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THE ADIABATIC ELASTIC MODULI OF SINGLE-CRYSTAL
ALPHA URANIUM AT 25°C

by

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THE ADIABATIC ELASTIC MODULI OF SINGLE-CRYSTAL ALPHA URANIUM AT 25°C

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ABSTRACT

The 9 single-crystal elastic moduli pertaining to principal crystallographic axes of alpha uranium at 25°C were determined from measurements of high-frequency wave velocities for 21 modes in seven single-crystal specimens, using the phase-comparison method of McSkimin.⁽¹⁾ A special orienting and lapping technique was used for preparing the specimens from small single crystals. The probable errors in each measurement, computed from estimated uncertainties, agree well with the internal consistency of the data. The stiffness moduli, in units of 10^{12} dynes/cm², are as follows:

$$\begin{aligned}c_{11} &= 2.147 \pm 0.14\%, \quad c_{12} = 0.465 \pm 0.6\%, \quad c_{13} = 0.218 \pm 1.5\% \\c_{22} &= 1.986 \pm 0.14\%, \quad c_{23} = 1.076 \pm 0.34\%, \quad c_{33} = 2.671 \pm 0.14\% \\c_{44} &= 1.2444 \pm 0.10\%, \quad c_{55} = 0.7342 \pm 0.10\% \\c_{66} &= 0.7433 \pm 0.10\%\end{aligned}$$

From these results the elastic compliances, compressibilities and Poisson's ratios have been computed for the principal axes, and the variations with crystal direction of the stiffness moduli, Young's moduli and rigidity moduli have been plotted. The nature of the anisotropy for the different moduli indicate that the nearest neighbor interatomic bonds are considerably stiffer than the next nearest bonds, which are, however, only slightly larger in interatomic distance.

I. INTRODUCTION

Although alpha uranium is quite unique among metals because of its orthorhombic symmetry, the crystal structure can be described simply as an assembly of parallel corrugated layers.⁽²⁾ The anisotropy of the thermal expansion,⁽³⁾ electrical resistivity,⁽⁴⁾ and plastic deformation⁽⁵⁾ is, however, not of the type expected in a layer structure, and indicates a more complex distribution of interatomic binding forces. The present study of the single-crystal elastic moduli was undertaken in an effort to clarify the anisotropy of these forces as well as to measure the various elastic parameters needed for theoretical treatments of other anisotropic properties.

The results given in this report are the adiabatic elastic moduli as computed from the velocities of high-frequency ultrasonic waves in single crystals of reasonably good perfection. Because of the small dimensions of the available crystals, the phase-comparison technique and equipment of H. J. McSkimin at Bell Telephone Laboratories, Murray Hill, New Jersey, was used for the wave velocity measurements. All of the wave velocity data given here are due to Messrs. McSkimin and T. B. Bateman of B.T.L. The necessary specimens were prepared at ANL. (12,32,33)

In addition to describing the technique and results of the measurements, this report gives a brief review of the mathematical treatment of elastic stresses and strains for the purpose of defining the various elastic moduli and parameters and to provide the principles of the dynamic method of measurement. The discussion gives a correlation of the measurement with the interatomic bonds and describes the anisotropy of certain dilational and shear moduli.

II. REVIEW OF ELASTIC STRESS-STRAIN RELATIONS IN ORTHORHOMBIC CRYSTALS

A. Relation of Elastic Moduli to Stress and Strain Components

The classical theory of elastic stress-strain relations in anisotropic media is treated in various textbooks and reviews.^(6,7) For small elastic strains the relations can be described completely by the following linear forms of Hooke's law:

$$\begin{aligned}
 T_x &= c_{11} e_{xx} + c_{12} e_{yy} + c_{13} e_{zz} + c_{14} e_{zy} + c_{15} e_{zx} + c_{16} e_{xy} \\
 T_y &= c_{12} e_{xx} + c_{22} e_{yy} + c_{23} e_{zz} + c_{24} e_{zy} + c_{25} e_{zx} + c_{26} e_{xy} \\
 T_z &= c_{13} e_{xx} + c_{23} e_{yy} + c_{33} e_{zz} + c_{34} e_{zy} + c_{35} e_{zx} + c_{36} e_{xy} \\
 T_{zy} &= c_{14} e_{xx} + c_{24} e_{yy} + c_{34} e_{zz} + c_{44} e_{zy} + c_{45} e_{zx} + c_{46} e_{xy} \\
 T_{zx} &= c_{15} e_{xx} + c_{25} e_{yy} + c_{35} e_{zz} + c_{45} e_{zy} + c_{55} e_{zx} + c_{56} e_{xy} \\
 T_{xy} &= c_{16} e_{xx} + c_{26} e_{yy} + c_{36} e_{zz} + c_{46} e_{zy} + c_{56} e_{zx} + c_{66} e_{xy}
 \end{aligned} \tag{1}$$

or

$$\begin{aligned}
 e_{xx} &= s_{11} T_x + s_{12} T_y + s_{13} T_z + s_{14} T_{zy} + s_{15} T_{zx} + s_{16} T_{xy} \\
 e_{yy} &= s_{12} T_x + s_{22} T_y + s_{23} T_z + s_{24} T_{zy} + s_{25} T_{zx} + s_{26} T_{xy} \\
 e_{zz} &= s_{13} T_x + s_{23} T_y + s_{33} T_z + s_{34} T_{zy} + s_{35} T_{zx} + s_{36} T_{xy} \\
 e_{zy} &= s_{14} T_x + s_{24} T_y + s_{34} T_z + s_{44} T_{zy} + s_{45} T_{zx} + s_{46} T_{xy} \\
 e_{zx} &= s_{15} T_x + s_{25} T_y + s_{35} T_z + s_{45} T_{zy} + s_{55} T_{zx} + s_{56} T_{xy} \\
 e_{xy} &= s_{16} T_x + s_{26} T_y + s_{36} T_z + s_{46} T_{zy} + s_{56} T_{zx} + s_{66} T_{xy},
 \end{aligned} \tag{2}$$

where

x , y and z are orthogonal directions in a Cartesian coordinate system, T_x , T_y , T_z , T_{zy} , T_{zx} and T_{xy} are the six possible components of stress, e_{xx} , e_{yy} , e_{zz} , e_{zy} , e_{zx} and e_{xy} are the six possible components of strain and the c_{ij} and s_{ij} are respectively defined as stiffness and compliance moduli.

The physical meaning of the subscripts can be seen by separating the matrix formed by the coefficients into 4 groups:

$$\begin{array}{c}
 \begin{array}{c} 1 \end{array} \begin{array}{|c|c|c|c|} \hline C_{11} & C_{12} & C_{13} & C_{14} \\ \hline C_{12} & C_{22} & C_{23} & C_{24} \\ \hline C_{13} & C_{23} & C_{33} & C_{34} \\ \hline \end{array} \begin{array}{c} 2 \end{array} \begin{array}{|c|c|c|c|} \hline C_{15} & C_{16} & C_{25} & C_{26} \\ \hline C_{25} & C_{26} & C_{35} & C_{36} \\ \hline C_{35} & C_{36} & C_{45} & C_{46} \\ \hline \end{array} \\
 \begin{array}{c} 3 \end{array} \begin{array}{|c|c|c|c|} \hline C_{14} & C_{24} & C_{34} & C_{44} \\ \hline C_{24} & C_{25} & C_{35} & C_{45} \\ \hline C_{34} & C_{35} & C_{45} & C_{55} \\ \hline \end{array} \begin{array}{c} 4 \end{array} \begin{array}{|c|c|c|c|} \hline C_{16} & C_{26} & C_{36} & C_{46} \\ \hline C_{26} & C_{36} & C_{46} & C_{56} \\ \hline C_{36} & C_{46} & C_{56} & C_{66} \\ \hline \end{array}
 \end{array} \quad (3)$$

By referring back to equations (1) it can be seen that Group 1 (upper left) relates dilational strains, e_{xx} , e_{yy} and e_{zz} , to dilational stresses, T_x , T_y and T_z . The moduli which lie along the diagonal of the matrix of Group 1, c_{11} , c_{22} and c_{33} , relate dilational strains e_{xx} , e_{yy} and e_{zz} to T_x , T_y and T_z , respectively. The off-diagonal elements, c_{12} , c_{13} and c_{23} , relate dilational strains to dilational stresses in perpendicular directions.

Groups 2 and 3 are the moduli which relate shear strain to dilational stress, and dilational strains to shear stresses.

The Group 4 moduli are those relating shear strains and shear stresses. As in the case of Group 1, the stresses and strains related by the diagonal moduli of this group are of the same orientation. For example, c_{44} relates the shear strain in the y direction of the z plane, e_{zy} , to the shear stress in the same plane and direction, T_{zy} . Since $T_{zy} = T_{yz}$, it is seen that c_{44} also applies to shear in z direction of the y plane. The off-diagonal elements of this group relate shear strains and shear stresses which differ in orientation by 90° .

When neither x , y nor z refer to symmetry axes or planes of a crystal, all 21 of the moduli may be independent and nonzero. When x , y and z refer to the principal crystallographic axes of a crystal which has greater than triclinic symmetry, the number of independent moduli is reduced due to (1) certain moduli becoming zero in value and (2) certain moduli becoming simply related. For orthorhombic symmetry the number of independent moduli is decreased to 9, because of the disappearance of Groups 2 and 3 and the offdiagonal elements of Group 4. The elastic stiffness modulus matrix for alpha uranium (orthorhombic system) with reference to the principal axes is then as follows:

$$\begin{bmatrix}
 c_{11} & c_{12} & c_{13} & 0 & 0 & 0 \\
 c_{12} & c_{22} & c_{23} & 0 & 0 & 0 \\
 c_{13} & c_{23} & c_{33} & 0 & 0 & 0 \\
 0 & 0 & 0 & c_{44} & 0 & 0 \\
 0 & 0 & 0 & 0 & c_{55} & 0 \\
 0 & 0 & 0 & 0 & 0 & c_{66}
 \end{bmatrix}
 \quad (4)$$

The elastic compliance matrix, s_{ij} , reduces in the same way to give:

$$\begin{bmatrix}
 s_{11} & s_{12} & s_{13} & 0 & 0 & 0 \\
 s_{12} & s_{22} & s_{23} & 0 & 0 & 0 \\
 s_{13} & s_{23} & s_{33} & 0 & 0 & 0 \\
 0 & 0 & 0 & s_{44} & 0 & 0 \\
 0 & 0 & 0 & 0 & s_{55} & 0 \\
 0 & 0 & 0 & 0 & 0 & s_{66}
 \end{bmatrix}
 \quad (5)$$

B. Transformation of Axes

It is stated above that the values of the c_{ij} , or of the s_{ij} , for a given crystal depend on the orientation of the reference axes with respect to the structure of the crystal. To assist in discussion, it is convenient to designate the moduli which refer to the principal crystal axes as the unprimed c_{ij} or s_{ij} . For the general case, then, the moduli will be primed. In some cases it is convenient to refer to the principal axes, $[100]$, $[010]$ and $[001]$, as x , y and z and to use x' , y' and z' for the general case.

An important and convenient property of the elastic moduli matrix is the interdependence of the matrices which are based on different reference systems. Using the similarity transformation procedure of matrix algebra, one can easily form expressions for any primed modulus in terms of the unprimed moduli and the direction cosines of x' , y' and z' with respect to x , y and z . The reverse is also true, i.e., the unprimed moduli can be calculated from a knowledge of the primed moduli and direction cosines.

C. Relation between Stiffness and Compliance Moduli

Since the c_{ij} and s_{ij} both relate stress components to strain components they are related to each other. Using matrix algebra, it can be shown that the c matrix and the s matrix are related as inverse matrices:

$$[c][s] = [1] \quad , \quad (6)$$

where $[1]$ is the 6×6 identity matrix with all elements zero except for the diagonal elements, which are all equal to unity. Equation (6) gives 36 relations of the form

$$\left. \begin{aligned} c_{11}s_{11} + c_{12}s_{12} + c_{13}s_{13} + c_{14}s_{14} + c_{15}s_{15} + c_{16}s_{16} &= 1 \\ c_{11}s_{12} + c_{12}s_{22} + c_{13}s_{23} + c_{14}s_{24} + c_{15}s_{25} + c_{16}s_{26} &= 0 \end{aligned} \right\} \quad (7)$$

Equations for any s in terms of the c 's, or any c in terms of the compliances, s , can be obtained by simultaneous solution of the above relations. By means of determinants, the equations can be put in the form

$$s_{ij} = \frac{\pm |c|_{ij}}{|c|} \quad , \quad (8)$$

where i may or may not equal j , where $\pm |c|_{ij}$ is the cofactor of the element c_{ij} , and where $|c|$ is the complete determinant of the c matrix.

When the reference axes are the principal directions in an orthorhombic crystal, equations (8) become simplified so that the shear stiffness moduli and the corresponding shear compliance moduli are merely reciprocals of each other, i.e., $s_{44} = 1/c_{44}$. Furthermore, due to factoring, the denominator of equations (8) is merely the 3×3 determinant formed by the elements of Group 1 in (3) and the cofactors are formed by 2×2 determinants involving Group 1 only.

D. Relation of Moduli to Engineering Parameters

In the field of engineering elasticity, various parameters are encountered which are used as standard measures of resistance of materials to elastic strain, e.g., Young's modulus, rigidity modulus, Poisson's ratio and bulk modulus. It is of interest here to consider the relations between the stiffness moduli, compliance moduli and the above-mentioned parameters by inspection of the Hooke's law equations given in (1) and (2).

From (1) it can be seen that c_{11} is the ratio T_x/e_{xx} when all other strains are zero. It is obvious, then, that conditions for applying, or measuring c_{11} , are ordinarily not encountered where the stress is due to a static load. In the usual tensile test, for example, extension in the direction

of the stress is always coupled to contractions, or expansion in some cases, in lateral directions. From (2) it can be seen that T_x/e_{xx} in this case, where x is the direction of pull, is defined by $1/s_{11}$ and corresponds to the definition of Young's modulus. In general, then, Young's modulus, $E_{x'}$, is given by

$$E_{x'} = 1/s_{11}' \quad (9)$$

and is equal to the ratio of dilational stress to dilational elastic strain in the direction of the applied stress when the coupled dilation strains are permitted. It should be noted that $E_{x'}$ will always be smaller than c_{11}' .

Again from (1), it can be seen that c_{12} , c_{13} and c_{23} are not directly related to Poisson's ratio, since they are a measure of the uncoupled lateral dilations. The compliances s_{12} , s_{13} and s_{23} do, however, give directly the ratio of lateral dilation to stress in a perpendicular direction. For example, s_{12} defines the dilational strain in the y direction, e_{yy} , which is produced by a stress T_x . Poisson's ratio for this case, which is defined as

$$\sigma_{12} = -e_{yy}/e_{xx} \text{ for } T_x, \quad (10)$$

is then

$$\sigma_{12} = -s_{12}/s_{11} = -(E_x s_{12}) \quad (11)$$

Since the rigidity modulus, G , is defined as the ratio of shear stress to shear strain in the same planes and directions when coupling strains are present, it is simply related to the compliance moduli as follows:

$$G_{x'y'} = 1/s_{66}', \text{ etc.} \quad (12)$$

The modulus G bears a simple relation to the stiffness moduli c_{44} , c_{55} and c_{66} in the orthorhombic case, only when the reference axes are principal crystallographic axes, since

$$\begin{aligned} c_{66} &= 1/s_{66}, \text{ etc.}; \\ G_{xy} &= c_{66} = 1/s_{66}, \text{ etc.} \end{aligned} \quad (13)$$

The bulk modulus can be defined as the ratio of uniform hydrostatic pressure to fractional volume change, $\Delta V/V$. The compressibility is the reciprocal of the bulk modulus, and may be thought of as a bulk compliance modulus. By resolving the total volume change into strains e_{xx} , e_{yy} and e_{zz} , the following relations are obtained from (2):

$$e_{xx}/T_x = s_{11} + s_{12} + s_{13} \quad (14)$$

$$e_{yy}/T_y = s_{22} + s_{12} + s_{23} \quad (15)$$

$$e_{zz}/T_z = s_{33} + s_{13} + s_{23} \quad (16)$$

where $T_x = T_y = T_z =$ hydrostatic pressure. Each of the above equations defines a linear compressibility in the respective directions x , y and z . The volume compressibility is defined as the sum total of (14), (15) and (16):

$$\beta_V = s_{11} + s_{22} + s_{33} + 2(s_{12} + s_{13} + s_{23}) \quad (17)$$

III. MEASUREMENTS OF STIFFNESS MODULI

A. Relation of Stiffness Moduli to Elastic Waves in Crystals

Because the c_{ij} are individually related to uncoupled strains they are more useful than the s_{ij} in the computation of energy changes in crystals caused by mechanical strains and thermal effects in crystals. The elastic waves produced by thermal energy are assumed to travel through the lattice as plane waves, and in the approximation to harmonic motion, the frequencies of vibration can be calculated from a knowledge of the wave velocities propagated in all directions of the crystal at low temperatures.⁽⁸⁾ For plane elastic waves traveling in a crystal with dimensions which are large compared with the wave length, the wave velocities propagated in any direction can be rigorously related to the c_{ij} through the classical theory of vibrations due to Christoffel (given in reference 6). Because of these relations and because of the relatively recent development of electronic methods for producing acoustic wave pulses of high frequency, the c_{ij} have assumed major importance in the measurement of elastic properties. Using methods based on high-frequency pulses, the assumption of an infinite medium can be closely approached so as to permit very accurate measurements of the stiffness moduli. Accurate derivations of the engineering parameters which are calculated from the s_{ij} are then permissible.

Since the relations between the c_{ij} and the velocities of ultrasonic plane waves are intimately related to the measurements described in this report, the mathematical derivations of the relations will be briefly treated here. More complete treatments of vibrations in anisotropic solids are given in references (6) and (9). For this brief derivation we assume a plane wave of high frequency propagating in a direction x' in a crystal. The general equations of motion simplify to

$$\left. \begin{aligned} F_{x'} &= \rho \frac{\partial^2 u}{\partial t^2} = \frac{\partial T_{x'}}{\partial x'} \\ F_{y'} &= \rho \frac{\partial^2 v}{\partial t^2} = \frac{\partial T_{x'y'}}{\partial x'} \\ F_{z'} &= \rho \frac{\partial^2 w}{\partial t^2} = \frac{\partial T_{x'z'}}{\partial x'} \end{aligned} \right\}, \quad (18)$$

where $F_{x'}$, etc., are the components of force in the x' , y' , and z' directions, ρ equals the density of medium, u , v and w are components of displacement resolved along x' , y' and z' , time is given by t , and $\partial x'$ is the increment of movement of the wave front in the x' direction. The instantaneous stress can be resolved into the dilational component $T_{x'}$, producing u , the shear stress component $T_{x'y'}$, producing v , and the shear stress component $T_{x'z'}$, producing w .

Expressing the stresses in terms of the strains from equations (1), there are obtained relations such as

$$\frac{\partial T_{x'x'}}{\partial x'} = c_{11} \frac{\partial e_{x'x'}}{\partial x'} + c_{12} \frac{\partial e_{x'y'}}{\partial x'} + c_{15} \frac{\partial e_{x'z'}}{\partial x'} \quad (19)$$

Since

$$\left. \begin{aligned} e_{x'x'} &= \frac{\partial u}{\partial x'} \\ e_{x'y'} &= \frac{\partial v}{\partial x'} + \frac{\partial u}{\partial y'} \\ e_{x'z'} &= \frac{\partial w}{\partial x'} + \frac{\partial u}{\partial z'} \end{aligned} \right\}, \quad (20)$$

equations (18) become

$$\left. \begin{aligned} \rho \frac{\partial^2 u}{\partial t^2} &= c_{11} \frac{\partial^2 u}{\partial x'^2} + c_{16} \frac{\partial^2 v}{\partial x'^2} + c_{15} \frac{\partial^2 w}{\partial x'^2} \\ \rho \frac{\partial^2 v}{\partial t^2} &= c_{16} \frac{\partial^2 u}{\partial x'^2} + c_{66} \frac{\partial^2 v}{\partial x'^2} + c_{56} \frac{\partial^2 w}{\partial x'^2} \\ \rho \frac{\partial^2 w}{\partial t^2} &= c_{15} \frac{\partial^2 u}{\partial x'^2} + c_{56} \frac{\partial^2 v}{\partial x'^2} + c_{55} \frac{\partial^2 w}{\partial x'^2} \end{aligned} \right\} \quad (21)$$

Assuming that particle displacements can be represented as harmonic oscillations, the components u , v and w are then given by the following equations:

$$\left. \begin{aligned} u &= U e^{2\pi i(ft - \frac{x'}{\lambda})} \\ v &= V e^{2\pi i(ft - \frac{x'}{\lambda})} \\ w &= W e^{2\pi i(ft - \frac{x'}{\lambda})} \end{aligned} \right\} \quad (22)$$

where f equals the frequency of the propagated wave, λ equals the wave length, and $i = \sqrt{-1}$.

Differentiation of equations (22) to form

$$\left. \begin{aligned} \frac{\partial^2 u}{\partial t^2}, \frac{\partial^2 v}{\partial t^2}, \frac{\partial^2 w}{\partial t^2} \\ \frac{\partial^2 u}{\partial x^2}, \frac{\partial^2 v}{\partial x^2}, \frac{\partial^2 w}{\partial x^2} \end{aligned} \right\} \quad (23)$$

and substitution into (21) give

$$\left. \begin{aligned} \rho U(2\pi f)^2 &= \left(\frac{2\pi i}{\lambda}\right)^2 [Uc_{11} + Vc_{16} + Wc_{15}] \\ \rho V(2\pi f)^2 &= \left(\frac{2\pi i}{\lambda}\right)^2 [Uc_{16} + Vc_{66} + Wc_{56}] \\ \rho W(2\pi f)^2 &= \left(\frac{2\pi i}{\lambda}\right)^2 [Uc_{15} + Vc_{56} + Wc_{55}] \end{aligned} \right\} \quad (24)$$

which reduce to

$$\left. \begin{aligned} U \rho (f\lambda)^2 &= U\rho S^2 = Uc_{11} + Vc_{16} + Wc_{15} \\ V \rho (f\lambda)^2 &= V\rho S^2 = Uc_{16} + Vc_{66} + Wc_{56} \\ W \rho (f\lambda)^2 &= W\rho S^2 = Uc_{15} + Vc_{56} + Wc_{55} \end{aligned} \right\} \quad (25)$$

where $S = f\lambda$ is the phase velocity of the wave propagated in x' direction. A slight rearrangement gives the usual form of homogeneous simultaneous equations:

$$\left. \begin{aligned} (c'_{11} - \rho S^2)U + c'_{16}V + c'_{15}W &= 0 \\ c'_{16}U + (c'_{66} - \rho S^2)V + c'_{56}W &= 0 \\ c'_{15}U + c'_{56}V + (c'_{55} - \rho S^2)W &= 0 \end{aligned} \right\} \quad (26)$$

In order for U, V and W, which correspond to direction cosines, to have solutions other than zero, it can be shown that the determinant formed by the coefficients of U, V and W must equal zero:

$$\begin{vmatrix} (c_{11} - \rho S^2) & c_{16} & c_{15} \\ c_{16} & (c_{66} - \rho S^2) & c_{56} \\ c_{15} & c_{56} & (c_{55} - \rho S^2) \end{vmatrix} = 0 \quad (27)$$

The latter equation has 3 solutions for ρS^2 in terms of the elastic moduli. If each solution for ρS^2 and the appropriate numbers for c_{ij} are substituted back into equation (26), it can be seen that there are 3 separate nonzero sets of U, V and W each corresponding to a given velocity, S. These sets of U, V, W and S correspond physically to the 3 normal modes for propagation in direction x' . In general, the mode with the largest value of S will be that which is close to a longitudinal mode (U close to 1, V and W close to zero for a unit strain). In the following sections this mode will be henceforth referred to as quasi-longitudinal. The remaining two velocities are those of the quasi-shear modes where the displacement directions defined by U, V and W are respectively 90° from that of the quasi-longitudinal mode and 90° from each other.

B. General Application of Pulse Techniques

The derivation of the stiffness moduli, c_{ij} , is based entirely on the solutions to equation (27), which give the relations between a measured wave velocity and the c_{ij} . Since the c_{ij} can be algebraically found in terms of the c_{ij} and the direction cosines of the primed reference axes with respect to the principal crystal axes, the experimental problem is to measure separately each of the 3 wave velocities for a given x' direction and obtain 3 equations for the c_{ij} . For orthorhombic crystals there are nine c_{ij} ; consequently, at least 9 independent equations for the c_{ij} are required for complete solution. This means that, in general, measuring the wave velocities for each wave mode for 3 different x' directions is sufficient. (In special cases three x' directions are not sufficient, as 2 modes may be determined by the same stiffness moduli.)

The physical arrangement for measuring ultrasonic wave velocities by the various modifications of the pulse technique are generally similar. A plate-shaped piezoelectric transducer is coupled to a flat face on a single crystal specimen with a seal material which is suitable for transmitting the acoustic waves produced when the transducer is excited by an electric field. The specimen has a second flat face which is parallel to the coupled face. A pulsed train of high-frequency plane waves travels through the specimen and, in some cases, is partially transmitted to a second piezoelectric crystal

which is coupled to the second face and acts as a detector. The electrical impulses produced by the detector crystal are amplified and rectified for viewing the transmitted signals and subsequently arriving echo signals on an oscilloscope screen. The most direct method for measuring the velocity of the wave through the crystal is to measure the time delay between the pulse trigger and the first detected signal or the time between echo signals, knowing the path length of the propagated wave. A slight modification of this method is to use the same piezoelectric crystal as source and detector.

The experimental conditions and type of apparatus needed for measurements of high accuracy depend considerably on the dimensions of the specimen. If the lateral dimensions of the specimen are small, say less than 1 cm on edge, the frequency of the propagated wave should be relatively high, so as to approach the infinite-plate conditions upon which the correlation of wave velocity and elastic moduli is based. In practice, one finds a variation in measured wave velocity with frequency which decreases as the frequency is increased.

C. Method and Procedure for Alpha Uranium

The fractional error in the velocity as measured from delay time increases with decreasing specimen thickness. For specimens less than 3 mm in thickness the accuracy of delay-time measurements is severely limited. In such cases the phase-comparison method, developed at the Bell Laboratories, has been shown to be a very precise means of measuring the velocity of pulsed waves.⁽¹⁾ In this method a series of pulsed high-frequency wave trains are permitted to overlap and phase-interfere in the specimen. The frequency is varied until all of the waves in the specimen are in phase with each other, as detected by the scheme described below. The wave velocity is then computed from the usual resonance relation:

$$S = 2tf_n/n \quad , \quad (28)$$

where f_n is the "in phase" frequency, t is the specimen thickness, n equals the number of wavelengths in $2t$, and $2t/n$ equals λ , the wavelength.

The scheme for detecting the frequency at which the specimen waves are in phase is best shown with reference to Figure 1, a block diagram of the measuring circuit, and Figure 2, which shows some of the details of the buffer-specimen assembly specifically designed for small specimens. The output from a stable continuous wave oscillator is periodically gated to provide a series of wave trains. By means of X-cut transducers (for longitudinal waves), or Y-cut transducers (for shear waves), repeat trains of mechanical waves are set up in the buffer rod, the construction of which is described below. Some of the energy passes through the seal (the nature of which is also described below) and reflects back and forth within the specimen, giving rise to a series of pulses which are detected by the

transducer that was originally the transmitter. A larger share of the energy is, however, reflected from the buffer to seal interface and gives rise to a large amplitude signal which precedes the echo signals on the observed oscilloscope pattern. The succession of reflections are shown schematically in Figure 3a. If the pulse is lengthened, the successive reflections can phase-interfere with each other giving rise to a variation with time in the amplitude of the strong interface signal. From transmission line theory, it can be shown, however, the the frequencies which place the specimen echoes, E_t and $E_{t'}$, in phase also place E_b , the strong interface reflection, out of phase with E_t , etc. At such "critical" frequencies the strong interface signal amplitudes are systematically changed with time; this gives rise to a characteristic oscilloscope pattern when the time scale is compressed. Further details on use of the method are given in references 1 and 10.

Although the buffer rod is not a necessary part of the method, it adds greatly to the precision and accuracy of measurement when the specimens are of small dimensions. At high frequencies it is desirable to have a small active area at the center of the specimen face so that edge distortions are reduced. This can be obtained by beveling the end of the buffer, as shown in Figure 2. The bevel also serves to reduce the amplitude of the strong interface reflection and to maintain a smaller E_b/E_t ratio, which assists in phase comparison and in evaluating the seal-coupling effect discussed below.

For propagating longitudinal waves, the buffer was a quartz crystal, $\frac{1}{2}$ in. diameter x 2 in. long, with length along the x axis of the quartz. The transducer, also X-cut quartz with a 20-Mc/sec natural frequency, was affixed to one end of the buffer with standard silver paste solder. The specimen was attached to the tapered end of the buffer, which had a face, $\frac{1}{8}$ in. x $\frac{1}{8}$ in. square, ground flat with 303- $\frac{1}{2}$ emery finish. For transmitting longitudinal waves from buffer to specimen, a thin seal of silicone oil (Dow Corning DC703) was quite efficient.

For shear wave propagation, the unit consisted of a fused quartz buffer rod to which a Y-cut quartz transducer of 13-Mc/sec material frequency was pasted, again with silver solder. Although silicone oil was in some cases adequate for transmitting energy between the buffer and specimen, a stiffer liquid is more desirable. "Nonaq" stopcock grease was found generally satisfactory. Dow Chemical resin 276-V9 (poly- α -methyl styrene) was also very satisfactory as a shear wave seal; however, it has the possible disadvantage of requiring heat to permit the seal to form.

Each unit was attached to a brass flange for mounting in a chamber surrounded by a brass water-cooling jacket. The water was pumped through the jacket from a constant temperature reservoir, and the temperature very close to the specimen was monitored to maintain a temperature of $25 \pm 0.1^\circ\text{C}$.

The procedure of measurements was as follows:

- a) One specimen face was sealed to the tapered end of the buffer rod in either the longitudinal or shear wave unit.
- b) The rf oscillator was then triggered so that repeated wave trains with a certain harmonic of the oscillator frequency were propagated in the specimen, and the pattern of the envelope of the received wave trains was observed on the oscilloscope.
- c) The oscillator frequency was varied within the band until the "in phase" pattern appeared.
- d) The frequency at which the "in phase" pattern appeared was measured with a maximum error of ± 500 cps, absolute, or $\pm 0.005\%$. The maximum scatter in reproducing a given measurement was about 0.015%. This scatter was due to the uncertainty in defining the "in phase" pattern.
- e) After recording the "in phase" frequency, f_n , the oscillator frequency was changed and the frequencies corresponding to subsequently observed "in phase" patterns were recorded.
- f) The number of wavelengths in $2t$ [see equation (28)] at any "in phase" frequency was then computed from the relation

$$n_o = f_o / \Delta f \quad , \quad (29)$$

where f_o is the "in phase" frequency and Δf is the frequency separation between f_o and f_1 , or $f_{N+1} - f_N$.

- g) Equation (28) must be considered an approximation for S , the phase velocity, since the small shift in phase due to reflection at the specimen seal interface is not considered. This effectively means that the wavelength associated with a given f_n is $(2t/n) + (\gamma/360)$, where γ is the phase angle associated with the reflection and is denoted as δ in Figure 3b. When the specimen thickness is large, the correction can be assumed to be insignificant. For small t , however, significant errors may be involved by disregarding γ , and equation (28) should be revised:

$$S = \frac{2t f_n}{n + (\gamma/360)} \quad (30)$$

From transmission line theory one can evaluate γ from a measurement of the amplitude ratio E_t/E_b and knowledge of the seal impedance, Z_o , buffer rod impedance, Z_s , and specimen impedance, Z_r .⁽¹⁾ Figure 4 shows plots of γ versus E_t/E_b for longitudinal waves for various values of

Z_o (typical of seals used), using typical values of Z_r and Z_s for uranium and fused silica, respectively. As a consequence of the high impedance for uranium, γ remains relatively small regardless of Z_o ; it can be safely assumed that corrections for shear waves are equally small and that Z_o need not be accurately known. Furthermore, since the measured E_t/E_b ratios were of the order of 0.03 to 0.05, in the present case γ was quite small. For a total phase shift in the specimen of $(360^\circ \cdot n)$, where n is of the order of 200, a maximum relative error of 0.01% in the velocity is obtained for $\gamma = 8^\circ$. Consequently, the correction was disregarded in the computations, but is included in the estimation of possible errors in Section VI. It should be noted, however, that large n values (high frequencies) are important to prevent large errors in disregarding γ .

IV. SPECIMEN PREPARATION

A. Basis for Selection of Orientations

In connection with equation (27) it was pointed out that the derivation of the 9 independent c_{ij} for orthorhombic symmetry requires the measurement of 9 different mode velocities, where each of the c_{ij} is involved in at least one of the mode stiffnesses. It would then be sufficient to prepare 3 single crystals in the form of plates with 2 parallel faces, or planes, each providing a different crystal direction for wave propagation. With a knowledge of the orientations and the velocities of each of the 3 modes in each crystal, one obtains the 9 necessary simultaneous equations. There are, however, some objections to this somewhat random selection of orientations. The first objection pertains to the propagation of errors in the moduli derived from the simultaneous solution, since each c_{ij} is derived from 9 different velocity measurements. Secondly, the direction cosines of the normal to a plane which is not 90° to a principal plane cannot in general be as accurately measured by either photographic or spectrometer X-ray techniques, as more simple orientations.

Two other objections are presented which pertain to cross coupling of the longitudinal and shear modes when x' , the wave propagation direction, is not judiciously chosen. First, in such cases all six elements of the determinant of equation (27) are nonzero, and solutions for ρS^2 must be derived by perturbation methods.⁽¹¹⁾ Secondly, it is generally advantageous, from the experimental viewpoint, to work with pure longitudinal or shear modes. If the propagated wave is, for example, quasi-longitudinal, some of the energy is lost to the coupled quasi-shear modes; this results in less reflected wave energy and possible greater detection difficulties. This consideration may be particularly important for small specimens where difficulties due to energy loss are already great because of the small seal areas and because of the high wave frequencies necessary for accuracy.

It is obvious, then, from the above discussion, that one can increase the accuracy of the derived c_{ij} by choosing the specimen face orientations so that the measured velocities are governed by the fewest c_{ij} , and the propagated modes are as pure as possible. The ideal orientation would then be that for which the elements c_{16} , c_{15} and c_{36} are all zero and where c_{11} , c_{66} and c_{55} each correspond to a single unprimed c_{ij} . The appropriate orientations for alpha uranium are then easily derived by inspection of the general equations for the c_{ij} involved, which are as follows:

$$c_{11} = \alpha_1^4 c_{11} + \alpha_2^4 c_{22} + \alpha_3^4 c_{33} + 2\alpha_1^2 \alpha_2^2 (c_{12} + 2c_{66}) + 2\alpha_1^2 \alpha_3^2 (c_{13} + 2c_{55}) + 2\alpha_2^2 \alpha_3^2 (c_{23} + 2c_{44}) \quad (31a)$$

$$c_{66} = (\alpha_1 \beta_1)^2 c_{11} + (\alpha_2 \beta_2)^2 c_{22} + (\alpha_3 \beta_3)^2 c_{33} + 2(\alpha_1 \alpha_2 \beta_1 \beta_2) c_{12} \\ + 2(\alpha_1 \alpha_3 \beta_1 \beta_3) c_{13} + 2(\alpha_2 \alpha_3 \beta_2 \beta_3) c_{23} + [(\alpha_1 \beta_2)^2 + (\alpha_2 \beta_1)^2 \\ + 2\alpha_1 \alpha_2 \beta_1 \beta_2] c_{66} + [(\alpha_1 \beta_3)^2 + (\alpha_3 \beta_1)^2 + 2\alpha_1 \alpha_3 \beta_1 \beta_3] c_{55} \\ + [(\alpha_2 \beta_3)^2 + (\alpha_3 \beta_2)^2 + 2\alpha_2 \alpha_3 \beta_2 \beta_3] c_{44} \quad (31b)$$

$$c_{55} = (\alpha_1 \gamma_1)^2 c_{11} + (\alpha_2 \gamma_2)^2 c_{22} + (\alpha_3 \gamma_3)^2 c_{33} + 2(\alpha_1 \alpha_2 \gamma_1 \gamma_2) c_{12} \\ + 2(\alpha_1 \alpha_3 \gamma_1 \gamma_3) c_{13} + 2(\alpha_2 \alpha_3 \gamma_2 \gamma_3) c_{23} \\ + [(\alpha_1 \gamma_2)^2 + (\alpha_2 \gamma_1)^2 + 2\alpha_1 \alpha_2 \gamma_1 \gamma_2] c_{66} \\ + [(\alpha_1 \gamma_3)^2 + (\alpha_3 \gamma_1)^2 + 2\alpha_1 \alpha_3 \gamma_1 \gamma_3] c_{55} \\ + [(\alpha_2 \gamma_3)^2 + (\alpha_3 \gamma_2)^2 + 2\alpha_2 \alpha_3 \gamma_2 \gamma_3] c_{44} \quad (31c)$$

$$c_{16} = \alpha_1^3 \beta_1 c_{11} + \alpha_2^3 \beta_2 c_{22} + \alpha_3^3 \beta_3 c_{33} + \alpha_1 \alpha_2 (\alpha_2 \beta_1 + \alpha_1 \beta_2) (c_{12} + 2c_{66}) \\ + \alpha_1 \alpha_3 (\alpha_3 \beta_1 + \alpha_1 \beta_3) (c_{13} + 2c_{55}) + \alpha_2 \alpha_3 (\alpha_2 \beta_3 + \alpha_3 \beta_2) (c_{23} + 2c_{44}) \quad (31d)$$

$$c_{15} = \alpha_1^3 \gamma_1 c_{11} + \alpha_2^3 \gamma_2 c_{22} + \alpha_3^3 \gamma_3 c_{33} + \alpha_1 \alpha_2 (\alpha_2 \gamma_1 + \alpha_1 \gamma_2) (c_{12} + 2c_{66}) \\ + \alpha_1 \alpha_3 (\alpha_3 \gamma_1 + \alpha_1 \gamma_3) (c_{13} + 2c_{55}) + \alpha_2 \alpha_3 (\alpha_2 \gamma_3 + \alpha_3 \gamma_2) (c_{23} + 2c_{44}) \quad (31e)$$

$$c_{56} = \alpha_1^2 \beta_1 \gamma_1 c_{11} + \alpha_2^2 \beta_2 \gamma_2 c_{22} + \alpha_3^2 \beta_3 \gamma_3 c_{33} + \alpha_1 \alpha_2 (\beta_2 \gamma_1 + \beta_1 \gamma_2) (c_{12} + c_{66}) \\ + \alpha_1 \alpha_3 (\beta_3 \gamma_1 + \beta_1 \gamma_3) (c_{13} + c_{55}) + \alpha_2 \alpha_3 (\beta_3 \gamma_2 + \beta_2 \gamma_3) (c_{23} + c_{44}) \\ + (\alpha_1^2 \beta_2 \gamma_2 + \alpha_2^2 \beta_1 \gamma_1) c_{66} + (\alpha_1^2 \beta_3 \gamma_3 + \alpha_3^2 \beta_1 \gamma_1) c_{55} \\ + (\alpha_2^2 \beta_3 \gamma_3 + \alpha_3^2 \beta_2 \gamma_2) c_{44} \quad , \quad (31f)$$

where the α , β and γ are direction cosines of the 3 primed axes with respect to the principal axes in the following scheme:

	x	y	z
x'	α_1	α_2	α_3
y'	β_1	β_2	β_3
z'	γ_1	γ_2	γ_3

(32)

1. Orientations for $c_{11}, c_{22}, c_{33}, c_{44}, c_{55}$ and c_{66} : When $\mathbf{x}' = \mathbf{x}$, or for wave propagation in $[100]$, the direction cosines, α_i , have the following values:

$$\alpha_1 = 1$$

$$\alpha_2 = \alpha_3 = \beta_1 = \gamma_1 = 0$$

Since y' and z' can assume any orientation in the $[100]$ plane, as indicated in Figure 5, then $\beta_2, \beta_3, \gamma_2$ and γ_3 can be given as trigonometric functions of the rotation angle θ , where

$$\beta_2 = \cos \theta$$

$$\beta_3 = -\sin \theta$$

$$\gamma_2 = \sin \theta$$

$$\gamma_3 = \cos \theta$$

The elements of equation (27) then become

$$c'_{11} = c_{11}$$

$$c'_{66} = c_{66} \cos^2 \theta + c_{55} \sin^2 \theta$$

$$c'_{55} = c_{66} \sin^2 \theta + c_{55} \cos^2 \theta \quad (33)$$

$$c'_{56} = \sin \theta \cos \theta (c_{66} - c_{55})$$

$$c'_{16} = c'_{15} = 0$$

and the cubic equation (27) is then

$$(c'_{11} - \rho S^2) [(c'_{66} - \rho S^2)(c'_{55} - \rho S^2) - (c'_{56})^2] = 0 \quad (34)$$

The factor on the left gives one root immediately:

$$\rho S^2 = c'_{11} = c_{11} \quad , \quad (35)$$

obviously corresponding to the stiffness of the longitudinal mode. The other 2 solutions are obtained from the quadratic equation:

$$2\rho S^2 = c'_{55} + c'_{66} \pm [(c'_{55} + c'_{66}) - 4(c'_{55}c'_{66} - (c'_{56})^2)^{1/2}] \quad (36)$$

This equation can be solved uniquely by means of the relations between c'_{ij} and c_{ij} , given above, to obtain

$$\begin{aligned}
 2\rho S^2 &= c_{55} + c_{66} \pm [(c_{55} + c_{66})^2 - 4c_{55}c_{66} (\cos^2\theta + \sin^2\theta)^2]^{1/2} \\
 &= c_{55} + c_{66} \pm (c_{55} - c_{66}) \quad ,
 \end{aligned}
 \tag{37}$$

which gives the 2 solutions

$$\rho S^2 = c_{55} \tag{38}$$

and

$$\rho S^2 = c_{66} \tag{39}$$

These correspond to the 2 pure shear waves with shear displacements in the [001] and [010] directions, respectively.

For $x' = y$, or wave propagation in [010],

$$\alpha_2 = 1$$

$$\alpha_1 = \alpha_3 = \beta_2 = \gamma_2 = 0 \quad ,$$

and x and y are interchanged in Figure 5 to give:

$$\beta_1 = \cos \theta$$

$$\beta_3 = -\sin \theta$$

$$\gamma_1 = \sin \theta$$

$$\gamma_3 = \cos \theta \quad .$$

The solutions to equation (27) are then

$$\rho S^2 = c_{22}, \text{ longitudinal mode stiffness;} \tag{40}$$

$$\rho S^2 = c_{44}, \text{ shear mode stiffness with shear displacement in [001];} \tag{41}$$

and

$$\rho S^2 = c_{66}, \text{ shear mode stiffness with shear displacement in [100].} \tag{42}$$

For $x' = z$, or wave propagation in [001],

$$\alpha_3 = 1$$

$$\alpha_1 = \alpha_2 = \beta_3 = \gamma_3 = 0 \quad ,$$

and interchanging x and z in Figure 5 gives

$$\beta_1 = -\sin \theta$$

$$\beta_2 = \cos \theta$$

$$\gamma_1 = \cos \theta$$

$$\gamma_2 = \sin \theta$$

The solutions to equation (27) are then

$$\rho S^2 = c_{33} = \text{longitudinal mode stiffness;} \quad (43)$$

$$\rho S^2 = c_{44} = \text{shear mode stiffness, for shear displacement in [010];} \quad (44)$$

$$\rho S^2 = c_{55} = \text{shear mode stiffness, for shear displacement in [100];} \quad (45)$$

To summarize the analyses for wave propagation in the principal crystallographic directions:

- 1) only pure modes can be propagated;
- 2) each mode velocity is related to a single unprimed elastic modulus, c_{ij} ;
- 3) only the diagonal moduli, c_{11} , c_{22} , c_{33} , c_{44} , c_{55} and c_{66} , can be derived from these orientations;
- 4) one obtains 2 independent measurements of each shear modulus.

2. Orientations for Deriving c_{12} , c_{13} and c_{23} :

a. Derivation of c_{12} : The above analysis has shown that none of the cross-coupling moduli, c_{12} , etc., can be derived from wave propagation along a principal orthorhombic crystal axis. Furthermore, inspection of the Hooke's Law equations (1) shows that these moduli cannot be directly measured, i.e., one cannot obtain c_{12} , etc., without prior knowledge of certain diagonal moduli.

The first approach to selection of orientations is then to eliminate as many direction cosines as possible while providing for solutions to the secular equations (27) which include at least one of the cross-coupling moduli. The best that can be done in this respect is to make x' a nonprincipal direction in a principal plane. For example, if x' is 90° to $[001]$ (Figure 6), the following direction cosines matrix applies:

	x	y	z
x'	cos θ	sin θ	0
y'	-sin θ	cos θ	0
z'	0	0	1

or:

$$\alpha_1 = \cos\theta$$

$$\alpha_2 = \sin\theta$$

$$\beta_1 = -\sin\theta$$

$$\beta_2 = \cos\theta$$

$$\gamma_3 = 1$$

$$\gamma_3 = \beta_3 = \gamma_1 = \gamma_2 = 0 \quad .$$

Referring to equations (27) and (31),

$$c_{15}^1 = c_{56}^1 = 0 \quad ,$$

and equation (27) reduces to

$$(c_{55}^1 - \rho S^2) [(c_{11}^1 - \rho S^2)(c_{66}^1 - \rho S^2) - (c_{16}^1)^2] = 0 \quad , \quad (46)$$

where

$$\begin{aligned} c_{55}^1 &= c_{55} \cos^2\theta + c_{44} \sin^2\theta \\ c_{11}^1 &= c_{11} \cos^4\theta + c_{22} \sin^4\theta + 2 \sin^2\theta \cos^2\theta (c_{12} + 2c_{66}) \\ c_{66}^1 &= c_{66} (\cos^2\theta - \sin^2\theta)^2 + \sin^2\theta \cos^2\theta (c_{11} + c_{22} - 2c_{12}) \\ c_{16}^1 &= \sin^3\theta \cos\theta (c_{22} - c_{12} - 2c_{66}) - \cos^3\theta \sin\theta (c_{11} - c_{12} - 2c_{66}) \end{aligned} \quad (47)$$

The obvious root:

$$\rho S^2 = c_{55}^1 = c_{55} \cos^2\theta + c_{44} \sin^2\theta \quad , \quad (48)$$

is that of the pure shear mode with displacement in the [001] direction. The remaining 2 roots are obtained from the quadratic formula

$$2\rho S^2 = c'_{11} + c'_{66} \pm [(c'_{66} - c'_{11})^2 + 4(c'_{16})^2]^{1/2}, \quad (49)$$

which gives

$$2\rho S^2 = c_{66} + c_{11} \cos^2\theta + c_{22} \sin^2\theta + [M]^{1/2} \quad (50a)$$

$$2\rho S^2 = c_{66} + c_{11} \cos^2\theta + c_{22} \sin^2\theta - [M]^{1/2} \quad (50b)$$

Since the solution (50a) is obviously a larger quantity than (50b), one can say most certainly that these solutions correspond to the quasi-longitudinal and quasi-shear modes, respectively. The problem of the displacement directions for these are discussed in a later section. The present intent is to derive the expressions for the modulus c_{12} , which is included in the quantity M . The modulus c_{12} can be obtained from either solution by using the relations obtained upon rearrangement of equations (50):

$$M = (c'_{66} - c'_{11})^2 + 4(c'_{16})^2 = [2\rho S^2 - (c'_{11} + c'_{66})]^2 \quad (51)$$

$$(c'_{66})^2 - 2c_{11}c'_{66} + (c'_{11})^2 + 4(c'_{16})^2 = 4(\rho S^2)^2 - 4\rho S^2(c'_{11} + c'_{66}) + (c'_{11})^2 + 2c_{11}c'_{66} + (c'_{66})^2, \quad (52)$$

giving

$$(c'_{16})^2 - c'_{11}c'_{66} = (\rho S^2)^2 - \rho S^2(c'_{11} + c'_{66}). \quad (53)$$

It can be shown that the variables containing c_{12} are all on the left side of (53), since:

$$c'_{11} + c'_{66} = c_{66} + c_{11} \cos^2\theta + c_{22} \sin^2\theta. \quad (54)$$

With the use of equations (39) the left side of (53) becomes

$$\begin{aligned} & \sin^2\theta \cos^2\theta (c_{12} + c_{66})^2 - c_{66} (c_{66} \sin^2\theta \cos^2\theta + c_{11} \cos^4\theta + c_{22} \sin^4\theta) \\ & - c_{11}c_{22} \sin^2\theta \cos^2\theta. \end{aligned}$$

Further isolating c_{12} and factoring gives

$$\begin{aligned} \sin^2\theta \cos^2\theta (c_{12} + c_{66})^2 &= [\rho S^2 - (c_{11} \cos^2\theta + c_{66} \sin^2\theta)] \\ & \quad [\rho S^2 - (c_{66} \cos^2\theta + c_{22} \sin^2\theta)] \end{aligned} \quad (55)$$

The final form, which is obtained by taking square roots of both sides, is then

$$c_{12} = \pm \frac{1}{\sin\theta \cos\theta} \left\{ [\rho S^2 - (c_{11} \cos^2\theta + c_{66} \sin^2\theta)] [\rho S^2 - (c_{66} \cos^2\theta + c_{22} \sin^2\theta)] \right\}^{1/2} - c_{66} \quad (56)$$

There are 3 points to be noted in connection with equation (56):

- a) There is an ambiguity in the algebraic sign of the right-hand side of the equation which arises from the square root operation. This ambiguity indicates that c_{12} may be negative. This possibility and the possibilities of c_{13} and c_{23} being negative are, however, precluded by arguments given in Appendix B.
- b) Using the positive algebraic sign, the same value of c_{12} is theoretically obtained using either the ρS^2 determined from the quasi-longitudinal mode velocity or the quasi-shear mode velocity. In practice this property provides a cross check on each of the c_{ij} (where $i \neq j$).
- c) The possible error in deriving c_{12} may be quite large, since 5 different velocity measurements are involved. In some cases the fractional error in the c_{ij} due to small errors in measuring θ can be quite large. Evaluation of errors is discussed further in a later section.
- b. Selection of rotation angle: The above analysis shows that the modulus c_{12} can be derived from wave propagation in any nonprincipal direction in the (001) plane, and, in fact, this type of orientation provides the most direct relationship between c_{12} and a measured ρS^2 . There remains, however, a choice in the angle between the propagation direction and the [100] and [010] in this plane.

In most cases, the direction 45° to the principal axes is used to minimize the fractional error in the right side of equation (55) for a given absolute error in velocity determinations, i.e., the absolute value of the differences is largest at 45° . This orientation also considerably simplifies the mathematics due to the equality of cosine and sine, and for crystals of cubic symmetry there is no cross coupling between modes in the [110] direction. In considering this problem for the present case, the energy losses in the specimen were considered to be the more important factor; consequently, it was felt that the orientation should be that between 30° and 60° which gives the least coupling between the quasi-longitudinal and quasi-shear modes. In this range of θ the fractional error, discussed above, is not greatly different from that at 45° . Some calculations were then made to find the range of orientation which provided the least coupling. The nature of the calculations are presented in Appendix A.

c. Derivations of c_{13} : The procedure for deriving the best orientations and equations for determining the moduli c_{13} and c_{23} is precisely the same as given above for c_{12} .

For deriving c_{13} the propagation direction x' is best chosen in the $[010]$ plane, as shown schematically in Figure 7. The direction cosine matrix is

	x	y	z
x'	$\sin \theta$	0	$\cos \theta$
y'	0	1	0
z'	$\cos \theta$	0	$-\sin \theta$

It should be noted here that the symbol θ is used as a general rotation angle. In the derivation of c_{12} , θ was the angle between x' and x . In the present case θ is the angle between x' and z .

Using the above cosines in equations (31) gives

$$c'_{15} = c'_{56} = 0 \quad ,$$

and equation (27) reduces to:

$$(c'_{66} - \rho S^2) [(c'_{11} - \rho S^2)(c'_{55} - \rho S^2) - (c'_{15})^2] = 0 \quad , \quad (57)$$

where

$$\left. \begin{aligned} c'_{66} &= c_{44} \cos^2 \theta + c_{66} \sin^2 \theta = \text{Stiffness of pure shear mode} \\ &\quad \text{with displacement in } [010] \text{ direction.} \\ c'_{11} &= c_{11} \sin^4 \theta + c_{33} \cos^4 \theta + 2 \sin^2 \theta \cos^2 \theta (c_{13} + 2c_{55}) \\ c'_{55} &= \sin^2 \theta \cos^2 \theta (c_{11} + c_{33} - 2c_{13}) + c_{55} (\cos^2 \theta - \sin^2 \theta)^2 \\ c'_{15} &= \sin^3 \theta \cos \theta (c_{11} - c_{13} - 2c_{55}) - \cos^3 \theta \sin \theta (c_{33} - c_{13} - 2c_{55}) \end{aligned} \right\} \quad (58)$$

The quadratic form gives the 2 roots

$$2 \rho S^2 = c_{55} + c_{33} \cos^2 \theta + c_{11} \sin^2 \theta + [N]^{\frac{1}{2}} \quad (59a)$$

$$2 \rho S^2 = c_{55} + c_{33} \cos^2 \theta + c_{11} \sin^2 \theta - [N]^{\frac{1}{2}} \quad , \quad (59b)$$

where [N] contains the modulus c_{13} , as given by

$$N = (c'_{55} - c'_{11})^2 + 4(c'_{15})^2 ,$$

and (59a and b) are, respectively, the quasi-longitudinal and quasi-shear mode stiffnesses. By rearrangement, the equation for c_{13} is obtained:

$$c_{13} = \pm \frac{1}{\sin \theta \cos \theta} [(\rho S^2 - c_{33} \cos^2 \theta - c_{55} \sin^2 \theta)(\rho S^2 - c_{55} \cos^2 \theta - c_{11} \sin^2 \theta)]^{\frac{1}{2}} - c_{55} . \quad (60)$$

As in the case of c_{12} , the positive sign is used (see Appendix B), and ρS^2 is that obtained from measurements of either of the coupled mode velocities.

d. Derivation of c_{23} : For deriving the modulus c_{23} , an x' direction in the (100) plane is most suitable (Figure 8). The cosine matrix becomes

	x	y	z
x'	0	$\sin \theta$	$\cos \theta$
y'	1	0	0
z'	0	$\cos \theta$	$-\sin \theta$

and equations (31) give

$$c'_{16} = c'_{56} = 0 .$$

Equation (27) becomes

$$(c'_{66} - \rho S^2) [(c'_{11} - \rho S^2)(c'_{55} - \rho S^2) - (c'_{15})^2] = 0 , \quad (61)$$

where

$$\left. \begin{aligned} c'_{66} &= c_{55} \cos^2 \theta + c_{66} \sin^2 \theta = \text{Stiffness of pure shear mode with displacement in [100] direction.} \\ c'_{11} &= c_{22} \sin^4 \theta + c_{33} \cos^4 \theta + 2 \sin^2 \theta \cos^2 \theta (c_{23} + 2c_{44}) \\ c'_{55} &= \sin^2 \theta \cos^2 \theta (c_{22} + c_{33} - 2c_{23}) + c_{44} (\cos^2 \theta - \sin^2 \theta)^2 \\ c'_{15} &= \sin^3 \theta \cos \theta (c_{22} - c_{23} - 2c_{44}) - \cos^3 \theta \sin \theta (c_{33} - c_{23} - 2c_{44}) . \end{aligned} \right\} \quad (62)$$

The 2 roots obtained from the quadratic form are then

$$\rho S^2 = c_{44} + c_{33} \cos^2 \theta + c_{22} \sin^2 \theta + [P]^{\frac{1}{2}} \quad (63a)$$

$$\rho S^2 = c_{55} + c_{33} \cos^2 \theta + c_{22} \sin^2 \theta - [P]^{\frac{1}{2}} \quad (63b)$$

where the modulus c_{23} is contained in $P = (c_{55}' - c_{11}')^2 + 4(c_{15}')^2$, and (63a and b) are, respectively, the quasi-longitudinal and quasi-shear mode stiffnesses. The quantity c_{23} is then derived from the relation

$$c_{23} = \pm \frac{1}{\sin \theta \cos \theta} [(\rho S^2 - c_{33} \cos^2 \theta - c_{44} \sin^2 \theta) (\rho S^2 - c_{44} \cos^2 \theta - c_{22} \sin^2 \theta)^{\frac{1}{2}} - c_{44}] \quad (64)$$

As in the case of c_{12} and c_{13} , the positive sign only is used to derive c_{13} , for reasons given in Appendix B, and ρS^2 is obtained from measurements of either coupled mode.

B. Preparation of Specimens

1. Physical Requirements:

From the preceding sections, the nature of the specimen requirements should be clear. For the measurement technique 2 flat and parallel faces are necessary. To assist in obtaining the most accurate moduli values, 6 different sets of faces are required, each of which corresponds to one of the following indices: (100), (010), (001), (hko), (hol), (okl). The specimens were prepared from single crystal rods obtained by the grain-coarsening technique.⁽¹²⁾ These crystals are free of gross substructure, such as produced by phase-change methods,⁽¹³⁾ but are unfortunately limited in dimensions, with maximum diameters of approximately 5 mm and lengths up to 1 cm.* Since the lateral dimensions (the face areas) should be as large as possible to aid in energy transmission and to permit use of the infinite-plate mathematics, it is clear that at least 6 specimens were necessary, i.e., 2 sets of faces on one specimen are not practical.

* To obtain 5-mm diameter crystals, the procedure described in reference 12 was modified by including the NaK siliconizing technique described in reference 32. The difficulties encountered in attempting to prepare 1-cm diameter crystals are discussed in reference 33.

The specific objectives involved in the preparation of suitable specimens were as follows:

- 1) preparation of a plane on the crystal which corresponds accurately to a preselected crystal plane,
- 2) preparation of a second face which was accurately parallel to the first,
- 3) make both faces reasonably flat, corresponding to relatively straight and widely spaced interference fringes with optical flat inspection, and
- 4) keep the depth of the work-hardened, or distorted, material to a minimum.

The apparatus and procedures for accomplishing the objectives are given below.

2. Orienting Procedure:

The equipment used in aligning the crystals for producing the desired face orientations was relatively simple. The rods were attached with wax to a goniometer stage where the central axis of the goniometer was in line with an X-ray collimator set up for back-reflection Laue photographs (Figure 9). After two or three photographs were taken, the crystal plane to which the ground face was to be parallel was rotated so that it was parallel to the base circle of the goniometer (or perpendicular to the central axis). The goniometer, with the rod in its final orientation, was then removed from the X-ray machine stage and set vertically so that the base circle was parallel to a support base (Figure 10). A table with a hole at the center of the table top was then set on the support base so that the crystal protruded through the hole. The table legs were made so that the table top was parallel to the base circle of the goniometer. A 1-in. diameter, hollow steel cylinder with parallel ends was placed on the table top so that it surrounded the crystal (Figure 11). A cold-setting, liquid, synthetic resin was then poured into the cylinder to fill the cavity and fix the crystal in its orientation relative to the end faces of the cylinder. The liquid flowing into perforations in the cylinder wall produced a keying effect upon solidification.

3. Grinding of Faces:

Because of the above procedure, the ends of the steel cylinders, in which the crystals were mounted, were reference planes to which the crystal faces were to be parallel. One end of the mount was then ground away on rotating waterproof abrasive paper (silicon carbide 80 to 400 mesh)

to produce a face on the crystal which was slightly smaller in area than was desired in the final specimen. The face produced was roughly close to parallel to the opposite end of the mount. To improve the parallelism, so that the ground face could subsequently be used as a reference plane for grinding of the second face, the ground surface of the mount was lapped with 14-micron grit diamond paste in the hand-lapping apparatus described below. In this apparatus, the lapped face was made parallel to the reference face to within 0.0001 in. over the 1-in. diameter.

After the second face had been produced and made parallel to the first, the crystal was removed from the mount. One of the faces was then electropolished in an alcohol base, phosphoric acid electrolyte to remove the surface layer which had been cold worked during grinding. When X-ray photographs indicated complete removal of the disturbed metal, the specimen was mounted for further lapping.

4. Lapping:

The lapping device (Figure 12) was similar to a type of machine which recently has been used for obtaining concentration gradient data for diffusion studies, where the samples to be analyzed are from parallel planes in a specimen.⁽¹⁴⁾ The specimen was attached to a rod supported vertically by a yoke. The specimen face was moved across the lap by sliding the yoke over a horizontal plane on which the lap was set. Contact between the specimen face and the abrasive was maintained by vertical motion of the supported rod. The lap remained stationary at the center of a surface plate, and the yoke was manually moved over the well-lubricated surface. The specimens were attached with either a double-backed tape seal or by cementing with the synthetic resin to a pedestal which screwed into the base of the vertical rod.

The lapping was carried out in 3 steps using 14-micron, 3-micron, and 1-micron diamond paste, respectively, and electropolishing between steps until X-ray Laue patterns indicated the removal of most of the effects of prior lapping. Because electropolishing produced a convex surface, it was necessary to stagger the schedule to preserve a flat face as reference for maintaining the orientation.

During lapping with the finer abrasives, there was some difficulty in preventing deep scratches. Pyrex glass laps were advantageous in comparison with metal laps in this respect; however, it was equally important that the whole of the specimen face was in contact with the lap. This was the primary reason for removing the specimens from the steel mounts used during grinding.

Each lapping step was carried out until the flatness, as determined by interference fringes from sodium light, could not be improved by further lapping with the abrasive used. Parallelism was also checked after each step by thickness measurements over the specimen width of a Sheffield visual comparator.

5. Description of Specimens:

Seven specimens were prepared by the above procedure; the orientations and dimensions of these are listed in Table I. For Crystals A through F, the final faces gave interference fringes which were relatively straight, and were spaced evenly and wide enough apart to be easily inspected for irregularities. Each of these crystals was uniform in thickness to an estimated 0.00001 in. Crystal G became too thin to handle for correcting certain irregularities in flatness. The indicated thicknesses were obtained by comparison with gauge blocks to a precision of 0.00002 in., or 0.0005 mm., for all but Crystal G.

The orientations noted (Table I) for Crystals A, B and C were determined from back-reflection Laue patterns. For these three crystals, the Laue technique is sufficiently precise, since deviations in alignment of the principal axes with the face normals are easy to detect from the films, using the straightness of the lines formed by the principal zone reflections as measures. The effect of possible deviations of the order of $1/2^\circ$ are included in the discussion of uncertainties in the values of the moduli (Section VI). Such deviations are shown to have small effects on the relative errors in the diagonal moduli.

For the rotated cuts, Crystals D, E, F and G, the accuracy of the film technique is not adequate for careful measurement of the rotation angles, θ_D , θ_E , θ_F and θ_G , although deviations of the plane normal from a principal zone can be reasonably well fixed, again by the straightness of the line formed by the principal zone reflections. Consequently, the rotation angles for all four crystals were determined using a goniometer technique similar to that described by Bond and Armstrong.⁽¹⁵⁾ The principle of the method and a description of the precision of results are given in Appendix C. The measured rotation angles for each crystal are given in Table I.

The extent of crystal imperfections at the specimen faces following the final lapping step were estimated from Laue patterns. In all cases these patterns indicated a disturbed metal surface layer of the order of 1 micron in thickness. In addition to the imperfections due to plate preparation, thin twin bands are generally found in crystals grown by the technique used here. The bands are few in number and apparently contribute very little to the losses of energy in the propagated waves; however, their presence is taken into account in considering the homogeneity of the specimens and the precision of measurement, as discussed in Section VI.

The shapes and, to some extent, the dimensions of each specimen were governed largely by the grain-growth habits during formation of the single-crystal rods. The $[010]$ directions are, in general, closely aligned with the rod axes. Consequently, the plate faces varied in shape depending on the orientations. Some examples of the specimen shapes are shown in Figure 13.

V. RESULTS

A. Velocities of Propagation

1. Treatment of Data: Propagation velocities, S , for each of the three modes of vibration in each of the 7 crystals were obtained by the procedure outlined in Section IIIB. An example of the velocity computations, using equations (28) and (29), is given in Table II for the longitudinal mode in Crystal C. In the first column is the index number N of the "in phase" frequencies observed within a frequency band. The first set of data is that for the 6th harmonic, ranging from 56.886 to 74.952 Mc/sec. The frequencies f_n at which the "in phase" pattern was observed are listed in the second column. It should be noted that in this case f_n corresponding to each successive in-phase pattern up to number 22 was recorded. The 23rd through 26th were observed and counted, and the 27th was recorded.

The 3rd column of Table II gives the frequency separation, Δf , between each successive f_n . For the first 22 f_n this is obtained by merely subtracting the zeroth from the first, etc. For 22 through 27 the Δf is the average obtained by subtracting the 22nd f_n from the 27th and dividing by the number 5.

The Δf which is used for computing n from equation (29),

$$n = f_n / \Delta f,$$

is that obtained by averaging all of the Δf 's in column 2; this average is recorded in column 3. Column 4 is then the result of dividing each f_n by the average Δf . Although this result is not strictly an integer, the error, e , is small and the integral number of wavelengths in $2t$ (t = sample thickness) is easily obtained without ambiguity. Column 5, then, gives the wave velocity, S , computed from the f_n and the whole number n .

The data in the subsequent part of the table are for the measurements at frequencies within the 9th, 13th, 15th and 18th harmonics. At the higher frequencies, the recognition of the characteristic pattern became more difficult due to shortening of the band width, greater specimen losses and smaller separation between the f_n . As a consequence, only the more definite f_n 's were recorded. Only one reading was obtained in the 180-Mc/sec range. The n for this f_n was obtained from the average Δf in the preceding band.

It should be noted that the frequency range covered in this example is not typical of the remaining measurements. The other 2 modes in Crystal C also gave fairly good patterns at the high frequencies (above 100 Mc/sec). With the other crystals, however, measurements were more difficult in this frequency range, and in some cases they were not possible.

2. Determination of Limit Velocity: The computed velocities for all 21 modes are plotted in graphical form as a function of measured frequency in Figures 14 through 20. In these curves the general tendency for the computed velocity to decrease with increasing frequency is obvious; all curves show some indication of this type of velocity dispersion. This effect is common to interferometric methods for acoustic wave-velocity measurements in liquids where the sound source does not completely cover the area of the plate, and it is explained on the basis of diffraction of the propagated wave by radial strains emanating from the beam to the lateral edges.⁽¹⁶⁾ The diffraction produces an aperiodicity in the propagated wave which causes an apparent increase in wave length for a given frequency. In the present case, this effect would increase the frequency at which the wave nodes terminate at the specimen faces; however, as the frequency is increased over a wide range, the effect of the diffraction decreases and the computed velocity approaches the true, or unperturbed free space, velocity.

In terms of the graphical variation, the free space velocity is the limit velocity as the frequency approaches infinity. Since the total fractional variations of velocities were relatively small, the limit velocity for a given mode could be approximated by extrapolating graphically to the high-frequency ranges. However, since the best fit curves were very close to the analytical form of the equation relating apparent and true velocities in an ideal wave guide:

$$\frac{1}{S_p^2} = \frac{1}{S^2} - \left(\frac{K}{2\pi} \right)^2 \frac{1}{f^2}, \quad (65)$$

where S_p equals apparent velocity, S equals free space velocity, f equals frequency and $(K/2\pi)^2$ is the slope of straight lines, $1/S_p^2$ vs. $1/f^2$, this equation was used to provide a consistent manner for deriving S , i.e., S^2 is the intercept at $1/f^2 = 0$ on the $1/S_p^2$ ordinate. Although this formula is an approximation, the error in S due to this procedure is at most 0.02%, which is relatively small compared to other sources of error discussed in Section VI. Table III gives the limiting velocities from each set of measurements which were used for deriving the stiffness moduli.

B. Calculation of Stiffness Moduli

1. Direct Computation: The most direct relations between the limit velocities and the elastic stiffness moduli were derived in Section IV above. Table IV gives the specific relations between each velocity and a modulus. The absolute values of the moduli were derived using a density at 25°C of 19.04 gm/cm³, which is that reported by M. H. Mueller from X-ray lattice parameter measurements with high-purity uranium.⁽¹⁷⁾ Bridge et al. obtained a 25°C density of 19.05 gm/cm³ from similar studies

using less pure uranium.⁽³⁾ Blumenthal also reports 19.05 gm/cm^3 from hydrostatic weighing measurements on high purity uranium.⁽¹⁸⁾ Mueller's value is, however, considered most accurate, and, in view of the other values, it is probably correct to within 0.03%.

The moduli determined from the relations given in the 3rd column in Table IV are tabulated in column 4, rounded to 4 decimal places. From these relations we obtain one value each for c_{11} , c_{22} and c_{33} and 2 values for each shear modulus, c_{44} , c_{55} and c_{66} . The average values for the shear moduli, which are given in Table V, were used in computing the cross-coupling moduli, c_{12} , c_{13} and c_{23} . Since Crystals F and G were almost of duplicate orientation, we give 4 values for c_{12} and 2 values each for c_{13} and c_{23} . As was the case for the shear moduli, the different values of the cross-coupling moduli do not agree, due to errors in velocity and/or orientation measurements. The problem of obtaining the best value for each of the 9 moduli is discussed in Section VI.

2. Indirect Computation: Although the most direct equations, given in Table IV, give the most accurate moduli, it is possible to compute all 9 moduli using only the data from the rotated cuts, Crystals D, E and F or G, and thereby obtain an independent check on the velocities, S. To do this, the data from Crystal G were disregarded and the remaining 6 equations given in Table VII were used. The first 3 are the relations between the pure shear mode velocity in the rotated cuts and the shear moduli given in equations (48), (58) and (62), and the second 3 are the sums of the 2 roots of the quadratic formulæ given in equations (50), (59) and (63). The first set of equations give:

$$\left. \begin{aligned} c_{44} &= \frac{q_{12}(\sin\theta_E \cos\theta_F)^2 - q_{15}(\sin\theta_D \cos\theta_F)^2 + q_{18}(\sin\theta_D \cos\theta_E)^2}{H} \\ c_{55} &= \frac{q_{12}(\sin\theta_F \sin\theta_E)^2 + q_{15}(\sin\theta_F \sin\theta_D)^2 + q_{18}(\sin\theta_E \cos\theta_D)^2}{H} \\ c_{66} &= \frac{q_{12}(\sin\theta_F \cos\theta_E)^2 + q_{15}(\cos\theta_F \cos\theta_D)^2 - q_{18}(\cos\theta_D \cos\theta_E)^2}{H} \end{aligned} \right\}, \quad (66)$$

where $q_{12} = \rho S_{12}^2$, $q_{15} = \rho S_{15}^2$, $q_{18} = \rho S_{18}^2$ and $H = (\cos\theta_D \cos\theta_F \sin\theta_E)^2 + (\cos\theta_E \sin\theta_F \sin\theta_D)^2$.

The values obtained from equations (66) were used to compute c_{11} , c_{22} and c_{33} from the following relations:

$$\left. \begin{aligned} c_{11} &= \frac{K_F(\cos\theta_D \sin\theta_E)^2 - K_E(\cos\theta_D \sin\theta_F)^2 + K_D(\sin\theta_E \cos\theta_F)^2}{H} \\ c_{22} &= \frac{K_F(\cos\theta_E \sin\theta_D)^2 + K_E(\cos\theta_D \cos\theta_F)^2 - K_D(\cos\theta_E \cos\theta_F)^2}{H} \\ c_{33} &= \frac{K_F(\sin\theta_D \sin\theta_E)^2 + K_E(\sin\theta_D \sin\theta_F)^2 + K_D(\sin\theta_E \cos\theta_F)^2}{H} \end{aligned} \right\}, (67)$$

where $K_F = \rho(S_{16}^2 + S_{17}^2) - c_{66}$, $K_E = \rho(S_{13}^2 + S_{14}^2) - c_{44}$ and $K_D = \rho(S_{10}^2 + S_{11}^2) - c_{55}$.

The moduli c_{12} , c_{13} and c_{23} were computed using the relations of Table IV. The values computed from either the quasi-longitudinal or quasi-shear stiffness will now be the same. Alternatively, one could use the following relations:

$$\cos\theta_D \sin\theta_D (c_{13} + c_{55}) = [\alpha_{11} \alpha_{33} - q_{10} \cdot q_{11}]^{\frac{1}{2}},$$

where α_{11} equals $c_{11} \sin^2\theta_D + c_{55} \cos^2\theta_D$, α_{33} equals $c_{55} \sin^2\theta_D + c_{33} \cos^2\theta_D$ and q_{10} equals ρS_{10}^2 , etc.;

$$\cos\theta_F \sin\theta_F (c_{12} + c_{66}) = [\beta_{11} \beta_{22} - (q_{16})(q_{17})]^{\frac{1}{2}}, \quad (68)$$

where β_{11} equals $c_{66} \sin^2\theta_F + c_{11} \cos^2\theta_F$ and β_{22} equals $c_{22} \sin^2\theta_F + c_{66} \cos^2\theta_F$; and

$$\cos\theta_E \sin\theta_E (c_{23} + c_{44}) = [\gamma_{22} \gamma_{33} - q_{13} \cdot q_{14}]^{\frac{1}{2}},$$

where γ_{22} equals $c_{22} \sin^2\theta_E + c_{44} \cos^2\theta_E$ and γ_{33} equals $c_{44} \sin^2\theta_E + c_{33} \cos^2\theta_E$.

The modulus values computed from the above equations are given in Table VIII.

VI. ERRORS IN COMPUTED MODULI

This section is concerned with the inaccuracies in the computed moduli caused by accumulation of accidental errors in the measured variables. Although the cross checks afforded by the duplicate measurements of the shear moduli, the indirect measurements of all 9 moduli (given in Table VII), and various other relations give a fair idea of the precision in measurement, they are not sufficient in number for a reliable estimate of the probable error in any one measurement. To this end it seems best to estimate the possible errors in each variable, sum up these estimated errors, and calculate a probable error which can subsequently be tested using the cross checking relations.

A. Estimated Errors in Given Velocities

The errors involved in the stated limiting velocities arise from the following sources:

1. frequency measurement,
2. specimen thickness,
3. phase shift at the specimen to buffer seal, and
4. extrapolation to limiting velocity.

The estimated error due to each of the above factors is discussed below.

1. Frequency Measurement: It is estimated that the maximum error in absolute frequency was ± 500 cps; this gives a maximum relative error of $500/10^7$, or $\pm 0.005\%$. To this must be added the error in reproducing the frequency corresponding to a given "in phase" pattern. Tests on this factor show a maximum error of $\pm 0.015\%$. The total maximum error for a given f_n is $\pm 0.020\%$.
2. Specimen Thickness: The uncertainty in specimen thickness, as measured on the sheffield comparator, was estimated to be $\pm \frac{1}{4}$ of a division on the scale, or approximately 0.000025 in., which, considering the thinnest specimen, gives a maximum possible error of 0.035% . This uncertainty is caused primarily by small deviations from parallelism of the specimen faces, or to local deviations from flatness in the faces.
3. Phase-shift Error: The possible effect of the shift in phase of the wave reflecting from the seal interface is discussed in Section III, where it is stated that this effect was neglected in the calculation of the phase velocities. The correction factor $\gamma/360$, to be used in equation (30), was then at most 0.025 , or the maximum relative error in the denominator of equation (28) is approximately 0.02% .

4. Dispersion Error: Since the total variation of velocity with frequency was at most 0.05% and generally less than 0.02%, an estimated possible error of 0.01% seems adequate.

B. Errors Due to Structural Imperfections and Orientation Uncertainty

In addition to the errors in the measured velocities, it is desirable to consider the error in assuming that the given velocities are representative of the specimen material and orientation. In this consideration, the localized dispersion of twin boundaries, included small grains, second-phase particles and generally dispersed crystal imperfections are taken into account. In addition, the inaccuracies due to orientation measurement errors are considered.

1. Structural Imperfections: Although there is little theoretical basis for estimating a quantitative error caused by the various possible inhomogeneities in the crystals, it is quite probable that they were relatively insignificant, since the X-ray diffraction patterns showed no evidence of dislocation boundaries and the microstructures observed showed at most some widely dispersed second-phase particles and a few very thin twin bands in certain crystals. One might expect that the twin bands and the associated dislocation arrays formed by the coherence with the matrix crystal would have the largest effects on velocity. Since the room temperature twin bands in uranium are most often of [130] type, one might further expect a dependence of the twin band effect on the orientation of the crystal faces. The various cross checks on the moduli discussed above indicate that the crystal had very little effect on the derived modulus, or on the velocity from which the moduli were determined. Consequently, it seems safe to assume a possible error of 0.01% due to imperfections.

2. Effect of Possible Orientation Errors: The relative error in a modulus, which result from errors in orientation measurement, depends considerably on which modulus is considered. For the diagonal moduli, c_{ii} , the error in the modulus is a direct result of the error in correlating a single wave velocity with orientation. When the procedure for attaining the alignment is considered, it appears safe to assume a maximum possible error of $\frac{1}{2}^\circ$. To determine the error caused by a $\frac{1}{2}^\circ$ misalignment, one can use the general equations for c_{11} , etc., given in equations (31) and methodically compute the effect on ρS^2 of all directions of misalignment. For example, if the direction wave propagation in Crystal C was inclined $\frac{1}{2}^\circ$ from [001] in the direction of [010], the computed primed stiffnesses [from equations (62)] are:

$$c'_{11} = c_{22}(0.6 \times 10^{-12}) + c_{33}(0.99984) + 2(c_{23} + 2c_{44})(76 \times 10^{-6})$$

$$\approx (\sim 0 + 2.6707 + 0.0005) \times 10^{12} = 2.6712 \times 10^{12}$$

$$c_{55} = 1.2442 \times 10^{12}$$

$$c_{66} = 1.2442 \times 10^{12}$$

$$c_{15} = 3.2 \times 10^{-3}$$

Since the coupling coefficient c_{15} enters into the expression for ρS^2 as $(c_{15})^2$, it can safely be neglected; this gives, from equation (61),

$$\rho S_L^2 = 2.6712 \times 10^{12}$$

$$\rho S_G^2 = 1.2442 \times 10^{12}$$

$$\rho S_S^2 = 0.7341 \times 10^{12}$$

In comparing the above results with the computed moduli, c_{33} , c_{44} and c_{55} , respectively, the effect of the $\frac{1}{2}^\circ$ misalignment towards $[010]$ can be evaluated with the conclusions that such a misalignment would give a c_{33} value which is too high by 1 part in 26,000, a c_{44} value too low by 2 parts in 12,000 and a c_{55} value which is essentially the same as for perfect alignment. These small and relatively insignificant errors are typical of the errors caused by all types of $\frac{1}{2}^\circ$ misalignments in Crystals C, B and A. Consequently, an estimated maximum possible error of 0.01% in each ρS^2 , due to $\frac{1}{2}^\circ$ errors in orientation, is considered more than adequate.

The effect of errors in orientation measurement for Crystals D, E and F on the derived values of c_{12} , c_{13} and c_{23} is of greater importance and requires a more detailed analysis. This is considered in Section VI D.

C. Summation of Estimated Errors in Diagonal Moduli

Table VI summarizes the estimates of maximum errors in the variables which affect the wave velocity computation. The assumption that the probable error is equal to the mean square error gives a probable error in velocity measurement of 0.05%, or an estimated probable error in each ρS^2 of 0.14%, when possible errors in density and errors due to imperfections and misalignments are considered. This result implies that the directly determined values for c_{11} , c_{22} and c_{33} have probable errors of 0.14%. Since 2 values were directly computed for each shear modulus, the average of the 2 values for c_{44} , c_{55} and c_{66} has an estimated probable error of 0.10%. The best values for the diagonal moduli are given in Table V.

D. Probable Errors in the Cross-coupling Moduli

The estimated probable errors in the cross-coupling moduli were also computed from the law of propagation of errors. Since the equations involved here are not instantly obvious, they are given below.

The procedure for evaluating $\Delta c_{12}/c_{12}$ is as follows:

$$\Delta c_{12} = \left[\left(\frac{\partial c_{12}}{\partial c_{11}} \Delta c_{11} \right)^2 + \left(\frac{\partial c_{12}}{\partial c_{22}} \Delta c_{22} \right)^2 + \left(\frac{\partial c_{12}}{\partial c_{66}} \Delta c_{66} \right)^2 + \left(\frac{\partial c_{12}}{\partial \rho S^2} \Delta \rho S^2 \right)^2 + \left(\frac{\partial c_{12}}{\partial \theta_F} \Delta \theta_F \right)^2 \right]^{\frac{1}{2}}. \quad (69)$$

The partial differentials, as obtained from equation (56), are

$$\begin{aligned} \frac{\partial c_{12}}{\partial c_{11}} &= \frac{(c_{22} \sin^2 \theta + c_{66} \cos^2 \theta - \rho S^2)}{2(c_{12} + c_{66}) \sin^2 \theta} \\ \frac{\partial c_{12}}{\partial c_{22}} &= \frac{(c_{11} \cos^2 \theta + c_{66} \sin^2 \theta - \rho S^2)}{2(c_{12} + c_{66}) \cos^2 \theta} \\ \frac{\partial c_{12}}{\partial c_{66}} &= \left(\frac{\sin \theta}{\cos \theta} \right)^2 \frac{\partial c_{12}}{\partial c_{11}} + \left(\frac{\cos \theta}{\sin \theta} \right)^2 \frac{\partial c_{12}}{\partial c_{22}} - 1 \\ \frac{\partial c_{12}}{\partial \rho S^2} &= - \left[\frac{1}{\cos^2 \theta} \frac{\partial c_{12}}{\partial c_{11}} + \frac{1}{\sin^2 \theta} \frac{\partial c_{12}}{\partial c_{22}} \right] \\ \frac{\partial c_{12}}{\partial \theta} &= (2 \cos \theta \sin \theta) \left[\frac{1}{\cos^2 \theta} \frac{\partial c_{12}}{\partial c_{11}} (c_{66} - c_{11}) + \frac{1}{\sin^2 \theta} \frac{\partial c_{12}}{\partial c_{22}} (c_{22} - c_{66}) \right. \\ &\quad \left. - \frac{\cos^2 \theta - \sin^2 \theta}{2 \cos^2 \theta \sin^2 \theta} (c_{12} + c_{66}) \right] \end{aligned} \quad (70)$$

where θ equals θ_F and ρS^2 is that from the given quasi-longitudinal wave in S_{16} or the quasi-shear wave, S_{17} . To evaluate equation (69), the following estimated errors in the variables are pertinent:

$$\Delta c_{66} = 0.0010 \text{ } c_{66}$$

$$\Delta \theta_F = 0.00116 \text{ radian}$$

$$\Delta c_{11} = 0.0014 \text{ } c_{11}$$

$$\Delta c_{22} = 0.0014 c_{22}$$

$$\Delta \rho S^2 = 0.0014 \rho S^2 \quad .$$

For c_{12} evaluated from the quasi-longitudinal wave in Crystal F, the partial differentials for evaluating Δc_{12} are as follows:

$$\frac{\partial c_{12}}{\partial c_{11}} = - 0.535$$

$$\frac{\partial c_{12}}{\partial c_{22}} = - 0.468$$

$$\frac{\partial c_{12}}{\partial c_{66}} = - 2.003$$

$$\frac{\partial c_{12}}{\partial \rho S_{16}^2} = + 2.006$$

$$\frac{\partial c_{12}}{\partial \theta_F} = 0.340 \times 10^{12} \text{ dynes/cm}^2 \quad ,$$

which gives a relative error of 1.36%.

For c_{12} evaluated from the quasi-shear wave, the partial differentials have the following values:

$$\frac{\partial c_{12}}{\partial c_{11}} = 0.566$$

$$\frac{\partial c_{12}}{\partial c_{22}} = 0.647$$

$$\frac{\partial c_{12}}{\partial c_{66}} = + 0.28$$

$$\frac{\partial c_{12}}{\partial \rho S_{17}^2} = 2.439$$

$$\frac{\partial c_{12}}{\partial \theta_F} = 0.019 \times 10^{12} \text{ dynes/cm}^2 \quad ,$$

and the relative error in c_{12} is 0.71%. It is of interest to note here that c_{12} calculated from the quasi-shear velocity is considerably less dependent on the values of c_{66} and θ_F than in the previous calculation of c_{12} . Because of

this and the smaller value of ρS^2 involved, the estimated probable error, $\Delta c_{12} = 0.0033 \times 10^{12}$ dynes/cm², is almost lower by a factor of 2 than that obtained previously.

The equations and numerical values used in the evaluations of Δc_{13} are as follows:

$$\Delta c_{13} = \left[\left(\frac{\partial c_{13}}{\partial c_{33}} \Delta c_{33} \right)^2 + \left(\frac{\partial c_{13}}{\partial c_{11}} \Delta c_{11} \right)^2 + \left(\frac{\partial c_{13}}{\partial c_{55}} \Delta c_{55} \right)^2 + \left(\frac{\partial c_{13}}{\partial \rho S^2} \Delta \rho S^2 \right)^2 + \left(\frac{\partial c_{13}}{\partial \theta_D} \Delta \theta_D \right)^2 \right]^{\frac{1}{2}}, \quad (71)$$

where

$$\begin{aligned} \frac{\partial c_{13}}{\partial c_{33}} &= \frac{(c_{11} \sin^2 \theta + c_{55} \cos^2 \theta - \rho S^2) \cos^2 \theta}{2(c_{13} + c_{55})(\cos \theta \cdot \sin \theta)^2} = -0.636 \text{ and } +0.393 \rho S^2 \\ \frac{\partial c_{13}}{\partial c_{11}} &= \frac{(c_{33} \cos^2 \theta + c_{55} \sin^2 \theta - \rho S^2) \sin^2 \theta}{2(c_{13} + c_{55})(\cos \theta \cdot \sin \theta)^2} = -0.374 \text{ and } +0.637 \rho S^2 \\ \frac{\partial c_{13}}{\partial c_{55}} &= (\tan^2 \theta) \frac{\partial c_{13}}{\partial c_{33}} + (\cot^2 \theta) \frac{\partial c_{13}}{\partial c_{11}} - 1 = -2.010 \text{ and } +0.030 \\ \frac{\partial c_{13}}{\partial \rho S^2} &= -\frac{1}{\cos^2 \theta} \frac{\partial c_{13}}{\partial c_{33}} - \frac{1}{\sin^2 \theta} \frac{\partial c_{13}}{\partial c_{11}} = +2.020 \text{ and } -2.060 \\ \frac{\partial c_{13}}{\partial \theta} &= (2 \cos \theta \sin \theta) \left[\frac{1}{\cos^2 \theta} \frac{\partial c_{13}}{\partial c_{33}} (c_{55} - c_{33}) + \frac{1}{\sin^2 \theta} \frac{\partial c_{13}}{\partial c_{11}} (c_{11} - c_{55}) - \frac{\cos^2 \theta - \sin^2 \theta}{2 \cos^2 \theta \sin^2 \theta} (c_{13} + c_{55}) \right] = 1.408 \text{ and } +0.278, \end{aligned}$$

where the first result is that obtained using ρS_{QL}^2 , the second is that from ρS_S^2 , and $\theta = \theta_D$. Assuming relative errors of 0.14% for c_{33} and c_{11} and 0.10% for c_{55} and $\Delta \theta_D = 4$ minutes, or 1.16×10^{-3} radian, gives

$$\frac{\Delta c_{13}}{c_{13}} = 3.07\% \text{ as computed from } \rho S_{I0}^2,$$

$$\frac{\Delta c_{13}}{c_{13}} = 1.78\% \text{ as computed from } \rho S_{I1}^2.$$

For the error in c_{23} , the following relations and numerical values pertain:

$$\Delta c_{23} = \left[\left(\frac{\partial c_{23}}{\partial c_{33}} \Delta c_{33} \right)^2 + \left(\frac{\partial c_{23}}{\partial c_{22}} \Delta c_{22} \right)^2 + \left(\frac{\partial c_{23}}{\partial c_{44}} \Delta c_{44} \right)^2 + \left(\frac{\partial c_{23}}{\partial \rho S^2} \Delta \rho S^2 \right)^2 + \left(\frac{\partial c_{23}}{\partial \theta_E} \Delta \theta_L \right)^2 \right]^{\frac{1}{2}}$$

$$\frac{\partial c_{23}}{\partial c_{33}} = \frac{(c_{22} \sin^2 \theta + c_{44} \cos^2 \theta - \rho S^2) \cos^2 \theta}{2(c_{23} + c_{44})(\cos \theta \cdot \sin \theta)^2} = -0.9307 \text{ and } 0.508$$

$$\frac{\partial c_{23}}{\partial c_{22}} = \frac{(c_{33} \cos^2 \theta + c_{44} \sin^2 \theta - \rho S^2) \cos^2 \theta}{2(c_{23} + c_{44})(\cos \theta \cdot \sin \theta)^2} = -0.2668 \text{ and } 0.4910 \quad (72)$$

$$\frac{\partial c_{23}}{\partial c_{44}} = \tan^2 \theta \frac{\partial c_{23}}{\partial c_{33}} + \cot^2 \theta \frac{\partial c_{23}}{\partial c_{22}} = 1 = -2.00 \text{ and } 0.198$$

$$\frac{\partial c_{23}}{\partial \rho S^2} = -\frac{1}{\cos^2 \theta} \frac{\partial c_{23}}{\partial c_{33}} - \frac{1}{\sin^2 \theta} \frac{\partial c_{23}}{\partial c_{22}} = +2.198 \text{ and } -2.198$$

$$\frac{\partial c_{23}}{\partial \theta} = 2(\cos \theta \cdot \sin \theta) \left[\frac{1}{\cos^2 \theta} \frac{\partial c_{23}}{\partial c_{33}} (c_{44} - c_{33}) + \frac{1}{\sin^2 \theta} \frac{\partial c_{23}}{\partial c_{22}} (c_{22} - c_{44}) - \frac{\cos^2 \theta - \sin^2 \theta}{2 \cos^2 \theta \sin^2 \theta} (c_{23} + c_{44}) \right] = 1.232 \text{ and } -0.202$$

Again, assuming 0.14% relative errors for c_{33} and c_{22} , and 0.10% for c_{44} , and $\Delta \theta_E = 4$ minutes gives

$$\frac{\Delta c_{23}}{c_{23}} = 0.95\% \text{ as computed from } \rho S_{13}^2,$$

$$\frac{\Delta c_{23}}{c_{23}} = 0.4\% \text{ as computed from } \rho S_{14}^2.$$

The above computations show that the cross-coupling moduli computed from the quasi-shear stiffness are, in general, subject to less error than that computed from the quasi-longitudinal mode. To obtain the best value for each modulus, the weighted mean values were computed from the relation

$$c_{ij} = \frac{W_1 (c_{ij})_1 + W_2 (c_{ij})_2}{W_1 + W_2}, \quad (73)$$

where W_1 and W_2 are the relative weights of each computed c_{ij} , and they are obtained from the relation

$$\frac{W_1}{W_2} = \frac{(\Delta c_{ij})_2^2}{(\Delta c_{ij})_1^2} \quad (74)$$

When either computed value is given a weight of unity, $W = 1$, the other can be computed. Substituting into (73) then gives the best value for the c_{ij} , which has a probable error

$$(\Delta c_{ij})_{\text{mean}} = \frac{(\Delta c_{ij})_W = 1}{\sqrt{W_1 + W_2}}, \quad (75)$$

where the numerator is the probable error of the value with $W = 1$. The results of these computations for c_{12} , c_{13} and c_{23} are given in Table V.

E. Comparison of Estimated Errors and Cross Checks

The above calculations of probable error are based on an estimated 0.07% probable error in each velocity measurement. This estimate can then be tested by comparison of the various cross checks afforded by the data.

The most direct check is to compare the best shear modulus values with those given from each determination. The deviation of each value from the average values are all smaller than the 0.14% estimated error. The largest deviations, 0.11%, occur for c_{44} .

Indirect checks are obtained by evaluating ρS^2 for each of the equations of Table VII, using the best values of the moduli, and comparing these with the directly observed values. The differences are given in Table VII. In all cases these differences are smaller than the probable errors associated with the computed values. This close agreement is reflected in the individual modulus values obtained by simultaneous solution of the equations, which appear in Table VIII. The probable errors in each modulus computed by this method are relatively large, due to propagation of errors (column 2, Table VIII). The deviations from the best values are, however, quite small, and tend to indicate that the estimated errors are much more than necessary.

Instead of comparing the observed and calculated ρS^2 values in the equations in Table VII, an alternative cross check is to compute the rotation angles, θ , by using observed moduli and compare these with the observed X-ray diffraction measurements, which are given in Table I and discussed in Appendix C. By rearranging the eight equations in Table VII, the following relations for rotation angles are obtained:

$$\left. \begin{aligned}
 \cos^2 \theta_D &= \frac{\rho S_{12}^2 - c_{66}}{c_{44} - c_{66}} \text{ and } \frac{\rho(S_{10}^2 + S_{11}^2) - (c_{11} + c_{55})}{c_{33} - c_{11}} \\
 \cos^2 \theta_E &= \frac{\rho S_{15}^2 - c_{66}}{c_{55} - c_{66}} \text{ and } \frac{\rho(S_{13}^2 + S_{14}^2) - (c_{22} + c_{44})}{c_{33} - c_{22}} \\
 \cos^2 \theta_F &= \frac{\rho S_{18}^2 - c_{44}}{c_{55} - c_{44}} \text{ and } \frac{\rho(S_{16}^2 + S_{17}^2) - (c_{22} + c_{66})}{c_{11} - c_{22}} \\
 \cos^2 \theta_G &= \frac{\rho S_{21}^2 - c_{44}}{c_{55} - c_{44}} \text{ and } \frac{\rho(S_{19}^2 + S_{20}^2) - (c_{22} - c_{66})}{c_{11} - c_{22}}
 \end{aligned} \right\} \quad (76)$$

The rotation angles calculated from the above equations, by using observed ρS^2 values and best values of diagonal moduli, are listed in Table IX. Also listed are the probable errors in each θ , which were calculated on the basis of 0.14% error in each ρS^2 and each of the moduli involved. It will be noted that certain of the computed values are subject to very large errors because of the small difference between moduli which make up the denominator, i.e., where $(c_{55} - c_{66})$ and $(c_{11} - c_{22})$ appear. Otherwise, the computed values can be considered quite accurate, and do, indeed, compare quite favorably to the values measured by the X-ray spectrometer technique. In fact, when these computed values are compared with rotation angles estimated from back reflection Laue films (also given in Table IX), the computed θ 's are closer to the spectrometer results.

VII. ELASTIC COMPLIANCE MODULI AND ELASTIC PARAMETERS

The mathematical relations between the stiffness moduli, compliance moduli and the more commonly used elastic parameters are given in Section II. The computed s_{ij} values derived from the best values of the c_{ij} are given in Table X. The values of the parameters, Young's modulus, Poisson's ratio and compressibilities are given in Table XI.

The range of values for Young's moduli in the 3 principal directions agree rather well with the range of values reported for polycrystalline uranium, as determined from dynamic measurements.^(19,20,21) These latter values range from 1.6 to 2.0×10^{12} dynes/cm². Although the reports on the polycrystalline measurements do supply some information as to heat treatment of the samples, the preferred orientations are, in general, not given. Consequently, close comparisons of the polycrystalline and single-crystal values are, for the most part, not possible. The best basis for comparison seems to be one in which the theoretical values for wave velocities in infinitely extended isotropic polycrystalline uranium are calculated from the models of Voigt and Reuss.^(22,23) Voigt's equations are derived on the assumption of a uniform strain in an aggregate of small randomly dispersed grains and, for the present case, give the following isotropic wave velocities of longitudinal and shear waves:

$$S_L = 3.49 \times 10^5 \text{ cm/sec}$$

$$S_S = 2.15 \times 10^5 \text{ cm/sec}$$

Reuss's equations, which are based on uniform stress in a polycrystalline aggregate, give the following isotropic wave velocities:

$$S_L = 3.40 \times 10^5 \text{ cm/sec}$$

$$S_S = 2.06 \times 10^5 \text{ cm/sec}$$

Because of the assumptions involved in each of the above methods, the results give the maximum and minimum values, respectively, and the most correct values probably are between those given above. For comparison with existing data for polycrystalline alpha uranium, it is necessary to choose the published values for samples which have been heated into the beta phase and cooled so as to randomize the orientations of the individual grains. It should be noted that conditions of the heat treatment will influence the degree of randomization and, since these conditions are not always stated, the comparison loses some of its significance. Nevertheless, among the existing data for beta-treated uranium, the above values agree quite well with the data of Laquer⁽²⁴⁾ for beta-treated and annealed samples, and the data of Kammer *et al.*,⁽¹⁹⁾ where the treatment after beta heating is unknown. This comparison is treated more critically in references 24 and 25.

P. R. Morris has measured the longitudinal wave velocities in large uranium fuel elements for which preferred orientations were obtained by X-ray diffraction. (26) He reports a relatively high correlation between the observed wave velocities and those calculated from the single-crystal moduli (communicated to him prior to this report) by an approximation method.

The computed bulk modulus is approximately 11% greater than the isothermal bulk modulus reported by Bridgman in 1931. (27) More recent adiabatic measurements give values of 1.08 and 1.26×10^{12} dynes/cm², which are in good agreement with the value 1.11×10^{12} dynes/cm² given here. (28)

VIII. CORRELATION WITH STRUCTURE

A. Dilational Properties

An interesting product of this study is the variation of the elastic stiffnesses with crystal direction. As in the case of crystals of higher symmetry, such calculations give the degree of anisotropy of the elastic properties. In this case, however, one obtains the additional interesting information concerning the directions of maximum and minimum stiffness which are not entirely dictated by the symmetry of the crystal, and, consequently, should depend to large degree on the interatomic bond directions.

Figures 21a, b and c show the variations of the dilational stiffness, c'_{11} , and Young's modulus, $E' = 1/s'_{11}$, in directions within the principal planes. The c'_{11} variations come from equation (31); the E' variations are computed from a similar equation for s'_{11} :

$$s_{11} = \alpha_1^4 s_{11} + \alpha_2^4 s_{22} + \alpha_3^4 s_{33} + \alpha_1^2 \alpha_2^2 (2s_{12} + s_{66}) + \alpha_1^2 \alpha_3^2 (2s_{13} + s_{55}) + \alpha_2^2 \alpha_3^2 (2s_{23} + s_{44}) \quad (77)$$

The directions of the maxima and minima were obtained by maximizing the first derivatives of the equations for c'_{11} and s'_{11} . For the (100) plane, for example, $\alpha_1 = 0$, and the equation for c'_{11} becomes that given in equations (62). The derivative of c'_{11} with respect to the angle of rotation, θ , from [001] to [010] is

$$\frac{dc'_{11}}{d\theta} = 4(\sin\theta \cos\theta)[(c_{22} - c_{23} - 2c_{44}) \sin^2\theta - (c_{33} - c_{23} - 2c_{44}) \cos^2\theta] \quad (78)$$

Setting $dc'_{11}/d\theta$ equal to 0 gives extreme values for c'_{11} at

$$\begin{aligned} \theta_M &= 0^\circ \\ \theta_M &= 90^\circ \end{aligned} \quad ,$$

and at an intermediate value, θ_M , where

$$\tan^2 \theta_M = \frac{c_{33} - c_{23} - 2c_{44}}{c_{22} - c_{23} - 2c_{44}} \quad (79)$$

or

$$\theta_M = 36^\circ 58'$$

It can be seen that the intermediate θ_M is dependent primarily on the difference between c_{33} and c_{22} and that for cubic crystals where, $c_{33} = c_{22}$, θ_M is automatically 45° .

For E in the (100) plane the extremes are again at $\theta = 0^\circ$, $\theta = 90^\circ$, and the angle of the intermediate maximum is given by

$$\tan^2 \theta = \frac{2s_{33} - 2s_{23} - s_{44}}{2s_{22} - 2s_{23} - s_{44}} \quad (80)$$

or

$$\theta = 38^\circ 35'$$

From the plot of c_{11} and E in the (100) plane, Figure 21a, it can be seen that the values at the [001] direction and [010] represent minima in the plane, and the very significant maxima closely coincide at approximately 37° and 38° , respectively.

The (010) plane (see Figure 21b) also exhibits extremes intermediate to the [001] and [100] directions in this plane. The stiffness c_{11} decreases rather sharply from maximum at [001] to a minimum at approximately $55\frac{1}{2}^\circ$ from [001] and rises slowly to a second maximum at [100]. Young's modulus is quite similar in the form of the variation; however, because E_{001} and E_{100} are very close to being equal, the rather significant minimum occurs very close to 45° between the 2 principal directions.

In contrast to the above cases, the (001) plane (see Figure 21c) is relatively isotropic in c_{11} ; a slight minimum is exhibited at about 23° from [010]. Young's modulus, however, shows a rather significant but continuous decrease from [100] to [010].

Figures 22a and b show the mode of variation of the dilational moduli c_{11} and E among all directions in the crystal. The figures represent quadrants of a standard stereographic projection on the (001) plane and include the equivalent of all possible directions in the orthorhombic lattice. By substituting stiffness values of Young's moduli in place of Miller indices, the distribution of modulus values can be shown by iso-stiffness or iso-Young's modulus contours. In both plots it can be seen that the maximum in the (100) plane is indeed the highest value among all directions. The [100] direction also represents a peak in both moduli, but approximately $\frac{1}{3}$ smaller in height. In each diagram there are 2 low points or "craters," one between [100] and [001] and a second near [010], for c_{11} and at [010] for E.

The general features of the c_{11} and E variations are somewhat as one would expect from a simple correlation with interatomic directions and relative interatomic distances in the alpha uranium structure, shown in Figure 23. The nearest neighbor direction is, according to Jacobs and Warren(2) and as verified by Tucker,(29) approximately 27° from [001] in

the (100) plane; this corresponds closely to the high peak in Figure 21a at 37° from [001] in this plane. The second nearest neighbor is in the [100] direction, corresponding to the other peak in the distribution diagram Figure 22. The fact that the former peak is the larger is also as expected from consideration of interatomic distances alone. In fact, the ratio of the two peak stiffnesses, $(c_{11})_{\max}/c_{11}$, corresponds very closely to the ratios of the respective Pauling bond numbers for the first and second nearest neighbors.⁽³⁰⁾ According to Pauling, these bond numbers are the number of shared electron pairs involved in a covalent bond and are related to the covalent radii of the atoms by the following equation:

$$R(1) - R(n) = 0.300 \log n ,$$

where $R(1)$ is the single valent radius corrected to coordination number of 12 and $R(n)$ is the radius for a bond number n . Assuming 5.78 bonding electrons for the uranium atom in the metal and $R(1) = 1.421 \text{ \AA}$, the bond numbers for the structure of Figure 23 are 1.36 and 0.96 for the first and second neighbors, respectively. The ratio, $1.36/0.96 = 1.42$, is surprisingly close to the ratio $(c_{11})_{\max}/c_{11} = 1.40$. At the present time the significance (if any) of this coincidence is difficult to evaluate. The significant difference between $(c_{11})_{\max}$ and c_{11} does, however, suggest that Pauling's description⁽³⁰⁾ of the lattice as an assembly of strings, which are weakly bound laterally, is more appropriate than the analogy to layer lattices, suggested by others.⁽²⁹⁾

Another interesting feature of the correlation with structure shows up in the computed Poisson's ratios given in Table XI. These come out to be quite abnormally high between [001] and [010], abnormally low between [001] and [100], and slightly below normal between [100] and [010]. In addition, one finds, when calculating Poisson's ratios between directions close to that of the nearest neighbor bond and any direction at right angles to it, that coupling with dilations in this direction are also abnormally low. As a consequence, the differences between the stiffnesses, c_{11} , and the Young's moduli, E , which can be seen from Figures 21a and b, are small (about 5%) in the [100] and in the high peak direction and relatively large in other directions (24% at [001] and 25% at [010]). It should further be noted that the small value of s_{13} and the large negative value of s_{23} make the [100] direction more compressible under hydrostatic pressure than either [010] or [001].

B. Anisotropy of the Shear Moduli

The computed shear moduli also show an interesting correlation with structure which again seems to reflect the large influence of the nearest neighbor bonds on the elasticity. Since a shear modulus pertains to a given plane, as well as to a direction in the plane, the computations and the presentation of its anisotropy are more complicated than in the case of the

dilation modulus. To compute a shear modulus c_{66} [see equation (31), for example], it is necessary to consider not only the coordinates of $\mathbf{x}'$ in terms of x, y and z , as was the case for c_{11} , but the y' coordinates must also be defined. Consequently, even an incomplete survey which considers only important low-index planes is a laborious and somewhat meaningless task unless one is interested in a particular shear direction in a given plane. In case one has such an interest in c_{66} , the equation given under (31) can be used where α_i and β_i are obtained from the relations between the normal to the plane, $\mathbf{x}'$, the direction involved, y' , and the principal axes, as given in (32). In the case of the shear elasticity, the rigidity modulus, G , is generally of greater interest than the shear stiffness and can be determined from the relation

$$G = 1/s_{66} \quad ,$$

where:

$$\begin{aligned} s_{66} &= 4[(\alpha_1\beta_1)^2 s_{11} + (\alpha_2\beta_3)^2 s_{22} + (\alpha_3\beta_3)^2 s_{33} + 2\alpha_1\alpha_2\beta_1\beta_2 s_{12} \\ &\quad + 2\alpha_1\alpha_3\beta_1\beta_3 s_{13} + 2\alpha_2\alpha_3\beta_2\beta_3 s_{23}] + [(\alpha_1\beta_2)^2 + (\alpha_2\beta_1)^2] \\ &\quad = 2\alpha_1\alpha_2\beta_1\beta_2 s_{66} + [(\alpha_1\beta_3)^2 + (\alpha_3\beta_1)^2 + 2\alpha_1\alpha_3\beta_1\beta_3] s_{55} \\ &\quad + [(\alpha_3\beta_3)^2 + (\alpha_3\beta_2)^2 + 2\alpha_2\alpha_3\beta_2\beta_3] s_{44} \end{aligned} \quad (81)$$

For the purpose of correlating the shear properties with crystal structure, it seemed advisable and adequate to survey the maximum and minimum rigidity moduli in a number of planes in the crystal. Such data lends itself to presentation in the form of the stereographic projection, as in the dilational moduli, so that the mode of variation in G throughout the crystal can be better seen. The general scheme used for mapping the stereographic projection and for computing the extremes for G in each plane involved 3 rotations, as shown in Figure 24. The first 2 rotations, ψ and θ , were for the purpose of systematically locating the plane normal, which is arbitrarily designated $\mathbf{x}''$ to denote 2 transformations from $\mathbf{x}$, and the 3rd rotation, ϕ , was to systematically change the direction in the plane. The final G values are then derived from the modulus $1/s_{66}''$, where s_{66}'' is derived from rotation about $\mathbf{x}''$ by the relation

$$s_{66}'' = s_{66}'' \cos^2 \phi + 2 \sin \phi \cos \phi s_{56}'' + s_{55}'' \sin^2 \phi \quad , \quad (82)$$

where ϕ is the angle of rotation from y'' to y''' . The extreme values of G , in the plane $\mathbf{x}''$, are obtained by differentiating (82) with respect to ϕ and maximizing; this gives the relation

$$\tan 2\phi_M = \frac{2s_{56}^{11}}{s_{66}^{11} - s_{55}^{11}}; \text{ or } \phi_M = \frac{1}{2} \arctan \frac{2s_{56}^{11}}{s_{66}^{11} - s_{55}^{11}}, \quad (83)$$

where ϕ_M is the rotation angle which gives a maximum or minimum G , depending on which of the 2 values of ϕ_M is used. The 2 values of ϕ_M satisfying (83) are 90° apart.

The double-primed variables s_{66}^{11} , s_{56}^{11} and s_{55}^{11} are computed from the primed moduli s_{ij}^1 . Since the transformation from primed to double-primed axes involved a simple rotation, in this case about y' , the relations for s_{ij}^{11} can be given in terms of a single rotation angle as follows

$$\left. \begin{aligned} s_{66}^{11} &= s_{66}^{11} \cos^2 \theta + 2s_{46}^1 \sin \theta \cos \theta + s_{44}^1 \sin^2 \theta \\ s_{56}^{11} &= 2 \sin^2 \theta \cos \theta (s_{34}^1 - s_{14}^1) + 2 \cos^2 \theta \sin \theta (s_{36}^1 - s_{16}^1) \\ &\quad - [\sin^3 \theta - \cos^2 \theta \sin \theta] s_{45}^1 + [\cos^3 \theta - \sin^2 \theta \cos \theta] s_{56}^1 \\ s_{55}^{11} &= 4 \sin^2 \theta \cos^2 \theta [s_{11}^{12} - 2s_{13}^1 + s_{33}^1] + (\cos^2 \theta - \sin^2 \theta)^2 s_{55}^1 \\ &\quad + 4 [\sin^3 \theta \cos \theta - \cos^3 \theta \sin \theta] (s_{15}^1 - s_{35}^1) \end{aligned} \right\} \cdot \quad (84)$$

The relations for computing s_{ij}^1 from the fundamental s_{ij} are also simple, since the transformation from the principal axes x , y and z , or $[100]$, $[010]$ and $[001]$, was a simple rotation about the z axis. This gives the following relations for the s_{ij}^1 involved in computing s_{ij}^{11} :

$$\left. \begin{aligned} s_{11}^1 &= s_{11} \cos^4 \psi + s_{22} \sin^4 \psi + \sin^2 \psi \cos^2 \psi (2s_{12} + s_{66}) \\ s_{33}^1 &= s_{33} \\ s_{44}^1 &= s_{44} \cos^2 \psi + s_{55} \sin^2 \psi \\ s_{55}^1 &= s_{55} \cos^2 \psi + s_{44} \sin^2 \psi \\ s_{66}^1 &= 4 \sin^2 \psi \cos^2 \psi [s_{11} + s_{22} - 2s_{12}] + (\cos^2 \psi - \sin^2 \psi)^2 s_{66} \\ s_{13}^1 &= s_{13} \cos^2 \psi + s_{23} \sin^2 \psi \\ s_{16}^1 &= \sin \psi \cos \psi [2s_{22} \sin^2 \psi - 2s_{11} \cos^2 \psi + (\cos^2 \psi - \sin^2 \psi) (s_{66} + 2s_{12})] \\ s_{36}^1 &= 2 \sin \psi \cos \psi (s_{23} - s_{13}) \\ s_{45}^1 &= \sin \psi \cos \psi (s_{44} - s_{55}) \\ s_{14}^1 = s_{15}^1 &= s_{34}^1 = s_{35}^1 = s_{46}^1 = s_{56}^1 = 0 \end{aligned} \right\} \quad (85)$$

The above procedure lends itself to providing a simple program for computing on the IBM 704 automatic computer. The program (Met. 110) consists of the above nonzero equations for s' , s'' , s''' , equation (83) for computing ϕ_M , and the values for s_{ij} . The program is divided into one case for each increment of rotation ψ above $[001]$. Each case is in turn split into increments, or subcases, of rotation θ about y' . Since the s'_{ij} are the same for each subcase, the computations for s''_{ij} as a function of θ are simplified. After computing θ_M from the s''_{ij} , the machine computes the 2 values $s'''_{\max}$ and $s'''_{\min}$ as the required output.

The results of the computations are given here in various forms.

Figures 25a and b show how $G_{\max}$ and $G_{\min}$, respectively, vary from plane to plane in the crystal. The maximum G in the crystal is that corresponding to c_{44} so that 2 peaks of equal magnitude in the distribution of $G_{\max}$ occur at (001) and (010) . The most significant changes in G with change in orientation of the plane occurs in the $[100]$ zone, where the maximum G becomes smallest in the (023) plane $[100]$ direction. The smallest minimum G 's also occur in planes close to this zone. An absolute minimum G is calculated to occur in the (154) plane in a direction normal to the $(1\bar{5}4)$ plane, with a modulus of 0.593×10^{12} dynes/cm².

Table XII gives the maximum and minimum G moduli for a number of low index planes classified according to crystal zones of $[hko]$ type. The approximate directions of the extreme moduli in the plane are given in the 3rd and 4th columns of the table. Where these directions coincide closely (within 1°) to simple indices, these are given in the table. In many cases, however, the simple designation is not possible and the direction is then described by the angle of rotation from some reference direction in the plane; a positive angle denotes that a clockwise rotation about the plane normal is necessary to bring the direction being described into coincidence with the reference direction. The right hand column gives the ratio $G_{\max}/G_{\min}$, which is an index of the anisotropy.

The variation of the ratio $G_{\max}/G_{\min}$ is schematically plotted in the stereographic projection of Figure 25c. The dominant feature of the plot is the peak index of 1.72 in a plane which is almost equally inclined to the 3 principal planes. Two indices of almost the same magnitude, 1.69 and 1.67, occur at the (001) and (010) planes. There are five planes which are isotropic in G , all of which lie in principal crystallographic zones. In the $\{100\}$ zone the isotropic planes are approximately (052) and (045) , 26° and 1° from (010) , respectively. The first mentioned corresponds to the plane which contains both the nearest and next nearest neighbors, atoms A, B, C and D in Figure 23. The latter is almost precisely normal to the direction of the nearest neighbor atoms, AC.

In the $[010]$ zone there are also 2 planes isotropic in G , one which is $7^\circ 10'$ from (100) and the other $53\frac{1}{2}^\circ$ from (100) . The latter is about 4° from (102) , which contains atoms B, C, B' and C' in Figure 23. The isotropic plane in the $[001]$ zone is approximately 8° to (100) .

IX. SUMMARY AND CONCLUSIONS

The high-frequency ultrasonic wave velocities of 21 modes of propagation in 7 single crystals of alpha uranium have been measured at 25°C by a phase-comparison technique. These data were sufficient to give at least two determinations of each of the 9 independent elastic stiffness moduli. The internal consistency of the data is quite good, considering the small dimensions of the specimens, and this indicates that the technique can be used to great advantage for high-frequency measurements with small specimens.

From a correlation of the results with the crystal structure, it appears that the two nearest neighbor bonds have a predominating influence on the anisotropy of the elastic properties.

The anisotropy, as calculated from the results, of the elastic stiffness c_{11} , Young's modulus, E , and rigidity modulus, G , are presented.

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XI. APPENDIX A

Basis for Selecting Rotation Angles in Crystals D, E, F and G

In Section IV A2 the problem of selecting the optimum rotation angle for rotated cuts of orthorhombic crystals is discussed. In the present case the object was to select rotation angles close to 45° for which the coupling between the quasi-longitudinal and quasi-shear modes was a minimum. For the perfectly uncoupled case (where the wave propagation direction, x' , is such that c'_{15} , c'_{16} and c'_{56} are zero) the three possible modes have displacement components (uoo), (ovo) and (oow) with respect to the x' , y' and z' reference axes. When, however, x' is not in a principal plane, but is rotated from the principal axes in that plane, the off-diagonal moduli, c'_{ij} , are, in general, nonzero, with values depending on the rotation angle and the unprimed elastic moduli. If the rotation axis is called z' , as in Figure 6, c'_{16} may be nonzero and, from equation (26), the displacement components u and v may appear in either the quasi-longitudinal or quasi-shear waves with the following ratios:

$$\frac{v}{u} = \frac{\rho S^2 - c'_{11}}{c'_{16}} = \frac{c'_{16}}{\rho S^2 - c'_{66}} \quad (86)$$

The objective of the selection was then to make the ratio v/u as small as possible when ρS^2 corresponds to the stiffness of the quasi-longitudinal mode. Since the moduli c_{12} , c_{13} and c_{23} were not known at the time, the procedure involved calculation of (86) with the c_{ij} and rotation angle θ as variable parameters. For x' in the (001) plane (Crystal F, for example), c'_{11} , c'_{66} and c'_{16} were calculated for various values of θ in equations (47) using c_{12} values which varied from 0.5 to 2.5×10^{12} dynes/cm². The longitudinal wave stiffness was then calculated from equation (49), and equation (86) was evaluated for each case using a code assembled for the AVIDAC computer (Met 11). Some of the results are plotted in Figure 26, which shows how v/u varied with the rotation angle for various values of c_{12} . It can be seen that for $\theta = 45 \pm 2^\circ$ the ratio v/u is small regardless of the c_{12} value. Consequently, x' for Crystal F was made close to 45° .

For x' in the (010) plane (Crystal D), the results were quite similar to the above case. The ratios v/u were all somewhat larger than shown in Figure 26; however, the mean value of v/u and the differences between v/u for c_{13} values of 0.5 and 2.0×10^{12} dynes/cm² were smallest for x' between 45 and 50° from [001].

For x' in the (100) plane (Crystal E), the v/u values were all quite large at $\theta = 45^\circ$ compared to the significantly small values at $\theta = 37^\circ$ from [001] for c_{23} values of 0.5 to 2.0×10^{12} dynes/cm².

It may be of interest here to note the actual v/u , once the c_{ij} were obtained. Since the measured value of c_{13} is considerably below the range of values considered in the above computations, the v/u ratio calculated for θ_D in Table I and the given value of c_{13} is somewhat larger than aimed for, and it gives a $7\frac{1}{2}^\circ$ deviation between the displacement and propagation directions for the longitudinal wave. This is to be compared with approximately 0° and $1\frac{1}{2}^\circ$ deviations in Crystals E and F, respectively. In view of the fact that no particular experimental difficulty was associated with Crystal D, the consideration given here in selecting the rotation angle seems doubtful in value.

XII. APPENDIX B

Algebraic Sign of the Cross-coupling Moduli

Equations (56), (60) and (64) of the text are quadratic in that there are two possible solutions for c_{12} , c_{13} and c_{23} , respectively, which satisfy the given equations. Use of the positive sign in all 3 equations gives the positive values; these have been accepted as correct for reasons to be given here. Using the negative sign, on the other hand, gives the following negative values:

$$c_{12} = -1.95 \times 10^{12} \text{ dynes/cm}^2$$

$$c_{13} = -1.69 \times 10^{12} \text{ dynes/cm}^2$$

$$c_{23} = -3.56 \times 10^{12} \text{ dynes/cm}^2$$

Elasticity theory in itself does not give a generally rigorous basis for solving the ambiguity. One may use the relationships between the elastic moduli which must be satisfied for a thermodynamically stable crystal;⁽³¹⁾ for orthorhombic symmetry, these are

$$c_{11}c_{22} > c_{12}^2; \quad c_{11}c_{33} > c_{13}^2; \quad c_{22}c_{33} > c_{23}^2$$

The relations involving c_{12} and c_{13} are satisfied in the present case by using either the positive or negative values, although only the positive value of c_{23} proves correct on this basis.

A more rigorous treatment, which can be used for solving all the ambiguities, is closely allied to the discussion of Appendix A, where it is shown that the use of the positive c_{ij} gives relatively small values for the angle of deviation between the directions of longitudinal wave propagation and corresponding directions of particle displacement. These same positive values give the expected large v/u ratios when the ρS^2 for the quasi-shear modes are used in equation (84). The possible negative values of the c_{ij} given above, result in totally different values of c'_{11} and c'_{16} which, when combined with the observed ρS^2 , give large values for v/u for quasi-longitudinal waves and relatively small values for the quasi-shear modes. In Crystal E, for example, the deviation between directions of propagation and particle displacement for the longitudinal mode becomes 72° , when the negative value of c_{23} is used; whereas, for the shear mode, it is only 18° . These latter computations, when combined with the experimental arrangement, would imply that the longitudinal transducer produced primarily the wave with quasi-shear mode velocity, whereas the shear transducer produced primarily the longitudinal wave. These implications are inconsistent with logic for a number of reasons, and, consequently, it must be assumed that the positive c_{ij} are correct.

XIII. APPENDIX C

Procedure and Results of Rotation Angle Measurement

A. Principle

The crystal is mounted on an X-ray diffractometer so that one face is parallel to the tangent of the focusing circle XPG (Figure 27) and a principal crystal axis, [100], [010] or [001], is normal to the plane of the spectrometer table. The position of G, the X-ray detector, is set to receive X rays diffracted from the crystal at an angle μ (where $\sin \mu = n\lambda/2d$)* corresponding to the d value for a given low-index crystal plane which is inclined 90° to the table and some unknown angle δ from the crystal face. Upon rotation about the vertical principal axis, P, of δ degrees, the crystal plane corresponding to the critical d value is moved into diffraction position and the Geiger counter indicates a maximum diffracted intensity (Figure 27b). At this point the position of an indicator at point O' with reference to a circular scale is recorded to the nearest 0.01° . The crystal is then rotated 180° about the horizontal axis O'P and then about the vertical axis, P, until the maximum diffracted intensity is again observed (Figures 27c and d). The magnitude of the last vertical rotation is 2δ , or twice the inclination of the low-index plane to the crystal face. Since the inclination of the low-index plane to the principal axis is well established from the lattice parameters, the angle between the crystal face and either of the 2 horizontal principal planes can be readily obtained.

B. Apparatus

The measurements were carried out on an XRD-5 G.E. spectrometer. A G.E. single-crystal orienter provided the means for rotating about the vertical axis, P, and it measured these rotations to the nearest 0.01° . A specially constructed piece was necessary for supporting the specimen with its faces perpendicular to the table and to provide the horizontal rotation axis, OP; the design of the stage is shown in Figure 28. The specimen, with the exposed face heavily electropolished to remove distortions due to lapping, was mounted on a carefully prepared surface perpendicular to the axis OP. The shaft which rotated the specimen about OP had a straight transverse hole drilled through it. The specimen was mounted so that the principal crystal axis, which was parallel to crystal face, was vertical and parallel to the hole in the shaft. The locating dowel maintained the vertical alignment of the hole. The stage was grooved in its bottom plane so that it could be moved along a track parallel to the focusing circle of the spectrometer.

*Because we have used θ to define the rotation angle, the Bragg angle is denoted here as μ .

C. Results

The procedure involved, first, setting the spectrometer to the proper diffraction angle 2μ (or 2θ in commonly used notation) which corresponded to a low-index plane that was inclined within 20° of the crystal face and, second, observing the diffracted intensity as the specimen was rotated about the vertical axis P. When the rotation angle was such as to give intensities much higher than the background, the relative intensities were recorded for each 0.02° increment, so as to establish the angular location of the maximum. As an example, the data for the (240) reflection from Crystal F, with rotation about the [001] direction, is given in Table XIII.

From examination of the data, the peak intensities were at 178.22° for the first setting and 178.81° for the second setting. The angle 2δ was then

$$178.81^\circ - 178.22^\circ = 0.59^\circ$$

and

$$\delta \simeq 18'$$

At this point, there is an apparent ambiguity in the direction of the inclination which is resolved from a knowledge of the directions of the [100] and [010] axes with respect to the geometry of each setting. From Laue photographs, it was known that the [100] axis for the first setting was oriented in a direction which could be described as a clockwise rotation about P from the normal to the crystal face. Since the rotation necessary to bring the (240) plane in reflecting position was counterclockwise, it is deduced that the (240) plane had lesser inclination to the (100) plane than the crystal face. The angle θ_F , which describes the inclination of the crystal face to the (100) plane, is therefore

$$44^\circ 13' + 18' = 44^\circ 31'$$

where $44^\circ 13'$ is the angle between (240) and (100).

The angle θ_F was also obtained from (130) and (220) reflections, giving values of $44^\circ 27'$ and $44^\circ 28'$, respectively. The cause of the differences is not easily apparent; however, the internal precision is quite adequate for the purpose of reducing the effect of orientation errors on the calculation of C_{12} .

A summary of the rotation angle measurements obtained from the various reflections for all of rotated cuts is given in Table XIV.

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TABLE I
Description of Uranium Single Crystal Plates

Crystal	Thickness (mm)	Approximate Lateral Dimensions (mm)	Inclination of Wave Propagation Direction to Principal Axes		
			[100]	[010]	[001]
A	2.9162	$5\frac{1}{2} \times 4\frac{1}{2}$	0°		
B	3.9185	ellipse - $4\frac{1}{2} \times 5$		0°	
C	2.7991	$6 \times 4\frac{1}{2}$			0°
D	2.6040	$4\frac{1}{2} \times 6$		90°	45°32'
E	2.9766	5×6	90°		36°
F	1.7960	ellipse 4×5	44°29'		90°
G	1.7168	ellipse 4×5	46°19'		90°

TABLE II

Data for Longitudinal Wave along [001] Axis (Crystal C) at 25°C

N	f_N (Mc/sec)	$\Delta f =$ $f_{N+1} - f_N$	Average Δf	$\frac{n + e}{f_N \Delta f \text{ avg.}}$	$S = 2f_N \frac{n}{n}$ (10^5 cm/sec)	
Sixth Harmonic						
0	9.481 x 6	0.1120 x 6	0.111504 x 6 = 0.66902	85 + 0.028	3.74655	
1	9.593	0.1115		86 + 0.032	3.74673	
2	9.7045	0.1115		87 + 0.032	3.74671	
3	9.816	0.1115		88 + 0.032	3.74670	
4	9.927	0.1110		89 + 0.028	3.74649	
5	10.039	0.1120		90 + 0.032	3.74666	
6	10.1505	0.1115		91 + 0.032	3.74665	
7	10.262	0.1115		92 + 0.032	3.74663	
8	10.3735	0.1115		93 + 0.032	3.74661	
9	10.4850	0.1115		94 + 0.032	3.74660	
10	10.596	0.1110		95 + 0.027	3.74641	
11	10.707	0.1110		96 + 0.023	3.74622	
12	10.819	0.1120		97 + 0.027	3.74638	
13	10.930	0.1110		98 + 0.023	3.74620	
14	11.0415	0.1115		99 + 0.023	3.74619	
15	11.153	0.1115		100 + 0.023	3.74618	
16	11.265	0.1120		101 + 0.028	3.74633	
17	11.376	0.1110		102 + 0.023	3.74616	
18	11.4875	0.1115		103 + 0.023	3.74615	
19	11.599	0.1115		104 + 0.023	3.74614	
20	11.710	0.1110		105 + 0.019	3.74597	
21	11.822	0.1120		106 + 0.023	3.74612	
22	11.934	0.1120		107 + 0.027	3.74627	
27	12.492	0.1116		112 + 0.023	3.74637	
Ninth Harmonic						
0	9.889 x 9	0.0740 x 9		0.074325 x 9 = 0.668925	133 + 0.05	3.74618
2	10.037	0.0740 x 9			135 + 0.04	3.74592
4	10.186	0.0745	137 + 0.05		3.74603	
6	10.334	0.0740	139 + 0.03		3.74577	
8	10.4835	0.0748	141 + 0.04		3.74608	
10	10.632	0.0743	143 + 0.05		3.74599	
12	10.7805	0.0743	145 + 0.05		3.74590	
14	10.929	0.0743	147 + 0.04		3.74585	
16	11.078	0.0745	149 + 0.05		3.74596	
18	11.2265	0.0743	151 + 0.05		3.74591	
20	11.375	0.0743	153 + 0.04		3.74583	
22	10.3715 x 10	0.0744 x 9	155 + 0.05		3.74589	
22	10.505 x 10	0.0742	157 + 0.04		3.74578	
Thirteenth Harmonic						
0	10.6026 x 13	0.0513 x 13	0.051520 x 13 = 0.66976		206 - 0.21	3.74573
2	10.705				208 - 0.22	3.74577
4	10.8085				210 - 0.21	3.74560
5	10.860				211 - 0.21	3.74565
6	10.911				212 - 0.22	3.74570
7	10.963	0.0520			213 - 0.21	3.74549
Fifteenth Harmonic						
0	9.456 x 15	0.0445 x 15	0.04462 x 15 = 0.66930		212 - 0.08	3.74599
2	9.545	0.0445 x 15		214 - 0.08	3.74554	
4	9.6345	0.0448		216 - 0.08	3.74558	
6	9.7235	0.0445		218 - 0.08	3.74537	
8	9.813	0.0448		220 - 0.08	3.74568	
9	9.858	0.0450		221 - 0.07	3.74570	
10	9.9015	0.0435		222 - 0.09	3.74522	
11	9.947	0.0455		223 - 0.07	3.74575	
12	9.991	0.0440		224 - 0.09	3.74552	
14	10.081	0.0450		225 - 0.07	3.74581	
Eighteenth Harmonic						
0	10.036 x 18			270 - 0.10	3.74558	

TABLE III

Limiting Velocities of Ultrasonic Waves in Alpha Uranium

Velocity Number	Crystal	Type Mode	Direction of Particle Motion	Velocity (cm/sec)
1	A	long	[100]	335,835
2	A	shear	[010]	197,585
3	A	shear	[001]	196,388
4	B	long	[010]	322,942
5	B	shear	[100]	197,587
6	B	shear	[001]	255,514
7	C	long	[001]	374,548
8	C	shear	[100]	196,350
9	C	shear	[010]	255,792
10	D	quasi-long	in (010) plane*	328,850
11	D	quasi-shear	in (010) plane**	238,085
12	D	shear	[010]	227,980
13	E	quasi-long	in (100) plane*	396,630
14	E	quasi-shear	in (100) plane**	189,565
15	E	shear	[100]	196,810
16	F	quasi-long	in (001) plane*	324,980
17	F	quasi-shear	in (001) plane**	204,890
18	F	shear	[001]	227,535
19	G	quasi-long	in (001) plane*	324,920
20	G	quasi-shear	in (001) plane**	205,040
21	G	shear	[001]	229,325

*Close to propagation direction (see Table I).

**Close to 90° from propagation direction.

See Section VI-A for note on uncertainties.

TABLE IV

Elastic Stiffness Moduli for Alpha Uranium as Directly Computed from the Velocities of Table III

Modulus	Crystal from which Derived	Equation for Derivation*	Value (10^{12} dynes/cm ²)	Estimated Probable Error (%)
C ₁₁	A	ρS_1^2 (subscript refers to velocity number in Table II)	2.1474	0.14
C ₂₂	B	ρS_4^2	1.9857	0.14
C ₃₃	C	ρS_7^2	2.6711	0.14
C ₄₄	B	ρS_6^2	1.2431	0.14
C ₄₄	C	ρS_9^2	1.2458	0.14
C ₅₅	A	ρS_3^2	0.7343	0.14
C ₅₅	C	ρS_8^2	0.7341	0.14
C ₆₆	A	ρS_2^2	0.7433	0.14
C ₆₆	B	ρX_5^2	0.7433	0.14
C ₁₂	F	$1/sc [(s^2 c_{22} + c^2 c_{66} - \rho S_{16}^2)(s^2 c_{66} + c^2 c_{11} - \rho S_{16}^2)]^{1/2} - c_{66}^{**}$	0.4631	1.36
C ₁₂	F	Using ρS_{17}^2 in above equation	0.4649	0.71
C ₁₂	G	Using ρS_{19}^2 in above equation	0.4718	1.36
C ₁₂	G	Using ρS_{20}^2 in above equation	0.463	0.71
C ₁₃	D	$1/sc [(s^2 c_{11} + c^2 c_{55} - \rho S_{10}^2)(s^2 c_{55} + c^2 c_{33} - \rho S_{10}^2)]^{1/2} - c_{55}$	0.2179	3.76
C ₁₃	D	Using ρS_{11}^2 in above equation	0.2185	1.82
C ₂₃	E	$1/sc [(s^2 c_{22} + c^2 c_{44} - \rho S_{13}^2)(s^2 c_{44} + c^2 c_{33} - \rho S_{13}^2)]^{1/2} - c_{44}$	1.0795	0.95
C ₂₃	E	Using ρS_{14}^2 in above equation	1.0748	0.40

*Density, $\rho = 19.04$ gm/cm³**s = sin θ , c = cos θ , where θ is the angle of rotation for each crystal listed in Table I.

TABLE V

Weighted Mean Values for
Stiffness Moduli in
Alpha Uranium

Modulus	Value (10^{12} dynes/cm ²)
c ₁₁	$2.1474 \pm 0.14\%$ *
c ₂₂	$1.9857 \pm 0.14\%$
c ₃₃	$2.6711 \pm 0.14\%$
c ₄₄	$1.2444 \pm 0.10\%$
c ₅₅	$0.7342 \pm 0.10\%$
c ₆₆	$0.7433 \pm 0.10\%$
c ₁₂	$0.4646 \pm 0.58\%$
c ₁₃	$0.2183 \pm 1.50\%$
c ₂₃	$1.0755 \pm 0.34\%$

*Given to 4 decimal
places for comparison
with Table VIII.

TABLE VI

Estimated Maximum Errors in
Variables Affecting Velocity
Computation

Error	Value (%)
Frequency measurement (f_n)	0.02
Thickness (t)	0.035
Reflection phase shift (γ)	0.02
Dispersion	0.01
Imperfections in crystals	0.01
Total	0.095
Mean Square	~ 0.05

TABLE VII

Equations for Cross Checking

Equation	Difference between Calculated and Observed ρS^2 Values (%)
$\rho S_{12}^2 = c_{66} + \cos^2\theta_D (c_{44} - c_{66})$	0.04
$\rho S_{15}^2 = c_{66} + \cos^2\theta_E (c_{55} - c_{66})$	0.01
$\rho S_{18}^2 = c_{44} + \cos^2\theta_F (c_{55} - c_{44})$	0.11
$\rho S_{21}^2 = c_{44} + \cos^2\theta_G (c_{55} - c_{44})$ *	0.03
$\rho (S_{10}^2 + S_{11}^2) = c_{11} + c_{55} + \cos^2\theta_D (c_{33} - c_{11})$	0.01
$\rho (S_{13}^2 + S_{14}^2) = c_{22} + c_{44} + \cos^2\theta_E (c_{33} - c_{22})$	0.06
$\rho (S_{16}^2 + S_{17}^2) = c_{22} + c_{66} + \cos^2\theta_F (c_{11} - c_{22})$	0.04
$\rho (S_{19}^2 + S_{20}^2) = c_{22} + c_{66} + \cos^2\theta_G (c_{11} - c_{22})$ *	0.16

*Equations which were not used for indirect derivation of
diagonal moduli.

TABLE VIII
Moduli Determined from Data of Crystals D, E and F

Modulus	Value (10^{12} dynes/cm ²)	Calculated Probable Error (%)	Deviation from Values Given in Table VI (%)
C ₁₁	2.1448*	0.45	-0.12
C ₂₂	1.9868	0.58	+0.05
C ₃₃	2.6712	0.30	+0.00
C ₄₄	1.2456	0.20	+0.10
C ₅₅	0.7348	0.25	+0.08
C ₆₆	0.7424	0.39	-0.12
C ₁₂	0.4638	>1	-0.19
C ₁₃	0.2174	>1	-0.41
C ₂₃	1.0747	>1	-0.08

*Given to 4 decimal places for comparison with Table V.

TABLE IX

Comparison of Observed and Calculated Rotation Angles, θ

Crystal	Reference Axis: θ is Angle between Propagation Direction and	Value Indicated by Spectrometer	Value Indicated by Laue Patterns	Calculated Values	
				Shear Mode Velocity	Coupled Mode Velocity
D	[001]	45°32' $\pm$ 4'	45°50'	45°29' $\pm$ 10'	45°34' $\pm$ 30'
E	[001]	36°7' $\pm$ 4'	36°	39° $\pm$ 6°	35°55' $\pm$ 28'
F	[100]	44°29' $\pm$ 4'	44°40'	44°36' $\pm$ 9'	45°55' $\pm$ 1 $\frac{1}{4}$ °
G	[100]	46°19' $\pm$ 4'	45°46'	46°17' $\pm$ 9'	44°55' $\pm$ 1 $\frac{1}{4}$ °

TABLE X

Compliance Moduli Derived
from Weighted Mean Values
of Stiffness Moduli

Modulus	Value (10^{-12} cm ² /dynes)
s_{11}	0.491
s_{22}	0.673
s_{33}	0.479
s_{44}	0.804
s_{55}	1.362
s_{66}	1.345
s_{12}	-0.119
s_{13}	+0.008
s_{23}	-0.261

TABLE XI

Elastic Parameters

Parameter	Relation to Moduli	Value	General Definition
E_{100}	$1/s_{11}$	2.038×10^{12} dynes/cm ²	Young's Modulus
E_{010}	$1/s_{22}$	1.483×10^{12} dynes/cm ²	
E_{001}	$1/s_{33}$	2.084×10^{12} dynes/cm ²	
σ_{12}	$-s_{12}/s_{11}$	+0.243	$\frac{\text{Extensional strain in } [100]}{\text{Compressional strain in } [010]}$
σ_{21}		+0.177	
σ_{13}		-0.017	Linear compressibility in [100]
σ_{31}		-0.017	
σ_{23}		+0.390	
σ_{32}		+0.548	Volume Compressibility Bulk Modulus
β_{100}	$s_{11} + s_{12} + s_{13}$	0.380×10^{-12} cm ² /dyne	
β_{010}	$s_{22} + s_{12} + s_{23}$	0.292×10^{-12} cm ² /dyne	
β_{001}	$s_{33} + s_{13} + s_{23}$	0.225×10^{-12} cm ² /dyne	
β_V	$\beta_{100} + \beta_{010} + \beta_{001}$	0.897×10^{-12} cm ² /dyne	
K	$1/\beta_V$	1.115×10^{12} dynes/cm ²	

TABLE XII

Maximum and Minimum Rigidity Moduli, G, for Various Crystal Planes in Alpha Uranium

Crystal Plane (Miller Index or Rotation Angle*)	Rigidity Modulus (10 ¹² dynes/cm ²)	Direction of Extremes		Crystal Plane (Miller Index or Rotation Angle*)	Rigidity Modulus (10 ¹² dynes/cm ²)	Direction of Extremes	
		Max	Min			Max	Min
100 ($\psi = 0$) $\theta = 7^\circ 10'$ 101 102 $\theta = 53\frac{1}{2}^\circ$ 103 106 001 ($\theta = 90$) $\psi = 8^\circ, \theta = 0$	0.743 0.748 0.947 1.035 1.007 1.065 1.191 1.244	010 isotropic isotropic 010 010 010 010 010	001	1.012 1.000 1.145 1.064 1.000 1.125 1.500 1.695	1.012 1.000 1.145 1.064 1.000 1.125 1.500 1.695	isotropic isotropic isotropic isotropic isotropic isotropic isotropic isotropic	0.741 0.741 0.741 0.741 0.741 0.741 0.741 0.741
210 ($\psi = 13.7^\circ$) 211 212 213 214	0.751 0.859 1.004 1.083 1.068	0.737 0.718 0.754 0.820 0.892	001 324 -30° to 101 -6° to 513 -25° to 201	1.019 1.196 1.331 1.320 1.197	1.019 1.196 1.331 1.320 1.197	120 7° to 011 -6° to 021 142 121	0.751 0.859 1.004 1.083 1.068
110 ($\psi = 26^\circ$) 221 111 223 113	0.796 0.913 1.056 1.108 0.986	0.725 0.680 0.678 0.718 0.772	001 212 312 412 512 31° to 110	1.098 1.343 1.558 1.543 1.397 1.132	1.098 1.343 1.558 1.543 1.397 1.132	6° to 122 143 032 153 512 -58° to 110	0.796 0.913 1.056 1.108 0.986
120 ($\psi = 44.2^\circ$) 121 243 122 123 125	0.927 1.051 1.063 1.031 0.941 0.900	0.714 0.628 0.617 0.632 0.703 0.813	001 614 47° to 210 -42° to 210 12° to 011 10° to 511 12° to 131	1.298 1.673 1.723 1.631 1.339 1.107	1.298 1.673 1.723 1.631 1.339 1.107	210 4° to 111 -42° to 210 12° to 011 032 16° to 501	0.927 1.051 1.063 1.031 0.941 0.900

TABLE XIII

Relative Diffracted X-ray Intensity of (240) Reflection
versus Angle of Rotation about [001] Axis in
Crystal F

Crystal Setting	Rotation Indi- cator Reading, degrees	Relative Intensity of Diffracted Beam, counts per 10 sec
First	178.15 178.18 178.20 178.22 178.24 178.26 178.28 178.30 178.32 178.34	1337 1374 1390 1396 1393 1380 1367 1352 1320 1282
180° about horizontal axis, OP, from first setting	178.71 178.73 178.75 178.77 178.79 178.81 178.83 178.85 178.87 178.89	3887 3971 4016 4054 4089 4098 4071 4043 3994 3920

TABLE XIV

Results of Rotation Angle Measurements
by X-ray Spectrometer Method

Crystal D

Reflection	δ	Angle between Reflecting Plane and (001)	θ_D
204	4°36'	40°58'	45°34'
202	-14°36'	60°04'	45°28'

Crystal E

Reflection	δ	Angle between Reflecting Plane and (001)	θ_E
023	6°44'	29°23'	36°07'
022	-4°03'	40°10'	36°07'
043	-12°20'	48°24'	36°04'

Crystal F

Reflection	δ	Angle between Reflecting Plane and (100)	θ_F
240	0°18'	44°13'	44°31'
130	-11°08'	55°35'	44°27'
220	18°32'	25°56'	44°28'

Crystal G

Reflection	δ	Angle between Reflecting Plane and (100)	θ_G
240	2°05'	44°13'	46°18'
130	-9°16'	55°35'	46°19'
220	20°25'	25°56'	46°21'

Figure 1

Measuring Circuit Block Diagram

Figure 2

Sketch of Buffer Unit Used for Phase Comparison Method

Figure 3

Schematic Analysis of Phase Relations between Successive Echoes in Buffer Rod Unit. (a) Incident Wave (E_i), First Reflection (E_b) and Transmitted Waves (E_t, E_t'). (b) Vector Diagram when Phase Shift in Specimen is $2\pi n$ Radians.

Figure 4

Curves for Evaluating the Reflection Phase Angle, γ , from Detected Signal Amplitudes, for Uranium Specimen. Impedances, Z , are given in units of mechanical ohms per cm^2 .

Figure 5

Scheme of Reference Axes Used for Equations (33). X' is normal to faces of Crystal A.

Figure 6

Scheme of Reference Axes Used for Equations (47). X' is normal to faces of Crystal F.

Figure 7

Scheme of Reference Axes Used for Equations (58). X' is normal to faces of Crystal D.

Figure 8

Scheme of Reference Axes Used for Equations (62). X' is normal to faces of Crystal E.

22,811

Figure 9

Arrangement on X-ray Machine for Orienting Crystal Relative to Goniometer Base.

Figure 10
Fixture for Holding Goniometer during Mounting of Crystal

22,812

Figure 11
Steel Mount and Table in Preparation for Pouring of Resin

22,814

Figure 12
Schematic Sketch of Manually
Operated Lapping Machine

22,819

22,820

22,821

Figure 13
Crystals A, E and F, which Show Typical Specimen Shapes

Figure 14

Wave Velocity versus Frequency for the Three Wave Modes in Crystal A. Curves are based on Equation (65).

Figure 15

Wave Velocity versus Frequency for the Three Wave Modes in Crystal B. Curves are based on Equation (65).

Figure 16

Wave Velocity versus Frequency for the Three Wave Modes in Crystal C. Curves are based on Equation (65).

Figure 17

Wave Velocity versus Frequency for the Three Wave Modes in Crystal D. Curves are based on Equation (65).

Figure 18

Wave Velocity versus Frequency for the Three Wave Modes in Crystal E. Curves are based on Equation (65).

Figure 19

Wave Velocity versus Frequency for the Three Wave Modes in Crystal F. Curves are based on Equation (65)

Figure 20

Wave Velocity versus Frequency for the Three Wave Modes in Crystal G. Curves are based on Equation (65)

Figure 21

Variation of c'_{11} and E in Principal Planes of Alpha Uranium. a) (100) plane, b) (010) plane and c) (001) plane

Figure 22

Variation of c_{11} , (a) and E, (b), among Crystallographic Directions in Alpha Uranium. Curves are iso-stiffness contours plotted on quadrant of (001) standard projection. Numbers are units of 10^{12} dynes/cm².

Figure 23

Crystal Structure of Alpha Uranium at 25°C.

Figure 24

Transformations Used for Systematically Computing Anisotropy of Shear Moduli. First rotation, ψ , is about z ; second, θ , is about y' ; third, ϕ is about x'' .

Figure 25

Variation among Crystal Planes of
 (a) Maximum Rigidity Modulus for
 the Plane, $G_{\max}$, (b) Minimum Rigidity
 Modulus for the Plane, $G_{\min}$, and
 (c) Anisotropy in Plane, $G_{\max}/G_{\min}$.
 Curves show distribution of indicated
 modulus values in units of 10^{12} dynes/
 cm^2 among the planes as they would
 appear on (001) standard projection.

Figure 26

Ratio of Displacement Components in y' to
 x' Directions, v/u , for Propagation of Quasi-
 Longitudinal Wave in x' Direction in (001)
 Plane, where x' is θ degrees from [100]
 Direction. Curves are for 3 different
 assumed values of c_{12} . $v/u = 0$ for pure
 longitudinal wave.

Figure 27

Steps in the Procedure for Measuring Rotation Angles, θ , for Crystals D, E, F and G on X-ray Diffractometer. (A) Crystal is set up tangent to circle formed by X-ray source (X), center (P) and counter (G), with principal rotation axis normal to table. (B) Crystal is rotated about P through angle δ to bring plane with Bragg angle, μ , tangent to circle. (C) Crystal is then rotated 180° about O'P. (D) Crystal is again rotated about P through angle 2δ to bring reflecting plane tangent to circle.

Figure 28

Sketch of Stage Used for Supporting and Rotating Crystal during Rotation Angle Measurement.