Reprinted from: The Journal of the Acoustical Society of America

10.6 **Received 16 April 1971**

Determination of Third- and Fourth-Order Longitudinal Elastic Constants by Shock Compression Techniques—Application to Sapphire and Fused Quartz*

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A number of solids sustain large elastic compressions under shock-wave loading. In these solids, measurements of the stress and compression in the direction of shock propagation can be used to calculate both third- and fourth-order longitudinal elastic constants if measurements are carried out over a wide range of compressions. Only limited measurements of fourth-order constants have been previously determined by other techniques. Determinations of third-order constants under these large elastic compressions afford the opportunity to test the applicability of the finite-strain formulation of constitutive relations. A general method for calculating these third- and fourth-order constants is presented and applied to shock compression data for sapphire and fused quartz. For sapphire, it is found that $C_{111}\approx C_{333}=-(3.3\pm0.3)\times10^4$ kbar and $C_{1111}\approx C_{3333}=+(5.0\pm1.5)\times10^5$ kbar. For fused quartz, it is found that $C_{111}=+(5.5\pm0.1)\times10^3$ kbar and $C_{1111}=+(110\pm10)\times10^3$ kbar. The technique and method of analysis seem generally applicable to solids that exhibit elastic limits of a few percent of their longitudinal elastic constants.

INTRODUCTION

When subjected to shock-wave compression, a number of solids are observed to exhibit unusually large elastic limits. Noting that large elastic compressions can be achieved under shock compression, Fowles[1] expressed the finite-strain high-order elastic constant theory in terms suitable for analysis of elastic shock-compression data. He proposed that longitudinal fourth-order elastic constants could be computed from shock-compression data if the second- and third-order elastic constants were known. From this analysis, the longitudinal fourth-order elastic constants of α quartz were computed from the shock-compression data in the elastic range, i.e., below the Hugoniot elastic limit. The present paper extends the analysis of shock-compression data to the determination of both third- and fourth-order longitudinal elastic constants.

A number of measurements of third-order elastic constants have been accomplished with static compression techniques, including measurements on Ge,[2–6] MgO,[2] Si,[4,6] fused quartz,[2] α quartz,[7] and sapphire.[8] Measurements of these third-order constants are of both fundamental and applied interest. The third-order constants are associated with anharmonicity of a crystal lattice; hence, they may be used to calculate generalized Grüneisen parameters.[9] Furthermore, quantitative descriptions of acoustic amplification at microwave frequencies in solids[10,11] requires knowledge of the third-order elastic constants. If piezoelectric solids are used for amplification, high-order piezoelectric constants are also important.[12–14] The attenuation in microwave delay lines is influenced by the Akhiezer phonon–phonon interaction mechanism,[15] which can be calculated from the third-order elastic constants.

Only a limited number of fourth-order elastic constant measurements have been accomplished and there is no established technique for their determination. In addition to Fowles's measurements, fourth-order constants of several cesium halides have been measured by ultrasonic techniques,[16] and several combinations of fourth-order constants of fused quartz have been determined in uniaxial tension experiments.[17] Fourth-order constants have not yet been required for interpretation of microwave phenomena, but it has been suggested that harmonic generation in stressed crystals could be used to determine fourth-order constants.[18]

Although, at present, only longitudinal elastic constants can be determined from shock-compression measurements, it appears that these longitudinal constants are often of interest. Furthermore, the determination of the third-order constant under large compressions permits a test of the formulation of the finite-strain theory.

Synthetic single-crystal Al_2O_3, i.e., sapphire, exhibits a Hugoniot elastic limit as high as 210 kbar. Hence, high-order longitudinal elastic constants for sapphire can be determined from the shock-compression data. Although third-order constants for sapphire were calculated from shock-compression data in a recent paper,[19] it appears that a more extensive analysis of the data will permit calculation of both third- and fourth-order constants. Furthermore, recent data on fused quartz reported by Barker and Hollenbach[20] permit similar calculations for fused quartz to 38 kbar.

It is the object of this paper to develop a general method for determining third- and fourth-order longitudinal elastic constants from elastic shock-compression data and to use this method to determine the values of these constants for sapphire and fused quartz from the shock-compression data of Graham and Brooks[19] and Barker and Hollenbach.[20] Although the theory expressed by Fowles[1] is used without modification, the method of data analysis is modified so that both third- and fourth-order constants can be determined. The method seems generally applicable to solids that exhibit elastic limits of a few percent of their longitudinal elastic constants.

The paper is organized in the following way: Following the expression of the shock-compression relations and comments on experimental capability, the stress–volume relations are expressed in terms of high-order elastic constants; the method of relating the experimental observations to the high-order constants are then discussed and applied to the shock-compression data; the constants are then compared to similar values obtained ultrasonically from static compression experiments; in conclusion, the general applicability of the method to other solids is discussed.

I. BACKGROUND

A. Shock-Compression Experiments

The most common technique for determining third-order constants involves measurements of the change in transit time of an ultrasonic wave when a solid is subjected to hydrostatic pressures up to about 10 kbar or uniaxial stresses up to about 2 kbar.[8,21] Second-harmonic generation at microwave frequencies has also been used to determine third-order longitudinal constants.[11] Although shock-compression measurements are most frequently accomplished at pressures greater than 100 kbar,[22] techniques utilizing the planar impact of samples with flat-faced projectiles permit shock-compression measurements at stresses as low as 1 kbar.[23,24] When the impact experiment is employed, stresses can be applied to samples on a continuously increasing amplitude scale up to hundreds of kilobars. Instrumentation has been developed that is capable of determining small compression changes and numerous Hugoniot elastic limits have been measured.[25] These observations reveal that a number of solids undergo large compressions while remaining elastic; in particular, Z-cut α quartz has a Hugoniot elastic limit of 150 kbar and X-cut α quartz has a value of 60 kbar, while Z-cut sapphire has a value of 210 kbar and X-cut sapphire has a value of 150 kbar. The possibility of determining high-order constants from shock-compression experiments is a result of the large elastic compressions that some solids exhibit, and the experimental capability for routinely measuring shock compressions throughout the elastic compression range.

When a solid is rapidly compressed ($<10^{-8}$ sec) over a large planar area, the inertial response of the sample produces a well-defined state of one-dimensional strain in the direction of shock propagation.[22] Measurements of shock-wave velocity and the particle velocity behind the shock front are accomplished in a region of the sample that is far enough removed from the boundaries such that unloading from boundaries does not occur in the time of interest to the experiment.

Assuming that materials respond elastically, that the shock profiles are steady in time and one dimensional, and that the shock front moves into an unstressed medium at rest, conservation of momentum[22] results in the equation

$$\sigma_x = \rho_0 U u, \tag{1}$$

where σ_x is the component of stress in the shock propagation direction taken along the x axis, ρ_0 is the initial density, U is the shock-wave velocity, and u is the particle velocity imparted by the shock front. In the low stress limit, $U=(C_{xx}/\rho_0)^{\frac{1}{2}}$, where C_{xx} is the adiabatic second-order longitudinal elastic constant for compression along the direction of wave propagation.

From the conservation of mass, it can be shown that[22]

$$\eta = \Delta V/V_0 = u/U, \tag{2}$$

where η is the linear compression, ΔV is the change in specific volume imparted by the shock front, and V_0 is the original specific volume. From these relations, it is apparent that measurements of shock velocity for various particle velocities over a wide range of compressions, up to the Hugoniot elastic limit, permit determination of the elastic-longitudinal-stress versus compression relation. In the elastic range, the stress configuration is well known and the compressions are isentropic to a close approximation.

Various experimental configurations can be utilized to determine the σ_x, η relation. When a projectile impact experiment is utilized, the most effective experimental procedure is to achieve the symmetric impact of the sample with a projectile facing of the same material. Under these conditions, the particle velocity impacted to the sample is exactly one-half the measured projectile impact velocity. The projectile impact velocity can be routinely measured to $\pm 0.1\%$.[26] If explosive loading techniques are utilized, measurements of free surface velocities give a measure of particle velocity to about $\approx 3\%$.

The shock-wave propagation velocity through the sample may be detected by optical techniques,[27] Sandia quartz gauge techniques,[28] charged pins,[29] or electrical response measurements from the shock-loaded samples.[23] An accuracy of about $\pm 1\%$ is normally achieved. For impact experiments utilizing impacts produced by a quartz gauge[28] and shock arrival times indicated by quartz gauges, shock-velocity accuracies of $\pm 0.5\%$ are achieved. Shock-velocity measurements with the Sandia velocity interferometer[30] can achieve accuracies of a few tenths of 1%. These experimental capabilities indicate that the best precision of shock-velocity measurements approach those achieved ultrasonically. However, the nominal accuracy of $\pm 1\%$ achieved in most shock experiments is suitable to establish accurate (2%–6%) high-order constants when large compressions are achieved.

B. High-Order Elastic Constants

Fowles[1] has expressed the finite elastic strain theory, as developed by Thurston,[31] in terms of the one-dimensional compressions achieved in the shock-compression experiment. An expansion of strain energy in a power series in finite strain, P_x, gives the result that:

$$\sigma_x=(V/V_0)[C_{xx}P_x+\tfrac{1}{2}C_{xxx}P_x^2+\tfrac{1}{6}C_{xxxx}P_x^3], \quad (3)$$

when the expansion is arbitrarily terminated at the cubic term. In Eq. 3, σ_x is the longitudinal component of stress in the shock propagation direction taken to be the x axis, C_{xx} is the usual adiabatic second-order elastic constant for uniaxial compression in the shock direction, C_{xxx} is the longitudinal third-order elastic constant, C_{xxxx} is the longitudinal fourth-order elastic constant, and $P_x\equiv\eta[(\eta/2)-1]$.

The shock-compression experiments provide values for σ_x and η that can be used in Eq. 3 to evaluate those longitudinal elastic constants that contribute significantly to the magnitude of the stress. The magnitude of the contribution of a given constant depends very strongly upon the magnitude of the compression; high-order contributions at typical static compressions of less than 0.1% are negligible, whereas high-order contributions for typical elastic shock compressions of a few percent are pronounced. It is apparent from Eqs. 1–3 that the shock-compression measurements determine only those longitudinal constants in the direction of shock propagation.

Because of the somewhat arbitrary definition of strain utilized in Thurston's finite strain development, it is possible that alternate formulations[32,33] may give a better representation to compression data. In any event, however, it is useful to determine the extent to which the data can be represented by these constants.

Unlike the static compression measurements that provide isothermal derivatives of adiabatic constants, the elastic shock compression measurements provide adiabatic derivatives of adiabatic constants. However, the difference between the two thermodynamic conditions is not significant for the present accuracies achieved in the third-order constant measurements.

II. DATA ANALYSIS

Examination of Eq. 3 indicates that a single σ_x, η measurement cannot be used to calculate a high-order constant unless all lower-order constants are known. Even though a single experiment cannot distinguish between various high-order elastic constants, the best fit to data obtained over a wide range of compressions must accommodate the various high-order contributions that are significant in different compression ranges. The principal contribution to the stress in the elastic compression range is the second-order constant, which is frequently known to a precision of $\pm 0.1\%$. Thus, it is appropriate to assume that the second-order contribution can always be calculated and proceed to determine third- and fourth-order constants.

At compressions of less than a few tenths of 1%, the third- and fourth-order contribution to the magnitude of the stress is too small to influence the data. As compressions become larger, third-order contributions become significant, whereas the fourth-order contribution is too small to influence the data. If elastic compressions of a few percent are achieved, the fourth-order contributions may become significant.

Although a cubic polynomial can be fit to the $\sigma_x V_0/V$ vs P_x relation, the iterative procedure given below has the feature of taking full advantage of the precisely known second-order constant before proceeding to successively less well characterized constants. Consider calculating an elastic constant, C_{xxx}^+, representing all contributions greater than second order, from observed σ_x vs η data. In this case, it follows from Eq. 3 that

$$C_{xxx}^+\equiv(2/P_x^2)[\sigma_x V_0/V-C_{xx}P_x], \quad (4)$$

where C_{xx} is taken to be the value obtained from ultrasonic measurements. If C_{xxxx} is zero, or the contribution of $C_{xxxx}P^3$ is negligible at the compression in question, $C_{xxx}^+=C_{xxx}$ and the third-order constant is determined. In all cases, however, C_{xxx}^+ can serve as the first approximation to C_{xxx}. Based on the shock-compression data, an iterative procedure can then be followed to give the best set of C_{xxx} and C_{xxxx} constants over the entire compression range. If data exist over a sufficiently large pressure range, the high-order longitudinal constants can be independently determined.

The iterative procedure follows a perturbation technique as follows:

(1) Calculate C_{xxx}^+ from Eq. 4 for all data.

(2) Compute a σ_x vs η relation from C_{xxx}^+ and C_{xx} and compare it to the observed σ_x vs η data.

(3) If C_{xxx}^+ gives a good fit to the data over the entire compression range, C_{xxxx} has a smaller value than can be observed and $C_{xxx}^+=C_{xxx}$.

TABLE I. Comparison of high-order elastic constants of sapphire.[a]

Method	Present work,[b] shock	Present work,[c] shock	Hankey *et al.*,[d] static ultrasonic	Gieske,[e] static ultrasonic	Carr,[f] 4.2°K, microwave
Maximum strain	4.0%	2.2%	0.1%	0.5%	$\sim 10^{-7}$
C_{111}	-3.3 ± 0.3	···	-3.87 ± 0.07	-3.9 ± 0.03	-3.8 ± 0.3
C_{333}	-3.3 ± 0.3	-3.25 ± 0.1	-3.34 ± 0.1	-3.1 ± 0.03	-2.1 ± 0.1
C_{1111}	50 ± 15	···	···	···	···
C_{3333}	50 ± 15	···	···	···	···

[a] Units are 10^4 kbar, temperatures are 25°C except for the work of Carr.
[b] Data from Ref. 19.
[c] Data from Ref. 20.
[d] Ref. 8.
[e] Ref. 36.
[f] Ref. 11.

(4) If systematic differences are observed, the C_{xxx}^+ value includes contributions from C_{xxxx}. Compute a value of C_{xxxx} from C_{xxx}^+ used in Eq. 3.

(5) Iterate with assumed values of C_{xxx} and C_{xxxx} until a good fit is obtained over the entire compression range.

(6) Express errors in the constants in terms of the range of values of constants which give an equally good fit to the observed experimental data.

For successful application of this technique, shock-compression data are required over a large compression range. Unfortunately, most elastic shock-compression data are limited to Hugoniot elastic limit points; however, the sapphire data of Graham and Brooks[19] extend from 0.3% to 4% compression and the sapphire data of Barker and Hollenbach[20] extend from 0.5% to 2.2%, while their fused quartz data extend from 0.2% to 10%. These data are used to compute third- and fourth-order longitudinal elastic constants for sapphire and fused quartz in Secs. III-A and III-B.

III. RESULTS

A. Sapphire

Before proceeding with detailed analysis of the sapphire data, several general features of the elastic shock-compression data on sapphire of Graham and Brooks[19] should be noted. The data were obtained from experiments in three crystallographic orientations, 0°, 90°, and 60°. The 0° and 90° orientations are "specific" crystallographic directions and purely longitudinal compression modes are achieved.[34] The C_{11} and C_{33} values differ by only 0.9%.[35] Thus, even though sapphire has trigonal symmetry, the longitudinal elastic constants do not vary significantly with orientation. This is an important consideration, since a number of experiments were also performed on natural-growth 60°-orientation samples. These samples are more readily available in large diameters required for shock-compression experiments. Since this orientation is not a specific direction, these samples would not be expected to exhibit pure longitudinal motion. However, since the longitudinal constants do not vary strongly with orientation, it might be expected that the nonlongitudinal contribution is small. A special experiment reported in Ref. 19 found that these 60° samples responded in a purely longitudinal mode within the experimental error. The data have the feature that, within the observed experimental scatter, all data from the three orientations fall on a common compression curve. Thus, to a good approximation, the high-order constants are independent of orientation for the three orientations investigated.

To analyze the data of Graham and Brooks, the stress-versus-compression data are smoothed by fitting to a quadratic polynomial with no restraints applied to the data. Following this, a mean value of $C_{xx}=4990$ kbar is chosen for all orientations[35] such that the second-order contribution is computed from the ultrasonic data. The analysis shows that $C_{xxx}^+=-3.6\times10^4$ kbar but that a good fit is not obtained over the entire compression range. Upon iteration, the high-order longitudinal constants are found to be $C_{111}\approx C_{333}=-(3.3\pm0.3)\times10^4$ kbar and $C_{1111}\approx C_{3333}=(5\pm1.5)\times10^5$ kbar. The errors are established by observing the range of values that give an equally good fit to the data within one standard deviation of the raw fit to the data. Because the second-order contribution dominates at smaller compressions, the high-order constants are determined by data in the range from 100 to 200 kbar.

The sapphire data of Barker and Hollenbach were obtained on 0° samples. Even though their shock-velocity measurements are more precise than those of Graham and Brooks, the compression range that they utilized is too small for significant contributions from the fourth-order constants. Analysis of their data gives: $C_{333}^+=C_{333}=-(3.25\pm0.1)\times10^4$ kbar.

The data are compared to third-order constants as determined by static stress and microwave techniques in Table I. The data at 25°C from all sources are found to be in reasonable agreement. The fourth-order constants of sapphire are obtained for the first time. It appears that the accuracy of the third-order constants obtained from the shock-compression data is comparable with that achieved in the static experiments. The low-temperature microwave second-harmonic-generation measurements of Carr[11] cannot be directly compared to

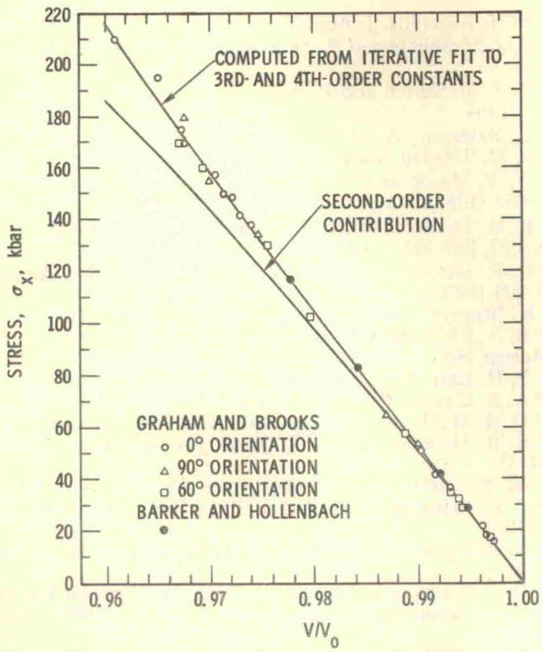

FIG. 1. The experimental shock-compression data for sapphire are plotted for comparison to the stress–volume relation computed from the third- and fourth-order elastic constants as determined in the text. The second-order constant contribution is taken from ultrasonic measurements of Gieske and Barsch.[35]

the room-temperature data but are included for completeness. The individual datum points of the various shock-compression experiments are compared to the stress–volume relation calculated from the high-order constants as computed from the data of Graham and Brooks in Fig. 1.

In analyzing the shock-compression data, it is interesting to note the compression ranges over which the various elastic constants made significant contributions to the stress amplitude. The second-order longitudinal constants give a good description to the observed stresses to compressions up to about 1%, whereas second- and third-order longitudinal constants give a good description to the observed stresses to compressions up to about 2%. The fourth-order contribution must be utilized to give a good description to the observed data from 2% to 4% compression. These observations emphasize the need for continuous data over a wide compression range if all the higher-order contributions are to be determined from shock-compression data.

B. Fused Quartz

Shock-compression studies of fused quartz have been accomplished from about 1 kbar to 65 kbar by Barker and Hollenbach[20] and up to 620 kbar by Wackerle.[37] At low pressures, the shock data show, in agreement with static compressibility measurements,[38] an increasing compressibility with pressure. This is contrary to the normal behavior of most solids and makes the Hugoniot elastic limit difficult to define. By employing both stress loading and stress unloading, Barker and Hollenbach found that fused quartz responds as an elastic solid to 38 kbar and, to a good approximation, the samples seemed elastic to 65 kbar.

Although the second-order constants of fused quartz have been measured by several investigators,[2,39] these constants are known to depend strongly on density.[7] Since they have not been measured on the fused quartz used in the shock-compression investigation, $\rho_0=2.201$ $g\cdot cm^{-3}$, the velocity of the leading edge of the stress pulse is used to determine the second-order constant. With the resulting value of $C_{11}=774.0$ kbar, the iterative procedure applied to the fused-quartz data to 38 kbar gives $C_{111}=+(5.5\pm0.1)\times10^3$ kbar and $C_{1111}=+(110\pm10)\times10^3$ kbar. The constants are compared to the ultrasonic data of Bogardus[2] in Table II.

The shock data above 38 kbar illustrate one particular problem with high-order constant measurements at large compressions. At stresses greater than 38 kbar, the bulk modulus begins to increase with pressure in the normal manner. This corresponds to the behavior observed by Bridgman[38] under static compression at 20 kbar since both the static and shock-compression results show that the change in bulk modulus begins to occur at a compression of 6%. Anderson and Dienes[40] have pointed out that a 6% volume compression would compress fused quartz to the volume of cristobalite, a well-known phase of SiO_2. Roy and Cohen[41] have observed a permanent densification and change in refractive index of fused silica for static pressures greater than 20 kbar. There is also evidence for a change in the strain dependence of the refractive index above 38 kbar from the shock data.[20] Thus, because there is evidence for a structural change at a volume compression of 6%, it appears that the high-order constant analysis should not be extended beyond 38 kbar.

The results indicate that the shock-compression technique yields a more accurate third-order constant than has heretofore been available. The fourth-order constant has been determined for the first time.

For the fused-quartz data, it is again instructive to consider the compression ranges over which the various-order elastic constants contribute significantly to the stress. The second-order constant gives a good fit up to

TABLE II. Comparison of high-order elastic constants of fused quartz.[a]

Method	Present work,[b] shock	Bogardus,[c] static ultrasonic
Maximum strain	6%	~0.2%
C_{111}	5.5± 0.1	5.3±0.4
C_{1111}	110.0±10.0	...

[a] Units are 10^3 kbar. Temperatures are 25°C.
[b] Data from Ref. 20.
[c] Ref. 2.

compressions of 0.5%. Between 0.5% and 2.5%, the third-order constant gives a significant contribution, while the fourth-order constant gives a significant contribution for compressions from 2.5% to 6%.

IV. CONCLUSION

Analysis of the sapphire and fused-quartz shock-compression data demonstrate that the analytical method employed permits the determination of both third- and fourth-order longitudinal elastic constants. Although the third-order constants may be determined by other techniques, the fourth-order constants have been determined only by shock-compression techniques. The method is limited to solids that sustain large elastic compressions in uniaxial strain; however, a number of solids exhibit large Hugoniot elastic limits. The materials with known large Hugoniot elastic limits[25] include: sapphire, α quartz, MgO, Ge, Si, CdS, InSb, TiO_2, B_4C, BeO, and yttrium iron garnet. Thus, the method may be applied to a reasonably large number of solids of technical interest. Although shock-compression measurements have been performed on all these solids, the measurements are usually limited to several discrete stress–volume points, and these data are insufficient for the determination of third- and fourth-order constants

Even though there is some question as to the appropriateness of extending the theory to compression data to fourth order,[33] it is clear that the experimentally observed compressions of α quartz, sapphire, and fused quartz can be adequately described by the fourth-order constant development. Furthermore, it appears that shock-compression measurements can play a generally useful role in the determination of longitudinal third- and fourth-order elastic constants. If precise stress-versus-volume relations can be obtained under large elastic compressions, it appears that the finite-strain formulations can be given an evaluation. The present measurements are somewhat limited in accuracy, but it appears that the finite-strain formulation given here gives an appropriate description to both the ultrasonic and shock-compression data of sapphire and fused quartz.

ACKNOWLEDGMENT

The author is pleased to acknowledge very helpful discussions with L. W. Davison and J. C. Crawford and the very helpful review of the manuscript by W. B. Gauster.

* Work supported by the U. S. Atomic Energy Commission.

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