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# Superconductivity of α-Uranium and Uranium Compounds at High Pressure

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The pressure dependence of the superconducting transition temperature $T_c$ of $\alpha$-uranium has been measured to a maximum pressure of 22 kbar and constitutes an important extension of the pressure range of previous measurements. It has been observed that $T_c$ increases rapidly as a function of pressure up to ~9 kbar, passes through a broad maximum, and then decreases. A possible explanation of this behavior is offered on the basis of a pressure-induced transformation in the electronic properties of uranium. Data are also reported for observations of the $T_c$ under pressure of the compounds $U_6Fe$ and $U_6Mn$ and the solid solution alloy $U_{0.85}Mo_{0.15}$.

## INTRODUCTION

MEASUREMENTS of the pressure dependence of the superconducting transition temperature of $\alpha$-uranium, reported earlier,[1] identified uranium with the small group (Tl,[2,3] La,[4] Ti,[5] Zr,[6] V,[7] and U [1]) of superconducting elements for which $T_c$ increases with a decrease of volume. Since the report of these measurements, the superconductivity of $\alpha$-uranium has been the subject of extensive investigation and speculation.[8–13] Possibly the most important fact to emerge from this work is that all previously reported[1,14] superconducting transition temperatures at zero pressure, ranging from 0.5 to above 1°K, which were magnetically or resistively determined were not associated with bulk superconductivity, but instead resulted from a connected network of superconducting filaments. The first evidence for this conclusion can be found in the specific-heat data of Dempesy, Gordon, and Romer[15] for $U^{238}$ down to 0.15°K, which failed to show the characteristic anomaly associated with the transition to the superconducting state. Unfortunately, they made no attempt to detect a magnetic transition in this sample and so very reasonably concluded that the lack of superconductivity above 0.15°K was associated specifically with their particular sample rather than a property of $\alpha$-uranium. However, recent,[10,11] more extensive specific-heat measurements on samples exhibiting magnetic transitions have also failed to show any evidence of bulk superconductivity, not only at the magnetic transition, but from the measurements of Phillips and Ho[11] even down to 0.1°K. Thus on the basis of these latter measurements, even the most recently reported[9] transition between 0.21 and 0.25°K

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for a good single crystal of $\alpha$-uranium is not associated with bulk superconductivity.

Two suggestions have been made[8,9] concerning the nature of the filaments responsible for the observed magnetic transitions of $\alpha$-U. Firstly, from work[8] on stabilized $\beta$-U (formed by the addition of 2 at.% of either Pt, Rh, or Cr) and from a consideration of earlier work[16,17] on stabilized $\gamma$ U-Mo alloys, Matthias *et al.* postulated that the filaments consisted of impurity-stabilized networks of $\beta$ and $\gamma$ phases. However, this suggestion has been criticized by Howlett[13] on the basis that there is no metallurgical evidence for the presence of such stabilized phases in high purity $\alpha$-U. In our opinion, the pressure dependence of the transition temperatures of the uranium compounds and the stabilized $\beta$ and $\gamma$ phases, which we have measured and report here, is inconsistent with the possibility of filaments of these alloys being responsible for the observed pressure dependence of $T_c$ for $\alpha$-U. Secondly,[9] the observed transitions were associated with filaments from two distinct origins, depending on the temperature range of the observed transition. Thus the first postulate of stabilized $\beta$- or $\gamma$-phase filaments was retained for transitions above 0.8°K, whilst transitions below 0.8°K were considered to arise from strain filaments produced by the highly anisotropic thermal expansion[18] of uranium at low temperatures. The presence of a network type of structure in uranium has been observed[13,19] in recent transmission electron microscopy investigations. However, it has not been possible to identify the nature of the material comprising the network and hence distinguish impurity-stabilized phases from regions of strain or dislocation.

Whilst the nature of the observed magnetic transitions at zero pressure suggests, and the zero-pressure specific-heat data confirms, filamentary rather than bulk superconductivity, it is difficult to believe that filamentary superconductivity, due to strain, is maintained up to 10 kbar since, by this pressure, the strain within the grains must be larger than that initially present at the grain boundaries. Recent specific-heat[12] measurements made on uranium at 10 kbar have, in fact, demonstrated that the superconducting transitions measured at high pressure are representative of bulk superconducting properties.

In view of the importance of pressure measurements in helping to understand the properties of uranium there was considerable incentive to extend the pressure range of the $T_c$ measurements beyond the previous[1] limit of 10 kbar. Thus $T_c$ has now been studied, as a function of pressure, to a maximum pressure of 22 kbar revealing a dramatic change in its pressure dependence.

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## EXPERIMENTAL METHOD

The two samples of $\alpha$-U, U9 and U10, used in the present investigation, were cut from separate lots of high purity Los Alamos uranium, kindly made available by Dr. C. E. Olsen. Both of these samples were taken from material used in the extensive heat-capacity measurements of Phillips and Ho,[11] the sample U10 coming from the same lot as the sample for which specific-heat measurements[12] at 10 kbar were made. The alloy samples were prepared by melting the required amounts of the constituents in an argon arc furnace.

The transition temperatures of the alloy samples were determined in the same high pressure apparatus as was used for the original measurements[1] on $\alpha$-uranium. A redesigned piston and cylinder assembly was used to extend the pressure range of the previous measurements on $\alpha$-uranium. The cylinder was fabricated from a hardened Be–Cu (Berylco 25) alloy and the $\frac{1}{4}$-in.-diam piston was unsupported tungsten carbide, tipped with high-density alumina. The alumina tip was supported by the cylinder walls and served to transmit the force from the carbide pistons to the sample assembly. Such an arrangement was necessary in order to reduce pickup in the detection coils from the superconductivity and weak ferromagnetism of the tungsten carbide. A small piece of tin was included in the sample assembly to serve as a direct low-temperature manometer. The pressure was calculated from the superconducting transition temperature of the tin using the absolute pressure-dependence data of Jennings and Swenson.[2] As these data only extend to 10 kbar, we were obliged to extrapolate their empirical relationship, which can be written ($P$ expressed in kbar) as,

$$\Delta T_c = -4.89\times10^{-2}P + 3.8\times10^{-4}P^2$$

in order to determine the higher pressures involved in the present study of $\alpha$-U. Powdered Teflon was used as the quasihydrostatic pressure-transmitting medium.

## RESULTS AND DISCUSSION

### (i) Compounds and Alloys

From the limited number of superconducting compounds of uranium we chose $U_6Fe$ and $U_6Mn$[16] in order to study and compare the effect of pressure upon the superconducting transition temperature of uranium compounds. No significant change in $T_c$ could be observed up to 10 kbar for either compound, though it is possible that a small effect was masked by the width of the transitions. The results are presented graphically in Fig. 1, where $T_c$ is plotted as a function of applied pressure. The solid vertical line indicates the width of a transition, which was determined from an extrapolation of the central linear portion of the transition curve.