

# HIGH PRESSURE STUDY OF THE 37K PHASE TRANSITION OF $\alpha$ -U SINGLE CRYSTAL BY AN AC CALORIMETRIC TECHNIQUE<sup>\*</sup>

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The 37K phase transition of  $\alpha$ -U single crystal was observed by a calorimetric technique to be suppressed by the application of hydrostatic compression. The rate of decrease of the transition temperature  $T_2$  with pressure  $P$  is  $dT_2/dP = -(3.6 \pm 0.2)\text{K/kbar}$ . Our results qualitatively support the phase diagram of  $\alpha$ -U single crystal proposed by Smith.

In an unstrained  $\alpha$ -U single crystal, there are three distinct phase transitions [1] with transition temperatures  $T_1$ ,  $T_2$  and  $T_3$  at 42K, 37K and 23K, respectively. The transitions are accompanied by anomalies in thermal expansion [1], elastic moduli [2], magnetic anisotropy [3] and specific heat [4]. From thermal expansion measurements, Steinitz et al. [1] concluded that the two transitions at 37K and 23K are first order but the 42K one is second order. Fisher and Dever [5], by measuring the elastic moduli, found  $dT_2/dP = -3.4\text{K/kbar}$  for the 42K transition. Later Smith [6] examined the hydrostatic pressure dependence of the superconducting transition temperature  $T_c$  of an  $\alpha$ -U single crystal and observed considerable structure in the  $T_c$  versus  $P$  curve. As shown in fig. 1,  $T_c$  rises rapidly with  $P$  but becomes  $P$ -independent between 6 and 8 kbar. For  $P > 8$  kbar,  $T_c$  increases again, reaches a maximum at 11.5 kbar and decreases with higher  $P$ . Based on these results and those of others [1, 5], Smith [6] proposed a phase diagram for single crystal  $\alpha$ -U shown in fig. 1. The three modifications of the low temperature phase are labelled  $\alpha_1$ ,  $\alpha_2$  and  $\alpha_3$ . The structure in the  $T_c$  versus  $P$  curve was thus attributed to the three phase transitions. The purpose of this experiment is to investigate the proposed phase

boundary between  $\alpha_1$  and  $\alpha_2$  modifications, i.e., to determine the  $P$ -effect on the 37K phase transition temperature  $T_2$ .

Crangle and Temperol [4] recently observed relatively large effect of the 37K phase transition on the specific heat of an  $\alpha$ -U single crystal at atmospheric pressure. Hence we decided to integrate the ac calorimetric method [7] into our high pressure clamp tech-

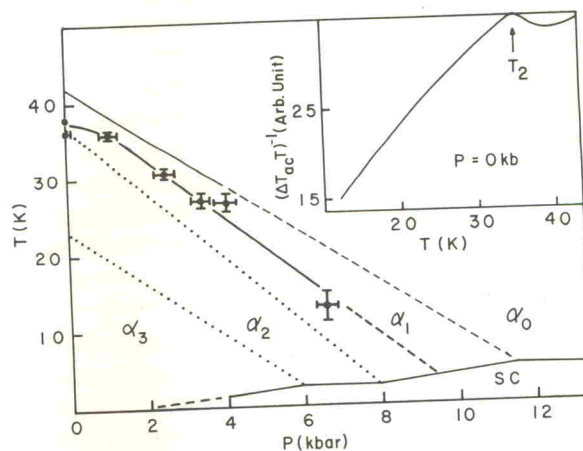


Fig. 1. Comparison of our results of the  $P$ -dependence of the 37K phase transition with the proposed phase diagram for  $\alpha$ -U single crystal by Smith [6]. The insert shows the  $T$ -dependence of the ratio of the relative specific heat to  $T$  at  $P=1$  bar. --- extrapolation; — denotes present work; - - - work by Fisher and Dever and .... the proposed phase boundaries by Smith. SC stands for the superconducting state.

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nique [8] for the detection of this phase transition under hydrostatic compression. The main idea is to confine the system as close as possible to the small sample leaving the high pressure surroundings almost thermally unperturbed. This can be achieved by properly adjusting different thermal links. The ac temperature response  $\Delta T_{ac}$  of a sample to an ac heat input is then approximately inversely proportional to its specific heat. The temperature dependence of the relative specific heat of a sample can thus be recorded continuously at different pressures.

Employing the method described above, we have investigated the pressure dependence of  $T_2$  up to 12 kbar. A sample of 1.6 mm dia.  $\times$  6.0 mm length was spark cut from an  $\alpha$ -U single crystal.  $\Delta T_{ac}$  was measured by a pair of Au+0.07% Fe-CHR thermal couples [9]. The pressure  $P$  was determined by a lead manometer [10] and the ambient temperature  $T$  by a pair of Au+0.07% Fe-CHR thermal couples and/or a Ge-thermometer depending on the  $T$ -range. Without the pressure medium inside the pressure cell the  $(\Delta T_{ac}T)^{-1}$  versus  $T$  curve clearly exhibited three peaks at 25.7 K, 36.8 K and 41.2 K corresponding to the three phase transitions observed by Steinitz et al. [1]. However we could detect only the 37 K transition with pressure medium present as depicted in the insert of fig. 1. This is consistent with the specific heat data at  $P=1$  bar showing that the 37 K transition gives rise to the largest anomaly. The temperature where the peak occurs was taken as the transition temperature  $T_2$ . We found that  $T_2$  is suppressed by the application of hydrostatic compression and the size of the anomaly decreases with  $P$ . No anomaly could be seen beyond 7 kbar. It should be noted that the decrease in the anomaly size could be due to one or more of the following: (a) a real effect, (b) the decrease in sensitivity of the ac temperature sensor under high pressure and at lower temperature, and (c) the increase in thermal coupling between the sample and its surroundings at high pressure and low temperature. The zero pressure  $T_2$  was found to be slightly lower ( $\sim 1$  K) after the removal of  $P=12$  kbar. In view of the highly sensitive effect of strain on the phase transitions of  $\alpha$ -U single crystal [5], this irreversible decrease in  $T_2$  may be caused by the possibly imperfect hydrostatic pressure environment. Our results were compared with the proposed phase boundary and given in fig. 1. The uncertainty in  $T_2$  at high  $P$  was mainly attributed to the

ill-defined anomaly peak due to its diminishing apparent size at higher  $P$ .  $T_2$  was found to decrease slowly and nonlinearly with  $P < 2$  kbar but almost linearly with  $P$  up to 7 kbar at a rate  $dT_2/dP = -(3.6 \pm 0.2)$  K/kbar. The phase boundary so obtained was represented by the heavy solid line in fig. 1. It lies slightly above the proposed boundary for  $P > 2$  kbar. Since  $dT_2/dP = -(3.6 \pm 0.2)$  K/kbar and the fractional volume change [1] due to the transition is  $-1.5 \times 10^{-4}$ , one can use the Clausius-Clapeyron relation and obtain a latent heat of  $(2.0 \pm 0.1)$  J/mole which is in good agreement with the value of  $(2.08 \pm 0.5)$  J/mole from atmospheric pressure specific heat measurements by Crangle and Temperol [4]. The linearly extrapolated  $T_2$  at  $P > 7$  kbar crosses the superconducting phase boundary in fig. 1 between 8 and 10 kbar where  $T_c$  increases again with  $P$ .

In conclusion we have successfully detected the 37 K phase transition in  $\alpha$ -U single crystal under hydrostatic compression by an ac calorimetric technique. It was found that  $T_2$  decreases with pressure and the extrapolated  $T_2$  crossed the superconducting phase boundary at a pressure where  $T_c$  behaves anomalously. This observation supports the suggestion by Smith that the structure in the  $T_c$  versus  $P$  curve at  $\sim 8$  kbar might be related to the 37 K transition of  $\alpha$ -U single crystal at high pressure.

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