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Akhiezer damping and the thermal conductivity of pure and impure dielectrics

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The ultrasonic attenuation of pure and impure dielectric crystals in the high-temperature or Akhiezer regime is derived in a relaxation-time approximation which includes both dispersion and anisotropy of all relevant thermal phonon parameters. The attenuation coefficient results are proportional to the product of the thermal conductivity for temperature gradients along the sound-wave propagation direction K_k times an average of the phonon-mode Grüneisen parameter weighted by K_k. Experimental data of the attenuation in the ultrahigh-frequency band at temperatures between 80 and 300 K, in pure crystals of Si, Ge, TiO_2, MgO, and SiO_2, are used to extract the temperature dependence of the effective ultrasonic Grüneisen parameter (UGP). For the first three crystals the temperature dependence of the UGP is qualitatively similar to that of the thermal GP squared, while its magnitude is in good agreement with calculations for the anisotropic continuum model. For neutron-irradiated quartz and for Ge-Si crystalline alloys the UGP should also be dependent on the concentration of defects or impurities. This dependence removes the discrepancies between the behavior of the thermal conductivity and of the ultrasonic attenuation of imperfect dielectrics. Independent available experimental evidence is presented in support of the present explanation of the above discrepancies.

I. INTRODUCTION

The absorption of sound waves in insulators for $\Omega\tau<1$ (Ω is the sound-wave angular frequency and τ is the thermal-phonon relaxation time), has been the subject of an extensive literature from Akhiezer's original paper[1] to more recent review articles and comments.[2-5] Concisely, sound dissipation is due to the modulation of the thermal-phonon frequencies caused by the strain of the sound wave. The perturbed phonons relax towards local equilibrium via anharmonic phonon-phonon collisions in an entropy-producing process which damps the wave. For $\hbar\Omega \ll kT$ (where T is the absolut temperature) a linearized-Boltzmann-equation treatment with heuristic assumptions as to the form of the collision integral is sufficient to calculate the absorption coefficient for all $\Omega\tau$.[4,6,7] Acoustic dispersion and anisotropies may be formally included in this treatment but the resulting general expression for the attenuation coefficient is too complex for direct application to the experimental data. The main difficulty being that one lacks sufficiently detailed knowledge about the dispersion and polarization dependence of such relevant microscopic quantities as the Grüneisen parameters (GP) and relaxation times of the thermal-phonon modes coupled to the external wave. To obtain manageable expressions for the attenuation one resorts to drastic approximations. For example, neglecting the wave-vector dependence of the mode GP, and disregarding local equilibrium effects, and for a constant phonon relaxation time equal to the thermal conductivity average, one obtains for the attenuation coefficient

$$\alpha=(3KT\Omega^2/\rho c^3 s^2)\gamma^2. \qquad (1)$$

Here K is the thermal conductivity of the material, s and c are the Debye average and wave velocities, respectively, ρ is the density, and γ^2 is an adjustable effective anharmonic parameter independent of T. Besides its simplicity, Eq. (1) relates the attenuation to the thermal conductivity, a fact that has been used to establish tendencies and to roughly predict the magnitude of the ultrasonic absorption, from known thermal and acoustic properties.[8-12] It is possible to estimate the value of γ^2 in the anisotropic-continuum model.[2,13] When applied to the experimental data of pure crystals, Eq. (1) agrees with the observed frequency dependence of the attenuation, but it fails to predict the correct temperature dependence unless γ^2 is also allowed to vary with temperature. The need for a temperature-dependent anharmonic parameter in the attenuation has been noted by several authors.[10,14,15]

An additional difficulty arises in the case of impure or imperfect crystals: the proportionality between the sound attenuation and the thermal conductivity predicted by Eq. (1) does not seem to hold. Measurements by Keller[16] and Lemanov *et al.*[17] in Ge-Si crystalline alloys show that the attenuation was a much weaker function of atomic composition than the thermal conductivity. Furthermore, the attenuation of longitudinal waves was markedly less sensitive to increases in concentration than that of shear waves. This discrepancy led Maris[18] to propose a phonon relaxation mechanism for the extreme impure limit whereby the attenuation was independent of the impurity (elastic) relaxation time. In a recently published paper Logachev[19] has pointed out some divergencies in the former work for the case of point impurity scattering.

It has been shown[20] that if in the general expression for the attenuation coefficient one averages the microscopic GP weighted by the mode thermal conductivity, one can obtain an expression for the attenuation coefficient formally identical to Eq. (1) without the need to introduce approximations about the anisotropy and dispersion of the various phonon properties. The magnitude and temperature dependence of the resultant effective "ultrasonic" Grüneisen parameter (UGP) are obtained from the attenuation data in the Ω^2 range. Such an approach, which had been previously proposed in the constant-phonon-relaxation-time approximation for cubic crystals,[14,21] gives some insight as to origin of the similarities or differences between the ultrasonic and thermal GPs with reference to the phonon modes which are effectively coupled to the sound wave. A direct application of this formulation to the case of Ge-Si alloys has explained the discrepancy between their thermal and acoustic properties by the change in the mean lattice anharmonicity due to increasing concentration of impurities. In addition, these changes were found to be larger for longitudinal than for shear waves.

In the present work we extend the application of the above formulation to a number of nominally pure dielectrics whose attenuation was found to vary as Ω^2 from 80 to 300 K. As the original treatment[20] was developed for a single (inelastic) relaxation time, we present here the results of its extension to the case when simultaneous elastic (e) and inelastic (i) scattering processes limit the thermal-phonon lifetime. In this way we are able to apply the results more appropriately to the examination of the attenuation of crystalline Si-Ge alloys and of neutron-irradiated quartz. Elastic and thermal data are also presented for other crystalline alloys systems which lend support to the proposed correlation between impurities or defects and the mean anharmonic properties of the lattice.

II. ATTENUATION COEFFICIENT FOR PURE AND IMPURE DIELECTRICS

Following the method of Woodruff and Ehrenreich[9] the attenuation coefficient of a sound wave of polarization j, radian frequency Ω, propagating in a crystal with velocity c_j along $\vec{k}$ can be written in the Akhiezer limit, $\Omega^2\tau_n^2 \ll 1$, as

$$\alpha_j = \frac{\Omega^2 T}{\rho c_j^3} \sum_{\vec{q},n} C_{\vec{q},n}\,\tau_n(\vec{q}) \left[\gamma_{nj}^2(\vec{q}) - \gamma\gamma_{nj}(\vec{q})\left(1 - \frac{\beta_n^2}{c_j^2}\right)\delta_{jL}\right]. \tag{2}$$

In this expression $C_{\vec{q},n}$ is the specific heat of the phonon mode $(\vec{q},n)$, $\beta_n = \vec{v}_n \cdot \hat{k}$ is the projection of the phonon velocity along the sound-wave propagation direction, $\gamma_{nj}(\vec{q})$ is the generalized GP which describes the change in the mode frequency caused by the strain of the sound wave,[22] γ is the calculated volume GP, and δ_{jL} is Kronecker's δ. In Eq. (2), terms multiplied by δ_{jL} arise from local equilibrium effects associated with longitudinal sound waves, the last term representing classical thermoelastic losses. Equation (2) can be written in a more concise form by performing a weighted average with the mode thermal conductivity for temperature gradients along the wave propagation direction,

$$K_k(\vec{q},n) = \tfrac{1}{3} C_{\vec{q},n}\,\beta_n^2 \tau_n, \tag{3}$$

and defining an ultrasonic mode GP as

$$\Gamma_{nj}^2(\vec{q}) = (s^2/\beta_n^2)\left[\gamma_{nj}^2 - \gamma\gamma_{nj}(1 - \beta_n^2/c_j^2)\,\delta_{jL}\right]. \tag{4}$$

We then have for the attenuation coefficient

$$\alpha_j = \frac{3K_k T\Omega^2}{\rho c_j^3 s^2}\,\Gamma_j^2, \quad \Omega^2\left(1 - \frac{\beta_n}{c_j}\right)^2 \tau_n^2 \ll 1, \tag{5}$$

where

$$\Gamma_j^2 = \sum_{\vec{q},n} \frac{K_k(\vec{q},n)\,\Gamma_{nj}^2(\vec{q})}{K_k} \tag{6}$$

and

$$K_k = \sum_{\vec{q},n} K_k(\vec{q},n). \tag{7}$$

As described in Sec. I, the above expression for the attenuation retains the simplicity of the original qualitative expression given by Eq. (1), and provides its formal justification. The new effective UGP, Γ_j^2, may be calculated if sufficient knowledge about the magnitude, dispersion, and anisotropy of its constituents is available. As this is not in general the case, it seems more fruitful to use the ultrasonic measurements to determine the temperature dependence and magnitude of Γ_j^2 for comparison with the volume GP or with predictions from specific models.

For impure dielectrics the main difference with the previous cases is that the collision integral in Boltzmann's equation will now have both inelastic and elastic relaxation terms for each phonon mode $(\vec{q},n)$:

$$\left(\frac{\partial f_n}{\partial t}\right)_{\text{coll}} = -(f_n - f'_{ni})\,\tau_i^{-1} - (f_n - f'_{ne})\,\tau_e^{-1}. \tag{8}$$

To simplify the notation the branch index n and the phonon wave vector $\vec{q}$ will not be included subsequently in the relaxation times. The equilibrium distribution function of the phonons in the presence of the perturbing strain $S_j(\vec{r},t)=S_j e^{i(\vec{k}\cdot\vec{r}-\Omega t)}$ is given by $f_n(\vec{q},\vec{r},t)$. As described by Woodruff and Ehrenreich and others[9,18,19] each scattering process will relax the distribution function $f_n(\vec{q},\vec{r},t)$ towards Planck's distributions f'_{ni}, f'_{ne} which are characterized by the perturbed energy of the phonons and their local "effective" temperatures T_i and T_e. The local effective temperature for elastic processes is assumed in general to depend on $\vec{q}$. By analogy with the pure dielectric case, we write the fractional changes in local temperature about the equilibrium crystal temperature T as

$$\begin{aligned} \delta T_i/T &= (\gamma'_j + i\gamma''_j)S_j(\vec{r},t),\\ \delta T_e/T &= [\xi'_q(\vec{q}) + i\xi''_j(\vec{q})]S_j(\vec{r},t). \end{aligned} \tag{9}$$

Calculating the attenuation coefficient by the method of Woodruff and Ehrenreich with $\tau^{-1}=\tau_i^{-1}+\tau_e^{-1}$ we obtain, in the approximation $\Omega^2(1-\beta_n/c_j)^2\tau^2 \ll 1$:

$$\alpha_j = \frac{\Omega^2 T}{\rho c_j^3}\sum_{\vec{q},n} C_{\vec{q},n}\tau\Big[\gamma_{nj}^2(\vec{q}) + \gamma_{nj}(\vec{q}) \times\left(\frac{\gamma'_j\tau}{\tau_i}+\frac{\xi'_j\tau}{\tau_e}+\frac{\gamma''_j}{\Omega\tau_i}+\frac{\xi''_j}{\Omega\tau_e}\right)\delta_{jL}\Big]. \tag{10}$$

The quantities γ'_j, γ''_j, ξ'_j, and ξ''_j are determined by the system of equations

$$\begin{aligned} \sum_{\vec{q},n}\hbar\omega_n(\vec{q})\left(\frac{\partial f_n}{\partial t}\right)^i_{\mathrm{coll}} &= 0,\\ \sum_{\vec{q},n}\hbar\omega_n(\vec{q})\left(\frac{\partial f_n}{\partial t}\right)^e_{\mathrm{coll}} &= 0, \end{aligned} \tag{11}$$

which connote conservation of the total energy of the phonon system under both elastic and inelastic scattering processes. For elastic collisions, Maris[18] and Logachev[19] made use of the invariance of the number of phonons at fixed frequency instead of the above constraints. We found it more convenient here to analyze the results with reference to the pure dielectric case where the energy-conservation condition is normally used. Some features of Eq. (10) are worth mentioning at this point: (a) the expression is symmetrical in τ_i and τ_e so that for $\tau_e\to\infty$ one recovers the attenuation for pure dielectrics. In this case, from the first part of Eq. (11) we obtain

$$\gamma'_j = -\gamma \quad \text{and} \quad \gamma''_j = \Omega\sum_{\vec{q},n} C_{\vec{q},n}\gamma_{nj}\beta_n^2\tau_n \Big/ c_j^2 \sum_{\vec{q},n} C_{\vec{q},n},$$

and substitution in Eq. (10) then yields Eq. (2). (b) For pure shear waves ($j=T$) the attenuation coefficient is formally identical to the corresponding expression for pure dielectrics except that τ is now the combined relaxation time $(\tau_i^{-1}+\tau_e^{-1})^{-1}$. In this instance the anharmonic parameter can readily be averaged with the mode thermal conductivity and the attenuation results are again proportional to K_k.

For longitudinal waves ($j=L$) local equilibrium effects yield the terms in parentheses in Eq. (10) which make the expression for the attenuation more cumbersome. However, since τ is a common factor under the summation sign, we may perform the average as described above for the pure dielectric case. We can then write the attenuation coefficient proportional to the appropriate component of the thermal conductivity times an effective UGP. To test the validity of this procedure we have solved Eq. (11) in several approximate ways[23] and found that the local equilibrium terms depend only on averages of the combined relaxation time τ. For example, for relaxation times independent of branch number n and for γ_{nj} dependent only on the direction of $\vec{q}$, the local equilibrium terms inside the square brackets of Eq. (10) have results proportional to:

$$-\langle\gamma_{nj}\rangle\langle\langle\tau\rangle\rangle + \langle\gamma_{nj}\rangle\gamma''/\Omega, \tag{12}$$

where the angular brackets $\langle\ \rangle$ and $\langle\langle\ \rangle\rangle$ represent specific-heat-weighted averages over n and $\vec{q}$, respectively. The first term in Eq. (12) is common to the pure dielectric case and the magnitude of the second term will give a contribution to the attenuation of the same order of the classical thermoelastic losses. Several other approximations make it possible to write simplified expressions for the attenuation coefficient.[19] As the validity of these approximations is difficult to assess for real solids, we prefer instead to use the averaging procedure referred to above and express the attenuation for *both* pure and impure dielectrics as in Eq. (5):

$$\alpha_j = (3K_k T\Omega^2/\rho c_j^3 s^2)\,\Gamma_j^2.$$

The effective UGP, Γ_j^2, which can be obtained from the attenuation data, will thus contain the effects of temperature and/or defect concentration on the average anharmonicity of the lattice as sampled by the sound waves.

In the applications that follow we will present, in addition to ultrasonic data, some independent experimental evidence which shows that the average anharmonicity in materials with impurities or defects does change markedly with concentration. For the cases presented, this change is stronger for longitudinal than for shear wave polarization.

III. EXPERIMENTAL METHODS

By conventional cryogenic pulse-echo techniques[24] we measured the temperature and frequency dependence of the ultrasonic attenuation in nominally pure crystals of Si, SiO_2, TiO_2, and MgO for various polarization and propagation directions. Nonresonant crystal holders[25] and CdS film transducers[26] provided sufficient flexibility to study the attenuation for different polarizations in a frequency range from 0.2 to 1.0 GHz at temperatures above 80 K. The magnitude of the attenuation coefficient was determined by fitting a calibrated exponential to the pulse pattern while its variation with temperature was obtained by the use of an automatic ultrasonic attenuation recorder.[27]

The frequency dependence of the attenuation coefficient was determined at both extremes of temperatures to ensure that for all samples measured the attenuation was proportional to Ω^2. In most cases the temperature dependence of the attenuation was determined at the lowest frequency of the experimental range for which the best pulse pattern was obtained.

Neutron irradiation of our quartz samples was carried out in the core irradiation facilities of the RV-1 research reactor with samples and dosimeters inside an aluminum can. Dose refers to intregrated thermal neutron flux.

IV. RESULTS AND APPLICATIONS

A. Pure dielectrics

Measurements of the frequency dependence of the attenuation at room temperature showed that in our samples the attenuation depends quadratically on frequency. Similar results have been reported for these and other dielectrics for frequencies up to 1 GHz.[5] The same frequency dependence was observed at liquid-nitrogen temperatures although for some samples a weakening of the frequency dependence is already noticeable above 0.7 GHz. For this reason the temperature dependence of the attenuation for a given material or polarization was studied at the lowest convenient frequency in the Ω^2 range.

The resulting effective UGP obtained by fitting Eq. (5) to our measurements or to data published by other authors[2,14] are shown in Figs. 1–4. Values for ρ, c_j, and s were taken from an unpublished compilation by Anderson.[28] The appropriate component of the thermal conductivity was used, when available, in the calculation of Γ_j^2 but this is not in general the case. For comparison purposes the squares of the measured thermal GPs[29-32] are also shown in the graphs.

With the exception of quartz and MgO, the temperature dependence of the Γ_j^2 and γ^2 results are qualitatively similar over most of the experimental range. Bearing in mind that one is comparing different averages of the GPs, it may be tentatively concluded that for these materials the contribution

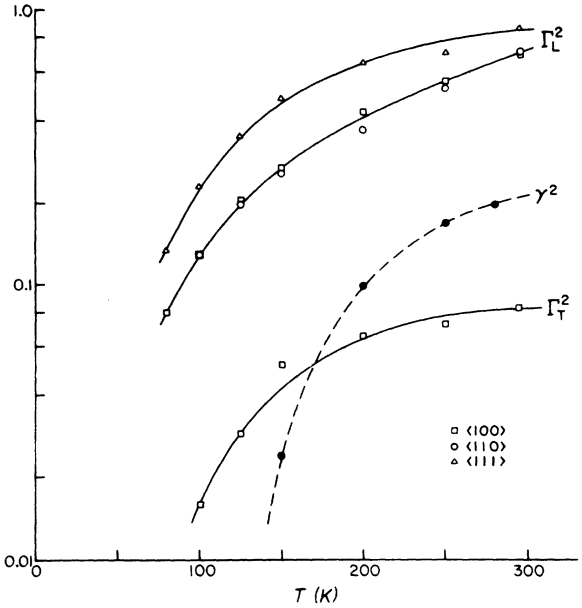

FIG. 1. Temperature dependence of the ultrasonic and thermal Grüneisen parameters of silicon. L and T indicate sound wave polarization.

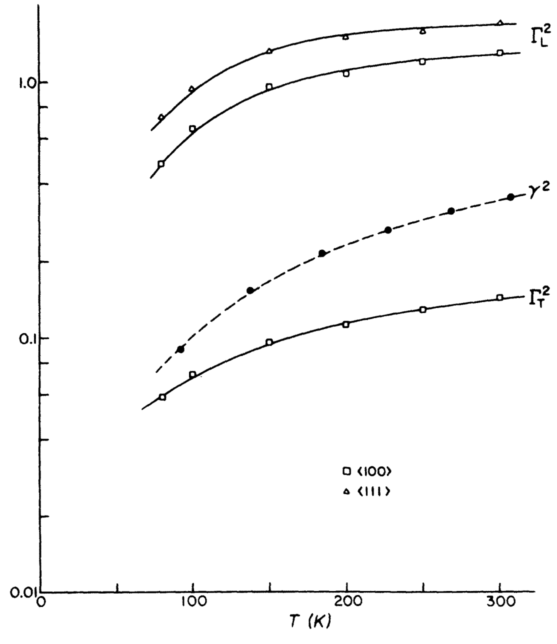

FIG. 2. Temperature dependence of the ultrasonic and thermal Grüneisen parameters of germanium (Refs. 2 and 14). L and T indicate sound wave polarization.

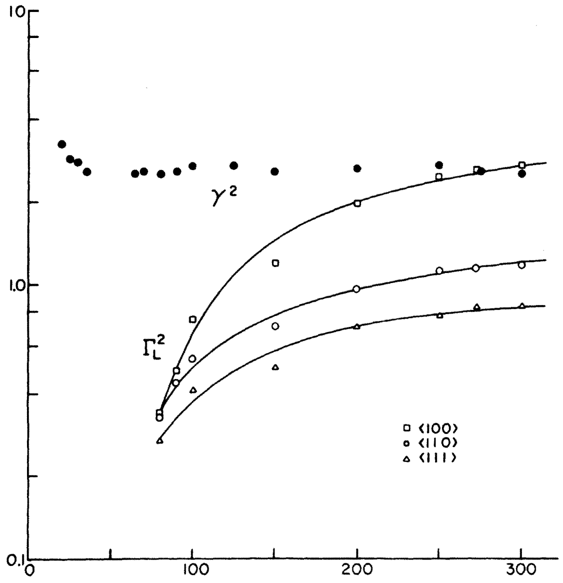

FIG. 3. Temperature dependence of the ultrasonic and thermal Grüneisen parameters of MgO for longitudinal waves.

to both parameters comes from the same branches of the phonon spectrum. This would not be the case for quartz and MgO.

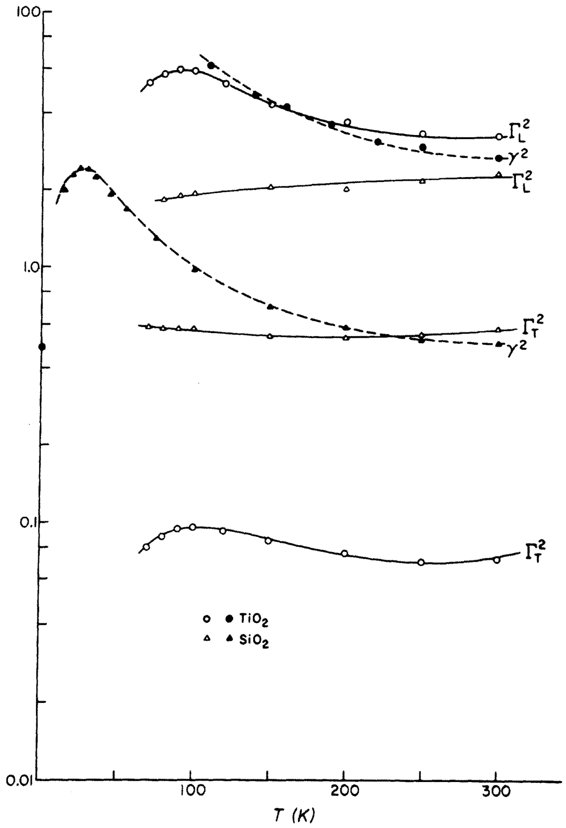

FIG. 4. Temperature dependence of the ultrasonic and thermal Grüneisen parameters. *L* stands for longitudinal waves along the *c* and *x* axes, respectively, and *T* indicates shear waves along the *c* and *AC* axes, respectively, of rutile and quartz crystals.

At room temperature one may make a comparison of the measured Γ_j^2 with its value calculated from elasticity theory by assuming that τ_n is independent of branch number n, and γ_{nj} depends only on the direction of the phonon wave vector and n. For this approximation, from Eqs. (2) or (5), Γ_j^2 is roughly equal to $\langle\gamma_{nj}^2\rangle$, the specific-heat-weighted average of the square mode GP. Local equilibrium terms are thus neglected in the attenuation of longitudinal waves. Table I compares the value of $\langle\gamma_{nj}^2\rangle$ calculated by the method of Mason and Bateman[2,13] with the value of Γ_j^2 obtained from the measured ultrasonic attenuation at room temperature for a number of crystals. Considering the limitation of the anisotropic continuum model, and that the various properties involved in the calculation of Γ_j^2 were not in general measured in the same

TABLE I. Comparison of the room-temperature Grüneisen parameter and its square mean, calculated for the anisotropic continuum model, and their values obtained from thermal and ultrasonic measurements.

Direction	Crystal	Longitudinal				Shear		References
		Γ^2	$\langle\gamma^2\rangle$	$\langle\gamma\rangle$	γ	Γ^2	$\langle\gamma^2\rangle$	
⟨100⟩	Si	0.70	0.71	0.50	0.45	0.08	0.17	2, 13, 29
	Ge	1.21	1.10	0.67	0.60	0.14	0.13, 0.35	2, 29, 30
	MgO	2.72	4.67	1.44	1.60	0.17	0.39	13, 31
⟨110⟩ [a]	CaF_2	1.56	1.83	1.15	1.90	0.33	0.20	5, 32, 33
	BaF_2	1.78	1.68	0.73	1.57	0.19	0.38	5, 33, 34
	SrF_2	1.23	1.18	0.93	1.58	0.28	0.05	5, 21, 33
c axis	TiO_2	3.24	···	0.70	1.64	0.07	···	35, 36
x axis	SiO_2	2.30	1.38	0.71	0.70	···	···	37
AC axis	SiO_2	···	···	···	···	0.57	0.62	37

[a] Shear polarization $\langle 1\,\bar{1}\,0\rangle$.

samples, the agreement is surprisingly good. By calculation we have found that for cubic crystals, both the values of Γ_j^2 and $\langle \gamma_{nj}^2 \rangle$ depend more strongly on the polarization (j) than on the sound wave propagation direction. Note that for longitudinal waves the disagreement between the values of Γ_j^2 and $\langle \gamma_{nj}^2 \rangle$ is at worst less than a factor of 2. For some cubic crystals this disagreement is so small that a mutual cancellation of the terms in the attenuation coefficient due to local equilibrium effects is suggested. For these cases, the observation of Lewis[11] that collinear phonon interactions (for which $\beta_n^2 \simeq c_j^2$) still make a large contribution to the attenuation of longitudinal waves in dielectrics at room temperature is a plausible explanation for the agreement found between the two anharmonic parameters.

It is interesting to note from Figs. 1-4 the difference between the temperature dependence of the thermal GP of Si and Ge and those of quartz and rutile: in the latter materials the thermal GP increases with decreasing temperature. Such an anomalous behavior of γ^2 has been attributed to the contribution that low-lying optical phonon modes make to the thermal expansion of these materials at relatively high temperatures.[38] Taking into account the contribution of optical modes to the anharmonicity of rutile, Shaner[39] has predicted that γ should pass through a peak around 100 K and then decrease to its elastic value of 0.7 as T approaches 0 K. The fact that the temperature dependence of Γ_j^2 for this material is in accordance with these predictions indicates that the sound wave in TiO_2 is effectively coupled to the optical modes. This coupling seems to be much weaker for quartz and MgO. Further evidence of the contribution that optical modes make to γ for some crystals is that its measured value is significantly greater than the value $\langle \gamma_{nj} \rangle$ calculated from the elastic constants. The corresponding columns of Table I indicate that this is the case for TiO_2 and the alkaline-earth fluorides at room temperature.

B. Imperfect or impure dielectrics

Figure 5 shows Γ_j^2 as a function of dose for neutron irradiated natural quartz obtained by fitting Eq. (5) to our data. Also shown in the figure are data of Bommel and Dransfeld[8] and of Fitzgerald *et al.*[40] at 90 K. The thermal conductivity used in the calculations was read from the smooth curve shown in the figure which is drawn through the experimental points reported by Berman[41] for natural irradiated quartz. Our results are only qualitative since the magnitudes of Γ_j^2 calculated from our data have been changed arbitrarily to coincide, at low doses, with that obtained from previous ultrasonic data over a wider dose range.[8,40] However, the increase in Γ_j^2 with the concentration of radiation-induced defects is quite evident. The general trend on the curves of Γ_j^2 vs dose indicates that the effect is greater for longitudinal than for shear waves, but more data are needed for the latter polarization at higher neutron doses.

To study the dependence of the effective UGP on impurity concentration we have selected measurements of the ultrasonic attenuation at room temperature in Ge-Si crystalline alloys by Keller[16] at 0.64 GHz, and by Lemanov *et al.*[17] at 1 GHz. The lower half of Fig. 6 shows the variation of the ultrasonic attenuation and thermal conductivity with atomic composition, and in its upper part Fig. 6 shows the resultant Γ_j^2 calculated from Eq. (5). Also shown for Ge-rich alloys is the volume GP used by Abeles *et al.*[42] to fit their thermal conductivity measurements. The dependence of Γ_j^2 on concentration is seen to be quite large, particularly for Si-rich alloys, and clearly stronger for longitudinal sound waves. For Ge-rich alloys the increase of Γ_j^2 with composition is weaker than in Si-rich alloys, but it is nevertheless four times greater in $Ge_{91}Si_{0.9}$ than in pure germanium.

There seems to be little direct experimental data

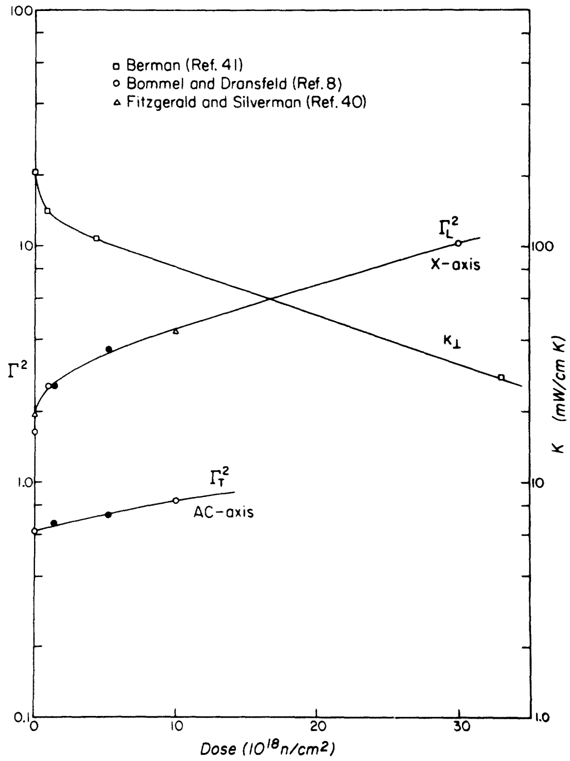

FIG. 5. Thermal conductivity and ultrasonic Grüneisen parameter of natural quartz as a function of neutron irradiation dose at 90 K. Filled circles represent our data. L and T indicate sound wave polarization.

on the effect of composition on the anharmonicity of Ge-Si crystals. The results of Abeles *et al.*[42] for the γ of Ge-rich alloys shown in Fig. 6 are perhaps too indirect to accurately reflect the changes. We have found in addition reports of the pressure derivatives of the elastic constants and of the third-order elastic constants of some crystalline alloy systems[43-46] from which one may calculate an average elastic GP.[47] The results of such calculations are shown in Fig. 7 for[43] Be-Cu and[44] Cu-Zn alloys. For the first system γ_e^2 changes much more with composition for longitudinal than for shear polarization even though the range of composition reported is only a few tenths of an atomic percent. The same behavior is observed for shear polarization in Cu-Zn, where 30-at.% Zn produces only a 10% change in γ_e^2. Measurements in Cu-Ni alloys[45] indicate a monotonic increase in the value of the third-order elastic constants from pure copper to pure nickel. For α-phase Cu-Al alloys[46] the change in the third-order elastic constants with composition reflects itself in a monotonic decrease in the calculated high-temperature GP of approximately 0.5% per at.% Al. This limited evidence is in general in agreement with our explanation of the discrepancies between the acoustic and thermal properties of crystals with defects in terms of a concentration-dependent effective UGP. Clearly, more experimental data are desirable on the effect of impurities or defects on the anharmonic properties of dielectrics, such as thermal expansion coefficients or third-order elastic constants.

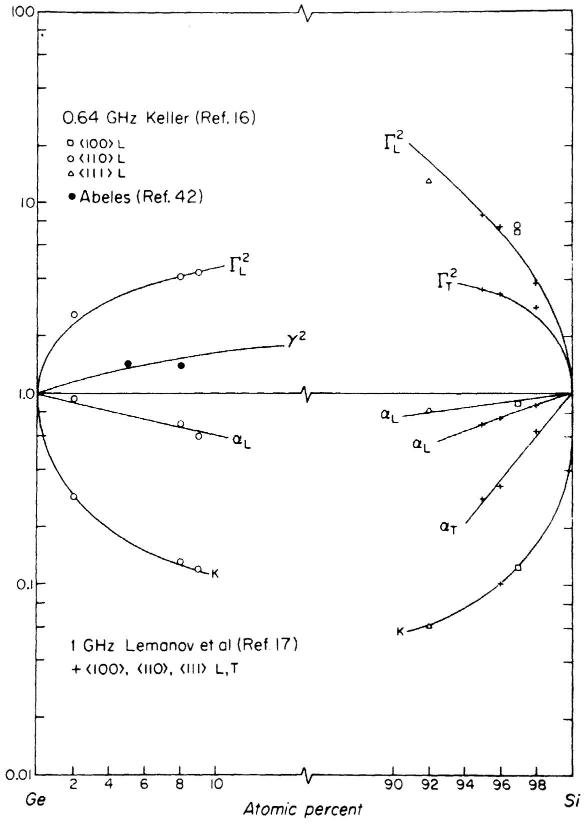

FIG. 6. Relative variation of the ultrasonic attenuation, thermal conductivity, and Grüneisen parameters with composition in Ge-Si crystalline alloys. *L* and *T* indicate sound wave polarization.

V. CONCLUSIONS

It is shown that the high-temperature ultrasonic attenuation in pure and impure dielectrics crystals have results proportional to the thermal conductivity for temperature gradients along the sound wave propagation direction times an average effective ultrasonic Grüneisen parameter, which is temperature dependent. For pure dielectrics the magnitude of this parameter compares well with that calculated from the elastic constants in the anisotropic continuum model, while its temperature dependence results are qualitatively similar, in some cases, to that of the square thermal Grüneisen parameter. For impure dielectrics the ultrasonic Grüneisen parameter should also be a function of the concentration of impurities or defects. This variation explains the discrepancies between the strong dependence of the thermal conductivity and the relative insensitiveness of the

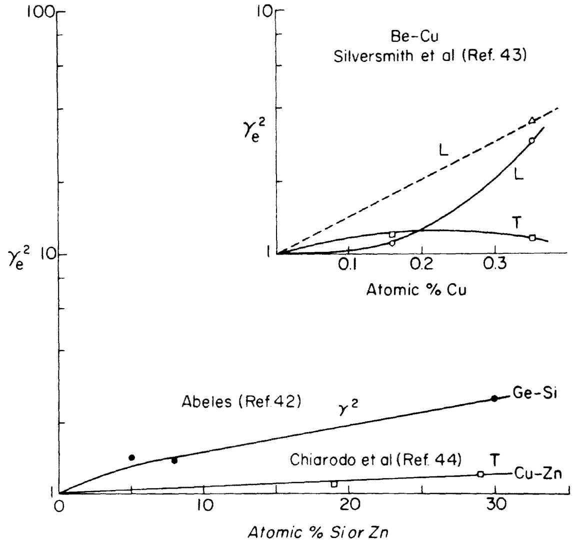

FIG. 7. Relative variation of the elastic Grüneisen parameter with composition in two crystalline alloys. *L* and *T* indicate sound wave polarization.

attenuation on the impurity concentration. Experimental data related to the variation with composition of the third-order elastic constants, and of the pressure derivatives of the second-order elastic constants in some crystalline alloys are in agreement with the present explanation of the above discrepancies.

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