

THERMAL EXPANSION OF POLYCRYSTALLINE UPt_2 , UPt_3 AND UPt_5

A. DE VISSER and J.J.M. FRANSE

Natuurkundig Laboratorium der Universiteit van Amsterdam, Valckenierstraat 65, 1018 XE Amsterdam, The Netherlands

The thermal expansion of the polycrystalline compounds UPt_2 , UPt_3 and UPt_5 was measured in the temperature range 1.4 up to 80 K. Of particular interest is the spin-fluctuation compound UPt_3 , which, after recently being discovered to behave as a heavy-fermion superconductor, nowadays receives much attention. Its thermal expansion reveals a large anomalous contribution below 50 K. This contribution appears as a low-temperature upturn in a plot of α/T versus T^2 , and is associated with spin fluctuations. In the compounds UPt_2 and UPt_5 a similar upturn appears, although less pronounced. The thermal expansion data are compared with previously published specific heat data on the same samples.

The uranium-platinum system offers the possibility to study the competition between itinerant and local behaviour of the 5f electrons in uranium intermetallics. Along this series the U-U distance varies from 3.65 Å in orthorhombic UPt , 3.81 Å in orthorhombic UPt_2 , 4.13 Å in hexagonal UPt_3 , to 5.25 Å in cubic UPt_5 , thus leading to a considerable decrease in overlap of the 5f orbitals.

Of particular interest is the compound UPt_3 , which exhibits unusual low-temperature properties in the magnetic susceptibility and magnetization [1], in the specific heat [1, 2] and in the electrical resistivity [3], that could properly be described in a spin-fluctuation model. Moreover the recent discovery of superconductivity ($T_c \approx 0.5$ K) [4], and the classification of UPt_3 as a heavy fermion system with an effective mass $m \approx 180m_0$ [2], has attracted much attention to this compound. In UPt_2 and UPt_5 spin-fluctuation effects are less pronounced.

In this paper we present thermal expansion measurements in the temperature range 1.4–80 K on the compounds UPt_2 , UPt_3 and UPt_5 . The experimental data are compared with previously published specific heat measurements on the same samples [1, 5], except for UPt_2 . The thermal expansion of equiatomic UPt was recently studied by Kamma et al. [6].

The polycrystalline compounds were prepared by arc melting the appropriate amounts of constituents in a titanium gettered argon atmosphere. After casting of the melt into a water-

cooled copper crucible, cylindrical samples were obtained with a diameter of 6 mm and a length of 10 mm. The as-cast samples were annealed in evacuated sealed silica tubes at 1000°C for a period of 10 days. Finally, all samples were machined by means of spark erosion into the proper shape: i.e. planparallel and with a hole of 2 mm diameter along the cylinder axis.

For the thermal expansion measurements a sensitive three-terminal capacitance method was used, with a detection limit for changes in length of 0.1 Å. A detailed description of the thermal expansion cell was given by Hölscher in his thesis [7]. Data were gathered stepwise: below 15 K each temperature step (0.3 K) was made three times, i.e. up, down and up again, whereas above 15 K only steps upwards (mounting up to 3 K) were made. The accuracy in the results decreased with increasing temperature due to small temperature gradients over the cell, and is limited to $2 \times 10^{-7} \text{ K}^{-1}$ in α near 80 K.

The experimental results, obtained within several runs, are presented in fig. 1 as $\alpha = 1/L(dL/dT)$ versus T . Relative length changes, $\Delta L/L$, were deduced by integration, see fig. 2.

The overall picture displays a rather large thermal expansion. Especially the compound UPt_3 reveals a large additional contribution below 50 K. The maximum in the α versus T plot roughly coincides with the maximum in the susceptibility (16 K) [1]. The broad nature of this peak gives once more evidence that the anomaly near 16 K cannot be attributed to an antifer-

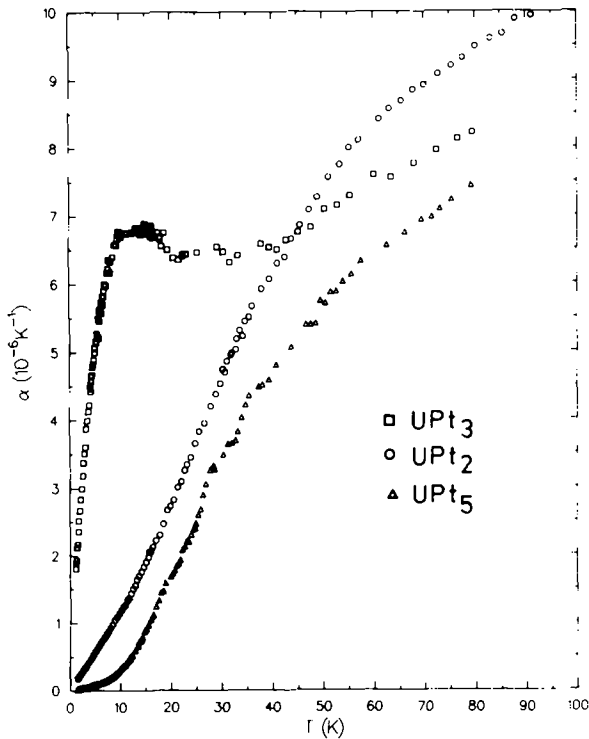


Fig. 1. The thermal expansion, $\alpha = 1/L(dL/dT)$, for polycrystalline UPt₃, UPt₂ and UPt₅.

romagnetic ordering of local moments.

In a first attempt to describe the data with an electron and phonon contribution, according to

$$\frac{\alpha}{T} = a + bT^2, \quad (1)$$

the data are presented as α/T versus T^2 in fig. 3. For UPt₃ such a description is far from appropriate: instead the additional contribution appears as a pronounced upturn. The analysis of the specific heat data of UPt₃ was successful with a $T^3 \ln T$ contribution, accounting for spin-fluctuation effects and leading to a similar upturn in a plot of C/T versus T^2 [2, 4, 5]. Assuming temperature independent Grüneisen parameters the spin-fluctuation contribution to the thermal expansion should follow the same temperature dependence. However, such an analysis leads yet not to satisfying results. This is probably due to a

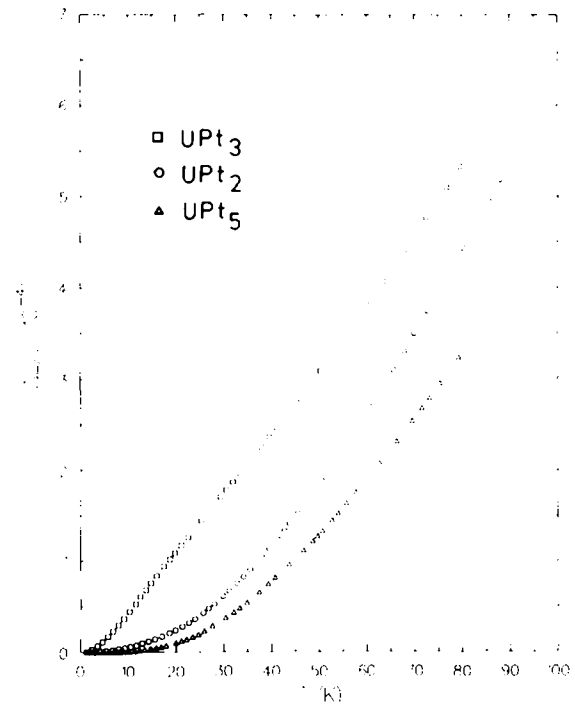


Fig. 2. The relative length changes, $\Delta L/L = (L(T) - L(1.4 \text{ K}))/L(1.4 \text{ K})$, for UPt₃, UPt₂ and UPt₅.

considerable amount of preferred orientation in the polycrystalline sample: preliminary thermal expansion measurements on a single crystalline sample reveal a large anisotropy between the basal plane and the hexagonal axis. Nevertheless we believe that the low-temperature upturn must be ascribed to spin-fluctuation effects.

A similar upturn, although small, can be observed for the compounds UPt₂ and UPt₅. These anomalies are found in the specific heat as well, and could also be ascribed to spin fluctuations [5]. In orthorhombic UPt₂ this contribution might be obscured due to preferred orientations too. For both compounds eq. (1) holds in the temperature interval $50 < T^2 < 250 \text{ K}^2$. Coefficients a and b are listed in table I, together with electron (γ) and phonon (β) coefficients from the specific heat analysis. Striking is the large electronic coefficient for UPt₂ and the large phonon coefficient for UPt₅.

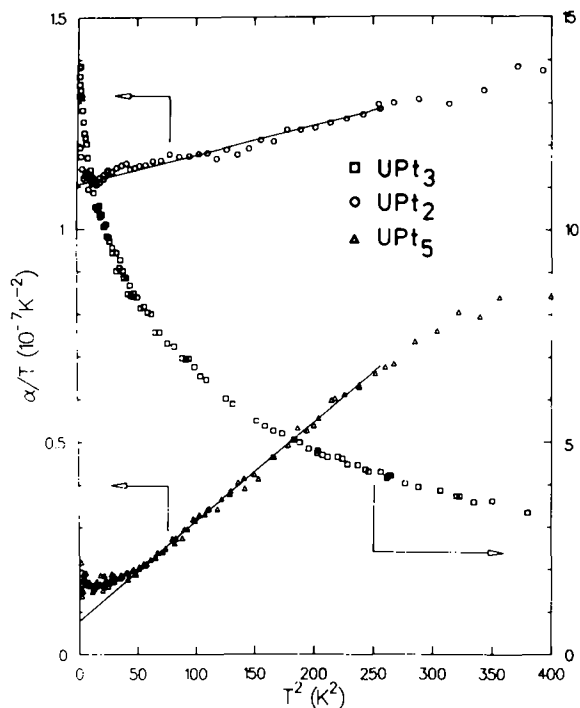


Fig. 3. The thermal expansion of UPt_2 , UPt_3 and UPt_5 plotted as α/T versus T^2 . The full lines denote fits with coefficients listed in table I. Note the different scales on the vertical axes.

The Grüneisen parameters $\Gamma_e = 3aV_m/(\kappa\gamma)$ and $\Gamma_{ph} = 3bV_m/(\kappa\beta)$ cannot be calculated due to the lack of compressibility data (κ will be about $1 \times 10^{-11} \text{ m}^2\text{N}^{-1}$). However, from the deduced $\kappa\Gamma_e$ values, see table I, it follows that Γ_e is at least one order of magnitude larger than Γ_{ph} . A similar

observation was made for equiatomic UPt [6]. With the above given estimate for the compressibility of the uranium-platinum compounds, the values for Γ_{ph} , calculated from the data in table I seem to be too small. Unfortunately, data on corresponding thorium-platinum or lanthanum-platinum compounds are not available. Therefore a more thorough analysis of the various contributions to the thermal expansion is, at this moment, not possible.

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Table I

Temperature coefficients and Grüneisen parameters from the thermal expansion data fitted to $\alpha/T = a + bT^2$ in the temperature interval $50 < T^2 < 250 \text{ K}^2$, and specific heat data fitted to $C/T = \gamma + \beta T^2$ in the same temperature interval (from ref. 5), for UPt_2 and UPt_5 . Data on UPt are taken from ref. 6. For UPt_3 the spin-fluctuation enhanced electronic coefficients are listed

Parameter	Units	UPt	UPt_2	UPt_3	UPt_5
a	10^{-8} K^{-2}	5.60	11.0	148	0.78
γ	$\text{mJ}/(\text{mol-f.u.} \cdot \text{K}^2)$	100	75	422	24
$\kappa\Gamma_e$	$10^{-11} \text{ m}^2/\text{N}$	4.4	14.8		6.0
b	10^{-10} K^{-4}	0.55	0.71		2.3
β	$\text{mJ}/(\text{mol-f.u.} \cdot \text{K}^4)$	1.4	1.0		4.2
$\kappa\Gamma_{ph}$	$10^{-11} \text{ m}^2/\text{N}$	0.3	0.7		1
V_m	$10^{-5} \text{ m}^3/\text{mol-f.u.}$	2.66	3.36	4.24	6.15