. The observed magnetic structure is closely related to the structure found in $U_2(Ru_{0.65}Rh_{0.35})_3Si_5$ [6]. There, the ordering is collinear, the stacking being identical to that of μ_x in $U_2Rh_3Si_5$. This structure is analogous to URh_2Si_2 and URu_2Si_2. Our results demonstrate the change of the magnetic structure in the series $U_2(Ru_{1-x}Rh_x)_3Si_5$, anticipated in Ref. [7].

In the temperature dependence of the lattice constants below T_N a smooth decrease of a′ and b and an increase of c proportional to μ^2 is found. The relative changes of the lattice constants are $\Delta a'/a' = -0.8(3) \times 10^{-4}$, $\Delta b/b = -3.4(2) \times 10^{-4}$ and $\Delta c/c = +4.2(3) \times 10^{-4}$ between T_N and 8.5 K. Within the accuracy of the experimental data, the unit cell volume is conserved. Although the magnitude of the specific heat anomaly indicates a first-order transition [3], no evidence for a structural transition producing the anomalous behaviour of the bulk properties at T_N could be obtained from the present data.

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