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Crystal and magnetic structure in the itinerant 5f antiferromagnet UCr_2Si_2

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Abstract

We have grown a single crystal of UCr_2Si_2 for the first time. UCr_2Si_2 exhibits a structural transition from the $ThCr_2Si_2$-type structure ($I4/mmm$) to the triclinic ($P\bar{1}$) structure below $T_a = 210$ K. An antiferromagnetic structure was determined by neutron diffraction below $T_N = 27$ K, with a propagation vector $Q = [-\frac{1}{2}, \frac{1}{2}, 0]$ and a magnetic moment 0.65 μ_B/U oriented along [0, 0, 1] direction.

UT_2X_2 (T: transition metal, X: Si, Ge) [1] is one of the traditional 5f-electron systems the magnetic and transport properties of which have been studied systematically. In UCr_2Si_2 the small number of d-electrons and the small distance between U atoms give rise to an enhanced itinerant character for the 5f-electrons [2]. In fact, this compound shows a strong anisotropic susceptibility which obeys the Curie–Weiss law with an effective moment 2.54 μ_B/U, this being much smaller than that of a uranium free ion. The electronic specific heat coefficient $\gamma = 80$ mJ K^{-2} mol^{-1} is the signature for heavy fermions with medium mass enhancement. An antiferromagnetic ordering was observed below $T_N = 27$ K. Recently, we have succeeded in growing single crystals of UCr_2Si_2 for the first time [3]. The purpose of this study is to examine the structural and magnetic properties of UCr_2Si_2 by means of neutron scattering.

Polycrystalline samples were grown by the arc-melting of stoichiometric amounts of the raw materials. Single crystals were grown by the Czochralski-pulling method in a tetra-arc furnace (see [3] for details of the sample preparation). Neutron scattering experiments were carried out at the research reactor JRR-3 at the Japan Atomic Energy Research Institute (JAERI). Powder diffraction patterns were recorded using a high resolution powder diffractometer (HRPD) in the reactor hall. The crystal and magnetic structure was studied in detail with a single crystal sample using the triple axis spectrometers LTAS and TAS-2 (see [3]).

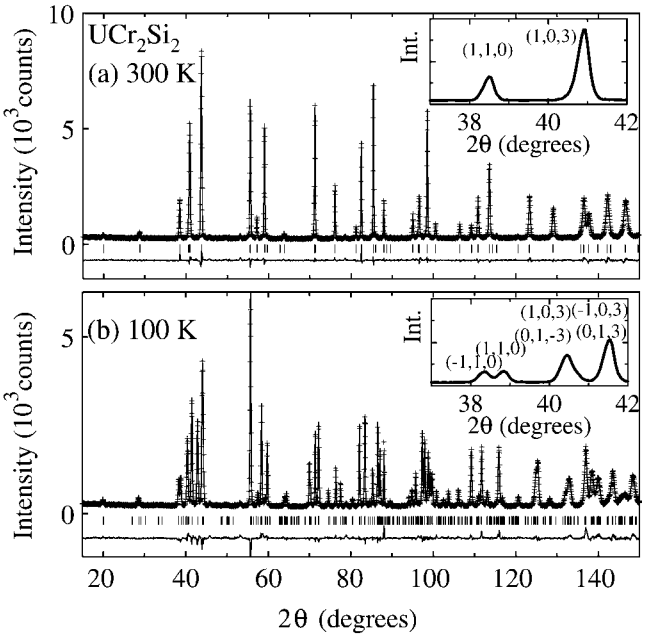

Figure 1. High resolution neutron powder diffraction pattern of UCr_2Si_2 at (a) 300 K and (b) 100 K. The solid curve on the main part of the graph is the calculated diffraction intensity after Rietvelt refinement. The insets show the diffraction pattern around the (1, 1, 0) and (1, 0, 3) peaks.

Table 1. Structural parameters of UCr_2Si_2. The number in parentheses following refined parameters represent the estimated standard deviations of the last significant digit(s)

Space group	Lattice parameter		Position			
T (K)	(nm, deg)	Atom (site)	x	y	z	B
$I4/mmm$	$a = b = 0.3913(1)$	U (2a)				0.271 886
300 K	$c = 1.0507(5)$	Cr (4d)				0.646 982
	$\alpha = \beta = \gamma = 90$	Si (4e)			0.386 31(5)	0.505 441
	$R_{wp} = 5.21$	$R_p = 3.97$	$R_e = 4.61$	$S = 1.1299$		
$P\bar{1}$	$a = 0.3906(5)$	U (1a)				0.2
	$b = 0.3903(6)$	U (1h)				0.2
	$c = 1.0506(2)$	Cr (2i)	0.521 34(0)	0.249 77(2)	0.030 8(1)	0.2
100 K	$\alpha = 91.49(6)$	Cr (2i)	−0.0320(3)	0.249 93(7)	0.472 01(3)	0.3
	$\beta = 88.51(3)$	Si (2i)	0.218 23(5)	0.385 67(5)	−0.017 6(0)	0.3
	$\gamma = 90.74(1)$	Si (2i)	0.510 53(9)	0.886 44(4)	0.486 75(1)	0.3
	$R_{wp} = 6.43$	$R_p = 4.95$	$R_e = 4.81$	$S = 1.3351$		

Figure 1(a) shows a neutron powder diffraction pattern at room temperature. The observed diffraction pattern was satisfactorily explained by the $ThCr_2Si_2$-type structure with space group $I4/mmm$. A very small amount of bcc-Cr was detected as an impurity phase. The obtained structural parameters after Rietvelt refinement are listed in table 1.

Figure 1(b) shows the neutron powder diffraction pattern at 100 K. We observed a clear splitting of the diffraction peaks, indicating a structural phase transition. The insets of figure 1

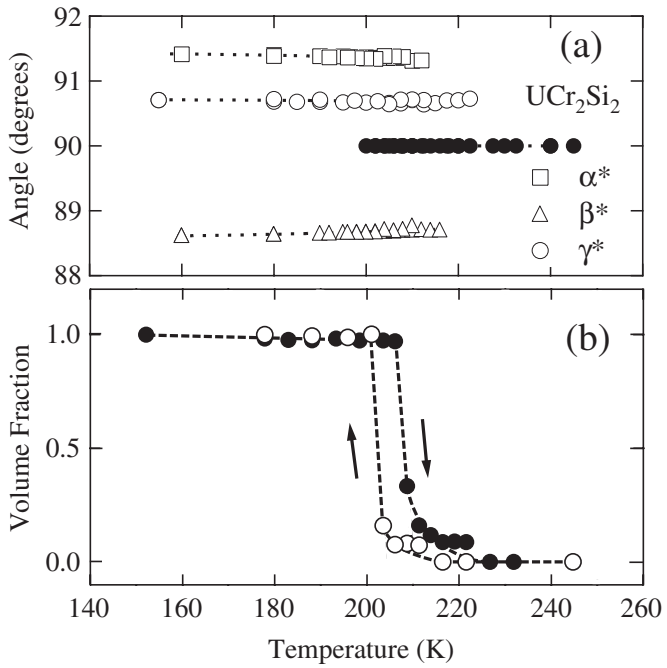

Figure 2. Temperature dependence of (a) the angles α^*, β^* and γ^* and (b) the volume fraction of the triclinic phase in UCr_2Si_2. Filled (open) circles indicate measurements where temperature was increasing (decreasing).

show the powder diffraction pattern around the (1, 1, 0) and (1, 0, 3) peaks. The splitting of these reflections implies that the low temperature phase has the triclinic structure. We found that the observed diffraction pattern was well explained by the model calculation with space group $P\bar{1}$ and crystal parameters as listed in table 1. The model calculation is in good agreement with the experimental data.

There is a large hysteresis of 20 K in angles with decreasing and increasing temperature, as shown in figure 2(a). Figure 2(b) indicates the temperature dependence of the volume fraction of triclinic phase estimated from the integrated intensity. The volume fraction shows a very sharp jump with a hysteresis of 5 K. These features are clear characteristics for a first-order transition.

The antiferromagnetic peaks were searched in the polycrystalline sample using the cold triple axis spectrometer LTAS. At 8 K we observed superlattice peaks at the scattering angles of the $(-\frac{1}{2}, \frac{1}{2}, 0)$ and $(-\frac{1}{2}, \frac{1}{2}, 1)$ peak positions, as shown in figure 3. These peaks disappear when the sample temperature is raised to 34 K (higher than $T_N = 27$ K). Therefore we concluded that these reflections are due to the antiferromagnetic ordering.

The magnetic scattering was studied in detail using a single crystal sample. Figure 4(a) shows the contour plot of the magnetic scattering intensity around the $(-\frac{3}{2}, \frac{3}{2}, 0)$ magnetic reflection in the $(h, k, 0)$ scattering plane. We observed two magnetic peaks due to the domain structure. The magnetic reflection indices were carefully assigned by taking the domain structure into consideration. Thus, we concluded that the antiferromagnetic wavevector is $Q = [-\frac{1}{2}, \frac{1}{2}, 0]$. It should be noted that the wavevector $[-\frac{1}{2}, \frac{1}{2}, 0]$ is not equivalent to $[\frac{1}{2}, \frac{1}{2}, 0]$ in the low temperature triclinic structure. The single-Q structure with $Q = [-\frac{1}{2}, \frac{1}{2}, 0]$ is the

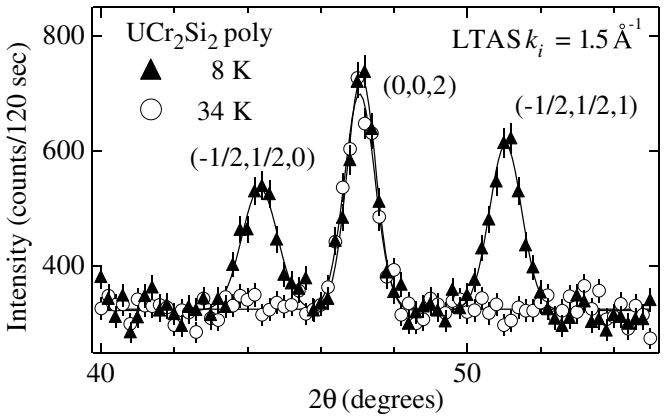

Figure 3. Powder diffraction pattern of UCr_2Si_2 at $T = 8$ and 34 K.

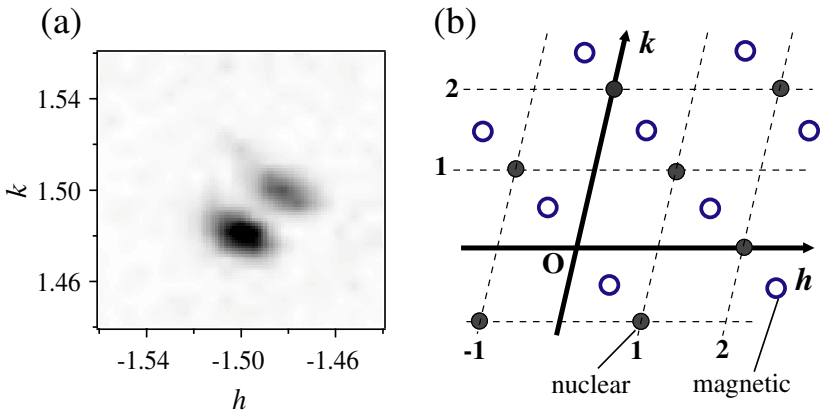

Figure 4. (a) Contour plot of the scattering intensity around $(-\frac{3}{2}, \frac{3}{2}, 0)$ magnetic reflections in UCr_2Si_2. (b) The position of the magnetic (filled circles) and nuclear (open circles) reflections in the $(h, k, 0)$ scattering plane.

reason for the observation of the two antiferromagnetic reflections around the $(-\frac{3}{2}, \frac{3}{2}, 0)$ peak position. The $(h, k, 0)$ plane and the positions of the nuclear and antiferromagnetic reflections are shown in the schematic representation in figure 4(b).

The observed integrated intensities of magnetic reflections were compared to the model calculation using the following equation

$$I_{\mathrm{mag}}(\boldsymbol{Q}) \propto |F_{\mathrm{mag}}(\boldsymbol{Q})|^2 f^2(\boldsymbol{Q})(\sin\alpha)^2 L(\theta), \tag{1}$$

where F_{mag} is the magnetic structure factor, f is the magnetic form factor of the uranium U^{3+} free ion, α is the angle between the ordered magnetic moment and the scattering vector Q and L is the Lorentz factor. The magnetic structure factor is unity in the present case, while the angle factor plays a dominant role for the relative intensities.

The experimental results are well explained by the model calculation with the ordered moment of uranium being 0.65 μ_{B}/U parallel to the [0, 0, 1] direction or the c-axis. From our experimental accuracy we cannot distinguish between the magnetic moment being parallel to the c-axis and it being perpendicular to the ab-plane.

In conclusion we have successfully grown a single crystal of the uranium intermetallic compound UCr_2Si_2 and measured the neutron scattering. Experimental results are summarized as follows:

(1) The crystal structure is found, as a result of the powder neutron diffraction experiment, to change from the $ThCr_2Si_2$-type structure ($I4/mmm$) to the triclinic structure ($P\bar{1}$) at the transition temperature $T_a = 210$ K.
(2) From the neutron scattering experiment on a single crystal sample, we determined an antiferromagnetic structure with propagation vector $Q = [-\frac{1}{2}, \frac{1}{2}, 0]$ and magnetic moment 0.65 μ_B/U oriented along [0, 0, 1] direction.

The distortion due to the structural change precludes the $[\frac{1}{2}, \frac{1}{2}, 0]$ magnetic domain which is equivalent to $[-\frac{1}{2}, \frac{1}{2}, 0]$ in the tetragonal structure.

Acknowledgments

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