by the stress-free equilibrium condition which is an essential part of the Cauchy relations. Of course, one will obtain the same value for the elastic constants whichever approach one uses, since they are basically the same. This can readily be verified by evaluating Cousins's expression for C_{IJK} for nearest-neighbor interactions only.

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APPENDIX

For convenience we quote equations from I that we use in the preceding text:

$$2V_{z}\hat{C}_{ij,kl} = -\sum_{n} \phi_{ij}^{on} X_{k}^{n} X_{l}^{n}; \qquad (A1)$$

$$2V_z \hat{C}_{ij,rs,kl} = -\sum_{n,p} \phi_{ijr}^{onp} X_s^p X_k^n X_l^n ; \qquad (A2)$$

$$C_{11} = \hat{C}_{11,11} - C_1 ,$$

$$C_{12} = 2\hat{C}_{12,12} - \hat{C}_{11,12} + C_1 ,$$

$$C_{44} = \hat{C}_{22,33} - C_1 ,$$

¹R. A. Coldwell-Horsfall, Phys. Rev. <u>129</u>, 22 (1963). ²C. S. G. Cousins, J. Phys. C <u>4</u>, 1117 (1971).

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$$C_{111} = \hat{C}_{11,11,11} - 3C_{11} ,$$

$$C_{112} = \hat{C}_{11,22,11} - C_{12} ,$$

$$C_{166} = \hat{C}_{11,12,12} - C_{12} - 2C_{66} ,$$

$$C_{123} = 2\hat{C}_{12,33,12} - \hat{C}_{11,33,22} + C_{12} ,$$

$$C_{456} = \hat{C}_{33,12,12} - C_{66} ,$$

$$C_{144} = 2\hat{C}_{12,23,13} - \hat{C}_{11,23,23} - C_{12} ;$$
(A3)

central-force constants:

$$\phi_{ijk}^{ooh} = X_i^h X_j^h X_k^h$$

$$\times \left(\frac{1}{r^3} \phi^{\prime\prime\prime}(r) - \frac{3}{r^4} \phi^{\prime\prime}(r) + \frac{3}{r^5} \phi^{\prime}(r)\right)$$

$$+ \left(X_i^h \delta_{jk} + X_j^h \delta_{ik} + X_k^h \delta_{ij}\right)$$

$$\times \left(\frac{1}{r^2} \phi^{\prime\prime}(r) - \frac{1}{r^3} \phi^{\prime}(r)\right)$$
(A4)

$$\phi_{ij}^{oh} = -X_i^h X_j^h \left(\frac{1}{r^2} \phi^{\prime\prime}(r) - \frac{1}{r^3} \phi^{\prime}(r)\right) \\ -\frac{1}{r} \phi^{\prime}(r) \delta_{ij} .$$

³M. Born and K. Huang, in *Dynamical Theory of Crys*tal Lattices (Clarendon, Oxford, 1956), p. 132.

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Ultrasonic Attenuation in Two Fluorite-Structure Crystals

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The attenuation of longitudinal ultrasonic waves originating from their interaction with the phonon gas and from other thermoelastic causes has been evaluated in CaF_2 and BaF_2 at 300 °K in the $\langle 100 \rangle$ and $\langle 110 \rangle$ directions using thermodynamic and elastic-constant data. The results obtained are in good agreement with the experimentally observed ones.

INTRODUCTION

For a thorough study of dielectric crystal behavior, extensive acoustic-attenuation measurements have been made in the recent past.¹⁻⁶ A major part of the attenuation in these crystals occurs because of the interaction of thermal phonons with ultrasonic waves. Generally for dielectric crystals, $\omega \tau_{\rm th} \ll 1$ at room temperature, where $\omega = 2\pi$ times the acoustic-wave frequency and $\tau_{\rm th}$ is thermal-phonon relaxation time. Now, because

of its very small relaxation time, the individual phonon loses its significance and an idea of the phonon gas having macroscopic parameters is described. The two types of attenuation in this region are (a) Akhieser loss caused by the relaxation of thermal energy between different phonon branches heated to different extents by ultrasonic waves and (b) thermoelastic loss caused by the relaxing flow of thermal energy from compressed and hotter regions towards the expanded and colder regions of the waves. (This type of loss takes

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TABLE I. Average Grüneisen constant and average square Grüneisen constant for longitudinal waves in CaF_2 and BaF_2 at 300 °K.

Direction	$\langle \gamma_i^j \rangle$	CaF_{2} $\langle \langle \gamma_{i}^{j} \rangle^{2} \rangle$	D	$\langle \gamma_i^j \rangle$	BaF_{2} $\langle (\gamma_{i}^{j})^{2} \rangle$	D
<pre>(100) (110)</pre>	1.151	1.683	8.246	0.734	1.222	8.749
	1.187	1.834	9.166	1.027	1.683	10.744

place for longitudinal waves only.) Here, we present the acoustic attenuation due to both these mechanisms for two diele tric crystals having a fluorite-type structure, viz., CaF_2 and BaF_2 , in the $\langle 100 \rangle$ and $\langle 110 \rangle$ directions at 300 °K. Further, the phonon viscosities in both these directions have been evaluated.

THEORY

The attenuation caused by the Akhieser loss⁷ mechanism is obtained from

,

$$\alpha = \frac{E_0(\frac{1}{3}D)}{2\rho V^3} \frac{\omega^2 \tau}{1+\omega^2 \tau^2}$$

which becomes for $\omega \tau \ll 1$

$$\alpha = \frac{E_0(\frac{1}{3}D)\omega^2 \tau_I}{2\rho V_I^3} \quad , \tag{1}$$

where E_0 is the thermal energy per unit volume, ω is the angular frequency of the acoustic wave, ρ is the density, and V_i is the velocity of longitudinal acoustic waves. The thermal relaxation time for longitudinal waves τ_i is twice that for shear waves^{2,3}:

$$\tau_I = 2\tau_s = 2 \frac{3K}{C_v \langle V \rangle^2} \quad , \tag{2}$$

where K is the thermal conductivity, C_{ν} is the specific heat per unit volume, and $\langle V \rangle$ is the Debye average velocity. The nonlinearity constant D in Eq. (1) is evaluated from third-order elastic moduli from

$$D = \frac{3}{E_0} \left(3 \sum_{i=1}^n E_i (\gamma_i^j)^2 - \langle \gamma_i^j \rangle^2 \rho C_v T \right) ,$$

where E_i is the thermal energy of *i*th phonon mode and γ_i^i are Grüneisen numbers for that particular mode and strain. The Debye approximation as-

TABLE II. Attenuation due to Akhieser (A) and thermoelastic (T) loss mechanisms in CaF₂ and BaF₂ at 300 °K. Units of (α/f^2) are 10⁻¹⁸ sec² cm⁻¹.

	Ca	F ₂	Ba	F ₂
Direction	$(\alpha/f^2)_A$	$(\alpha/f^2)_T$	$(\alpha/f^2)_A$	$(\alpha/f^2)_T$
(100)	2.84	0.11	9.60	0.22
〈110〉	3.16	0.12	11.78	0.42

sumes E_i to be equal for each mode. So using

$$\langle (\gamma_i^j)^2 \rangle = \frac{1}{n} \sum_{i=1}^n (\gamma_i^j)^2$$

one gets

$$D = 9 \langle (\gamma_i^j)^2 \rangle - 3 \langle \gamma_i^j \rangle^2 \rho C_v T / E_0 \quad . \tag{3}$$

The thermoelastic attenuation is calculated from

$$\alpha = \frac{\omega^2 \langle (\gamma_1^4) \rangle^2 KT}{2\rho V_l^5} \tag{4}$$

and the phonon viscosity associated with longitudinal waves is determined from

$$\eta_{l} = 2D_{l} \left(E_{0} K / C_{v} V_{l}^{2} \right) .$$
(5)

RESULTS AND DISCUSSION

The average Grüneisen constant $\langle \gamma_i^{4} \rangle$ and average square Grüneisen constant $\langle (\gamma_i^{4})^2 \rangle$ have been evaluated from elastic-constant data of Alterovitz and Gerlich⁸ and Gerlich.⁹ The values obtained for the $\langle 100 \rangle$ and $\langle 110 \rangle$ directions by averaging over 39 pure modes are given in Table I.

The C_v values of 2.65 and $1.993 \times 10^7 \text{ erg/cm}^3 \,^{\circ}\text{K}$ obtained from standard literature¹⁰ substituted into Eq. (2) give the value of the longitudinal thermal relaxation time τ_1 as 1.308 and 4.812×10⁻¹² sec for CaF₂ and BaF₂, respectively. From here, one can check that the condition $\omega \tau \ll 1$ holds well in the frequency range covered. The constant *D* obtained from Eq. (3) by taking the thermal-energy values to be 4.5 and 3.39×10⁹ erg/cm³ is of the same order as expected (Table I).

Figure 1 depicts the variation of thermal attenuation with the square of the frequency for the two crystals in the $\langle 100 \rangle$ and $\langle 110 \rangle$ directions. The calculated values are in good agreement with the experimentally observed values,¹¹ proving the validity of the idea of the phonon gas in the range $\omega \tau \ll 1$. Though the experimental attenuation in the two directions is the same, theoretically there is a little difference (Fig. 1). The total attenuation consists of two parts: the Akhieser loss and the thermoelastic attenuation. The attenuation due to the Akhieser loss mechanism is much larger than (about 30 times) that due to thermoelastic causes (see Table II). This shows that a major part of ultrasonic energy loss is used in achieving the equilibrium among various phonon branches and directions at different temperatures.

TABLE III. Phonon viscosity values. Unit is cP.

Direction	CaF_2	BaF ₂
〈100〉	1.923	4.255
$\langle 110 \rangle$	2.137	5.224

0.9

0,8

0.7 0.6 0.5

0.4

0.3

0.2

0.10

0.08

0.06

0.05

0.04

0.03

х (dB/cm) –

0

O

1

□- (100) 0- (110) }

BaF2

CaFo

5

6 7

▲- Gingist et al.⁽¹¹⁾ (Exptl.) [<100> and <110>]

o

Present values

O

2

Ż

 (Hz^2)

f²

0

Q

9 'n

9×10¹⁶

8





Since the Akhieser loss predominates over the other type of attenuation, it becomes essential to evaluate the phonon viscosity η_i , which is the controlling factor of this loss. The η_l values computed for the two directions are presented in Table III.

The evaluated attenuation obeys the variation of

¹W. P. Mason and T. B. Bateman, J. Acoust. Soc. Am. 36, 64 (1964). ²W. P. Mason, *Physical Acoustics* (Academic, New

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³W. P. Mason and T. B. Bateman, J. Acoust. Soc. Am. 40, 852 (1966).

⁴W. P. Mason, J. Acoust. Soc. Am. <u>42</u>, 2537 (1967).

⁵R. C. Hanson, J. Phys. Chem. Solids <u>28</u>, 475 (1967).

⁶M. F. Lewis, J. Acoust. Soc. Am. <u>44</u>, 713 (1968).

phonon viscosity and is of the same order for other dielectric crystals.

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⁹D. Gerlich, Phys. Rev. <u>168</u>, 947 (1968).

¹⁰American Institute of Physics Handbook (McGraw-Hill, New York, 1963).

¹¹A. D. Gingist, A. I. Morozove, and B. A. Stankoviski, Fiz. Tverd. Tela 11, 2313 (1968) [Sov. Phys. Solid State 11, 1868 (1969)].