Journal of Magnetism and Magnetic Materials 53 (1986) 309–312
North-Holland, Amsterdam

NEUTRON DIFFRACTION INVESTIGATION OF LONG-RANGE MAGNETIC ORDERING IN $TbCu_2Si_2$ AND $HoCu_2Si_2$

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Received 24 July 1985

The magnetic structures of $TbCu_2Si_2$ and $HoCu_2Si_2$ were determined by neutron diffraction. The crystal structure of both compounds is of the $ThCr_2Si_2$ type, space group 14/mmm. The magnetic unit cell at 4.2 K is described by a wavevector $k = (\frac{1}{2}, 0, \frac{1}{2})$. The Shubnikov space group is triclinic $P_a\,1(Sh_2)$. The magnetic order can be described as ferromagnetic (101) layers stacked antiferromagnetically with moments on rare-earth ions perpendicular to the *c*-axis.

1. Introduction

Continuing our neutron diffraction studies on the magnetic properties of $RECu_2Si_2$ intermetallic systems we report in this paper the results obtained for $TbCu_2Si_2$ and $HoCu_2Si_2$.

The structural and magnetic properties of $RECu_2Si_2$ compounds were published recently in a number of papers [1–4]. Only for $PrCu_2Si_2$ [5] and $DyCu_2Si_2$ [6] have their magnetic structures been determined by neutron diffraction studies. In $PrCu_2Si_2$ it is collinear antiferromagnetic of +-+- type, while in $DyCu_2Si_2$ it is autiferromagnetic with the magnetic in unit cell doubled along the *a*- and *c*-axes.

2. Experimental

The powder samples of $TbCu_2Si_2$ and $HoCu_2Si_2$ were prepared by arc-melting of stoichiometric amounts of high-purity elements. The samples were subsequently annealed in an argon atmosphere for 100 h at 1000 K. X-ray diffractograms of both compounds exhibited only lines characteristic of tetragonal body-centered structure of $ThCr_2Si_2$ type with the lattice parameters listed in table 1. They agree well with those given in ref. [1–4].

Neutron diffraction measurements at $\lambda = 0.1326$ nm were carried out on the powder diffractiometer DN-500 at the Świerk reactor EWA. The data were collected at room and liquid-helium temperatures.

The magnetic transition temperatures were determined by measuring the magnetic peak height as the cryostat was warming up slowly.

Refinement of the nuclear and magnetic data was performed using the Rietveld line profile analysis method. Nuclear scattering lengths were taken after ref. [7], magnetic form-factors for Tb^{3+} and Ho^{3+} ions after ref. [8].

3. Results

3.1. The crystal structures at 300 K

The reflections observed on the room-temperature neutron diffraction patterns were indexable in the quadratic system respecting the extinction condition for the space group 14/mmm – only peaks

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Table 1
Refined parameters from neutron intensities of $TbCu_2Si_2$ and $HoCu_2Si_2$ at 293 and 4.2 K. R_n, R_m, R_{wp} are the agreement values for nuclear, magnetic and weighted profile intensities, R_{exp} is the expected agreement value depending on the statistical accuracy of the data [7]. B is the overall temperature factor, ϕ_x, ϕ_z are the angles between the magnetic moment and the x, z-axes (tetr.)

	$TbCu_2Si_2$		$HoCu_2Si_2$	
	293 K	4.2 K	293 K	4.2 K
a(nm)	0.3982(1)	0.3982(1)	0.3952(1)	0.3931(1)
c(nm)	0.9961(3)	0.9982(5)	0.9971(3)	0.9926(8)
V(nm^3)	0.15794(13)	0.15828(16)	0.15573(13)	0.15338(22)
c/a	2.5015(14)	2.5070(21)	2.5230(14)	2.5250(27)
z_{Si}	0.3892(6)	0.3831(14)	0.3798(9)	0.3785(19)
B(nm^2)	0.009(3)	0.003(1)	0.005(2)	0.003(1)
$\mu(\mu_B)$		8.50(15)		8.22(37)
ϕ_x(deg)		23.4(1.3)		7.5(3.1)
ϕ_z(deg)		90		90
R_n(%)	4.4	2.6	8.12	6.7
R_m(%)	–	5.4	–	6.6
R_{wp}(%)	8.15	7.5	13.9	12.7
R_{exp}(%)	3.37	2.3	4.35	4.0

with indices $h + k + l = 2n$ are present. The intensities were calculated for the atoms distributed in the following sites:

Tb	or Ho	in 2(a)	0,0,0;	$\frac{1}{2},\frac{1}{2},\frac{1}{2}$;
Cu		in 4(d)	$\frac{1}{2},0,\frac{1}{4}$;	$0,\frac{1}{2},\frac{1}{4}$
			$\frac{1}{2},0,\frac{3}{4}$;	$0,\frac{1}{2},\frac{3}{4}$
Si		in 4(e)	$0,0,z$;	$0,0,\bar{z}$;
			$\frac{1}{2},\frac{1}{2},\frac{1}{2}+z$;	$\frac{1}{2},\frac{1}{2},\frac{1}{2}-z$.

The lattice parameters of the $ThCr_2Si_2$ unit cell and the positional parameter of silicon, z, were refined. The results are listed in table 1.

3.2. The magnetic structure at 4.2 K

The magnetic structures for both compounds were determined using neutron diffraction patterns obtained at 4.2 K. The magnetic superlattice reflections observed at this temperature were easily

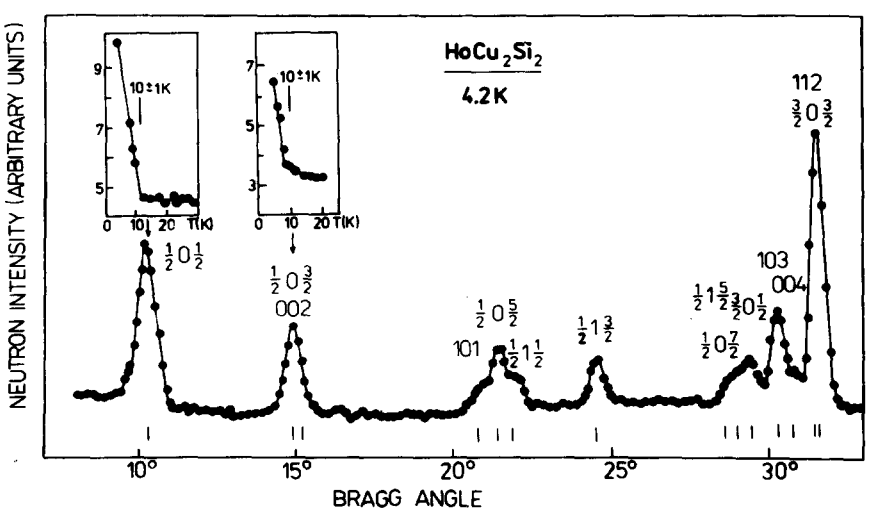

Fig. 1. Neutron diffraction pattern of $HoCu_2Si_2$ at 4.2 K. Peak height against temperature curve is shown in the inset.

Table 2
Calculated and observed neutron intensities of $TbCu_2Si_2$ and $HoCu_2Si_2$ at 4.2 K. The indices refer to the orthorhombic magnetic cell

hkl	$TbCu_2Si_2$			$HoCu_2Si_2$		
	I_{nuc}	I_{mag}	I_{obs}	I_{nuc}	I_{mag}	I_{obs}
101	–	103145	100513	–	49849	50338
103	–	52410	60652	–	23024	23457
004	5573	–	7109	4077	–	4882
012	264	–	190	587	–	1921
202	264	–	190	587	–	1921
105	–	25525	25848	–	10288	11552
111	–	30411	29526	–	6396	7016
113	–	27194	28743	–	6727	8118
210	20	–	0	122	–	875
107	–	13981	12910	–	5098	3608
115	–	21437	21137	–	5875	5307
301	–	11559	12753	–	4665	6343
016	11773	–	11223	8400	–	8400
206	11773	–	11212	8400	–	8400
008	8040	–	7820	4882	–	5715
303	–	10244	10314	–	3930	3954
214	81163	–	81210	45000	–	44980

indexed on a magnetic unit cell four times larger than the crystallographic one, i.e. doubled in the direction of the a-axis and along the c-axis. The corresponding magnetic wavevector is thus $k = (\frac{1}{2},0,\frac{1}{2})$. The observed magnetic reflections *hkl* shown in fig. 1 are indexed and an orthorhombic unit cell $a_1 = 2a$, $a_2 = a$, $c_1 = 2c$. Since the indices with $h = 2n + 1$ and $l = 2n + 1$ are present, the orthorhombic magnetic unit cell is B-centered and its a_1 and c_1 axes are anticentered.

The refinement of the structure parameters was carried out by the least-squares method. The data obtained are collected in table 1, and the calculated and observed intensities are compared in table 2.

The $(\frac{1}{2}\ 0\ \frac{1}{2})$ magnetic peak height versus temperature curve gives the transition temperatures at $(1\ 1\ \pm 1)$K and $(1\ 0\ \pm 1)$K for $TbCu_2Si_2$ and $HoCu_2Si_2$ respectively.

4. Conclusions

The magnetic structures of $TbCu_2Si_2$ and $HoCu_2Si_2$ are displayed in fig. 2. The relations

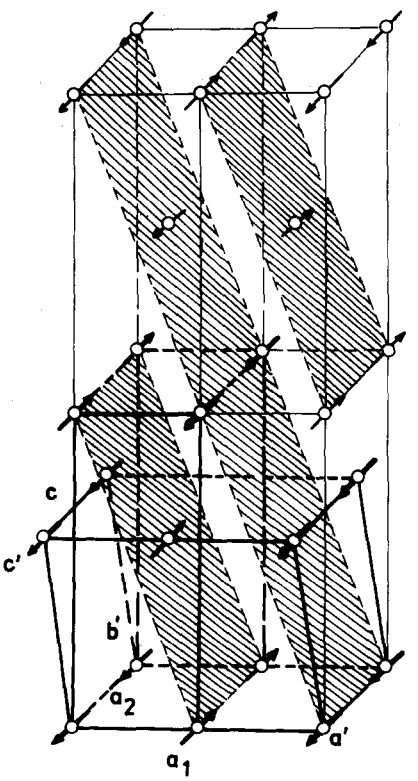

Fig. 2. Chemical cell (tetragonal) $a_1 = a_2$, c, $\theta_1 = \theta_2 = \theta_3 = 90°$. Magnetic cell (orthorhombic) $2a_1$, a_2, $2c$ and (triclinic) $a' = 2a_1$, $b' = a_2$, $c' = \frac{1}{2}(+a_1 + a_2 + c)$.

Table 3
The lattice constants, angles and volumes of the chemical tetragonal cell and the reduced triclinic magnetic cell of fig. 2 at 4.2 K

		$TbCu_2Si_2$	$HoCu_2Si_2$
chemical tetragonal	a(nm)	0.3982	0.3931
I-centered c(nm)		0.9982	0.9926
magnetic triclinic	a'(nm)	0.7964	0.7862
P_a	b'(nm)	0.3982	0.3931
a-anticentered	b'(nm)	0.5730	0.5688
	α'(deg)	110.3	110.2
	β'(deg)	110.3	110.2
	γ'(deg)	90.0	90.0

between the tetragonal body-centered crystallographic, the B-centered orthorhombic and the reduced triclinic unit cells is shown. The dimensions of the latter are also listed in table 3.

The observed magnetic structure belongs to the Shubnikov $P_a\bar{1}$ (Sh^7_2) space group [9], similar to the magnetic structures of isostructural $TbCu_2Ge_2$ and $HoCu_2Ge_2$ [10] and $DyCu_2Ge_2$ [11].

The observed magnetic ordering in $TbCu_2Si_2$ and $HoCu_2Si_2$ can be described as a sequence of ferromagnetic sheets coupled antiferromagnetically with the sequence +-+-. The sheet is the (101) plane of the tetragonal, $ThCr_2Si_2$ type unit cell. The magnetic moments are normal to the c-axis in both compounds studied, but make different angles with the a-axis: $(23.4 \pm 1.3)°$ and $(7.5 \pm 3.1)°$ in $TbCu_2Si_2$ and $HoCu_2Si_2$ respectively. The antiferromagnetic ordering determined in both compounds belongs to the AF type III according to the notation proposed in ref. [12].

The determined magnitude of the magnetic moment on the Tb^{3+} ion of $8.50(15)\mu_B$ is close to the free-ion value $9\mu_B$ whereas in $HoCu_2Si_2$ it is $8.82(57)\mu_B$, i.e. smaller than $10\mu_B$. This may be due to the lack of saturation at 4.2 K. No magnetic moment could be detected on the Cu ion within the accuracy of the powder neutron diffraction experiment.

Since the ordering temperatures for $RECu_2Si_2$ compounds are low, weak magnetic interactions of the RKKY type may be assumed. On the other hand interactions of the 4f shells with the lattice through the crystal fields seem to be more pronounced. This may be deduced from the analysis of the temperature dependence of the c/a ratios in the $RECu_2Si_2$ series which gave the correlation between the sign of the Stevens factor and the axis of easy magnetization [4]. For the compounds with RE = Sm, Er, Tm the magnetic moment was deduced to be parallel to the c-axis, while for RE = Pr, Nd, Tb, Dy, Ho it is normal to it. The directions of the Tb^{3+} and Ho^{3+} magnetic moments observed in our experiment agree with this conclusion.

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