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Ultrasonic investigation of the nonlinearity of fused silica for different hydroxyl-ion contents and homogeneities between 300 and 3°K

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The harmonic-generation technique was used to measure the third-order elastic constants C_{111} of four types of fused silica. The samples differed in amount of OH content and directional homogeneity. The results indicate that a relatively large OH content in fused silica may give rise to a relatively greater value of C_{111} and that C_{111} is a weak function of temperature regardless of OH content. The relationship between the experimentally defined ultrasonic nonlinearity parameter $\beta = -(3C_{11} + C_{111})/3C_{11}$ and the Grüneisen parameter γ also has been determined. The measured temperature dependence of β indicates that in the Debye model elastic constants other than C_{11} and C_{111} must dominate the strong temperature dependence of the Grüneisen parameter for fused silica at low temperatures.

I. INTRODUCTION

A number of the physical properties of solids result from the fact that solids are inherently nonlinear. Among these properties are thermal expansion, attenuation of high-frequency sound waves, heat conduction, and wave-form distortion of ultrasonic waves passing through a solid. These nonlinear effects arise because of anharmonicity of the interatomic potential.

The earliest investigations of anharmonicity in solids were pursued from more purely thermodynamic experimentation. For example, Fizeau[1] was among the earliest investigators to measure thermal expansion.

Grüneisen[2] developed the relationship between the thermal-expansion coefficient and a parameter which was assumed to be temperature independent (the Grüneisen parameter γ). Barron[3,4] and Sheard[5] were among the first to make calculations relating the Grüneisen parameter to elastic data. They limited their calculations to the cases of high and low temperatures. Collins,[6] Schuele and Smith,[7] Brugger and Fritz,[8] and Gerlich[9] have made more extensive calculations to determine the temperature dependence of the Grüneisen parameter from elastic data using the quasiharmonic Debye model of solids. Thus, the Grüneisen constant is a convenient link between thermal-expansion data and elastic data.

The agreement of the calculated dependence of the Grüneisen parameter as a function of temperature with values measured from thermal expansion has been limited because of the lack of availability of elastic data [particularly third-order elastic (TOE) constants] as functions of temperature. For many materials such as Cu, Ag, Au, Al, and Na qualitative agreement exists, but some materials exhibit an anomalous behavior of the thermal-expansion coefficient as a function of temperature. One such material is fused silica. Fused silica has a positive thermal-expansion coefficient down to a temperature of approximately 0.3 of its Debye temperature. There the thermal-expansion coefficient becomes negative and continues to grow more negative as the temperature is lowered.[10] This gives rise to the anomalous behavior of the Grüneisen parameter γ shown in Fig. 1, where it is seen that γ also becomes negative around 0.3 of the Debye temperature and continues to become more negative as the temperature approaches zero. This is in contrast with germanium, for which γ levels off as the temperature approaches zero and with quartz which maintains a positive γ for all temperatures. Several investigations[3-6,11-15] indicate that at least part of the explanation for the behavior of fused silica may be due to the dominance of transverse-acoustical modes at these temperatures; however, this hypothesis can be tested only as information on the behavior of TOE constants as functions of temperature is collected. Since the TOE constants determine how a large amplitude ultrasonic wave distorts as it propagates through the solid, TOE constants can be calculated from ultrasonic wave distortion. We have chosen to measure this wave-form distortion in order to understand more fully the unusual behavior of fused silica.

Ultrasonic wave distortion was initially observed in polycrystalline aluminum[16] and in several single crystals[17] with quartz transducers. Absolute amplitude measurements became possible with the development of the capacitive receiver,[18] and measurements at different temperatures became possible with a later refinement.[19] Several investigators[20-22] have used this method to calculate combinations of truly adiabatic TOE constants. Yost and Breazeale[23] combined the results of this meth-

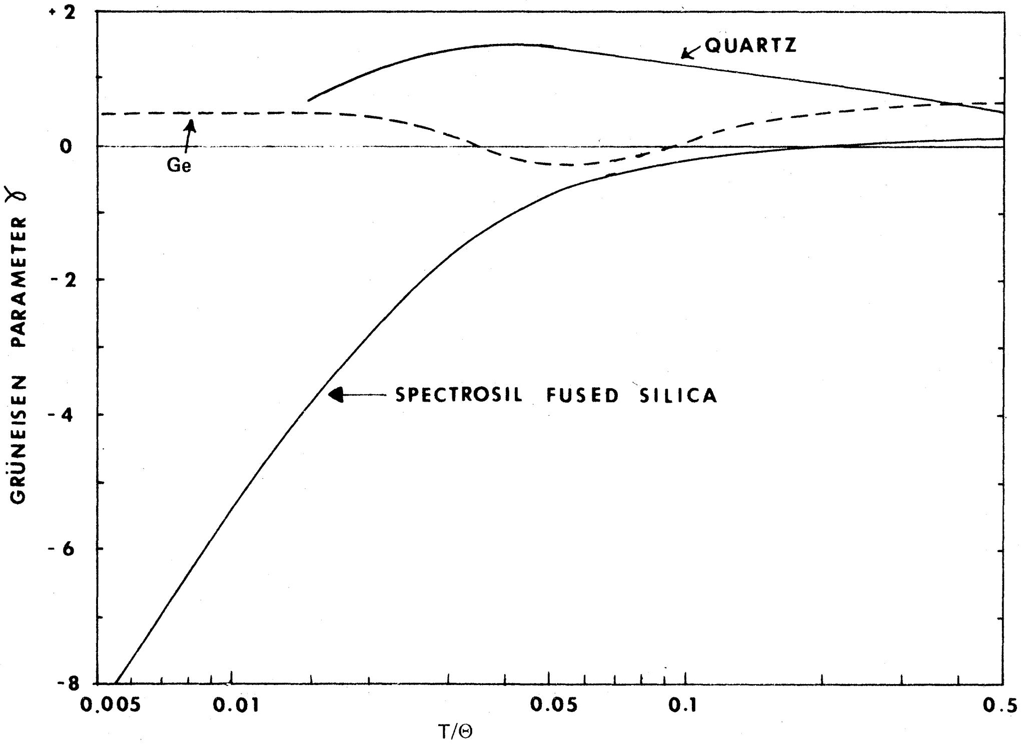

FIG. 1. Temperature dependence of the Grüneisen parameter for quartz, germanium, and spectrosil fused silica [from G. K. White, Cryogenic 4, 2 (1964)].

od with those of Dunham and Huntington[24] to obtain the first complete set of purely adiabatic TOE constants (of fused silica) at room temperature. Combinations of TOE constants of copper[25] and germanium[26] have been measured between room temperature and 77 °K. More recently, the measurements on germanium were extended to lower temperatures (3 °K).[27]

The purpose of the present investigation is to explore the results of anharmonicity more fully. To do this, we have measured the second harmonics of an initially sinusoidal ultrasonic wave in four different samples of fused silica between room temperature and 3 °K. From these data we have calculated the adiabatic TOE constant C_{111} as a function of temperature. Also, we have plotted the experimentally defined nonlinearity parameter $\beta = -(3C_{11}+C_{111}/3C_{11})$, because, as we show, there is a relationship between β and a Grüneisen parameter. The implications of the present results in light of the temperature dependence of the Grüneisen parameter are discussed.

II. THEORY

A. Ultrasonic nonlinearity in isotropic solids

For an isotropic solid pure mode propagation is possible for a longitudinal ultrasonic wave in any direction. In this special case the nonlinear wave equation takes the form[28]

$$\rho_0 \ddot{u} = C_{11}\frac{\partial^2 u}{\partial a^2} + (3C_{11}+C_{111})\frac{\partial u}{\partial a}\frac{\partial^2 u}{\partial a^2}, \tag{1}$$

where ρ_0 is the mass density of the undisturbed medium, u is the particle displacement, a is the Lagrangian (laboratory) coordinate in any direction, and C_{11} and C_{111} are second- and third-order elastic constants, respectively.

Assuming an initially pure sinusoidal disturbance at $a=0$, the solution to Eq. (1) may be written[28]

$$u = A_1 \sin(ka - \omega t) - [(3C_{11}+C_{111})/8C_{11}]A_1^2 k^2 \times a \cos 2(ka - \omega t), \tag{2}$$

where k is the propagation constant $2\pi/\lambda$, and ω is the angular frequency. Hence, an initial sinusoidal disturbance of amplitude A_1 distorts linearly with propagation distance a and generates a second harmonic of amplitude

$$A_2 = -[(3C_{11}+C_{111})/8C_{11}]A_1^2 k^2 a. \tag{3}$$

Solving Eq. (3) for C_{111} yields

$$C_{111} = -3C_{11}[\tfrac{8}{3}(A_2/A_1^2)(1/k^2 a)+1]. \tag{4}$$

All quantities on the right-hand side of this equation can be measured and C_{111} can be determined.

The quantity

$$\tfrac{8}{3}(A_2/A_1^2)(1/k^2a)=\beta \tag{5}$$

in Eq. (4) is a significant quantity in this study. If β is zero, then A_2 is zero and no second harmonic is generated. Since the generation of a second harmonic is a direct measure of the nonlinearity of a solid then β may be referred to as the ultrasonic nonlinearity parameter. For isotropic solids β is expressed in terms of the elastic constants as

$$\beta=-(3C_{11}+C_{111})/3C_{11}\,. \tag{6}$$

B. Grüneisen parameters and their relationship to β

1. Grüneisen parameters

The anharmonicity of solids also can be investigated from thermodynamic measurements (e.g., volume expansivity, variations in isothermal compressibility as a function of temperature, etc.). Among those investigating anharmonicity from these techniques it has become common to express the results in terms of the Grüneisen parameter γ, defined by[2]

$$\gamma=\alpha/K_TC_V=\alpha/K_SC_P\,, \tag{7}$$

where α is the thermal volume expansivity, K_T and K_S are the isothermal and isentropic compressibilities, respectively, and C_V and C_P are the isochoric and isobaric heat capacities, respectively.

A relationship between γ and the elastic constants can be found from calculations based on the assumption that[5,7-9] (i) the material behaves like continuous anisotropic medium, (ii) the nondispersive Debye model of specific heats is valid, and (iii) the generalized Grüneisen parameters depend on temperature only through the lattice dimensions.

If one defines the volume generalized Grüneisen parameter for the mode $(p,\vec{q})$ by

$$\gamma(p,\vec{q})=-\frac{V}{\omega(p,\vec{q})}\left(\frac{\partial\omega(p,\vec{q})}{\partial V}\right)_T, \tag{8}$$

where p is the polarization index $(p=1,2,3)$ corresponding to the three acoustical modes, $\vec{q}$ is the wave vector, V is the volume, T is the temperature, and $\omega(p,\vec{q})$ is the angular frequency of the mode. The Grüneisen parameter γ may then be expressed as[8]

$$\gamma=\sum_P\oint d\Omega\,\gamma(p,\hat{N})C(p,\hat{N})\Big/\sum_P\oint d\Omega\,C(p,\hat{N})\,, \tag{9}$$

where the integration is performed over the spatial direction $\hat{N}=\vec{q}/|\vec{q}|$ in the irreducible part of the Brillouin zone [in our case a sphere of radius $q_D=(6\pi^2/V_0)^{1/3}$, where V_0 is the volume per atom]. The specific heat of mode $(p,\hat{N})$ in Eq. (9) is given by

$$C(p,\hat{N})=\left(\frac{q_D}{2\pi}\right)^3\int_0^1 d\xi\,\frac{\xi^4[\theta(p,\hat{N})/T]^2}{\{\exp[\theta(p,\hat{N})/T]-1\}^2} \tag{10}$$

where $\theta(p,\hat{N})$ is the characteristic Debye temperature of mode $(p,\hat{N})$ given by

$$\theta(p,\hat{N})=(\hbar q_D/K)S(p,\hat{N})\,, \tag{11}$$

where K is the Boltzmann constant and

$$S(p,\hat{N})=\left(\frac{\partial\omega(p,\vec{q})}{\partial q}\right)_{q=0} \tag{12}$$

is the elastic wave speed of that mode.

It is convenient to introduce the *strain* generalized Grüneisen parameters $\gamma_{jk}(p,\vec{q})$ defined by

$$\gamma_{jk}(p,\vec{q})=-\left[\frac{1}{\omega(p,\vec{q})}\left(\frac{\partial\omega(p,\vec{q})}{\partial\eta_{jk}}\right)_T\right]_{\eta=0}, \tag{13}$$

where η is the Lagrangian strain tensor and the indices $j,k=1,2,3$. The *volume* generalized Grüneisen parameters can be expressed in terms of the *strain* generalized Grüneisen parameters by

$$\gamma(p,\vec{q})=\frac{1}{K_T}\sum_j\sum_{rs}S^T_{jjrs}\gamma_{rs}(p,\vec{q})\,, \tag{14}$$

where K_T is the isothermal compressibility and S^T_{jjrs} are the isothermal elastic compliance coefficients.

It has been shown[29] that $\gamma_{jk}(p,\vec{q})$ can be expressed in terms of the elastic constants by (Einstein notation)

$$\gamma_{jk}(p,\hat{N})=[1/2w(p,\hat{N})][2w(p,\hat{N})U_jU_k+(C_{jkmn}+C_{jkmnuv}U_uU_v)N_mN_n]\,, \tag{15}$$

where

$$w(p,\hat{N})=C_{munv}N_mN_nU_uU_v\,. \tag{16}$$

$\hat{N}$ gives the direction of wave propagation, $\hat{U}$ is a unit vector along the direction of polarization appropriate to p, and the subscripted C's are the second- and third-order elastic constants in the equations.

2. Relationship to the ultrasonic nonlinearity parameter

For pure longitudinal acoustic modes in an isotropic solid, Eq. (15) reduces to the set of relations (using Voigt[30] notation)

$${}_l\gamma_{11}=-(3C_{11}+C_{111})/2C_{11}\,, \tag{17}$$

$${}_l\gamma_{22}={}_l\gamma_{33}=-(C_{12}+C_{112})/2C_{11}\,, \tag{18}$$

and

$$ {}_l\gamma_{ij}=0,\quad i\neq j\,, \tag{19}$$

where the prescript l indicates longitudinal-acoustical modes. From Eq. (6) and (17) it is seen that ${}_l\gamma_{11}$ is related directly to the ultrasonic nonlinearity parameter β by

$$ {}_l\gamma_{11}=\tfrac{3}{2}\beta\,. \tag{20}$$

Hence, one of the strain generalized Grüneisen parameters is measured directly by the present harmonic-generation technique.

For an isotropic solid Eq. (9) reduces to the form

$$\gamma=(\gamma_l C_l+2\gamma_t C_t)/(C_l+2C_t)\,, \tag{21}$$

where

$$\gamma_l=+\tfrac{1}{2}\beta-(C_{12}+C_{112})/3C_{11} \tag{22}$$

is the longitudinal-mode *volume* generalized Grüneisen parameter obtained from Eq. (8), and C_l and C_t are the modal specific heats of Eq. (10). γ_t in Eq. (21) is the transverse-mode *volume* generalized Grüneisen parameter and is not easily expressed in terms of the ultrasonic nonlinearity parameter.

The significance of the relationship of the nonlinearity parameter β to the Grüneisen parameter γ is now established for the case of isotropic solids. It allows one to ascertain directly the contribution of the "longitudinal-mode" elastic constants C_{11} and C_{111} to the temperature dependence of the Grüneisen parameter when β is measured as a function of temperature.

III. EXPERIMENTAL TECHNIQUE

A. Apparatus and measurement techniques

The apparatus and measurement techniques for determining the amplitudes of fundamental and second-harmonic waves are described in detail elsewhere.[27] The amplitudes of the fundamental and the second-harmonic waves were measured absolutely in each sample at room temperature. These data were used in Eqs. (4) and (5) to determine room-temperature values of C_{111} and β. At all other temperatures the amplitudes were measured relative to the room-temperature values, and relative values of C_{111} and β were determined.

B. Second-order elastic constants

The second-order elastic (SOE) constant C_{11} appearing in Eq. (4) was determined from room-temperature longitudinal wave-velocity measurements, since

$$C_{11}=\rho_0 V^2\,, \tag{23}$$

where ρ_0 is the mass density of the unstrained solid. The measurements were made with capacitive transducers[31] which eliminated the problem of bond corrections. The temperature dependence of C_{11} was calculated from the data of A. Zarembowitch[32] who measured the SOE constants of Purposil fused silica as a function of temperature. The SOE constants of the Suprasil fused-silica samples used in these experiments are assumed to have the same relative temperature dependence as that of Purposil.

C. Samples

The fused-silica samples used in this work were manufactured under the commercial designation Suprasil. All samples were cylindrical, approximately 2.54 cm in diameter, and were coated on each end with copper to a thickness of approximately 1000 Å. The length of each sample, impurity content, and the measured ultrasonic wave velocities at room temperature (27 °C) are given in Table I. The symbol W indicates that the sample has only 5 ppm of OH, while the unlettered samples have OH contents of 1200 ppm. The numerical designation 1 or 2 indicates the degree of homogeneity of the sample. The designation 1 indicates a guaranteed strong homogeneity in all directions, whereas the designation 2 implies a guaranteed strong homogeneity only in the direction perpendicular to the sample surfaces.

IV. RESULTS

A. Room-temperature measurements

Accurate room-temperature values of the nonlinearity parameter β and of C_{111} were determined by an extrapolation technique which minimized the effect of attenuation on the results.[26] The nonlinearity parameter β was measured for different

TABLE I. Samples used in this study and pertinent properties.

Sample	Impurity content (ppm) OH	Fluorine	Chlorine	Sample length (cm)	Longitudinal ultrasonic velocity (10^5 cm/sec)
Suprasil *W*1	5	260	260	1.2590 ± 0.0003	5.9526 ± 0.0016
Suprasil *W*2	5	260	260	1.2587 ± 0.0002	5.9533 ± 0.0011
Suprasil 1	1200	130	130	1.2593 ± 0.0002	5.9363 ± 0.0012
Suprasil 2	1200	130	130	1.2586 ± 0.0002	5.9363 ± 0.0011

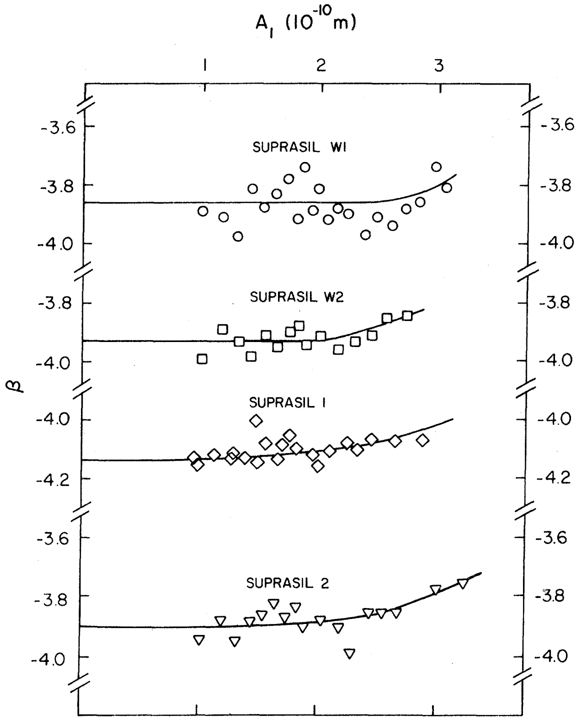

FIG. 2. Room-temperature plots of β as a function of the fundamental amplitude.

fundamental wave amplitudes A_1 for each sample. The resulting values of β were plotted as a function of A_1, as shown in Fig. 2. The solid curves are drawn through the data points in such a way that they approach $A_1 = 0$ with a horizontal tangent. Previous analysis[26] shows that this extrapolation corrects for the effect of attenuation. As can be seen the maximum magnitude of this correction is of the order of 5%.

The results of the extrapolated room-temperature measurements of β, C_{11}, and C_{111} are given in Table II. An examination of the table reveals that the SOE constants of the fused-silica samples with high OH content (Suprasil 1 and Suprasil 2) are lower than the samples with low OH content (Suprasil *W*1 and Suprasil *W*2). These results are consistent with those of Hetherington and Jack[33] who noted also that an increase in OH content caused an increase in mass density fluctuation as much as 3 parts in 10^3 in their experiments. Primak[34] suggested that in similar experiments by Fraser[35] in the ultrasonic range of frequencies the results reflect the effect of network cleavages induced by OH impurities.

Table II shows that β is negative for all fused-silica samples. This means that A_2 is also negative and that C_{111} is positive. Bains and Breazeale[36] have shown, using a phase-sensitive detector, that the harmonics of fused silica are generated out of phase with those generated by the same ultrasonic wave in copper. They conclude that the positive C_{111} of fused silica means that the solid becomes softer upon compression in contrast to copper which becomes stiffer upon compression.

According to Table II, Suprasil 1 has a substantially higher value of C_{111} than any of the other samples used in these experiments. It is tempting to infer that this is because of the high OH content of the sample (1200 ppm compared to 5 ppm for Suprasil *W*1 and Suprasil *W*2), but Suprasil 2 also has a high OH content (1200 ppm) and gives a value of C_{111} which lies between those of Suprasil *W*1 and Suprasil *W*2. The reason for this is not completely understood at this time. A check of all the samples with crossed polaroid sheets revealed that residual stresses were undetectable by this technique. Hence, residual stresses in a particular sample probably are not the cause of the difference.

A possible explanation may lie in the fact that even though Suprasil 2 is strongly homogeneous in the direction perpendicular to the sample faces, it may have inhomogeneities in other directions. If mass density variations existed parallel to the sample faces, then there would be an associated phase variation across the ultrasonic wave front. Such a situation leads to phase heterodyning which can cause significant changes in the amplitude of the electrical signal response.[37] Since this heterodyning is frequency dependent, fundamental and second-harmonic waves would be affected differently. The result could be a decrease in the measured value of C_{111} for Suprasil 2. If this were true, then one could consistently associate high C_{111} values with high OH content. Thus, the network cleavages introduced by the OH impurities

TABLE II. Measured room-temperature values of β, C_{11}, and C_{111}.[a]

Sample	β	C_{11} (10^{11} dyn/cm^2)	C_{111} (10^{12} dyn/cm^2)
Suprasil *W*1	-3.86 ± 0.071	7.8059 ± 0.0029	6.70 ± 0.124
Suprasil W_2	-3.93 ± 0.034	7.8078 ± 0.0020	6.85 ± 0.058
Suprasil 1	-4.14 ± 0.041	7.7633 ± 0.0016	7.31 ± 0.073
Suprasil 2	-3.90 ± 0.039	7.7633 ± 0.0014	6.75 ± 0.067

[a] The errors listed are the calculated standard deviations (random errors).

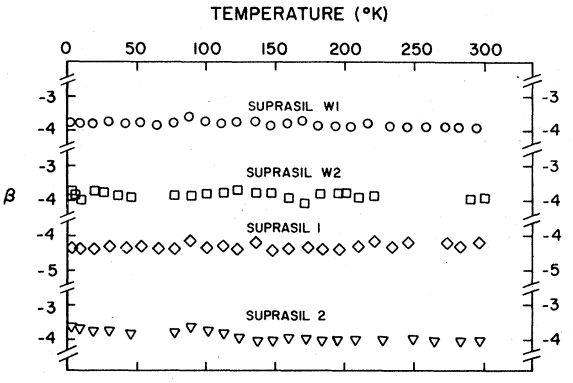

FIG. 3. Temperature dependence of β for the fused-silica samples.

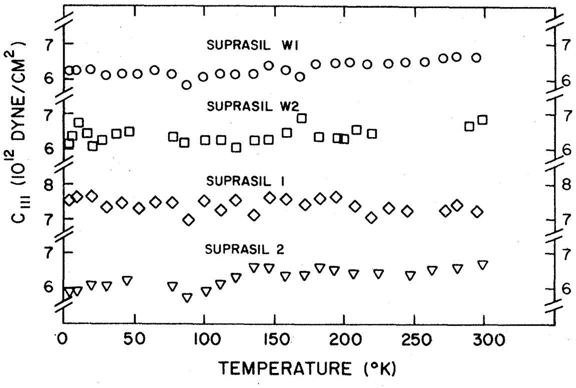

FIG. 4. Temperature dependence of C_{111} for the fused-silica samples.

which may be the cause of lower C_{11} values could also be the cause of higher C_{111} values.

B. Cryogenic measurements of β and its implications to the calculation of Grüneisen's γ

In making relative measurements with the cryogenic system, the quantity β is determined directly[27] [see Eq. (5)]. The resulting plots of β versus temperature in Fig. 3 indicate that β is virtually temperature independent. This means that the ${}_{l}\gamma_{11}$ component of the *strain* generalized Grüneisen parameter also is virtually temperature independent since ${}_{l}\gamma_{11}=\frac{3}{2}\beta$.

Therefore, the temperature variation of the experimentally determined Grüneisen parameter γ for fused silica shown in Fig. 1 must come from variations in the *volume* generalized Grüneisen parameter [see Eqs. (21) and (22)].

It is apparent from Eq. (21) that in order to obtain a negative value of the Grüneisen parameter at some temperature for fused silica, at least one of the *volume*-generalized Grüneisen parameters must be negative at that temperature since the model heat capacities C_l and C_t are always positive. Barron[3,4] and Blackman[11] have suggested theoretical models which for some volume generalized Grüneisen parameters, associated with certain transverse-vibrational modes, do become negative. These transverse modes necessarily involve elastic constants other than C_{11} and C_{111}.

C. Cryogenic measurements of C_{111}

The behavior of C_{111} as a function of temperature can be determined directly from that of β. Figure 4 is the resulting plot of C_{111} versus temperature for the four samples. These curves show greater scatter in the data points than was experienced in previous experiments with copper[25] and germanium.[26,27] Further, the estimate of random error due to temperature changes in the transducer bonds is ±3% and the standard error is no more than ±1%. Hence, the considerable amount of scatter would seem to indicate structural changes in the samples as a function of temperature. In large part, this may be due to the asymmetrical nature of the void surrounding the oxygen atom, which is a reflection of the random nature of the tetrahedral network. As the temperature is lowered, evidence exists[34] that the Si–O–Si bond angle changes due to the asymmetrical vibrations of the oxygen atoms. Thus, the tetrahedral network of fused silica below the quenching temperature is in a state of stress. The resulting states of strain are more accentuated for the weak impurity bonds and this makes them more efficient electron traps than the strong Si–O bonds. This means that the effect of temperature changes is to cause variations, especially in anharmonic-dependent effects since it is these effects that are most sensitive to variation in structural symmetry and strain perturbations introduced by impurity sites.

Although scatter in the cryogenic measurements is too large to attach a great amount of significance to individual data points, trends in the C_{111}-versus-temperature data are apparent. The C_{111} values of Suprasil *W*1, Suprasil *W*2, and Suprasil 2 all show a slight general decrease of C_{111} with decreasing temperature. The C_{111} values of Suprasil 1 on the other hand do not decrease with decreasing temperature. As with the room-temperature measurements, Suprasil 1 (with high OH content) seems to be an anomaly. Again however, Suprasil 2 (also with high OH content) seems to show the general C_{111}-versus-temperature behavior as Suprasil *W*1 and Suprasil *W*2 (both with low OH content). Following the explanation given for the room-temperature measurements, it is conceivable that the lack of strong homogeneity in the Suprasil 2 sample gives rise to large density fluctuations across the face of the ultrasonic beam. The change of temperature possibly may exaggerate these density fluctuations, thereby giving rise to the temperature dependence of C_{111} observed for Suprasil 2. The pronounced decrease of the C_{111} value of Suprasil 2 (Fig. 4) with decreasing temperature in the range 147–88 °K indicates that a strong structural change (and consequently density changes) may have taken place.

D. Conclusion

In conclusion, the present experimental determinations of β and C_{111} as functions of temperature allow an important inference to be made regarding the temperature dependence of the Grüneisen parameter for fused silica. If the quasiharmonic Debye model of a solid is assumed to hold for fused silica, then the relatively weak temperature dependence of β and C_{111} indicates that elastic constants, other than C_{11} and C_{111}, must dominate the strong temperature dependence of the experimentally determined Grüneisen parameter for fused silica at low temperatures. This is consistent with the suggestion of White and Birch[38] that transverse vibrations of fused silica are associated with the oxygen atoms of the tetrahedral network. The elastic constants associated with these transverse modes would be of the type C_{44}, C_{144}, and C_{456}, which do not enter into the measurements described here.

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