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Note

Temperature Dependence of the Lattice Constants and Thermal Expansion Coefficients of UMn_2Si_2 and UMn_2Ge_2 Compounds

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Lattice constants of polycrystalline UMn_2Si_2 and UMn_2Ge_2 samples were measured in the temperature range 80—300 K using the X-ray diffraction method. The anomalous behaviour of the lattice constant and the expansion coefficient, α_a of UMn_2Ge_2 was observed at 150 K. The magnetostriction contribution of uranium sublattice is responsible for these anomalies.

INTRODUCTION

Ternary MT_2X_2 compounds (M = RE, Th, U; T = 3d-, 4d-, 5d-transition metal; X = Si, Ge) have attracted great interest because of the variety in their magnetic properties. X-ray and neutron diffraction studies indicate that UMn_2Si_2 and UMn_2Ge_2 compounds have the tetragonal crystal structure of $ThCr_2Si_2$ type (space group $I4/mmm$) with atoms occupying the following sites: U- 2(a), Mn- 4(d) and Si or Ge- 4(e).[1]

The magnetometric and neutron diffraction data indicate that UMn_2Si_2 and UMn_2Ge_2 are ferromagnets with the Curie temperature points of 377 K and 390 K, respectively. At low temperatures, $T_{or} = 80$ K for UMn_2Si_2 and $T_{or} = 150$ K for UMn_2Ge_2, an additional phase transition is observed. Below these temperatures, the magnetic moments localized on uranium and manganese atoms are coupled ferromagnetically while above additional phase transition temperature only the Mn sublattice orders ferromagnetically.[2]

In this paper we report the temperature dependence of the lattice constants for polycrystalline samples of UMn_2Si_2 and UMn_2Ge_2.

RESULTS

Powder samples[2] were prepared by a combination of arc melting and the solid state diffusion technique. High purity components were melted and annealed for 100 hours at 800 °C and cooled down to room temperature. Diffraction patterns were collected using a DRON-3 powder diffractometer

(manufactured by Burieviestnik, USSR) with Co K_α radiation. Temperature variation over the 80—300 K range was achieved by means of a vapour cryostat with a thermoregulator (both produced by KRIOPAN, Poland).[3] In the whole investigated region, temperature stabilization was ± 1 K or better. The temperature was measured using a copper-constantan thermocouple placed near the sample surface.

For the polycrystalline UMn_2Si_2 and UMn_2Ge_2 samples the evolution of lattice constants was determined through the (028), (233), (136), (037), (235) and (138) peak positions. At high angles K_{α_1} and K_{α_2} radiation gave completely separated peaks, so both of them were used to determine the lattice constants. The difference between the lattice constants evaluated from peak positions of K_{α_1} and K_{α_2} radiation is negligible: the presented error is of three standard deviations and is the same for each diffraction pattern.

Figure 1 shows the temperature dependence of lattice parameters, unit cell volume and a/c ratio. For both compounds the lattice constants increase

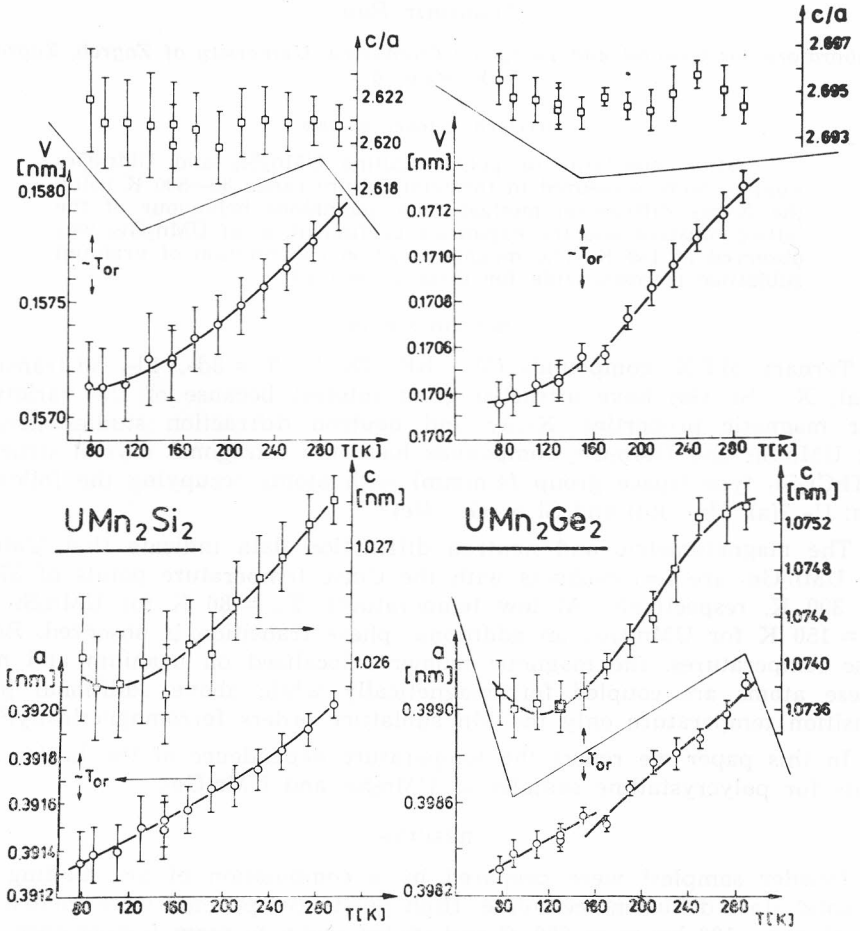

Figure 1. Temperature dependence of a and c lattice constants, cell volumes V and c/a ratio for UMn_2Si_2 and UMn_2Ge_2. Drawn lines are calculed by polynomials.

with increasing temperature. The temperature dependence of lattice parameters could be approximated using the well known Debye formula:

$$a\,(T) = a_0 + I_a \frac{T^4}{\Theta_a^3} \int_0^{\Theta_a/T} \frac{x^3\,dx}{e^x - 1}$$

$$c\,(T) = c_0 + I_c \frac{T^4}{\Theta_c^3} \int_0^{\Theta_c/T} \frac{x^3\,dx}{e^x - 1}$$

or by a third degree polynomial:

$$a\,(T) = a_0 + a_1T + a_2T^2 + a_3T^3$$

$$c\,(T) = c_0 + c_1T + c_2T^2 + c_3T^3$$

The last approximation[3,4] is confirmed by 3-dim chain model calculations[5] (of course $a_1\,(c_1) = 0$ for $T \rightarrow 0$ as a result of the 3^{rd} law of thermodynamics). Collected in Tables I and II, the values of a_i, c_i and a_0, c_0, I_a, I_c, Θ_a, Θ_c

TABLE I

The values of a_i *and* c_i *constants*

Compound	UMn_2Si_2	UMn_2Ge_2	
T (K)	80—300	80—150	150—300
a_0 (nm)	.39114(13)	.39811(4)	.39778(8)
a_1 (nm/K)	$2.9(2.4) \cdot 10^{-6}$	$2.76(36) \cdot 10^{-6}$	$4.48(33) \cdot 10^{-6}$
a_2 (nm/K^2)	$-5.6(1.4) \cdot 10^{-9}$	—	—
a_3 (nm/K^3)	$1.9(2.5) \cdot 10^{-11}$	—	—
c_0 (nm)	1.0269(10)	1.0762(5)	
c_1 (nm/K)	$-2.3(1.8) \cdot 10^{-5}$	$-5.4(1.0) \cdot 10^{-5}$	
c_2 (nm/K^2)	$1.4(9) \cdot 10^{-7}$	$3.3(6) \cdot 10^{-7}$	
c_3 (nm/K^3)	$-1.8(1.6) \cdot 10^{-10}$	$-5.3(1.1) \cdot 10^{-10}$	

parameters (respectively) were calculated using the least-square method. For the second procedure, the Debye function tables (for $x = \Theta/T > 10$)[6] and their rational approximation ($x \leq 10$)[7] were used. In the case of UMn_2Ge_2 the results of fitting by the polynomial, performed for $a\,(T)$ dependence, were improved after dividing the whole temperature range in two parts: 80—150 K and 150—300 K. Of the two fitting procedures, experimental data are better expressed by polynominals, but also the Debye method results are presented (Table II), because of the estimated Debye temperatures Θ.

For UMn_2Ge_2, the jump in the temperature dependence of the lattice parameter a was obtained ($\Delta a/a \cong 1.79 \cdot 10^{-4}$) near the temperature of 150 K. This fact is reflected in the calculated changes of the unit cell volume V ($\Delta V/V \cong 3.6 \cdot 10^{-4}$ at T_{or}). No hysteresis effects were observed for this transition region.

On the basis of functions $a\,(T)$ and $c\,(T)$ expressed by polynomials (see Table I), the temperature dependence of the thermal linear expansion coef-

TABLE II

The values of Debye formula constants

Compound	UMn_2Si_2	UMn_2Ge_2
T (K)	80—300	80—300
Θ_a (K)	438	508
a_0 (nm)	.39132(1)	.39832(1)
I_a (nm/K)	$1.24(4) \cdot 10^{-5}$	$1.61(6) \cdot 10^{-5}$
Θ_c (K)	904	807
c_0 (nm)	1.02573(5)	1.07352(6)
I_c (nm/K)	$6.5(4) \cdot 10^{-5}$	$6.7(4) \cdot 10^{-5}$

ficient ($\alpha_a = 1/a \; da/dt$ and $\alpha_c = 1/c \; dc/dT$) and the volume expansion coefficient ($\beta_V = 1/V \; dV/dT$) were calculated (see Figure 2). For UMn_2Ge_2 the anomaly in $\alpha_a(T)$ and $\beta_V(T)$ is present, whereas for UMn_2Si_2 all expansion coefficients are continuous in the observed temperature range. The estimated values of volume expansion coefficients at 100 K: $\beta_V(UMn_2Si_2) = 11 \cdot 10^{-6}$ K^{-1} and $\beta_V(UMn_2Ge_2) = 10 \cdot 10^{-6}$ K^{-1} can be compared with the reported one for isostructural URu_2Si_2 [8] β_V (100 K) $= 4 \cdot 10^{-6}$ K^{-1}.

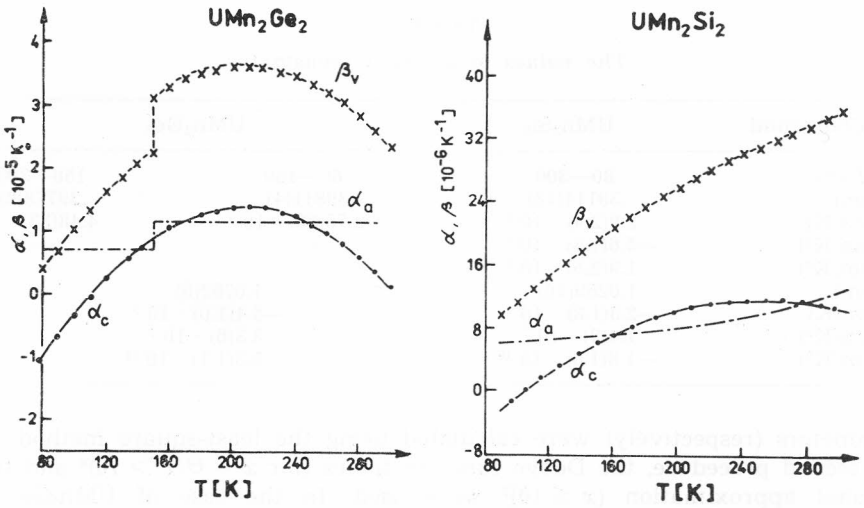

Figure 2. Calculated temperature dependence of thermal expansion coefficients: α_a, α_c and β_V for UMn_2Si_2 and UMn_2Ge_2.

Regarding the determined Debye temperatures two facts should be pointed out. Firstly, the observed anisotropy $\Theta_c/\Theta_a \cong 2$ agrees with the Mössbauer studies[9] results for $EuRu_2Ge_2$ ($\Theta_c/\Theta_a = 450$ K/180 K), but does not agree with the conclusions drawn from the X-ray refinement performed for isostructural compounds of $HoRu_2Si_2$ [10], $CeOs_2Si_2$ [11] or $TbRu_2Si_2$ [12]. Namely, the refined anisotropic temperature factors $U_{33} > U_{11}$ (U_{22}) indicate that one should rather observe $\Theta_c < \Theta_a$. Secondly, the Debye temperature values obtained here are larger than those determined by means of other methods for the same structural $ThCr_2Si_2$-type compounds (*e. g.* low temperature specific heat

measurement for $LaIr_2Si_2$ giving $\Theta_{iso} = 385$ K [13] or the mentioned Mössbauer studies for $EuRu_2Ge_2$ in the range of 4.1—600 K [9]).

CONCLUSION

Temperature dependence of the lattice constants and expansion coefficients confirms the magnetic phase transition in UMn_2Ge_2 at 150 K related to the disordering of magnetic moments in the uranium sublattice.

The thermal expansion coefficient in magnetically ordered materials is the sum of phonon, electron and magnetic components. The obtained results indicate that in UMn_2Ge_2 the magnetostriction contribution of the uranium sublattice is responsible for the anomalous behaviour of a (T) and α_a (T).

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Ovisnost dimenzije elementarne ćelije o temperaturi i koeficijent ekspanzije spojeva UMn_2Si_2 i UMn_2Ge_2

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Metodom rendgenske difrakcije izmjerene su dimenzije elementarne ćelije uzoraka UMn_2Si_2 i UMn_2Ge_2 u području temperature 80—300 K. Pri 150 K opaženo je anormalno ponašanje dimenzija elementarne ćelije i koeficijenta ekspanzije α_a spoja UMn_2Ge_2. Doprinos magnetostrikcije podrešetke urana odgovoran je za navedene anomalije.