HIGH PRESSURE STUDY OF THE 37 K PHASE TRANSITION OF α-U SINGLE CRYSTAL BY AN AC CALORIMETRIC TECHNIQUE☆

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The 37 K phase transition of α-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 $dT_2/dP = -(3.6 \pm 0.2)$ K/kbar. Our results qualitatively support the phase diagram of α-U single crystal proposed by Smith.

In an unstrained α-U single crystal, there are three distinct phase transitions [1] with transition temperatures T_1, T_2 and T_3 at 42 K, 37 K and 23 K, 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 37 K and 23 K are first order but the 42 K one is second order. Fisher and Dever [5], by measuring the elastic moduli, found $dT_2/dP = -3.4$ K/kbar for the 42 K transition. Later Smith [6] examined the hydrostatic pressure dependence of the superconducting transition temperature T_c of an α-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 α-U shown in fig. 1. The three modifications of the low temperature phase are labelled α_1, α_2 and α_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 α_1 and α_2 modifications, i.e., to determine the P-effect on the 37 K phase transition temperature T_2.

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

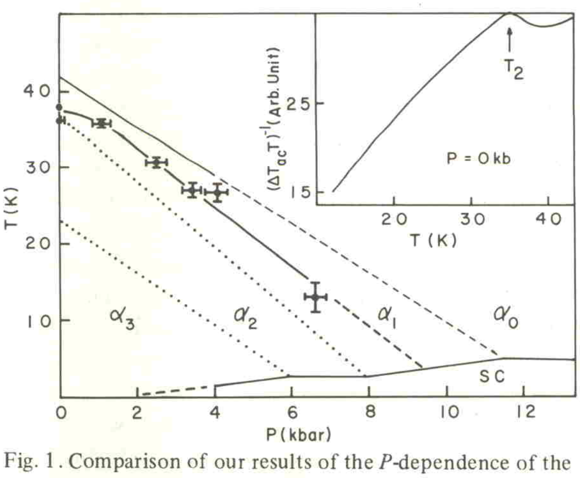

Fig. 1. Comparison of our results of the P-dependence of the 37 K phase transition with the proposed phase diagram for α-U single crystal by Smith [6]. The insert shows the T-dependence of the ratio of the relative specific heat to T at P=1 bar. – – – – extrapolation; ——— denotes present work; –.–.–. 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 proposerly adjusting different thermal links. The ac temperature response ΔT_{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. × 6.0 mm length was spark cut from an α-U single crystal. ΔT_{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 25.7K, 36.8K and 41.2K corresponding to the three phase transitions observed by Steinitz et al. [1]. However we could detect only the 37K 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 37K 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 supressed by the application of hydrostatic compression and the size of the anomaly decreases with P. No anomaly could be seen byond 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 (~ 1 K) after the removal of P=12 kbar. In view of the highly sensitive effect of strain on the phase transitions of α-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 -1.5×10^{-4}, one can use the Clausius-Clapeyron relation and obtain a latent heat of (2.0 ± 0.1) J/mole which is in good agreement with the value of (2.08 ± 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 37K phase transition in α-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 ~ 8 kbar might be related to the 37K transition of α-U single crystal at high pressure.

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References

[1] M.O. Steinitz, C.E. Burleson and J.A. Marcus, J. Appl. Phys. 41 (1971) 5057.
[2] E.S. Fisher and H.J. McSkimin, Phys. Rev. 124 (1961) 67.
[3] J.W. Ross and D.J. Lam, Phys. Rev. 165 (1968) 617.
[4] J. Crangle and J. Temperol, to be published.
[5] E.S. Fisher and D. Dever, Phys. Rev. 170 (1968) 607.
[6] T.F. Smith, Superconductivity in d- and f-band metals, ed. D.H. Douglass (1971) p. 310, and references therein.