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# EFFECT OF HYDROSTATIC PRESSURE ON THE LOW TEMPERATURE PHASE TRANSITION IN ALPHA-URANIUM*

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Measurements of single crystal elastic moduli of uranium at low temperatures and hydrostatic helium pressures to 4 kbar show that $dT_\alpha/dP = -3.4$°K/kbar, where $T_\alpha$ is the temperature of the phase transition that occurs at 42 ± 1°K at 1 bar. Extrapolation of the data indicates that $T_{c,\max}$, for superconductivity in uranium corresponds to $T_\alpha$ at 11.5 kbar. This result supports the concept that $dT_c/dP > 0$ in uranium is a result of the phase transition.

IN 1961 WE reported on the occurrence of a phase transition in uranium, at 43°K, that produced very large and clear anomalies in the temperature dependence of the elastic moduli in single crystals.[1] Subsequent studies of several physical properties[2-4] indicate that the transition from the phase above $T_\alpha \simeq 43$°K to the state at 4°K occurs in several distinct steps and gives rise to a domain-like structure[5] that is responsible for an increase in neutron intensities as diffracted by the crystal planes.

Correlation of this information with measurements of the transition to superconducting state in uranium suggest that the domain structure contains metastable filaments of the $\alpha$ phase which is the equilibrium phase about $T_\alpha$.[6,7] These filaments are presumed to account for the superconductivity detected by magnetization measurements at 1 bar pressures[6,7] whereas the bulk or the stable matrix is nonsuperconducting at $T > 0.1$°K, as indicated by heat capacity measurements.[8] The latter also show that uranium at 10 kbar is a bulk superconductor[8] with $T_c \sim 2$°K whereas recent[9] single crystal magnetization studies show that $dT_c/dP = 0.18$°K/kbar between 1 bar and 8 kbar, with $T_c$ at 1 bar of 0.20°K. Studies of polycrystalline samples showed $T_c$ increasing with pressure to 2.2°K near 10 kbar and then decreasing slightly from 12 kbar to 20 kbar.[7]

It has been proposed that $dT_c/dP > 0$ arises from a direct relationship between the anomalous negative volume thermal expansion at $T < T_\alpha$ and the number of localized 5*f* electrons, which act to suppress superconductivity.[6] The application of pressure at $T < T_\alpha$ increases $T_c$ by gradually decreasing the number of localized 5*f* electrons in either the bulk phase or in the filaments and the depopulation of the localized states is complete at 10 kbar, thus accounting for the specific heat anomaly associated with $T_c$ and the maximum in measured $T_c$. The implied effect of this model is that the major difference between the phases above and below $T_\alpha$ is gradually removed by pressure application and that $T_\alpha \to 0$°K near 10 kbar. This is not, however, consistent with a direct relation between volume and valence, since the compressibility at 4°K suggests that the excess volume due to anomalous thermal expansion is removed by about 4 kbar, or $dT_\alpha/dP \simeq 10$°K/kbar.

The model based on the electron–phonon interaction[10] accounts for $dT_c/dP$ on the basis of a

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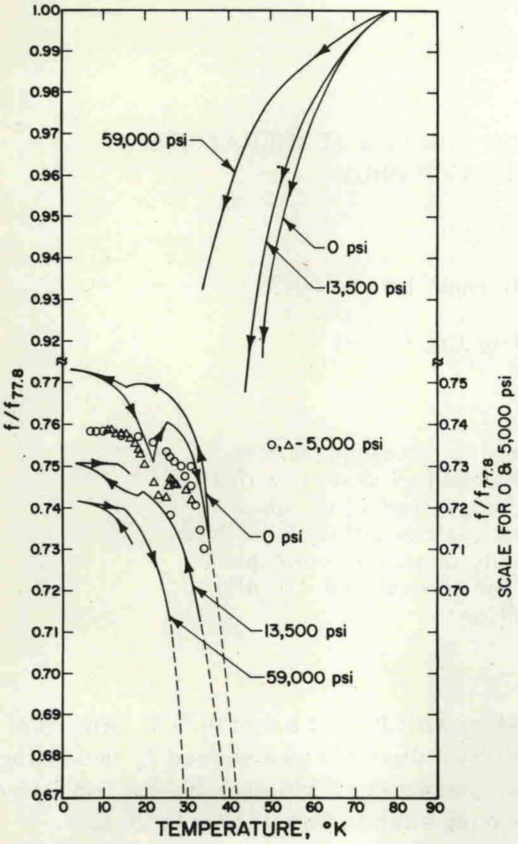

FIG. 1. Effects of pressure on the basic data for $C_{11}$ vs. temperature. ($C_{11} = \rho V_{[100]}$, $\rho$= density).

shift in energy of an *f* electron band relative to the Fermi level and an increasing hybridization and density of electronic states at the Fermi level. This model makes no predictions regarding $\mathrm{d}T_\alpha/\mathrm{d}P$, since there is no assumed distinction between the superconducting properties above or below $T_\alpha$, i.e., the filaments are not necessarily associated with the $\alpha$ phase.

The purpose of this paper is to report on the measurements of $\mathrm{d}T_\alpha/\mathrm{d}P$ from elastic modulus data. The experiments were made with the same pulsed ultrasonic technique and single crystals described in reference 2. The velocity change with temperature of longitudinal waves in the three principal directions of this orthorhombic crystal were measured under several different constant hydrostatic gas and solid helium pressures. The pressure bomb was of CuBe construction manufactured by Harwood Eng., Walpole. Mass.

The data are shown in Figs. 1, 2 and 3 for the [100], [010], and [001] directions, respectively. The ordinates for each figure are

$$\frac{f}{f_{77.8}} = \frac{V}{V_{77.8}} \cdot \frac{t_{77.8}}{t}$$

where *f* is a critical wave frequency for an

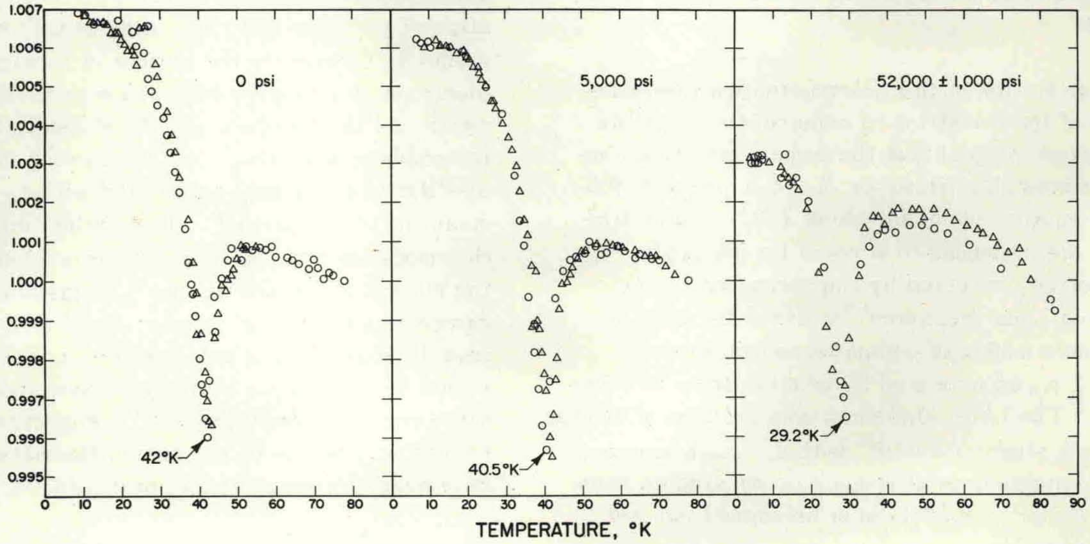

FIG. 2. Pressure effect on $T_\alpha$ as determined from basis data for $C_{22}$ vs. temperature. ($C_{22} = \rho V^2_{[010]}$).

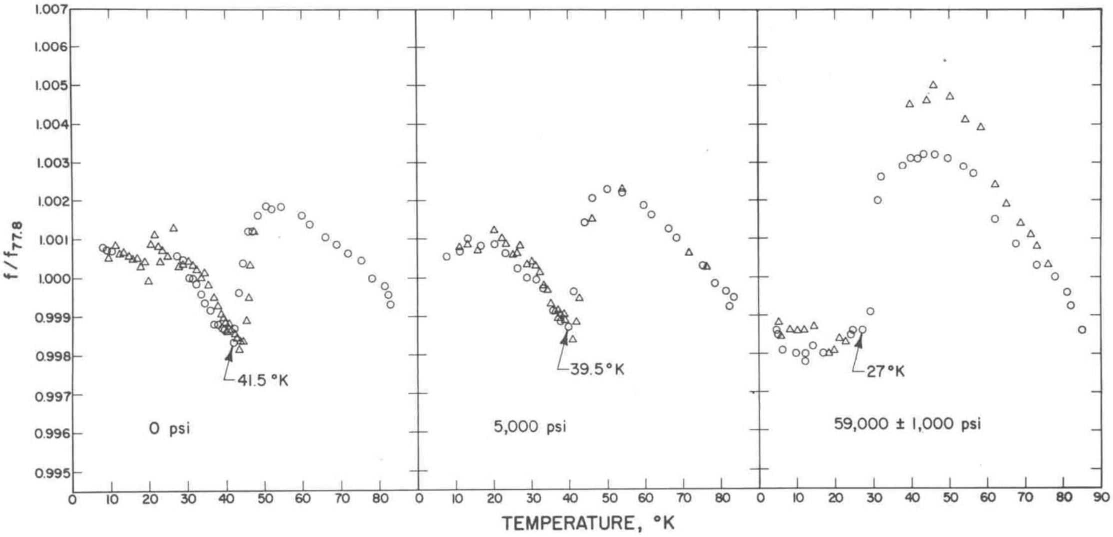

FIG. 3. Pressure effect on $T_\alpha$ as determined from basic $C_{33}$ vs. temperature data. ($C_{33}= \rho V^2_{[001]}$

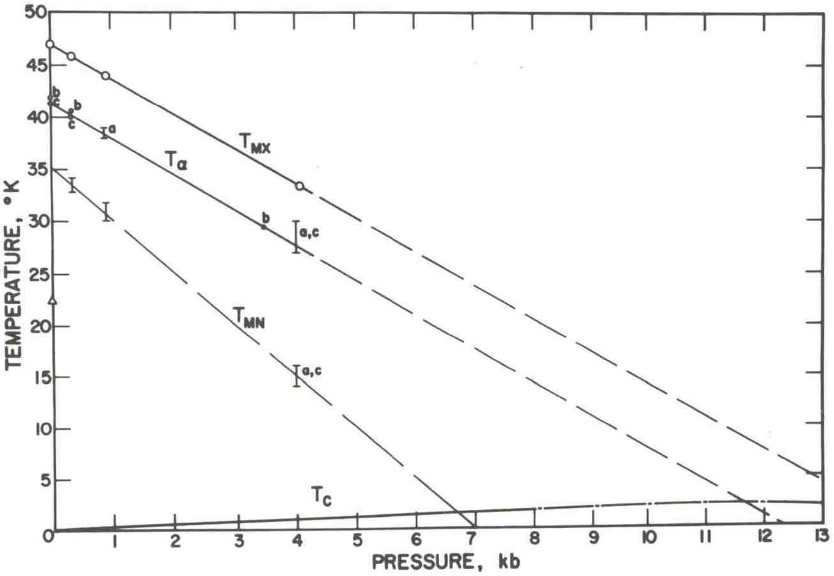

FIG. 4. Variations with hydrostatic pressure of $T_\alpha$ and the temperatures delineating the high acoustic attenuation for $C_{11}$ mode in uranium. The $T_c$ vs. pressure curve obtained from Reference 9 to 8 kbar and reference 7 at $p > 8$ kbar.

integral number of specimen waves, $V$ = wave velocity, $t$ = crystal thickness and the subscripted terms represent the basis data at 77.8°K. $T_\alpha$ is defined as the temperature at which there occurs an abrupt change in slope of the $f/f_{77.8}$ vs. $T$ curves. For the [100] data, Fig. 1, $T_\alpha$ cannot be directly observed because of the very large acoustic attenuation that is associated with the softness of this mode near the phase transition. The decrease with pressure in $T_\alpha$ and the temperature of high attentuation is, however, clearly indicated, as is the reduction of the hysteresis effects[2] at $T < T_\alpha$. The data for the [010] direction, Fig. 2, clearly show $T_\alpha$ at two different hydrostatic pressures as well as at 1 bar or 0 psi. The [001] data, Fig. 3, show that unambiguous shift in $T_\alpha$ between 1 bar and 0.35 kbar (5,000 psi). At 4.07 kbar, however, there is a clear change in the character of the data for [001] so that $T_\alpha$ is less well defined.

The data for $dT_\alpha/dP$ are summarized in Fig. 4, where the indicated $T_\alpha$ marked a, b and c refer to [100], [010] and [001], respectively. The points (a) were obtained by interpolation of the [100] data. The decrease in $T_\alpha$ with increasing pressure is very near linear, with $dT_\alpha/dP = -3.4°K/\text{kbar}$. The lines marked $T_{MX}$ and $T_{MN}$ delineate the temperature range at a given pressure over which the longitudinal [100] mode signal is lost to attenuation and the pressure range at a given $T$ that this mode velocity decreases with increasing pressure. Finally, the reported superconducting $T_c$ vs. pressure data are reproduced from reference 9 and 7. We arrive at the conclusion that the maximum $T_c$ occurs at or near the pressure at which $T_c = T_\alpha$. This is a rather different conclusion than is reached from a direct volume to valence relation. It is consistent, however, with the concept that $T_{c,\max}$ occurs when the electronic effects produced by cooling from $T_\alpha$ are completely reversed by ~ 10 kbar pressure or, similarly, ~ 10 kbar pressure is sufficient to retain the bulk superconducting $\alpha$ phase during cooling. It is not, however, necessarily inconsistent with an electron–phonon coupling model. The indications are that $T_{c,\max}$ occurs where $\omega_{[100]}$, the frequency for [100] longitudinal phonons, reaches a minimum value under hydrostatic pressure. In subsequent experiments we will attempt to measure the pressure dependence of the transverse mode velocities at 4°K and thereby estimate the changes in the whole phonon spectrum with pressure at superconducting temperatures.

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Messungen der elastischen Einkristallkonstanten von Uran bei tiefen Temperaturen und Helium-Drücken bis zu 4 kbar ergeben $dT_\alpha/dP = -3.4°K/kbar$, wobei $T_\alpha$ die Temperatur de Phasenumwandlung ist, die unter Normaldruck bei 42 ± 1°K auftritt. Eine Extrapolation/dieser Messungen zeigt, dass $T_{c,\max}$ die maximale kritische Temperatur für Supraleitung in Uran, mit $T_\alpha$ bei 11.5 kbar übereinstimmt. Dieses Resultat unterstutzt die Vorstellung, dass $dT_c/dP > 0$ in Uran eine Folge der Phasenumwandlung ist.