

Attenuation of Hypersound by Collinear Phonon Interactions in Quartz

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Low-temperature measurements of the attenuation of microwave phonons of longitudinal polarization along the x axis and of transverse polarization along the ac axis of natural quartz at 1.25 and 2.1 GHz are compared with calculations for a dispersive anisotropic continuum. By separating the analysis of the data into two temperature zones whose extent is determined by the relative magnitudes of $\Omega\tau$ and of the dispersion factor $\beta=v/c$, it was possible to obtain, respectively, estimates for the generalized Grüneisen parameters when $(1+\beta)^{-1} < \Omega\tau < (1-\beta)^{-1}$ and for the strength of the thermal-phonon interactions at low temperatures when $\Omega\tau > (1-\beta)^{-1}$. Very good agreement is obtained for the temperature and frequency dependence of the absorption coefficient by assuming that the transverse-wave attenuation is due to collisions with collinear transverse phonons. For longitudinal waves, evidence is in accordance with the assumption that the main contribution to the attenuation is from collisions with longitudinal *and* transverse thermal phonons. We find however, that the former process becomes increasingly predominant as the temperature is reduced. The experimental and theoretical Grüneisen parameters agree to better than a factor of 5. For large $\Omega\tau$ calculations predict a reduction in the temperature dependence of the absorption with moderate concentration of point impurities.

INTRODUCTION

Although the low-temperature hypersonic absorption in quartz has been the subject of several recent¹⁻⁴ experimental and theoretical studies, there is still uncertainty about the particular anharmonic interactions with thermal phonons which are responsible for the attenuation of longitudinal and transverse waves, as evidenced by the inability to account for the magnitude of the measured absorption of some of these waves.² The temperature and frequency dependence of the intrinsic absorption coefficient in quartz is common to a number of dielectrics. For hypersonic frequencies Ω greater than the inverse thermal-phonon relaxation time τ^{-1} , the coefficient shows a dependence close to the ΩT^4 predicted by Landau and Rumer⁵ for infinite τ ; at lower temperatures the frequency dependence disappears and the dependence on temperature may become as high as T^9 . This lower-temperature behavior of the absorption has been explained by including in the theoretical formulations a large but finite τ and normal (linear-chain) acoustic dispersion.

Recently Lewis and Patterson² interpreted their measurements of the attenuation of 9-GHz waves of various polarizations and propagation directions in natural quartz by means of a formulation due to Maris,¹ with very good results for the temperature dependence. The attenuation of transverse waves was attributed by these authors to two three-phonon processes: a Landau-Rumer process of the type $T+L \rightarrow L$, and a much less effective near-collinear process $T+T \rightarrow T$. They found, however, unexplained discrepancies in the magnitude of the absorption of waves of longitudinal polarization

along the x axis. The attenuation of longitudinal waves was attributed to the collinear processes $L+L \rightarrow L$, but the value of the calculated absorption for this polarization was found to be about two orders of magnitude smaller than measured. To explain this large discrepancy Purdom and Prohofs-ky⁴ have proposed the anharmonic process $L+T \rightarrow T$, where T stands for thermal phonons of a very dispersive transverse branch along the z axis of quartz. But even though a good fit was obtained with the 9-GHz experimental results, this approximation fails to account for the observed dependence of the absorption with frequency or $\Omega\tau^1$ in the temperature range valid for comparison.

It should be borne in mind, when considering discrepancies in absorption magnitude, that because of the low symmetry of quartz most theoretical formulations of the sound absorption treat anisotropy and dispersion in a simplified way which may lead to important errors. For example, King³ has shown that when elastic (second-order) anisotropy is taken into account in the averaging of the anharmonic coupling constants, interactions with thermal phonons of equal polarization are clearly predominant in the absorption of transverse waves in quartz in contradiction with the findings from simpler calculations.² For the case of longitudinal waves along the x axis of a trigonal crystal the simplified theories give, in terms of the second- and third-order elastic constants, an anharmonic coupling parameter for collinear interactions proportional to $(3C_{11}+C_{111})^2$ which, for quartz, is vanishingly small. However, a better account of crystalline anisotropy and dispersion may considerably alter this value.

Since knowledge of the dispersion and anisotropy

in quartz is insufficient, we do not intend in this work to consider these factors in any great complexity. Instead, we choose to interpret our experimental results on the attenuation of low-GHz waves in quartz by means of a simplified formulation for a dispersive anisotropic continuum, and *only* in terms of (near-) collinear three-phonon anharmonic interactions, which reduces considerably the difficulties related to anisotropy. We discuss in some detail the range of applicability of the dominant-phonon approximation and its effect on the magnitude of the anharmonic coupling constants deduced from the experimental absorption.

By making assumptions about the frequency and temperature dependence of the thermal-phonon relaxation time in quartz which seem appropriate at low temperatures we are able to obtain, through a suitable extension of the above formulation, a good fit to the experimental intrinsic absorption at the lowest temperatures and over a wide range of hypersonic frequencies such that $1 < \Omega\tau < kT/\hbar$ and, what may be of more relevance, our treatment makes it possible to obtain quantitative estimates of the interaction strengths for the thermal phonons from hypersonic measurements.

EXPERIMENTS

Samples used in these experiments consisted of natural Brazilian acoustic prisms ($4 \times 4 \times 12.5$ mm) commercially prepared to hypersonic specifications.⁶ Waves of both longitudinal and transverse polarizations were generated by conventional pulse-echo techniques in microwave resonators of the re-entrant coaxial type with the exciting rf field normal to the generating ends of the piezoelectric samples. Tuning was accomplished by a sliding metallic coaxial discontinuity which allowed the same resonator to be used at odd harmonics of its fundamental frequency (500 MHz). The tuning range for a given resonant mode was greater than 25%.

Attenuation was measured by matching the signal of a calibrated exponential generator⁷ triggered at $\frac{1}{2}$ of the repetition rate of the main microwave generator to the tail of the hypersonic echo train displayed on the screen of a dual-trace oscilloscope. This method could not be used with a very long echo pattern (absorption coefficients less than 8×10^{-3} dB/ μ sec fall outside the range of the exponential generator). In such cases we obtained the absorption coefficient by measuring photographs of the oscilloscope display on a micrometric profile projector. The attenuation was thus determined to better than 10%.

During the measurements the resonators containing the samples were surrounded by cool helium gas at atmospheric pressure and connected by means of a cold finger to the liquid-helium bath. The temperature was determined with a relative accuracy

of better than 0.05°K by means of a calibrated Ge thermometer⁸ placed next to the copper sample holder. Insufficient thermal contact between the crystal and thermometer introduces an error of no more than 0.5°K in the determination of the absolute sample temperature. The measured attenuation in dB/ μ sec for hypersonic waves along the x and ac axes is represented by the points in Figs. 1, 2, and 4 (1 Np = 8.68 dB). The temperature-independent residual attenuation observed below 10°K has been subtracted from the data presented in the graphs.

In order to compare the experimental results with the theory the thermal conductivity relaxation time is taken from Ref. 1. The density of quartz is taken as 2.65 g/cm^3 . Second- and third-order elastic constants to be used in the calculations of Grüneisen parameters are taken, respectively, from McSkimin *et al.*⁹ and Thurston *et al.*¹⁰ The Debye temperature for the different phonon modes is calculated by $T_D^i = \Theta v_i/v_m$, where Θ and v_m are the Debye temperature (545°K) and mean velocity (4.3 km/sec) in quartz as determined from the elastic moduli,¹¹ and v_i is the appropriate phonon velocity. This way of calculating T_D^i differs from that corresponding to a monatomic linear chain,² but is more consistent with our use of a Debye cutoff radius which takes into account the contents of the quartz unit cell. Velocities of longitudinal (L) and fast transverse (F) waves along the x axis and of slow transverse (S) waves along the ac axis of quartz are 5.7, 5.1, and 3.3 km/sec, respectively.¹² The calculated Debye temperatures are $T_D^L = 725$, $T_D^F = 650$, and $T_D^S = 420^\circ\text{K}$.

ATTENUATION BY COLLINEAR THREE-PHONON PROCESSES

The anharmonic model used in this work corresponds essentially to the anisotropic continuum¹³ where, for interactions of three phonons of equal polarization, Born-von Kármán dispersion¹⁴ is included in terms of the form v_i/c_j with c_j equal to the hypersonic wave velocity. In this approximation the absorption coefficient in dB/cm for a sound wave of polarization j interacting with thermal phonons of mode i is given by¹⁵

$$\alpha_j \approx \frac{8.68 \Omega k^4 T^4}{\rho c_j^3 h^3} \sum_i \frac{2\pi \gamma_{ij}^2}{v_i^3} I_i,$$

$$I_i = \int_0^{2\tau_D^i/\tau} \frac{x^4 e^x}{(e^x - 1)^2} \times [\arctan(1 + \beta_i) \Omega \tau_i - \arctan(1 - \beta_i)] dx. \quad (1)$$

This expression represents the leading terms for the case when $\Omega\tau_i > 1$. Here β_i stands for v_i/c_j , ρ is the density of the solid, and γ_{ij}^2 is an average of the generalized Grüneisen parameter for the in-

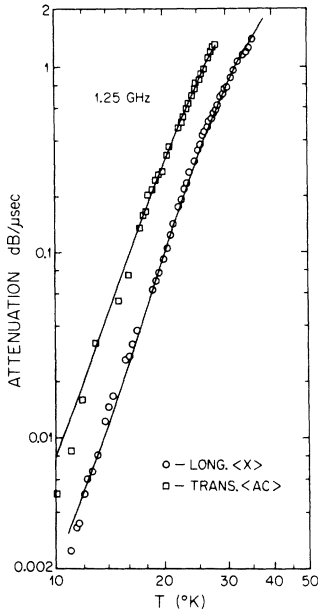


FIG. 1. Temperature-dependent absorption coefficient for longitudinal (x axis) and transverse (ac axis) waves in quartz at 1.25 GHz; solid curve calculated from Eqs. (4) and (6).

interaction.¹⁶ In the case of an anisotropic continuum this parameter is a function of the second- and third-order elastic constants and of the polar angles θ_i and ϕ_i of the thermal-phonon wave vector with respect to the sound-wave propagation direction. Since, however, only quasicollinear interactions ($\beta_i \leq 1$) are considered here, the terms in γ_{ij} proportional to $\sin\theta_i$ are neglected; furthermore, all other thermal-phonon parameters are taken as independent of θ_i and ϕ_i . k and h are, respectively, Boltzmann's and Planck's constants and x is the reduced thermal-phonon frequency $\hbar\omega_i/kT$. As discussed above, the dispersion is included when appropriate by taking $1 - \beta_i = \frac{1}{2}(\pi x T/2T_D^i)^2$.

In spite of the approximations already introduced the lack of knowledge about $\tau_i(x, T)$ excludes an exact evaluation of I_i . For the interpretation of the experimental results it will prove convenient and significant to make from the outset two approximations of the integral I_i . They predict, for intermediate and lowest temperatures, two distinct functional forms for the sound absorption coefficient both consistent with observations. The range of applicability of each approximation will be determined by the magnitude of $(1 \pm \beta_i)$ relative to $\Omega\tau_i$.

A. Landau-Rumer Zone: $(1 + \beta_i)^{-1} < \Omega\tau_i < (1 - \beta_i)^{-1}$

In this temperature range the term in brackets in I_i is nearly constant (for infinite τ_i it should be replaced by $\frac{1}{2}\pi$) and can be taken out from under the integral sign and evaluated by means of a dominant thermal-phonon approximation with $x = 3.83$, a value which maximizes the remaining integrand in I_i . One obtains in this case an absorption co-

efficient which can be conveniently written as

$$\alpha_j \approx \frac{8.68 \Omega k^4 T^4}{\rho c_j^3 h^3} \sum_i \frac{2\pi\gamma_{ij}^2}{v_i^3} J_4\left(\frac{2T_D^i}{\pi T}\right) \times \arctan \frac{2\beta_i \Omega \tau_i}{1 + (1 - \beta_i^2) \Omega^2 \tau_i^2}, \quad (2)$$

where τ_i represents the value of $\tau_i(x, T)$ at $x = 3.83$. For equal wave and phonon polarization, $1 - \beta_i^2 = (\pi x T/2T_D^i)^2$ should also be evaluated at the dominant-phonon reduced frequency. J_4 is the known transport integral of the specific heat C_v . In this regime, Eq. (2) will give an absorption coefficient which behaves close to the ΩT^4 law predicted by Landau and Rumer (LR). Since the magnitude of τ_i is here more important than its explicit functional form, good agreement with experiments for a number of insulators has been obtained^{1,2,15} by substituting for τ_i in Eq. (2) the thermal conductivity phonon relaxation time $\tau_K = 3K/v_m^2 C_v$.

We have found by computer calculations that Eq. (2) adapted to our experiments in quartz gives an absorption whose magnitude and temperature dependence are quite insensitive in the LR zone for values of x between 1 and 7. It has been found for a number of dielectrics that expressions similar to Eq. (2) agree well with experiments throughout the entire low-temperature range when the value of x is used as an adjustable parameter. For longitudinal waves in quartz¹⁷ values of x as high as 6 are necessary to obtain a good fit with the data to lowest temperatures (the best value for x will of course depend on the size of T_D^i used in $1 - \beta_i^2$). These large values of x have been taken in the literature as indicative of the importance of the frequency dependence of τ_i at temperatures at which $\Omega\tau$ is

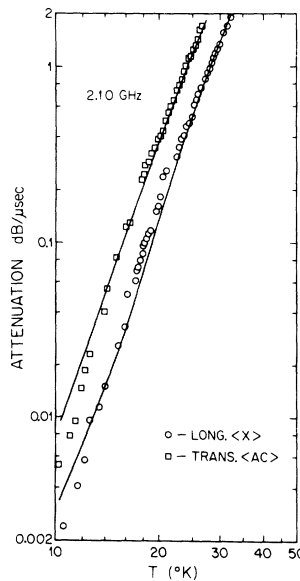


FIG. 2. Temperature-dependent absorption coefficient for longitudinal (x axis) and transverse (ac axis) waves in quartz at 2.10 GHz; solid curve calculated from Eqs. (4) and (6).

very large. The above procedure allows to account reasonably for the observed temperature dependence of the absorption. The use of a mean lifetime, however, at temperatures when τ_i may be rapidly varying with x (as for example when point impurity scattering of thermal phonons is important) is inadequate and, furthermore, excludes the possibility of making a better use of hypersonic measurements for the study of the strength of thermal-phonon interactions.

B. Large- $\Omega\tau$ Zone: $(1 - \beta_i)\Omega\tau_i > 1$

In this lower-temperature range the quantity in brackets in the expression for I_i is approximately $2\beta_i/(1 - \beta_i^2)\Omega\tau_i$, which can be integrated out in x if a functional form of $\tau_i(x, T)$ is known or assumed. For this latter purpose it is important to realize that this temperature range, wherein the absorption is rapidly varying with T , corresponds in a number of dielectrics including quartz to temperatures close to that at which the thermal conductivity is a maximum. It is well known that point impurity and normal (N) anharmonic scattering of thermal phonons become important for this range in limiting their lifetimes. Bearing in mind that the corresponding expressions for $\tau_i(x, T)$ are only approximate and of limited validity,¹⁸ we adopt for τ_i^{-1} the form $A_i x^4 T^4 + B_i x^{n_i} T^5$ (at these low temperatures $n_i = 2$ for longitudinal phonons; $n_i = 1$ for transverse phonons). When this expression for τ_i is substituted in the approximation for the arctangents given above and the integral I_i is calculated, one obtains for the intrinsic absorption at the lowest temperatures the expression

$$\alpha_j \approx \frac{8.68 k^4 T^4}{\rho c_j^3 h^3} \sum_i \frac{2\pi \gamma_{ij}^2}{v_i^3} \frac{2\beta_i}{(1 - \beta_i)^2} \times (A_i T^4 J_6 + B_i T^5 J_{2+n_i}) \quad (3)$$

Here $T \ll T_D^i$ and the transport integrals J_p take their asymptotic value $p! \zeta(p)$, where $\zeta(p)$ is the Riemann ζ function. For the dispersive case, $1 - \beta_i^2$ in Eq. (3) is independent of x and is equal to $(\pi T / 2T_D^i)^2$. For the nondispersive case, β_i is a constant but the index p of both transport integrals in the equation should be increased by two units. Equation (3) will later be shown to describe very

well the dependence on T of our lowest-temperature data. It is appropriate, however, to discuss at this time some of the general features of the resulting theoretical absorption. The equation predicts, in agreement with the experimental data at lowest temperatures, a frequency-independent absorption coefficient. For perfect crystals the B term of the expression will predominate, resulting in a T^7 or T^9 temperature dependence of α_j , depending on whether dispersion is to be included or not. This dependence will, however, reduce towards T^6 or T^8 , respectively, as the number of point imperfections or impurities increases moderately, so that the condition $\Omega\tau(1 - \beta_i) > 1$ is not violated. Clearly, once the experimental γ_{ij}^2 is determined from the analysis of the data in the LR zone, estimates for the values of A_i and B_i can be extracted from the best fit of Eq. (3) to the measured absorption in the lowest-temperature range. Such analysis will also allow the use of the hypersonic data for the study of the low-temperature scattering of thermal phonons in insulators with variable, but low, concentration of point impurities.

APPLICATIONS TO QUARTZ AND DISCUSSION

Longitudinal Waves along x Axis

The lower points in Figs. 1 and 2 show the measured intrinsic attenuation of hypersonic waves along this propagation direction in quartz. There is a well-defined zone where the coefficient varies as T^4 . Its extent is found to be from 24 to 31 °K for 1.25 GHz and from 25 to 31 °K for 2.10 GHz. The temperature dependence of the coefficient changes gradually with decreasing T in such a manner that it is nearly T^7 at the lower end of the range of measurements. At the same time the frequency dependence disappears at these lower temperatures (Fig. 4). These distinct dependences of the measured absorption are in accordance with our approximate theoretical formulation.

To compare our data with theory we assume that the attenuation of longitudinal waves along the x axis is due to two near-collinear interactions of the microwave phonons with longitudinal and fast transverse phonons: $L + L - L$ and $L + F - F$. The calculated absorption Eq. (2) for the LR zone can be written as

$$\alpha_L \approx 1.32 \times 10^{-18} \Omega T^4 \left[\gamma_{LL}^2 J_4 \left(\frac{462}{T} \right) \arctan \frac{2\Omega\tau_K}{1 + (T/462)^2 (x\Omega\tau_K)^2} + 1.4 \gamma_{LF}^2 J_4 \left(\frac{414}{T} \right) \arctan \frac{1.8\Omega\tau_K}{1 + 0.2(\Omega\tau_K)^2} \right] \quad (4)$$

Values for the generalized Grüneisen parameters were obtained from a least-squares fit of Eq. (4) to the experimental results in the LR zone at each

frequency with x as an adjustable parameter. The predicted T^4 zone for the first of the above processes was found to depend somewhat on the value of

x used in the equations, shifting to higher temperatures for larger x . The corresponding variation in the measured value of γ_{LL} is however less than 15% for x between 1 and 7. Since a clear T^4 absorption was experimentally observed we have chosen to fit the data with temperature intervals and values of x for which both terms inside the large square brackets in (4) show little variation with temperature. This choice corresponds to $x = 5$; $27.6 < T < 31.6$ °K for 1.25 GHz and $30 < T < 34$ °K for 2.10 GHz. For each frequency, $\Omega\tau_K(1 - \beta_i) < 1$ holds well in these temperature intervals for $x = 5$. Moreover, we find that with this value of x , Eq. (4) not only reproduces very well the temperature and frequency dependence of σ_j in the LR region, but seems also to fit reasonably well the experimental results to the lowest temperatures, as can be seen from the solid-line lower curves of Figs. 1 and 2. It is however, found on closer examination that the intrinsic attenuation is reproduced much better by the large- $\Omega\tau$ approximation of Eq. (3). The γ_{ij} obtained from the data are given in Table I for each process, together with the values calculated from the elastic constants. The measured γ_{LL} and γ_{LF} listed represent mean values, since it is found that their individual values for each frequency differ by less than 10%. The effect of including the process $L + F \rightarrow F$ is to reduce the previously reported discrepancy² between the calculated and measured γ_{LL} by a factor of 2. We think that the significance of the remaining difference cannot be properly discussed within the framework of the present model, since the small value of the calculated $\gamma_{LL} \propto 3C_{11} + C_{111}$ may reflect the simplified way in which its average in phonon-wave-vector space is performed here.

Figure 3 shows the relative contribution of the two aforementioned selected interactions as a function of temperature for $x = 5$. It is seen that the contribution of the process $L + F \rightarrow F$ becomes too small at the lowest temperatures and can be neglected in the large- $\Omega\tau$ zone. From the observed temperature dependence of the absorption, this

TABLE I. Generalized Grüneisen parameter values obtained from a least-squares fit of Eqs. (4) and (6) to the experimental results in the LR zone.

Wave polarization and propagation direction	Interaction process	Generalized Grüneisen parameter measured	Generalized Grüneisen parameter calculated
Longitudinal, x axis	$L + L \rightarrow L$	$\pm 1.26 (\pm 2.54^a)$	-0.28
Longitudinal, x axis	$L + F \rightarrow F$	± 1.16	-1.20
Transverse, ac axis	$S + S \rightarrow S$	$\pm 0.36 (\pm 0.36^a)$	

^aFrom Reference 2.

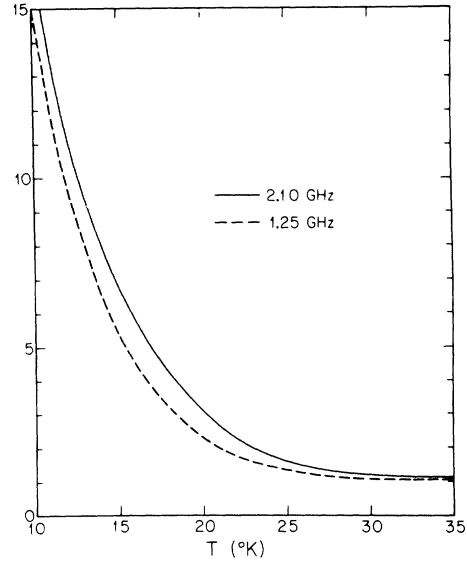


FIG. 3. Ratio of the temperature-dependent calculated absorption coefficient of the process $L + L \rightarrow L$ to that of process $L + F \rightarrow F$ as a function of temperature.

zone extends below 16 and 14 °K for the upper and lower experimental frequencies, respectively. The corresponding values of $\Omega\tau_K(1 - \beta_i)$ are greater than one for each case (for 9 GHz this low-temperature range would extend to 30 °K). The absorption coefficient Eq. (3) for the process $L + L \rightarrow L$ in the large- $\Omega\tau$ zone is written as

$$\alpha_L \approx 8.90 \times 10^{-13} [A_L T^6 6! \zeta(6) + B_L T^7 4! \zeta(4)]. \quad (5)$$

This expression is represented by the lower graph of Fig. 4. Best fit to the experimental points is obtained for values of A_L and B_L equal to 0.59 °K⁻⁴ sec⁻¹ and 4.70 °K⁻⁵ sec⁻¹, respectively. The predominance of the B_L term puts in evidence the quality of the crystals used in our measurements. The value of A_L agrees well with a value of 0.66 obtained from the work of Bergman *et al.* on the thermal conductivity and the natural occurrence of isotopic impurities in quartz.¹⁹ Figure 4 shows that Lewis and Patterson's² results at 9 GHz are in very good agreement with Eq. (5) and our measured γ_{LL} of Table I.

Transverse Waves along ac Axis

The measured intrinsic attenuation of transverse waves at our two frequencies is represented by the upper points in Figs. 1 and 2. The temperature dependence of the absorption shows the same behavior as for longitudinal waves along the x axis, and the experimental LR zone and large- $\Omega\tau$ zones coincide in extent with those determined for longitudinal waves and are also in good agreement with

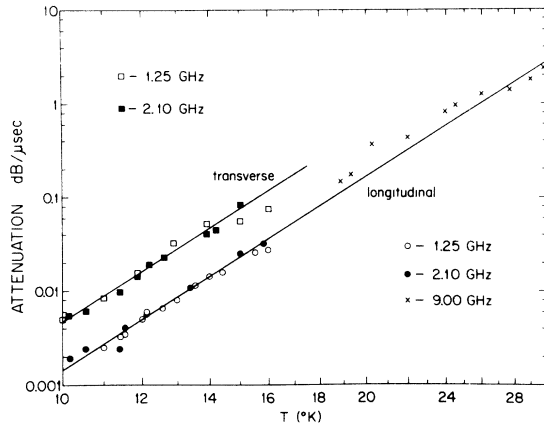


FIG. 4. Frequency-independent intrinsic attenuation in quartz in the temperature range below 30 °K. Solid lines calculated from Eqs. (5) and (7) show a $T^{6.9}$ dependence. \times represents Lewis and Patterson's results (Ref. 2).

the predictions from the magnitude of $\Omega\tau_K(1 \pm \beta_i)$. For this polarization $x=3$ gives the best least-squares fit to our data in the LR zone. To calculate the absorption we assume that transverse waves are attenuated by the single collinear process $S+S-S$, with an absorption coefficient given in the LR zone by

$$\alpha_s \approx 3.5 \times 10^{-17} \Omega T^4 \gamma_{SS}^2 J_4 \left(\frac{267}{T} \right) \times \arctan \frac{2\Omega\tau_K}{1 + (267/T)^2 (x\Omega\tau_K)^2}, \quad (6)$$

and in the large- $\Omega\tau$ zone by

$$\alpha_s \approx 1.13 \times 10^{-12} [A_T T^6 6! \zeta(6) + B_T T^7 3! \zeta(3)]. \quad (7)$$

The generalized Grüneisen parameter obtained from the best fit to the data in the LR zone is the same for both frequencies and is listed in Table I. It is not possible with the present model to calculate γ_{SS}^2 in a relatively simple way. Retaining terms in $\sin\theta$ in the averaging of $\gamma_{SS}(\theta, \phi)$ results in a leading term in the absorption containing the factor $(1 - \beta^2)$ which is negative for normal dispersion. This difficulty is also present for shear waves along other high-symmetry directions and may be eliminated when a more exact treatment of anisotropy is considered in the calculation of the average γ_{ij} 's in quartz.³ The solid-line upper curves in Figs. 1 and 2 show the calculated absorption for $x=3$ in the LR zone. As was true of longitudinal waves, we found that Eq. (6) could also be extended below its calculated range of applicability ($24^\circ\text{K} < T < 28^\circ\text{K}$ for 1.25 GHz and $24 < T < 26.8^\circ\text{K}$ for 2.10 GHz), but this extension again fails to

account correctly for the observed temperature dependence in the large- $\Omega\tau$ zone.

The results of a best fit of Eq. (7) to the data for temperatures in the range of large- $\Omega\tau$ (T below 16 and 14 °K, respectively, for the upper and lower frequencies) is shown in the upper curve of Fig. 4. Best agreement was obtained for $A_T = 0.37^\circ\text{K}^{-4} \text{sec}^{-1}$ and $B_T = 52.7^\circ\text{K}^{-5} \text{sec}^{-1}$. This lower value of A_T is consistent with the fact that our *ac* crystals were of better quality (gave a longer hypersonic echo train at 4 °K) than our *X* cuts. The larger value of B_T indicates that transverse thermal phonons in quartz have a shorter normal relaxation time τ_N than longitudinal thermal phonons. This is in accordance with the larger observed absorption for microwave frequency transverse phonons and also with findings derived from the thermal conductivity analyses in other materials which consider explicitly thermal conduction by both transverse and longitudinal phonons.¹⁸ For $x=3$ and 9 GHz, $\Omega\tau_K(1 - \beta_T)$ is greater than 1 from 10 to over 30 °K. The reported absorption² at this frequency and temperature range varies, however, only as $T^{4.3}$ which seems to indicate that the samples used in the hypersonic measurements may have impurities in sufficient concentration to exclude observation of the large- $\Omega\tau$ -zone absorption.

CONCLUSIONS

We have compared measurements of the low-temperature hypersonic absorption in quartz with calculations for a dispersive anisotropic continuum with finite thermal-phonon relaxation times. It was found possible both from theory and from the experimental results to identify two temperature ranges: (a) One is the Landau-Rumer zone, where the absorption coefficient behaves close to ΩT^4 , and where a dominant-phonon approximation which uses the thermal conductivity relaxation time accounts very well for the temperature and frequency dependence of the observed absorption. Here it is also possible to find reasonable estimates of the anharmonic coupling constant or generalized Grüneisen parameter for the effective phonon interactions. (b) The other is the zone where $\Omega\tau$ is very large and the dominant-phonon approximation was not found adequate. Here the higher-temperature dependence of the frequency-independent absorption is very well represented by an approximation which includes reasonable assumptions concerning the variation with temperature and frequency of the thermal-phonon relaxation times. This approximation has the further advantage that direct information about the strength of the interactions of thermal phonons could be obtained from the hypersonic data at the lowest temperatures. Discrepancies which remain between the magnitude of the

calculated and measured average Grüneisen parameters for some interactions are attributed to the inability of the simplified model to account appropriately for elastic and anharmonic anisotropy.

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Determination of the Elastic Constants of Xenon Single Crystals by Brillouin Scattering*

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Single crystals of xenon were grown and Brillouin spectra for various crystal orientations were obtained at a temperature of 156 °K. The spectra exhibited one longitudinal component and either one or two transverse components, depending on crystal orientation. From the observed Brillouin frequency shifts, the adiabatic elastic constants for xenon were evaluated. At 156 °K, $c_{11} = 2.98 \pm 0.05$, $c_{12} = 1.90 \pm 0.04$, $c_{44} = 1.48 \pm 0.04$ in units 10^{10} dyn/cm² and the elastic anisotropy was found to be 2.74 ± 0.30 . These values were compared with calculated values based on recent theories of lattice dynamics. Good agreement was obtained only with calculations that include all-neighbor interactions. It was also observed that the relative intensities of the Brillouin components were strongly dependent on crystal orientation. An analysis of this variation in intensity gave values of the ratios of the elasto-optic constants for xenon.

I. INTRODUCTION

For many years there has been considerable theoretical and experimental interest in the properties of the rare-gas crystals neon, argon, krypton, and xenon.¹ These elements crystallize in face-centered cubic lattices under the influence of van der Waals interatomic forces. The apparent simplicity of the van der Waals interaction makes these crystals attractive as examples of perfect monatomic lattices and many theories of their lattice dynamics have been developed. In part these theories have been tested against available experimental data on the bulk properties of the rare-gas

solids.^{1,2} Only recently have techniques for growing rare-gas crystals been developed to the state that single crystals a few millimeters in size can be grown.³ With such crystals, Simmons and co-workers⁴ have been able to obtain accurate measurements of the lattice parameter, isothermal compressibility, thermal expansion, and other thermodynamic properties. However, to properly analyze the dynamical properties of a lattice, and thus to probe the interatomic interaction, accurate measurements of the elastic constants are required. In spite of the improved techniques for growing rare-gas crystals, the elastic constants have been difficult to determine experimentally. For this