

# ULTRASONIC ATTENUATION IN DIELECTRIC CRYSTALS

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Ultrasonic attenuation and non-linearity parameters have been evaluated for dielectric crystals RbCl, RbBr and RbI in a wide temperature range. Basic physical parameters, nearest neighbour distance and hardness parameters of the substance and Coulomb and Born-Mayer potentials have been used to obtain ultrasonic attenuation due to phonon-viscosity mechanism along  $\langle 100 \rangle$  direction of propagation for longitudinal and shear waves. Results are as expected and it has been concluded that ultrasonic attenuation is a fundamental property of the substance.

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## 1. Introduction

Ultrasonic attenuation techniques have been used as a versatile tool for the determination of internal structure and other inherent properties of solids [1-5]. Ultrasonic attenuation studies have been made for metallic [6, 7], semiconducting [8, 9] and dielectric [10] substances. Of the three dominant interactions responsible for ultrasonic attenuation in solids, electron-lattice interaction is principal cause of acoustical dissipation in metals [8], at low temperatures, while phonon-phonon interaction is the main cause of acoustical attenuation in all types of solids at higher temperatures, provided the substance is perfect, non-ferromagnetic and non-ferroelectric as established by other workers [8, 11]. The prominent mechanism for ultrasonic attenuation in solids is the Akhiezer mechanism [12], in which the attenuation results because, during sound wave propagation through solids, equilibrium distribution of thermal phonons is disturbed, and reestablishment of equilibrium is a relaxational phenomenon, in which entropy is produced and energy is lost from the sound wave.

In the present communication, we have studied ultrasonic attenuation due to phonon-phonon interaction in rubidium halide crystals viz. rubidium chloride,

rubidium bromide and rubidium iodide at different temperatures viz. 100, 200, 300 and 400 K, using nearest neighbour distance, hardness parameter, Coulomb and Born-Mayer potentials and following Mason's approach [8]. The average Grüneisen parameters, nonlinearity parameters and attenuation coefficients for longitudinal and shear waves are evaluated along  $\langle 100 \rangle$  direction of propagation.

## 2. Theory

The theory consists of two parts. In the first part a temperature variation of second and third order elastic constants has been discussed, while in the next part ultrasonic attenuation due to phonon-phonon interaction at different temperatures is to be considered.

### 2.1. Theory of elastic constants

Assuming Coulomb or electrostatic potential ( $\pm e^2/r$ ) and Born-Mayer [13] type potential  $Q(r) = A \exp(-r/\rho)$ ,  $e$ ,  $r$ , and  $\rho$  being electronic charge, nearest neighbour distance and hardness parameter, and following Brugger's [14] definition of elastic constants at absolute zero, and taking interactions up to second nearest neighbours, the second and third order elastic constants (SOEC  $C_{ij}^0$  and TOEC  $C_{ijk}^0$  in Voigt notation) are obtained as shown in Table I. Various lattice sums are shown in Table II.

TABLE I

SOEC and TOEC at absolute zero.

$$\begin{aligned}
 C_{11}^0 &= (3/2)(e^2/r_0^4)S_5^2 + (1/\rho r_0)(1/r_0 + 1/\rho)Q(r_0) \\
 &\quad + (2/\rho r_0)(\sqrt{2}/2r_0 + 1/\rho)Q(\sqrt{2}r_0) \\
 C_{12}^0 &= C_{44}^0 = (3/2)(e^2/r_0^4)S_5^{1,1} + (1/\rho r_0)(\sqrt{2}/2r_0 + 1/\rho)Q(\sqrt{2}r_0) \\
 C_{111}^0 &= (15/2)(e^2/r_0^4)S_7^3 - (1/\rho)(3/r_0^2 + 3/\rho r_0 + 1/\rho^2)Q(r_0) \\
 &\quad - (1/2\rho)(3\sqrt{2}/r_0^2 + 6/\rho r_0 + 2\sqrt{2}/\rho^2)Q(\sqrt{2}r_0) \\
 C_{112}^0 &= C_{166}^0 = -(15/2)(e^2/r_0^4)S_7^{2,1} - (1/4\rho)(3\sqrt{2}/r_0^2 + 6/\rho r_0 + 2\sqrt{2}/\rho^2)Q(\sqrt{2}r_0) \\
 C_{123}^0 &= C_{144}^0 = C_{456}^0 = -(15/2)(e^2/r_0^4)S_7^{1,1,1}
 \end{aligned}$$

TABLE II

Various lattice sums.

$S_3^1 = -0.58252$	$S_5^2 = -1.04622$
$S_5^{1,1} = 0.23185$	$S_7^3 = -1.36852$
$S_7^{2,1} = 0.16115$	$S_7^{1,1,1} = -0.09045$

According to the anharmonic theory of lattice dynamics developed by Leibfried and Ludwig [15] or Hahn et al. [16], the change in elastic constant occurs due to change in vibrational energy of the crystal. Thus adding vibrational energy contributions we get SOEC and TOEC at required temperature as:

$$C_{ij}(T) = C_{ij}^0 + C_{ij}^{\text{vib.}} \quad \text{and} \quad C_{ijk}(T) = C_{ijk}^0 + C_{ijk}^{\text{vib.}} \quad (1)$$

This theory [17, 18] has been used earlier to obtain elastic constants. The vibrational energy contributions are presented in Table III. Various  $f^n$ 's and  $G_n$ 's are given in Table IV and Table V.

## 2.2. Theory of phonon-viscosity mechanism

SOEC's and TOEC's thus obtained are used to obtain Grüneisen parameters  $\langle \gamma_i^j \rangle$  and  $\langle \gamma_i^{j^2} \rangle$ , using Grüneisen tables [8] along  $\langle 100 \rangle$  direction of propagation for longitudinal and shear waves. These Grüneisen parameters are related with Mason's nonlinearity constant as:

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

where  $C_v$  is specific heat per unit volume,  $T$  absolute temperature,  $E_0$  is the energy of the crystal,  $\langle \gamma_i^j \rangle$  and  $\langle \gamma_i^{j^2} \rangle$  are average Grüneisen number and square average Grüneisen numbers over 39 pure modes for longitudinal wave and over 18 pure modes for shear waves.

Vibrational energy contributions. TABLE III

$C_{11}^{\text{vib.}} = f^{1,1} G_1^2 + f^2 G_2$
$C_{12}^{\text{vib.}} = f^{1,1} G_1^2 + f^2 G_{1,1}$
$C_{44}^{\text{vib.}} = f^2 G_{1,1}$
$C_{111}^{\text{vib.}} = f^{1,1,1} G_1^3 + 3f^{2,1} G_2 G_1 + f^3 G_3$
$C_{112}^{\text{vib.}} = f^{1,1,1} G_1^3 + f^{2,1} (2G_{1,1} + G_2) G_1 + f^3 G_{2,1}$
$C_{123}^{\text{vib.}} = f^{1,1,1} G_1^3 + 3f^{2,1} G_1 G_{1,1} + f^2 G_{1,1,1}$
$C_{144}^{\text{vib.}} = f^{2,1} G_1 G_{1,1} + f^3 G_{1,1,1}$
$C_{456}^{\text{vib.}} = f^3 G_{1,1,1}$
$C_{166}^{\text{vib.}} = f^{2,1} G_1 G_{1,1} + f^3 G_{2,1}$

 Expressions for various  $f^n$ 's.

TABLE IV

$f^2 = f^3 = (1/2r_0^3)(\hbar w_0/4) \coth X$
$f^{1,1} = f^{2,1} = -(1/2r_0^3)(\hbar w_0/48)(X/\sinh^2 X + \coth X)$
$f^{1,1,1} = (1/2r_0^3)(\hbar w_0/192)((\hbar w_0/kT)^2(\coth X/6 \sinh^2 X) + X/\sinh^2 X + \coth X)$
$X = \hbar w_0/2kT$ , $\hbar = h/2\pi$ , $k$ is the Boltzmann constant,
$w_0^2 = (1/M_+ + 1/M_-)(1/H_\rho r_0)$
$M_+$ and $M_-$ are masses of positive and negative ions

 Expressions for various  $G_n$ 's.

TABLE V

$G_1 = 2[(2 + \rho_0 - \rho_0^2)Q(r_0) + 2(\sqrt{2} + 2\rho_0 - \sqrt{2}\rho_0^2)Q(\sqrt{2}r_0)]H$
$G_2 = 2[(-6 - 6\rho_0 - \rho_0^2 + \rho_0^3)Q(r_0)]H + 2G_{1,1}$
$G_3 = 2[(30 + 30\rho_0 + 9\rho_0^2 - \rho_0^3 - \rho_0^4)Q(r_0)]H + 2G_{2,1}$
$G_{1,1} = (-3\sqrt{2} - 6\rho_0 - \sqrt{2}\rho_0^2 + 2\rho_0^3)Q(\sqrt{2}r_0)H$
$G_{2,1} = [(15/2)\sqrt{2} + 15\rho_0 + (9/2)\sqrt{2}\rho_0^2 - \rho_0^3 - \sqrt{2}\rho_0^4]Q(\sqrt{2}r_0)H$
$G_{1,1,1} = 0$
$H = [(\rho_0 - 2)Q(r_0) + 2(\rho_0 - \sqrt{2})Q(\sqrt{2}r_0)]^{-1}$
$\rho_0 = r_0/\rho$
$A = -3S_3^1(e^2/r_0^2)[6 \exp(-\rho_0) + 12\sqrt{2} \exp(-\rho_0\sqrt{2})]^{-1}$
$Q(r_0) = A \exp(-r_0/\rho)$ and $Q(\sqrt{2}r_0) = A \exp(-\sqrt{2}r_0/\rho)$

Using  $\langle \gamma_i^j \rangle$ ,  $\langle \gamma_i^{j^2} \rangle$  and nonlinearity constants following relations for phonon-viscosity loss are obtained [8] for ultrasonic frequencies in the range  $\omega\tau_{\text{th}} \ll 1$ :

$$(\alpha/f^2)_{p-p} = (2\pi)^2 E_0 (D/3) \tau_{\text{th}} / dv^3, \quad (3)$$

where  $d$  is density,  $\tau_{th}$  is thermal relaxation time for the exchange of acoustic and thermal energies, given as

$$(1/2)\tau_1 = \tau_s = \tau_{th} = 3K/C_v \bar{v}^2, \quad (4)$$

where  $v$  represents  $v_l$  (longitudinal wave velocity) or  $v_s$  (shear wave velocity). These are related with  $C_{ij}$ 's as:

$$v_l = (C_{11}/d)^{1/2} \text{ and } v_s = (C_{44}/d)^{1/2} \text{ and } 3/\bar{v}^3 = (1/v_l^3 + 2/v_s^3), \quad (5)$$

where  $\bar{v}$  is the Debye average velocity.

### 3. Evaluation

The constants  $C_{ij}$  and  $C_{ijk}$  at different temperatures are evaluated using Born-repulsive parameter (RbCl = 0.283, RbBr = 0.293, RbI = 0.298) all in  $\text{\AA}^0$  and nearest neighbour distance [19]. Using  $C_{ij}$  and  $C_{ijk}$  thus obtained, average Grüneisen numbers are computed along  $\langle 100 \rangle$  direction for longitudinal and transverse waves using Mason's tables [8]. The thermal relaxation time  $\tau_{th}$  is obtained using thermal conductivity [20] and specific heat [21] with the help of equations (4) and (5). Nonlinearity coupling constant  $D$  is obtained using specific heat and energy of the crystal [21] as a function of temperature and is shown in Table VI. Phonon-viscosity loss at different temperatures is obtained and is shown in Table VII and Fig. 1.

TABLE VI

$D_l$ ,  $D_s$  and  $D_l/D_s$  at different temperatures in rubidium halides along  $\langle 100 \rangle$  directions.

Substance	$D$	100 K	200 K	300 K	400 K
RbCl	$D_l$	43.56	42.88	40.63	38.78
	$D_s$	2.97	2.94	2.90	2.87
	$D_l/D_s$	14.62	14.57	13.97	13.49
RbBr	$D_l$	43.80	42.21	40.07	38.12
	$D_s$	2.98	2.94	2.91	2.87
	$D_l/D_s$	14.64	14.31	13.76	13.26
RbI	$D_l$	50.85	48.46	45.90	43.73
	$D_s$	2.91	2.88	2.84	2.82
	$D_l/D_s$	17.45	16.82	16.11	15.50

TABLE VII

Ultrasonic attenuation for longitudinal  $(\alpha/f^2)_l$  and shear wave  $(\alpha/f^2)_s$  in  $10^{-18} \text{ Np s}^2 \text{ cm}^{-1}$ .

Temp. K	RbCl		RbBr		RbI	
	$(\alpha/f^2)_l$	$(\alpha/f^2)_s$	$(\alpha/f^2)_l$	$(\alpha/f^2)_s$	$(\alpha/f^2)_l$	$(\alpha/f^2)_s$
100	4.46	2.09	19.60	8.68	48.90	23.60
200	9.07	4.00	27.32	13.51	65.81	35.10
300	11.62	6.32	32.10	17.50	76.30	45.40
400	14.24	8.45	36.45	21.76	89.20	58.60

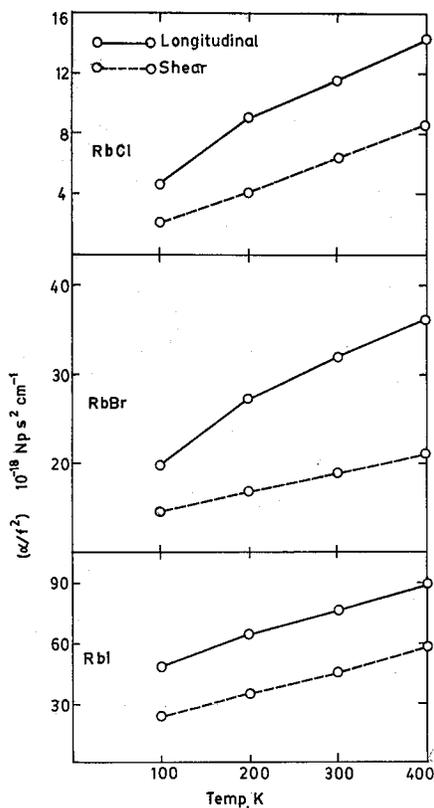


Fig. 1. Ultrasonic attenuation values for rubidium halides along  $\langle 100 \rangle$  direction for longitudinal and shear waves at different temperatures.

#### 4. Results and conclusions

The best check for the validity of the present investigation will be to compare nonlinearity coupling constants ( $D$ ) and phonon-viscosity loss  $(\alpha/f^2)_{p-p}$  with other similar solids.

##### 4.1. Nonlinearity coupling constant

The values of nonlinearity parameters for the three dielectric crystals are shown in Table VI. The nonlinearity parameters ratio ( $D_l/D_s$ ) lies between 14 to 17, which is as expected [22, 23]. Values of  $D_l$  and  $D_s$  are found to increase from RbCl to RbI. Also,  $D_l$  and  $D_s$  are decreasing with temperature along  $\langle 100 \rangle$  direction and  $D$  is larger for longitudinal wave than the shear wave like other alkali halide crystals [22].

#### 4.2. Phonon-viscosity loss

Temperature dependence of the ultrasonic attenuation due to phonon-phonon interaction for longitudinal and shear waves is shown in Table VII and Fig. 1. At 100 K magnitude of attenuation is the smallest, and as we go towards higher temperatures it increases and follows  $T^n$  law, where  $n$  lies between 0.40 and 0.55 for both longitudinal and shear waves. It is found that attenuation for shear wave is less than the longitudinal wave along  $\langle 100 \rangle$  direction of propagation, which is observed in other alkali halide crystals [22]. It has also been found that attenuation increases as molecular weight increases (from RbCl to RbI).

As the whole evaluation is done starting from the basic physical parameters of the substance and using electrostatic and Born-Mayer type potentials, it suffices to conclude that ultrasonic attenuation is a fundamental property of the substance.

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