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Effect of pressure on the Curie temperature of single-crystal UGe_2

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Abstract

The linear thermal expansion of single-crystal UGe_2 has been measured in the temperature range 4.2 K$<T<$300 K under high pressure up to 20 kbar. The linear thermal expansion coefficient α_i (K^{-1}) ($i=a$, b and c) exhibits a discontinuity near 50 K and a large anisotropy, which corresponds to ferromagnetic order. The discontinuous changes in the values of α_a, α_b and α_c at T_C are 25×10^{-6} K^{-1}, 40×10^{-6} K^{-1} and -10×10^{-6} K^{-1} respectively. It is found that T_C decreases with increasing pressure and becomes lower than 4.2 K at 20 kbar. The initial decreasing rate of T_C was estimated to be $dT_C/dP=-0.7$ K $kbar^{-1}$. The data are analysed briefly by assuming a weak ferromagnetic theory.

1. Introduction

Uranium compounds are well known to have 5f electrons exhibiting an intermediate character between the localized 4f electron system and the itinerant d electron system and to display heavy fermion behaviour. Application of pressure changes drastically the electronic state of these compounds because the hybridization between f electron states and the conduction band increases with a decrease in interatomic spacing [1, 2].

UGe_2 has an orthorhombic crystal structure having lattice constants $a=4.09$ Å, $b=15.20$ Å and $c=3.96$ Å [3]. It was reported that UGe_2 is a ferromagnetic compound with Curie temperature $T_C=52$ K and its physical behaviour exhibits a large anisotropy reflecting the non-cubic crystal structure [3].

In the present work we made an attempt to measure the thermal expansion $\Delta l/l$ and its temperature coefficient α (K^{-1}) of single-crystal UGe_2 under hydrostatic pressure up to 20 kbar in order to examine an instability of the ferromagnetism at high pressure.

2. Experimental procedure

Single-crystal UGe_2 was grown by a Czochralski pulling method. The details of sample preparation and characterization were reported elsewhere [3]. Thermal expansion was measured in the temperature range between 4.2 K and 300 K by means of strain gauge method (Kyowa Dengyo KFL-02-C1-11) [4]. The strain gauge was glued on a clean surface of the specimen in the direction parallel to each crystal axis (a, b and c axes). Copper (99.999%) was used as a reference material. We observed a difference in length between Cu and UGe_2. The hydrostatic pressure was generated by the piston–cylinder method up to 20 kbar. The pressure transmitting medium is a mixture of Fluorinert FC70 and FC77. The details of the high pressure apparatus were reported previously [5].

3. Results and discussion

Figure 1 shows the temperature dependence of the linear thermal expansion $(\Delta l/l)_a$, $(\Delta l/l)_b$ and $(\Delta l/l)_c$ along the a, b and c axes at ambient pressure. $(\Delta l/l)_a$ decreases gradually with decreasing temperature, but it begins

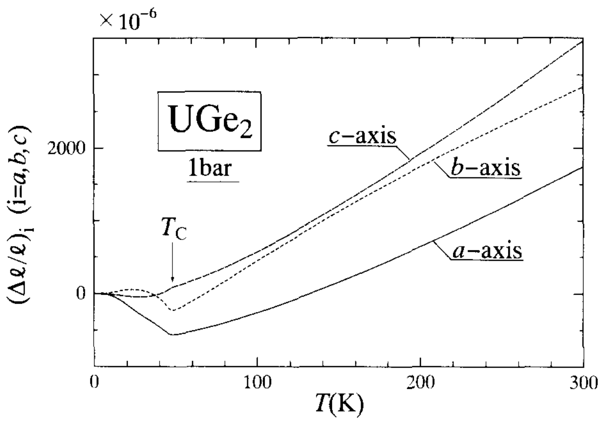

Fig. 1. Temperature dependence of linear thermal expansion $(\Delta l/l)_a$, $(\Delta l/l)_b$ and $(\Delta l/l)_c$ of single-crystal UGe_2 at ambient pressure. The Curie temperature T_C is shown by an arrow.

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to increase around 50 K (T_C). The anomalous increase is due to the ferromagnetic order. Below T_C, a small knee is observed around 25 K, which was also observed in the temperature dependence of thermoelectric power and electrical resistivity [3]. The temperature dependence of $(\Delta l/l)_b$ is almost the same as that of $(\Delta l/l)_a$ except for a broad maximum around 30 K. In the case of the c axis, the behaviour is different from those of the a and b axes below T_C, reflecting the orthorhombic crystal structure. Below 50 K, $(\Delta l/l)_c$ decreases with decreasing temperature.

We show in Fig. 2 the linear thermal expansion $(\Delta l/l)_c$ at high pressure. The anomaly due to ferromagnetic ordering becomes less prominent on application of pressure and then T_C also decreases.

Figure 3 shows the temperature dependence of the linear thermal expansion coefficient α_c under high pressure. α_c was obtained by differentiating $(\Delta l/l)_c$ with respect to T. On ferromagnetic transition, α_c increases discontinuously with $\Delta\alpha_c = -10\times10^{-6}\ K^{-1}$ at ambient pressure. At 20 kbar, no anomaly is found both in the thermal expansion and in the thermal expansion coefficient. This implies that T_C is lower than 4.2 K at 20 kbar.

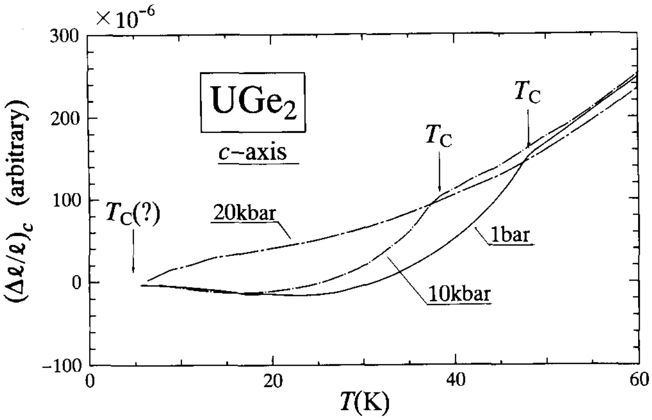

Fig. 2. Linear thermal expansion $(\Delta l/l)_c$ along the c axis as a function of temperature at various pressures. The Curie temperature T_C is shown by an arrow.

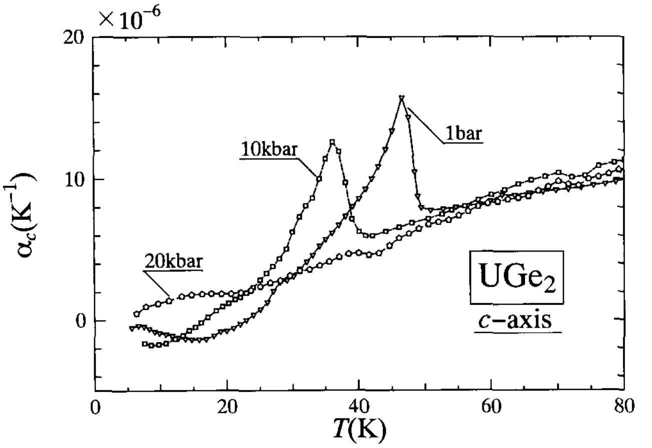

Fig. 3. Temperature dependence of linear thermal expansion coefficient α_c under high pressures.

To examine the temperature dependence of volume change, we calculate the fractional change $\omega \equiv \Delta V/V$ in volume as a function of temperature. ω is obtained from the following relation:

$$\omega \equiv \Delta V/V = (\Delta l/l)_a + (\Delta l/l)_b + (\Delta l/l)_c \quad (1)$$

The above relation is easily derived by using $V = l_a l_b l_c$. ω is shown in Fig. 4 as a function of temperature at various pressures. Although ω decreases with decreasing temperature, it begins to increase below T_C. The volume expansion is due to the spontaneous magnetization below T_C. To obtain the spontaneous volume magnetostriction $\omega_s(T)$, we estimate the volume in the paramagnetic state by fitting the data of $\omega(T > T_C)$ to the equation

$$\omega_{para}(T) = aT^2 + bT^4 \quad (2)$$

$\omega_s(T)$ is obtained by using the relation $\omega_s(T) = \omega(T) - \omega_{para}(T)$. T_C is defined as the temperature at which $\omega(T)$ deviates from $\omega_{para}(T)$.

We show in Fig. 5 the temperature dependence of ω_s under high pressure. The value of ω_s at ambient pressure is 1.23×10^{-3} at 0 K, which is smaller than that of Fe–Ni, or Fe–Pt Invar alloys (see for example ref. 6) and YMn_2 [7] by an order of magnitude. It is found that ω_s decreases on application of pressure and disappears above 20 kbar. Such behaviour was observed also in Invar alloys [8] and YMn_2 [9].

Figure 6 shows the pressure dependence of T_C. In the pressure range below 15 kbar, T_C decreases gradually with increasing pressure with the initial decreasing rate $\partial T_C/\partial P = -0.7\ K\ kbar^{-1}$. Above 15 kbar, T_C decreases rapidly with pressure. No discontinuous change in volume near T_C is observed at 20 kbar within experimental

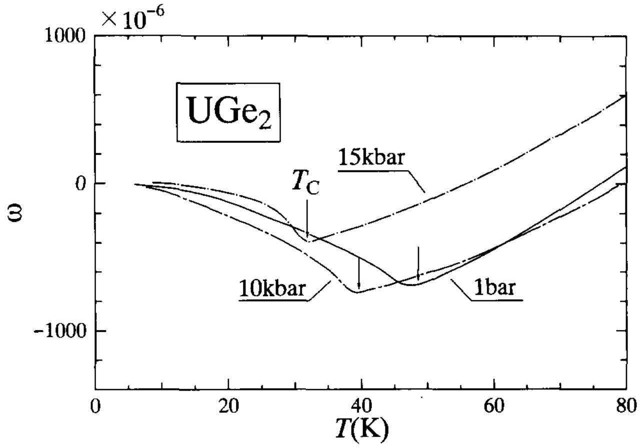

Fig. 4. Fractional change $\omega \equiv \Delta V/V$ in the volume as a function of temperature under high pressure. The Curie temperature T_C is shown by an arrow.

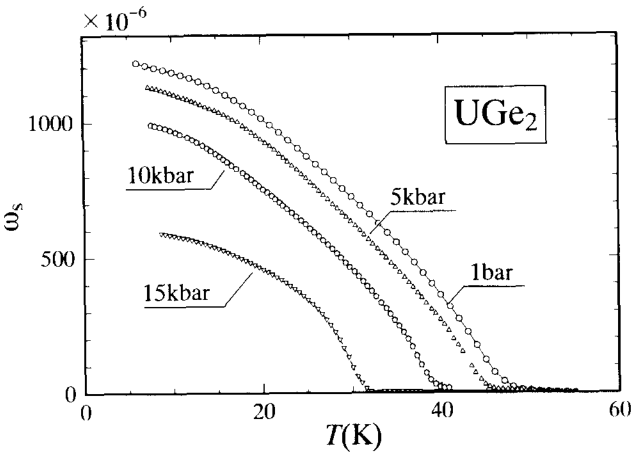

Fig. 5. Temperature dependence of spontaneous magnetostriction ω_s under high pressure.

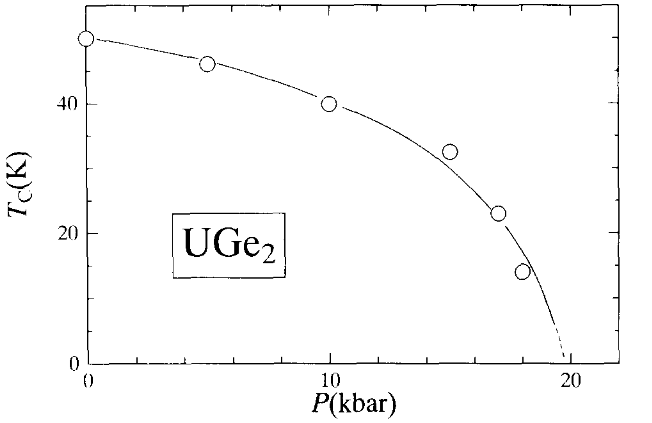

Fig. 6. Pressure dependence of Curie temperature T_C: ——, guide for the eye.

error. It is well known that the following expression is applicable for the pressure dependence of T_C of a weak ferromagnet [10]:

$$\left[\frac{T_C(P)}{T_C(0)}\right]^2 = 1 - \frac{P}{P_c} \qquad (3)$$

where $T_C(0)$ is T_C at ambient pressure and P_c is the critical pressure above which the system becomes paramagnetic.

Figure 7 shows the result of least-squares fitting of the observed data to eqn. (3). By taking $T_C(0) = 51.6$ K and $P_c = 21.7$ kbar, the present data are fitted well. Thus the pressure dependence of T_C is interpreted by the weak ferromagnetic model.

Finally we comment on the pressure dependence of T_C by using the Ehrenfest relation $dT_C/dP = TV\, \Delta\alpha/\Delta C$, where ΔC is the discontinuity in the specific heat at T_C. By substituting the observed value of $\Delta\alpha = 40 \times 10^{-6}$ K^{-1} and $\Delta C = -6$ J mol^{-1} K^{-1} [3], we

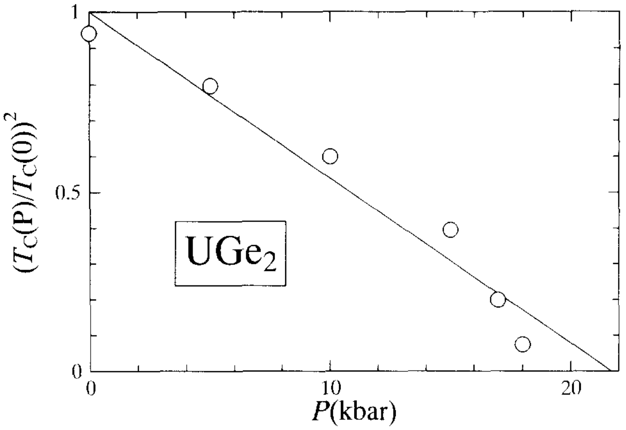

Fig. 7. Least-squares fitting of the present data to eqn. (3).

obtain $dT_C/dP = -1.2$ K $kbar^{-1}$. The estimated value of dT_C/dP is comparable with the observed value of -0.7 K $kbar^{-1}$.

4. Conclusion

We measured the thermal expansion of single-crystal UGe_2 under high pressure. The main results are summarized as follows.

(1) The thermal expansion coefficients α_i ($i = a$, b and c) exhibit discontinuities at T_C owing to ferromagnetic ordering.

(2) A clear magnetovolume effect is observed with the magnitude of spontaneous volume magnetostriction $\omega_s(0)$ at 0 K being 1.23×10^{-3}.

(3) The spontaneous volume magnetostriction ω_s decreases on applying pressure and disappears above 20 kbar.

(4) T_C decreases with increasing pressure with initial slope $\partial T_C/\partial P = -0.7$ K $kbar^{-1}$.

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