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Elastic properties of UO_2 at high pressure*

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The elastic constants of UO_2 have been measured as a function of pressure up to 20 kbar (2.0 GPa) at 23°C. To within experimental uncertainty, the constants are linear functions of pressure, and no pressure-induced phase transitions occur in this 20-kbar pressure range. The elastic constant data are used to estimate the various polycrystalline elastic moduli and their pressure dependences, and the zero-pressure moduli are compared with data from polycrystalline material. Isothermal moduli are calculated from the adiabatic values. The elastic-constant data are also used to discuss the separation of the 'pure' volume and temperature contributions to the (isobaric) temperature dependences of the elastic constants as well as to estimate the Grüneisen constant, Debye temperature, and equation of state of this material.

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I. INTRODUCTION

Uranium dioxide, UO_2, is a material of considerable technological importance because of its use in reactor fuel cells. Although the physical properties of UO_2 in both single-crystalline and polycrystalline forms have been quite extensively studied, especially at high temperature, there is little information available about the behavior of this material at high pressure. In particular, it has not been known whether UO_2 has any pressure-induced phase transitions, similar to those observed in other materials with the same (cubic fluorite) structure. In order to investigate the elastic properties of UO_2 at high pressure and to look for pressure-induced phase transitions we have made an ultrasonic determination of the pressure dependences of the elastic constants up to 20 kbar at room temperature. No phase transitions were found in this range. In Sec. II of this paper the experimental details of this work are presented. In Sec. III the elastic-constant data are presented and analyzed. In Sec. IV the results of several calculations using the elastic-constant data are discussed. These include the prediction of the elastic properties of polycrystals, the separation of "pure" temperature and volume effects, the transformation from adiabatic to isothermal properties, and estimates of the Grüneisen constant, Debye temperature, and of the equation of state.

II. EXPERIMENTAL

Since UO_2 has cubic symmetry, its elastic properties are determined by the three elastic constants C_{11}, C_{12}, and C_{44}. These three constants may be determined from the velocities of the three acoustic modes propagating along the [110] crystallographic direction. These velocities are given by the relations

$$\rho v_1^2 = C_{44}, \tag{1a}$$

$$\rho v_2^2 = \tfrac{1}{2}(C_{11} - C_{12}), \tag{1b}$$

$$\rho v_3^2 = \tfrac{1}{2}(C_{11} + C_{12} + 2C_{44}), \tag{1c}$$

where ρ is the density, v_i are the three velocities, and where $i=1$ represents the shear mode polarized along [001], $i=2$ represents the shear mode polarized along [1$\bar{1}$0], and $i=3$ represents the longitudinal mode. The UO_2 sample utilized in the ultrasonic experiments was prepared with pairs of [110] and [111] faces. The mode velocities along [111] are given by

$$\rho v_4^2 = \tfrac{1}{3}(C_{11} - C_{12} + C_{44}) \quad \text{(shear)}, \tag{2a}$$

$$\rho v_5^2 = \tfrac{1}{3}(C_{11} + 2C_{12} + 4C_{44}) \quad \text{(longitudinal)}. \tag{2b}$$

Measurements of these modes provided cross checks of the data obtained from the [110] orientation.

The UO_2 crystals used in this work were supplied by H. J. Anderson of the Westinghouse Hanford Company. Crystal growth, impurity analysis, and stochiometry are exactly the same as described by Wachtman *et al.*[1] The density was determined by weighing in CCl_4 to be equal to the theoretical x-ray value of 10.97 g/cm^3. For the sample used in the ultrasonic measurements, the [110] and [111] faces were oriented to within 0.5° by x-ray techniques, and each pair of faces was carefully lapped to be flat and parallel.

All the ultrasonic measurements were made by the pulse-superposition technique of McSkimin.[2] The instrumentation system included a pulse-sequence generator which utilizes logic circuitry to generate a number of sequential applied pulses to the sample at the repetition rate frequency f, but with a low over-all duty cycle.[3] Also included was a phase-sensitive peak detector with a servo feedback mechanism to automatically follow the superpositioning repetition rate as a function of pressure.[4]

For the measurements of the elastic constants at room temperature, transducers with fundamental frequencies of 10 and 20 MHz operating at either their fundamental or third harmonic frequencies were utilized. The use of these four frequencies plus the technique of McSkimin[5] allowed an unambiguous determination of the correct cycle-for-cycle superpositioning of the ultrasonic echoes. The high-pressure runs were made at 20 MHz, with some of the data checked at 60 MHz. In all cases the transducer-sample bond was phthalic anhydride-glycerin polymer. Hydrostatic pressure was generated by a standard Bridgman press with a 50-50 mixture of pentane and isopentane as the pressure fluid, and pressure was measured to an accuracy of 1% with a calibrated manganin coil.

TABLE I. Single-crystal adiabatic elastic parameters and their pressure and temperature derivatives.

Elastic parameter Q	Value of Q [a] This work	Wachtman *et al.* [b]	$\frac{dQ}{dp}$ [c]	$\frac{dQ}{dT}$ [d,e]
C_{11}	3.893 ± 0.017	3.96 ± 0.018	5.35 ± 0.24	-5.2
C_{12}	1.187 ± 0.017	1.21 ± 0.019	4.35 ± 0.24	-2.3
C_{44}	0.597 ± 0.003	0.641 ± 0.0017	1.62 ± 0.03	-1.1
S_{11}	3.00 ± 0.02	2.96 ± 0.010	-0.20 ± 0.05	...
S_{12}	-0.700 ± 0.012	-0.697 ± 0.008	-0.07 ± 0.01	...
S_{44}	16.75 ± 0.08	15.59 ± 0.041	-4.3 ± 0.1	...
κ_V	4.79 ± 0.03	4.70 ± 0.04	-1.03 ± 0.03	...

[a] C_{ij} in 10^{12} dyn/cm^2, S_{ij} and κ_V in 10^{-13} cm^2/dyn.

[b] Reference 1.

[c] dS_{ij}/dp in 10^{-24} cm^4/dyn^2.

[d] dC_{ij}/dT in 10^8 dyn/cm^2 °K.

[e] Reference 13.

III. RESULTS AND DATA ANALYSIS

A. Elastic constants under ambient conditions

The measurements of the repetition rate frequencies at room temperature and atmospheric pressure were reproducible to a few tenths of a percent when different rf frequencies and bonds were used. This slight lack of reproducibility is most likely due to nonideal bonds and is the largest source of error in the measurement of the absolute values of the elastic constants for the mode propagating along [110]. The values of the three elastic constants [determined from Eqs. (1)] and their estimated uncertainties are given in the second column of Table I. Also listed in that column are the derived elastic compliances S_{ij} and volume compressibility κ_V. All these elastic parameters are adiabatic parameters. The comparison of the present elastic constants with measurements of other workers will be discussed below.

As was already mentioned, the longitudinal and shear modes propagating along [111] were measured as cross checks of the data. It was found that the measured effective elastic constant for the longitudinal mode, Eq. (2b), agreed to within 0.1% with the value predicted using the C_{ij} obtained from the [110] propagation direction, Table I. The measured shear-mode effective elastic constant, Eq. (2a), however, was about 2% higher than predicted. The exact cause for this discrepancy is not known, but it may be associated with the fact that for this mode the energy flow direction (elastic Poynting vector) is not colinear with the propagation direction. It was noted that the echo pattern was not ideal for this mode and changed somewhat with change in the polarization direction. Therefore, measurements of this mode are probably considerably less reliable than for the other modes. The excellent agreement for the longitudinal mode along [111] indicates that the crystal was accurately oriented and that the correct cycle-for-cycle superpositions were used in the measurements.

B. Elastic constants at high pressure

The ultrasonic data for the three modes propagating along [110] are shown in Fig. 1. The quantity plotted is $[f(p)/f_0]^2$, where $f(p)$ is the repetition rate frequency (inverse pulse transit time) and f_0 is the value of $f(p)$ at $p=0$. The relation between the effective elastic constant $C'(p)$ for a given mode and the repetition rate frequency for that mode is

$$\frac{C'(p)}{C'_0} = \frac{V_0}{V(p)}\left(\frac{l(p)f(p)}{l_0 f_0}\right)^2 = \frac{l_0}{l(p)}\left(\frac{f(p)}{f(0)}\right)^2, \tag{3}$$

where V is the volume of the sample and l is the ultrasonic path length. Thus the data plots of Fig. 1 represent the reduced effective elastic constants (which are listed on the figure for each mode), but only to the extent that the compression of the sample can be ignored. To extract the pressure dependences of the individual elastic constants a method similar to the method of Cook[6] was used to calculate simultaneously and self-consistently both the dimensional and elastic-constant changes. The resulting pressure-dependent elastic

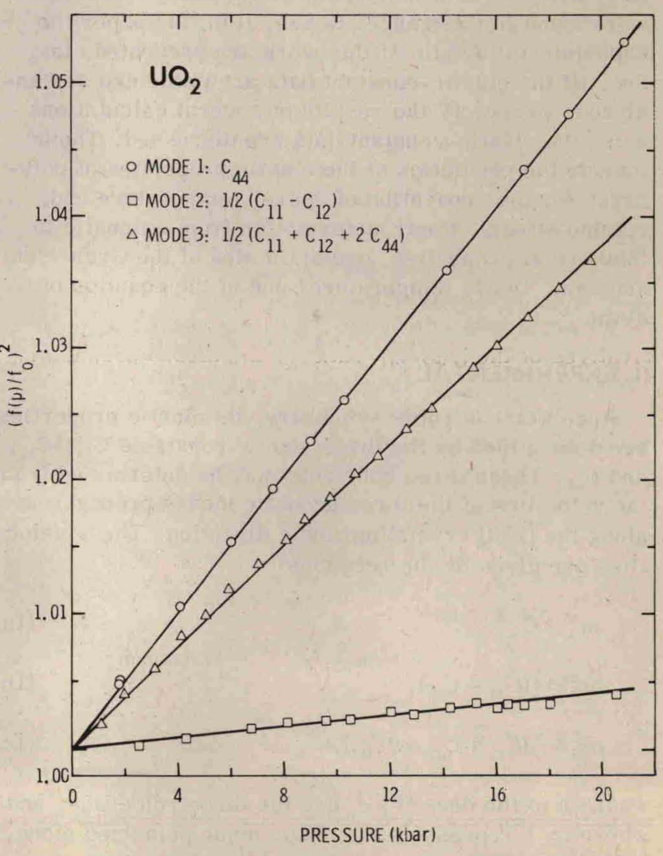

FIG. 1. Repetition rate data plotted as the square of the relative rate versus pressure.

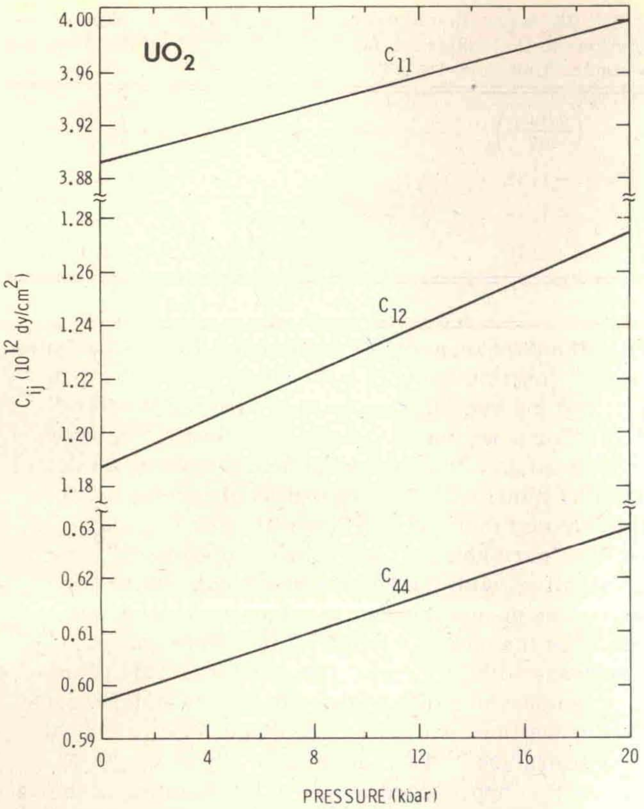

FIG. 2. Pressure dependences of the elastic constants.

constants are shown in Fig. 2. From Figs. 1 and 2 we see that the elastic properties of UO_2 appear to exhibit normal pressure dependences with no evidence for any anomalous softening of any elastic constant or for any pressure-induced phase transitions. Within experimental uncertainty the elastic constants vary linearly with pressure, at least up to the 20-kbar limit of the present experiment. The pressure derivatives of the various elastic parameters are listed in Table I with their experimental uncertainties.

The pressure-dependent elastic constants (determined as described above from the modes propagating along [110]) were used to predict the expected pressure derivatives of the $[f(p)/f_0]^2$ data for the modes propagating along [111]. For the longitudinal mode the agreement between prediction and the actual measurement is within 3%, which is well within the experimental uncertainties listed in Table I. For the shear mode there is a discrepancy of about 10%, but, again, the data for this mode are not considered as reliable as for the other modes.

IV. DISCUSSION

A. Elastic constants at ambient conditions and prediction of polycrystalline properties

The values of the elastic constants obtained under ambient conditions obtained in this work are shown in Table I along with the values obtained by Wachtman *et al.*[1] The agreement between these two sets of values is disappointingly poor, especially for C_{44}, where there is a 7% discrepancy which is well outside the range of the experimental uncertainties. The reason for the disagreement is not known, but there are several possibilities. In the first place, it has been found difficult to obtain consistent and reproducible values for other measured physical properties of UO_2 such as thermal expansion,[6] and this is apparently due to actual sample-to-sample variations. Thus, even though our crystal was from the same source as those used by Wachtman *et al.*,[1] the measured differences may be real. Wachtman *et al.* obtained a large range of values for the C_{ij} from their measurements on different samples and with different sound propagation directions. This may be partially due to sample differences. Another possible problem may be related to the fact that Wachtman *et al.* made their transit-time measurements on rectified pulses. Techniques using unrectified pulses (such as pulse superposition or pulse-echo overlap, for example) are generally regarded as significantly more reliable, as systematic errors which may occur when using rectified pulses are quite easily eliminated. A third source of error may be associated with the problem mentioned above of measuring the shear-mode velocity for [111] propagation. Noncolinearity of the acoustic energy flux and the propagation direction can, in some instances, cause distortion of the echoes leading to velocity errors. As Wachtman *et al.* did not measure velocities along high-symmetry directions, this effect could possibly have been a problem.

The single-crystal elastic constants can be used to estimate the bulk elastic properties of polycrystalline UO_2 by using the method of Hill[7] (Voigt-Reuss-Hill averaging scheme). Anderson[8] has found this method to give reliable values for the polycrystalline properties

TABLE II. Adiabatic elastic moduli of polycrystalline UO_2. K, G, and E in units of kbar.

Modulus M	VRH averages			Measured values		$\frac{dM}{dp}$
	This work	Wachtman *et al.* [a]	Marlowe [b]	Forlano *et al.* [c]	Igato and Domoto [d]	
K	2090	2130	2032	⋯	⋯	4.69
G	830	870	821	⋯	⋯	1.42
E	2210	2310	2172	2234	2196	3.90
σ	0.324	0.319	0.323	⋯	⋯	0.08 [e]

[a] Reference 1.
[b] Reference 9.
[c] Reference 10.
[d] Reference 11.
[e] Units of 10^{-12} cm^2/dyn.

of many materials. Predicted values for the bulk modulus K, shear modulus G, Young's modulus E, and Poisson's ratio σ are shown in Table II for the values of the C_{ij} obtained by us (first column) and by Wachtman *et al.* (second column). The last three columns show measured values (all extrapolated to zero porosity) as obtained by Marlowe,[9] Forlano *et al.*,[10] and Igata and Domoto.[11] The agreement between the measured values and the values predicted from the C_{ij} of Wachtman *et al.* is significantly worse. The pressure derivatives of K, G, E, and σ (predicted from the pressure dependences of the C_{ij}) are shown in the last column of Table II.

Anderson[8] has calculated values of "average" longitudinal and shear sound velocities for many oxides and it is interesting to compare the UO_2 results with the properties of the other oxides. We find average velocities (longitudinal and shear, respectively) of

$$\bar{v}_l = [(K + \tfrac{4}{3} G)/\rho]^{1/2} = 5.4\times 10^5 \text{ cm/sec},$$

and

$$\bar{v}_s = (G/\rho)^{1/2} = 2.8\times 10^5 \text{ cm/sec}.$$

The pressure derivatives of these velocities are $d\ln\bar{v}_l/dp = 1.0\times 10^{-3}$ kbar^{-1} and $d\ln\bar{v}_s/dp = 0.85\times 10^{-3}$ kbar^{-1}. Anderson[8] shows in his Fig. 2 the general trend for materials with a given kind of anion (oxides, fluorides, chlorides, bromides, iodides, and carbides) to have a decreasing value of $\bar{v}_l/\rho$ as a function of the mean atomic weight $\bar{M}$ (where $\bar{M} = M/n$ with M equal to molecular weight and n equal to number of atoms in the molecule). For the oxides considered by Anderson, $\bar{v}_l/\rho$ decreases anomalously rapidly compared to the other substances on the graph and in fact extrapolates to zero at $\bar{M} \approx 55$. For UO_2 we have $\bar{M} = 90$, and the value of $\bar{v}_l/\rho \approx 0.5$ is generally in line with the values for other materials with large values of $\bar{M}$, e.g., the iodides. Thus the general trends for the oxides with $\bar{M} \leq 41$ do not necessarily extrapolate to large values of $\bar{M}$. Anderson *et al.*[12] have shown that the elastic properties of many oxides can be parameterized as a function of $\bar{M}$ and ρ, but have cautioned that this approach may be too simplistic in general. The present results for UO_2 appear to justify this caution. For example, the UO_2 values of K and ρ do not fall anywhere near the values for other oxides on a double log plot of K versus ρ. (See, for example, Fig. 7 of Ref. 12.)

B. Separation of "pure" temperature and volume effects

The temperature dependences of the elastic constants C_{ij} between 4.2 and 300 °K have been measured by Brandt and Walker[13] in their study of the 31 °K antiferromagnetic phase transition. From their data we have determined the temperature derivatives of the C_{ij} at room temperature (296 °K). These derivatives, which are listed in Table I, can be considered as being composed of two separate contributions, namely the "pure volume" contribution (due to the thermal expansion of the sample) and a "pure temperature" contribution (due to anharmonic lattice effects at constant volume). The relation expressing this composition is[14]

$$\left(\frac{\partial \ln C_{ij}}{\partial T}\right)_p = -\frac{3\alpha}{\kappa_V}\left(\frac{\partial \ln C_{ij}}{\partial p}\right)_T + \left(\frac{\partial \ln C_{ij}}{\partial T}\right)_V, \tag{4}$$

TABLE III. Separation of "pure" temperature and volume contributions to the isobaric temperature derivatives of the elastic constants. Units are $10^{-4}/°K$.

C_{ij}	$\left(\frac{\partial \ln C_{ij}}{\partial T}\right)_p$	=	$-\frac{3\alpha}{\kappa_V}\left(\frac{\partial \ln C_{ij}}{\partial p}\right)_T$	+	$\left(\frac{\partial \ln C_{ij}}{\partial T}\right)_V$
C_{11}	−1.32	=	−0.65	−	0.67
C_{12}	−1.88	=	−1.72	−	0.16
C_{44}	+1.70	=	−1.27	+	2.97

where the first term on the right-hand side is the "pure volume" contribution with α the linear thermal expansion, and the second term is the "pure temperature" effect. For α we use the value $\alpha = 7.5\times 10^{-6}/°K$ which we obtained as a typical experimental value from data shown by Winslow.[6] The separation described by Eq. (4) is shown for the C_{ij} in Table III. For C_{11} and C_{12} both the "pure volume" and "pure temperature" effects are negative, with the volume effect dominating for C_{12}. For C_{44} the pure temperature effect is positive and dominates the isobaric temperature effect causing C_{44} to increase with increasing temperature. This anomalous temperature effect is due to the low-temperature phase transition, which has been discussed previously in the literature.[15] The softening of C_{44} as the 31 °K transition is approached is due to the coupling of the *xz* strain to the low-lying electronic states of the UO_2 ions (cooperative Jahn-Teller effect). It is the large temperature effect of this coupling which is responsible for the large and positive "pure temperature" effect for C_{44} shown in Table III.

C. Adiabatic-isothermal corrections

All of the elastic properties discussed so far have been adiabatic properties. For some applications it is desirable to have isothermal values of the various elastic moduli. The transformation between adiabatic and isothermal compliances (S^S_{ij} and S^T_{ij}, respectively) is given by the following equations:

$$\begin{aligned} S^T_{11} &= S^S_{11} + \frac{\alpha^2 T}{\rho C_p}, \\ S^T_{12} &= S^S_{12} + \frac{\alpha^2 T}{\rho C_p}, \\ S^T_{44} &= S^S_{44}, \end{aligned} \tag{5}$$

where C_p is the specific heat at constant pressure, for which we take the value[16] 15.3 cal/°K mol. Note that the following hold:

$$\begin{aligned} S^S_{11} - S^S_{12} &= S^T_{11} - S^T_{12}, \\ C^S_{11} - C^S_{12} &= C^T_{11} - C^T_{12}, \\ C^S_{44} &= C^T_{44}. \end{aligned} \tag{6}$$

We will discuss the adiabatic-isothermal correction only for the moduli K, G, E, and σ. For the bulk modulus we obtain from Eqs. (5) the result

$$\frac{1}{K^T} = \frac{1}{K^S} + \frac{9\alpha^2 T}{\rho C_p}, \tag{7}$$

which gives the isothermal bulk modulus $K^T = 2060$ kbar as compared to the adiabatic value (Table II) of $K^S = 2090$ kbar. For the shear modulus we have, because of Eq. (6), the results $G^S = G^T$. This result is obvious, because there is no thermoelastic effect associated with the shear mode. However, the point is emphasized here, because it apparently has not been realized by all workers.[9]

To calculate the Voigt-Reuss-Hill isothermal Young's modulus we first make separate adiabatic to isothermal corrections for the Voigt and Reuss expressions then take the average value of the resulting isothermal moduli. We obtain the isothermal Young's modulus of 2204 kbar compared to the adiabatic value of 2207 kbar. (The value of 2210 kbar given in Table II was rounded off to three significant digits. This was not done for the present calculation because the adiabatic to isothermal correction is so small.) The smallness of the correction is due to the fact that the value of the shear modulus G dominates the value of E and, as discussed above, there is no adiabatic to isothermal correction for G. For Poisson's ratio we find, using a similar procedure to that used for E, the isothermal value $\sigma^T = 0.322$ compared to the adiabatic value of $\sigma^S = 0.324$.

Marlowe[9] has published adiabatic to isothermal corrections for UO_2 based on the assumption (which he does not justify) that there is zero correction for σ. This procedure leads to a finite correction for G and a correction for E which is about three times larger than the one we have calculated. We believe that our procedure for calculating the adiabatic to isothermal corrections (zero correction for G) is better justified than Marlowe's. The corrections are, of course, quite small at room temperature.

D. Grüneisen constant, Debye temperature, and equation of state

Procedures for estimating the Grüneisen constant γ from single-crystal elastic-constant data have been extensively reviewed in the literature[17] and will not be discussed here in any detail. These procedures generally involve some approximations which cannot be rigorously justified. We will use what is probably the simplest (and least justifiable) approach, which is to assume an isotropic model with the values of $\overline{v}_l$ and $\overline{v}_s$ which were computed above as the sound velocities. In this approach we expect γ to lie between the values of γ_{LT} and γ_{HT} defined by the following equations:

$$\gamma_{\mathrm{LT}} = \frac{(\overline{v}_s/\overline{v}_l)^3\gamma_l + 2\gamma_s}{(\overline{v}_s/\overline{v}_l)^3 + 2},$$

$$\gamma_{\mathrm{HT}} = \tfrac{1}{3}(\gamma_l + 2\gamma_s). \tag{8}$$

In Eq. (8), γ_l and γ_s are defined as

$$\gamma_s = \frac{1}{3} + K^T\left(\frac{\partial \ln\overline{v}_s}{\partial p}\right)_T,$$
$$\gamma_l = \frac{1}{3} + K^T\left(\frac{\partial \ln\overline{v}_l}{\partial p}\right)_T \tag{9}$$

Using the values of $\overline{v}_l$, $\overline{v}_s$, and their pressure derivatives given above we obtain $\gamma_s = 2.1$ and $\gamma_l = 2.4$, which yield $\gamma_{\mathrm{HT}} = 2.2$ and $\gamma_{\mathrm{LT}} = 2.1$. The values of the thermal Grüneisen parameter reported in the literature have a range of values, possibly because of sample-to-sample variations. Hall[18] reports a room temperature value of 1.8 as calculated from data of Burdick and Parker[19] (thermal expansion) and of Rand and Kubaschewski[20] (heat capacity). Using the value of α quoted above and the heat-capacity data of Moore and Kelley[21] we obtain the same value of $\gamma_{\mathrm{th}} = 1.8$. Marlowe and Kaznoff[22] obtain values near 1.7, while Winslow[6] uses a value of 2.0.

Elastic data can be used to estimate the Debye temperature θ. The appropriate equation, which again holds for an isotropic model, is[17]

$$\theta = \frac{h}{k}\left(\frac{9\rho N}{4\pi M}\right)^{1/3}\left(\frac{2}{\overline{v}_s^3} + \frac{1}{\overline{v}_l^3}\right)^{-1/3}, \tag{10}$$

where h is Planck's constant, k is Bolzmann's constant, and N is Avogadro's number. Using the values of $\overline{v}_s$ and $\overline{v}_l$ obtained above we obtain a Debye temperature of $\theta = 385$ °K. Marlowe and Kaznoff[22] give a value of $\theta = 875$ °K computed from their elastic data. It is very difficult to understand this large discrepancy, since the elastic data are in reasonably good agreement. One possible explanation is that Marlowe and Kaznoff may have made an algebraic error in evaluating Eq. (10); in this regard note that the ratio of the two Debye temperatures is $875/385 \approx 10^{1/3}$. Jones *et al.*[16] obtain a value of $\theta = 160$ °K from low-temperature specific-heat data. It has been pointed out previously,[22,23] however, there is a magnetic contribution to the specific heat at low temperature due to the 31 °K antiferromagnetic transition which makes a reliable determination of the Debye temperature difficult. Dolling *et al.*[23] have fitted their neutron diffraction data on UO_2 to a shell model and have plotted the temperature dependence of the predicted Debye temperature between 0 and 500 °K. The 0 °K value for θ is $\theta = 395$ °K in good agreement with our value of $\theta = 385$ °K.

Anderson[24] has discussed the use of the pressure derivative of the isothermal bulk modulus dK^T/dp to estimate the high-pressure compression of solids via the Murnaghan equation of state. This equation is

$$\ln\frac{V}{V_0} = \left(\frac{dK^T}{dp}\right)^{-1}\ln\left(1 + p\,\frac{d\ln K^T}{dp}\right). \tag{11}$$

We have followed the procedure given by Anderson[24] to obtain dK^T/dp from our value of $dK^S/dp = 4.69$ (Table II). We obtain $dK^T/dp = 4.81$, which gives the following compression equation

$$V/V_0 = (1 + 0.00233p)^{-0.208}. \tag{12}$$

In summary, we have measured the elastic constants of UO_2 at high pressure, and these constants exhibit normal and linear increases with pressure up to 20 kbar. There is no evidence for a pressure-induced room-temperature phase transition up to this pressure. The data have been used to calculate various elastic properties of UO_2 and compared to other data and calculations where possible.

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