Theoretical Analysis of Acoustic Attenuation and Nonlinearity in Barium Monochalcogenides in B2 Phase

Rajendra Kumar SINGH, Rishi Pal SINGH, Manish Pratap SINGH and Sujeet Kumar CHAURASIA
Department of Physics, Banaras Hindu University, Varanasi-221005, INDIA
e-mail: rksingh_17@rediffmail.com

Abstract

Temperature dependence of ultrasonic attenuation due to phonon-phonon interaction and thermo-elastic loss have been studied in (CsCl-type) barium monochalcogenides (BaX, X= S, Se, Te), in the temperature range 50–500 K; for longitudinal and shear modes of propagation along <100>, <110> and <111> directions. Second and third order elastic constants have been evaluated using electrostatic and Born repulsive potentials and taking interactions up to next nearest neighbours. Gruneisen parameters, nonlinearity constants, nonlinearity constants ratios and viscous drag due to screw and edge dislocations have also been evaluated for longitudinal and shear waves at 300 K. In the present investigation, it has been found that phonon-phonon interaction is the dominant cause for ultrasonic attenuation. The possible implications of the results have been discussed.

Key Words: Barium chalcogenides, Ultrasonic attenuation, Phonon-phonon interaction.

PACS number: 43.35.cg; 62.20 Dc

1.   Introduction

Among wide band gap II-IV semiconductors, barium chalcogenides (BaX, X=S, Se, Te) are interesting in connection with optoelectronic applications involving blue light wavelength. Barium Chalcogenides form very important closed shell ionic systems crystallized in the NaCl (B1) type and CsCl (B2) type structures at ambient conditions. Alkaline earth chalcogenides are currently under intense investigations driven by their applications in light emitting diodes (LEDs) and laser diodes (LDs). It is expected that these compounds may provide new II-IV candidates for the fabrication of various electrical and optical devices [1, 2].

Experimental as well as theoretical work on different aspects of these compounds has been reported in the recent past [1–4]. No results are available on temperature dependent acoustical behavior of these chalcogenides viz. on ultrasonic attenuation due to phonon-phonon interaction, thermo-elastic loss, dislocation damping, Gruneisen parameter, non-linearity parameters and thermal relaxation time etc, which are very important to explain the microstructure and other related physical properties of these alkaline earth chalcogenides. In the present communication, second and third order elastic constants (obtained at different temperatures) have been used to evaluate Gruneisen parameters and non-linearity parameters along

different crystallographic directions viz. <100>, <110> and <111> for longitudinal and shear modes in the temperature range 50 K–500 K. Taking electrostatic and Born repulsive potentials and utilizing some parameters viz. nearest neighbours distance, hardness parameter; ultrasonic attenuation coefficients have been calculated at different temperatures.

2. Theory

2.1. Second and third order elastic moduli

Second and third order elastic constants (SOEC's and TOEC's), C_{ij}^0 and C_{ijk}^0 at 0 K have been obtained using electrostatic and Born-Mayer potentials and following Brugger's [5] definition of elastic constants. Repulsive parameter and nearest-neighbor distance have been used as input data and interaction up to next nearest-neighbours has been considered. According to Brugger's definition, n^{th} order elastic constant is defined as

$$C_{ijklmn.....} = (\partial^n u/\partial \varepsilon_{ij} \partial \varepsilon_{kl} \partial \varepsilon_{mn} \cdots). \tag{1}$$

For cubic crystals three independent SOEC's (C_{11}, C_{12} and C_{44}) and six independent TOEC's (C_{111}, C_{112}, C_{144}, C_{166}, C_{456} and C_{123}) occur. Using the theory discussed in [5, 6], SOEC & TOEC viz. C_{ij}^0 and C_{ijk}^0 obtained at 0 K are given as

$$C_{11}^0 = \frac{3e^2}{8r_0^4}S_5^{(2)} + \frac{3Q(r_1)}{qr_0}(\frac{\sqrt{3}}{qr_0} + \frac{1}{q}) + \frac{2Q(r_2)}{qr^0}(\frac{1}{2r_0} + \frac{1}{q})$$

$$C_{12}^0 = C_{44}^0 = \frac{3e^2}{8r_0^4}S_5^{(1,1)} + \frac{Q(r_2)}{qr_0}(\frac{1}{2r_0} + \frac{1}{q})$$

$$C_{111}^0 = -\frac{15e^2}{8r_0^4}S_7^{(3)} - \frac{Q(r_1)}{9q}(\frac{\sqrt{3}}{r_0^2} + \frac{3}{qr_0} + \frac{\sqrt{3}}{q^2}) - \frac{Q(r_1)}{2q}(\frac{3}{r_0^2} + \frac{6}{qr_0} + \frac{4}{q^2})$$

$$C_{112}^0 = C_{116}^0 = -\frac{15e^2}{8r_0^4}S_7^{(2,1)} - \frac{Q(r_1)}{9q}(\frac{\sqrt{3}}{r_0^2} + \frac{3}{qr_0} + \frac{\sqrt{3}}{q^2})$$

$$C_{123}^0 = C_{456}^0 = C_{144}^0 = -\frac{15e^2}{8r_0^4}S_7^{(111)} - \frac{Q(r_1)}{9q}(\frac{\sqrt{3}}{r_0^2} + \frac{3}{qr_0} + \frac{\sqrt{3}}{q^2}),$$

where the lattice sums are

$$S_1^0 = -Z_0 = -1.017678, \quad S_5^{(2)} = 0.354190, \quad S_5^{(1,1)} = 0.346708,$$

$$S_7^{(3)} = 0.540901, \quad S_7^{(2,1)} = -0.093356, \quad S_7^{(1,1,1)} - 0.16000$$

and

$$r_1 = \sqrt{3}r_0, \ r_2 = 2r_0.$$

According to lattice dynamics developed by Ludwig et al [7], temperature variation of SOEC and TOEC have been obtained by adding vibrational contribution to elastic constants, that is,

$$C_{ij}(T) = C_{ij}^0 + C_{ij}^{vib.} \tag{2}$$

$$C_{ijk}(T) = C_{ijk}^0 + C_{ijk}^{vib.} \tag{3}$$

Where $C_{ij}^{vib.}$ and $C_{ijk}^{vib.}$ are vibrational contribution to elastic constants. Explicity expressions for SOEC and TOEC at required temperature are given in [6]

2.2. Theory of acoustic attenuation

In the Akhiezer regime [8] ($\omega\tau \ll 1$) a sound wave passing through a solid can be attenuated by two processes. First, if the wave is longitudinal, periodic contractions and dilations in the solid induce a temperature wave via thermal expansion. Energy is dissipated by heat conduction between regions of different temperatures. This is called thermoelastic loss. Second, dissipation occurs as the gas of thermal phonons tries to reach an equilibrium characterized by a local (sound wave induced) strain. This is the internal friction mechanism.

The physical basis for obtaining attenuation coefficient is that the elastic constants contributed by thermal phonons relax [9-11]. The phonon contribution to the unrelaxed elastic constants is evaluated by taking into consideration the change in energy of the thermal phonons due to applied instantaneous strain. The frequency of each mode ν_i is changed by $\frac{\partial \nu_i}{\nu_i} = -\gamma_i^j S_j$, where γ_i^j is generalized Gruneisen parameter and S_j is instantaneous strain. It is assumed that all the phonons of a given direction of propagation and polarization have equal change in frequency. Then phonons of ith branch and j^{th} mode suffer a change in temperature $\frac{\Delta T_i}{T_0} = -\gamma_i^j S_j$, where T is temperature. A relaxed elastic constant is obtained after there is phonon-phonon coupling among various branches and the ΔT_i relax to a common temperature change, ΔT given by $\frac{\Delta T}{T} = - \langle \gamma_i^j \rangle S_j$; where $\langle \gamma_i^j \rangle$ is the average value of γ_i^j. Thermal relaxation time [11–13],

$$\tau = \tau_s = \frac{\tau_l}{2} = \frac{3K}{C_v <V>^2}, \tag{4}$$

Where K is thermal conductivity, C_v is specific heat per unit volume and $<V>$ is the Debye average velocity.

According to Mason and Batemann [11, 13], SOEC's and TOEC's are related by Gruneisen parameter γ_i^j and hence by non-linearity parameter D. Ultrasonic attenuation due to phonon-phonon interaction in Akhiezer regime ($\omega\tau << 1$) is given by [11–13]

$$(\alpha_{p-p})_l = \frac{2\pi^2 f^2 D_l E_0 \tau_l}{3dV_l^3} \tag{5}$$

$$(\alpha_{p-p})_s = \frac{2\pi^2 f^2 D_s E_0 \tau_s}{3dV_s^3}, \tag{6}$$

where non-linearity coupling constant [11–13]

$$D = 9 \left\langle \left(\gamma_i^j\right)^2 \right\rangle - \frac{3\left\langle \gamma_i^j \right\rangle^2 C_v T}{E_0}, \tag{7}$$

and $\left\langle \left(\gamma_i^j\right)^2 \right\rangle$ and $\left\langle \gamma_i^j \right\rangle^2$ are the square average and average square Gruneisen parameters, respectively; V is sound wave velocity, (V_l) for longitudinal waves and (V_s) for shear waves and d is density.

Propagation of sound wave through crystal produces compression and rarefactions as a result heat are transmitted from compressed region (at higher temperature) to rarefied region (at lower temperature) and hence thermoelastic loss occurs, which is given by.

$$\alpha_{th} = \frac{4\pi^2 f^2 \left\langle \gamma_i^j \right\rangle^2 KT}{2dV_L^5}. \tag{8}$$

Debye average velocity is given by

$$\frac{3}{(<V>)^3} = \frac{1}{V_L^3} + \frac{2}{V_S^3} \tag{9}$$

The Debye temperature is given by [14]

$$\Theta_D = \hbar < V > q_d/k_B, \tag{10}$$

where is $< V >$ Debye average velocity, k_B is Boltzmann constant and

$q_d = (6^2 N_a)^{1/3}$, where N_a is atom concentration.

Dislocation damping due to screw and edge dislocations also produces appreciable loss due to phonon-phonon interaction. The loss due to this mechanism can be obtained by multiplying dislocation viscosities by square of dislocation velocity. Dislocation damping due to screw and edge dislocations is given by [15]

$$\Lambda_{screw} = 0.071\eta \tag{11a}$$

$$\Lambda_{edge} = 0.053\eta/(1-\sigma^2) + 0.0079/(1-\sigma^2)(\mu/B)\chi, \tag{11b}$$

where

$$\chi = \eta_l - (4/3)\eta_s, \eta_l = E_0 D_l \tau/3, \eta_s, = E_0 D_s \tau/3$$

$$B = (C_{11} + 2C_{12})/3, \mu = (C_{11} - C_{12} + C_{44})/3$$

and

$$\sigma = C_{12}/(C_{11} + C_{12}),$$

where B, μ, η, σ and χ are the bulk modulus, shear modulus, phonon viscosity, Poisson's ratio and compressional viscosity respectively; and C_{ij}'s are second order elastic constants.

3. Result and Discussion

Second and Third order elastic constants (SOEC's and TOEC's) have been evaluated at different temperatures, taking lattice parameters from references [1–3] and hardness parameters 0.691 Å, 0.699 Å and 0.622 Å for BaS, BaSe and BaTe respectively, using the equations (2) and (3). Evaluated SOEC values are shown in Table 1 and compared with SOEC's values obtained by Local Density Approximation (LDA) and Generalized Gradient Approximation (GGA) model [1-3]. They are in good agreement with others except minor disagreement in C_{12} and C_{44} values, which may be attributed to lattice and hardness parameters used by us.

Table 1. Second order elastic constants (10^{12} dyne/cm^2) at 300 K.

Compound	Model	C_{22}	C_{12}	C_{44}
BaS	Present	1.13	0.04	0.03
	GGA	1.05[a]	0.11[a]	0.10[a]
	LDA	1.31[a]	0.13[a]	0.11[a]
BaSe	Present	1.12	0.03	0.90
	GGA	0.97[a]	0.10[a]	0.90[a]
	LDA	1.13[a]	0.12[a]	0.11[a]
BaTe	Present	1.10[a]	0.02[a]	0.02[a]
	GGA	0.80[a]	0.07[a]	0.03[a]
	LDA	0.95[a]	0.12[a]	0.42[a]

a: ref. [2].

Square average Gruneisen number ($< \gamma_i^{j2} >$) and average square Gruneisen number ($< \gamma_i^j >^2$), acoustic coupling constants D for longitudinal and shear wave and their ratios D_l/D_s, and D_l/D_s* along different directions of propagation and polarization are given in Table 2. Results are as expected [13, 16–18]. Viscous drag coefficients due to screw (Λ_{screw}) and edge dislocation (Λ_{edge}) have been evaluated using equations (11a) and (11b), as given in Table 3.

Table 2. Square Average and average square Gruneisen number for longitudinal $< \gamma_i^{j2} >_l$, $< \gamma_i^j >_l^2$ and shear $< \gamma_i^j >_s^2, < \gamma_i^j >_{s*}^2$ Waves, nonlinearity coupling constants D_l, D_s and nonlinearity coupling constants ratios D_l/D_s, D_l / D_s* at 300 K.

Compoun-d	Direction	$< \gamma_i^{j2} >_l$	$< \gamma_i^j >_l^2$	$< \gamma_i^j >_s^2$	$< \gamma_i^j >_{s*}^2$	D_l	D_s	D_{s*}	D_l /D_s	D_l/D_{s*}
BaS	100	0.94	0.17	0.04	–	7.82	0.37	–	20.81	–
	110	1.06	0.26	0.15	1.93	8.63	1.43	17.37	6.03	0.49
BaSe	100	0.90	0.24	0.04	–	7.27	0.43	–	16.79	–
	110	1.04	0.36	0.22	1.80	8.04	1.98	16.28	4.06	0.49
BaTe	100	1.68	1.28	0.30	–	10.63	2.73	–	3.88	–
	110	2.14	1.75	4.49	1.33	12.93	40.46	12.00	0.31	1.00

[l]for longitudinal wave

[s]for shear wave, polarized along [001]

[s*]for shear wave, polarized along [1 $\bar{1}$ 0]

Table 3. Viscous drag coefficient due to screw and edge dislocation at 300 K longitudinal (in cp) and shear (in mp.) waves.

compound	Λ_{screw} (cp)		Λ_{edge} (cp)	
	Long.	Shear	Long.	Shear
BaS	0.23	0.10	0.45	0.55
BaSe	0.29	0.17	0.60	0.79
BaTe	0.47	1.30	1.07	3.22

The ultrasonic attenuation due to phonon-phonon interactions for longitudinal, $(\alpha/f^2)_l$ and shear waves, $(\alpha/f^2)_s$ are evaluated using equations (5) and (6). The temperature variation of $(\alpha/f^2)_l$ and $(\alpha/f^2)_s$ along [100], [110] and [111] directions of propagation are shown in Figures (1)–(2) and ultrasonic attenuation due to thermoelastic loss $(\alpha/f^2)_{th}$, evaluated using equation (8) is shown in Figure 3. Thermal relaxation time is evaluated using equation (4). Temperature variation of thermal relaxation time is shown in Figure (4), which shows exponential decay according to relation $\tau = \tau_o \exp(-t/T)$, where τ_o and t are constants.
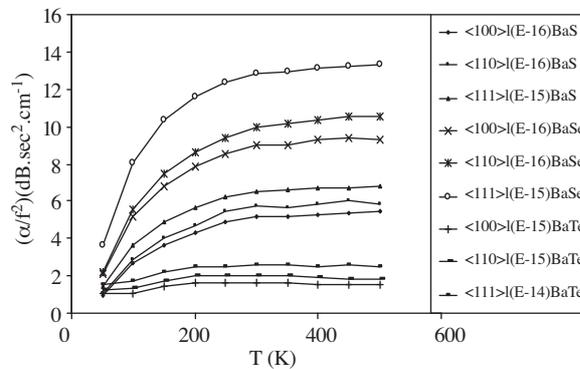
Figure 1. Temperature variation of $(\alpha/f^2)_l$ along different directio directions.
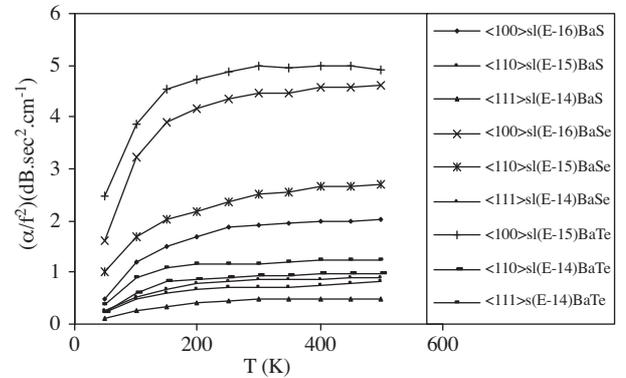
Figure 2. Temperature variation of $(\alpha/f^2)_s$ along different direction directions.

The attenuation due to phonon-phonon interaction for longitudinal and shear waves $(\alpha/f^2)_l$, $(\alpha/f^2)_s$ increase up to Θ_D and then become constants. When $(\Theta_D/T) \geq 1$, $(\alpha/f^2)_l$ and $(\alpha/f^2)_s$ increase and for values of temperature satisfying $(\Theta_D/T) < 1$, attenuation becomes nearly constant, because $[(\alpha/f^2)_l$ or $(\alpha/f^2)_s]$ due to p-p interaction is mainly affected by the specific heat, C_v (since (α/f^2) due to phonon-phonon interaction is related to C_v, through the relaxation time). For $(\Theta_D/T) \geq 1$, C_v increases and becomes nearly constant for the values satisfying $(\Theta_D/T) < 1$ (see Figure 5).
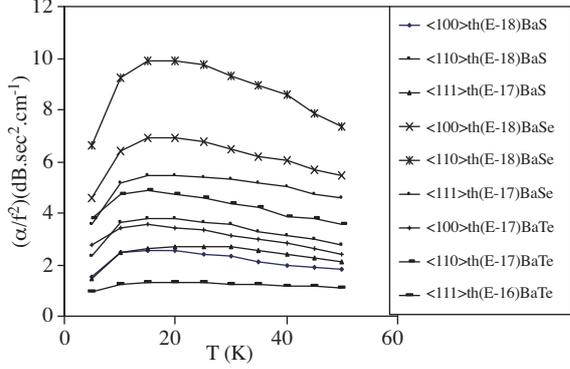
Figure 3. Temperature variation of $(\alpha/f^2)_{th}$ along diffrent direction.
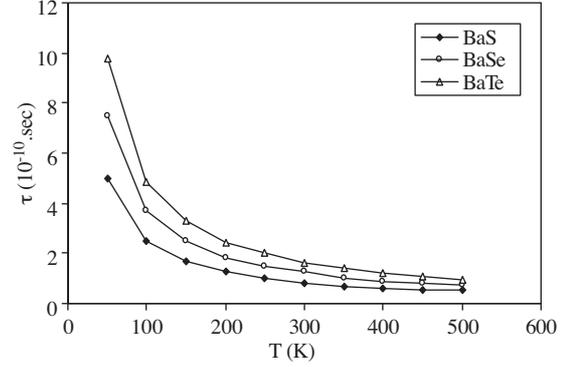
Figure 4. Temperature variation of thermal relaxation time, τ.
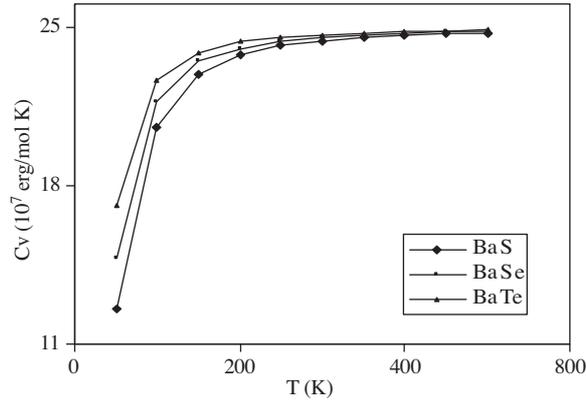
Figure 5. Temperature variation of specific heat, C_v.

$(\alpha/f^2)_{th}$ is directly proportional to thermal conductivity K, which depends on specific heat C_v and thermal relaxation time (see equation (4)). In low temperature range 50 K–200 K, compressional regions are at higher temperature and the rarefied regions are at low temperature, thus heat is transferred from compressional regions to rarefied regions resulting thermoelastic loss occurs. Thermoelastic loss increases in lesser temperature range 50 K–200 K, and then decreases slowly in the higher temperature range 200 K–500 K.

The value (α/f^2) is minimum for BaS and maximum for BaTe. The (α/f^2) depends upon Debye temperature (since Θ_D/T is taken as function of C_v and E_o). The Debye temperature is maximum for BaS and minimum for BaTe (Θ_D =200 K, 170 K and 143 K for BaS, BaSe and BaTe respectively, evaluated using equation (10)).Thus greater the Debye temperature, smaller will be attenuation. Debye temperature depends on the Debye average velocity $< V >$ and molecular weight, $M^{-1/3}$ through $(N/V)^{1/3}$ where N is Avogadro number and V ($V = M/d$, $M=$ mol. Wt. and $d=$ density) is volume. $< V >$ is maximum for BaS and minimum for BaTe ($< V > = 4.18$, 2.61 and 2.39 Km/sec respectively). Therefore larger $< V >$, smaller will be attenuation. Debye average velocity, $< V >$ decreases with increasing the molecular weight in these chalcogenide series due to increasing anion weight. The Θ_D and $< V >$ are SOEC,s dependent [14]

(equation (9) and (10)). Thus the increase in the value of $(\alpha/f^2)_l$, $(\alpha/f^2)_s$ and $(\alpha/f^2)_{th}$ from BaS to BaTe is mainly influenced by SOEC's values and Molecular weight.

Temperature variation of D_l and D_s along [100] and [110] directions of propagation are shown in Figures 6–9. Figures 6–9 depict that D_l increases with temperature and D_s decreases with temperature at small rate. This increase of D_l, increases the phonon viscosity which creates a greater resistance against the motion of moving dislocation for longitudinal wave. Value of D is amount of acoustical energy converted into thermal energy, which increase with temperature for longitudinal wave and decrease for shear wave.

On the basis of above discussion, we can say that attenuation in longitudinal wave is produced by phonon-phonon interaction, thermoelastic loss, dislocation damping and resistance created by D_l. On other hand attenuation in shear wave is produced by phonon-phonon interaction and dislocation damping.
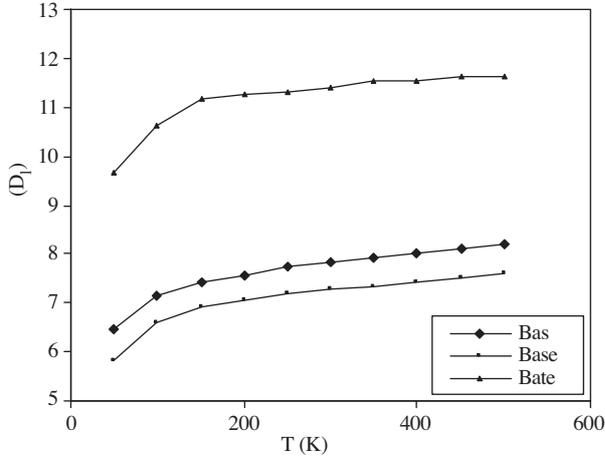
Figure 6. Temperature variation of Acoustic coupling constant, D_l for longitudinal wave along [100] direction.
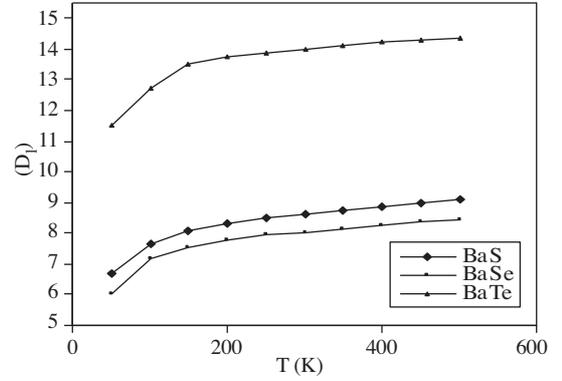
Figure 7. Temperature variation of Acoustic coupling constant, D_l for longitudinal wave along [110] direction.
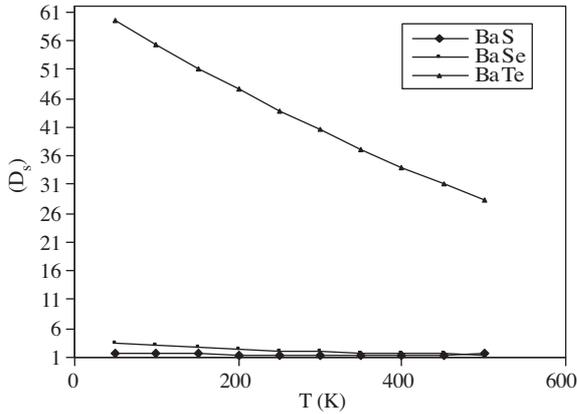
Figure 8. Temperature variation of Acoustic coupling constant, D_s for shear wave along [100] direction.
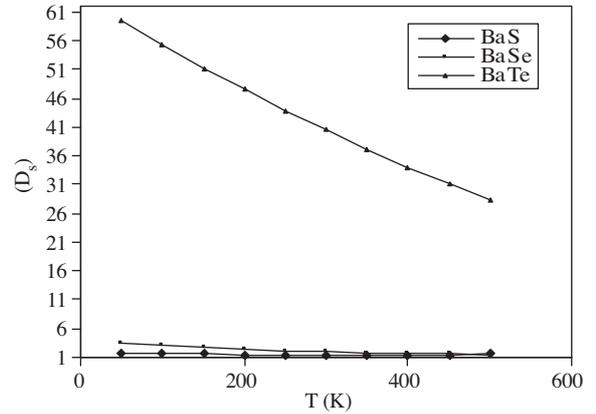
Figure 9. Temperature variation of Acoustic coupling constant, D_s for shear wave along [110] direction.

Acknowledgements

References

[1] Z. Charifi, H. Baoziz, F. El Haj Hassan and N. Bouarissa, *J. Phys. Condens. Matter*, **17**, (2005), 4083.

[2] A. Bouhemadou, R. Khenata, F. Zegrar, M. Sahnoun, H. Baltache, A. H. Resh, *Computational Material Science*, **38**, (2006), 263.

[3] F. El. Haj Hassan and H. Akbarzadeh, *Computational and Material Science*, **38**, (2006), 362.

[4] Phillip Cervantes, Quentin Williams, Michel Cote, Michael Rohlfing, Marvin L. Cohen, and Steven G. Louie *Phys. Rev. B*, **58**, (1998-I), 9793.

[5] K. Brugger, *Phys. Rev. A*, **133,** (1964), 1611.

[6] P. B. Ghate, *Phys. Rev. B*, **139**(5A), (1965), A1666.

[7] W. Ludwig and G. Leibfried, *Solid State Physics.*, (Academic Press New York, 12, 1967).

[8] A. Akhiezer, *J . Phys*, (USSR) **1,** (1939), 227.

[9] H. E. Bommel and K. Dransfield, *Phys. Rev.*, **177**, (1960), 145.

[10] A. B. Pippard, *Phil. Mag*, **45**, (1955), 1104.

[11] W. P. Mason, *Physical Acoustics*, Vol.III B, (Academic Press New York, 1965).

[12] W. P. Mason, and T. B. Bateman, *J. Acoust. Soc. Am.*, **36**, (1964), 645.

[13] W. P. Mason, *J. Acoust. Soc. Am.*, **42**, (1967), 253.

[14] Cz Jasiukiewicz., V. Karpus, *Solid State Communications*, **128**, (2003), 167.

[15] S. K. Kor, U. S. Tandon and G. Rai, *Phys. Rev. B*, **6**, (1972), 2195.

[16] S. K. Kor and R. K. Singh, *Acustica,* **79**, (1993), 292.

[17] S. K. Kor, K. Kailash, Shanker and P. Mehrotra, *J. Phys. Soc.Japan*, **56**, (1987), 2428.

[18] R. K. Singh and K. K. Pandey, *Acta Physica Polonica* A, **109**, (2006), 275.