

Acoustic Wave Propagation in Barium Monochalcogenides in the B1 Phase¹

R. K. Singh*, Rishi Pal Singh, M. P. Singh, and S. K. Chaurasia

Department of Physics, Banaras Hindu University, Varanasi-221005, India

*e-mail: adress:rk Singh_17@rediffmail.com

Received April 4, 2008

Abstract—Temperature dependence of ultrasonic attenuation due to phonon-phonon interaction and thermo-elastic loss have been studied in (NaCl-type) barium monochalcogenides [BaX, X = S, Se, Te], in the temperature range 50–500 K; for longitudinal and shear modes of propagation along $\langle 100 \rangle$, $\langle 110 \rangle$, $\langle 111 \rangle$ 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 results have been discussed.

PACS numbers: 43.20.Jr, 43.25.Dc, 43.35.Gk

DOI: 10.1134/S1063771009020067

1. INTRODUCTION

Among the wide band gap II–IV semiconductors, the barium chalcogenides [BaX, X = S, Se, Te] are interesting in connection with optoelectronic applications in blue light wavelength regime. The 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 made in the recent past [1–4]. No work has been made 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 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 $\langle 100 \rangle$, $\langle 110 \rangle$, $\langle 111 \rangle$ for

longitudinal and shear waves in the temperature range 50–500 K. Taking electrostatic and Born repulsive potentials and utilizing some parameters viz. nearest neighbours distance, hardness parameter and specific heat as a function of Debye temperature; ultrasonic attenuation coefficients have been calculated at different temperatures.

2. THEORETICAL APPROACH

2.1. Theory of Second and Third Order Elastic Constants

Second and third order elastic constants (SOEC and TOEC), C_{ij}^0 & 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-neighbour distance have been used as input data and interaction upto next nearest-neighbours has been considered. According to Brugger's definition, n th order elastic constant is defined as

$$C_{ijklmn\dots} = \left(\partial^n u / \partial \varepsilon_{ij} \partial \varepsilon_{kl} \partial \varepsilon_{mn} \dots \right) \quad (1)$$

where u is the crystal free-energy density and ε_{ij} is strain tensor. For cubic crystals three independent SOEC (C_{11} , C_{12} & C_{44}) and six independent TOEC (C_{111} , C_{112} , C_{144} , C_{166} , C_{456} & C_{123}) occur. Using the theory discussed in [5, 6], SOEC & TOEC viz. C_{ij}^0 & C_{ijk}^0 obtained at 0 K are given as:

¹ The text was submitted by the authors in English.

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

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

$$C_{111}^0 = -\frac{15e^2}{8r_0^4} S_7^{(3)} - \frac{Q(r_1)}{9q} \left(\frac{\sqrt{3}}{r_0^2} + \frac{3}{qr_0} + \frac{\sqrt{3}}{q^2} \right) - \frac{Q(r_2)}{2q} \left(\frac{3}{r_0^2} + \frac{6}{qr_0} + \frac{4}{q^2} \right),$$

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

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

Where the lattice sums are

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

$$S_5^{(1,1)} = 0.346708, S_7^{(3)} = 0.540901,$$

$$S_7^{(2,1)} = -0.093356, S_7^{(1,1,1)} = -0.16000,$$

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

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, SOEC and TOEC at any temperature are given by,

$$C_{ji}(T) = C_{ij}^0 + C_{ij}^{vib}, \quad (2)$$

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

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

2.2. Theory of Sound 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 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

v_i is changed by $\frac{\partial v_i}{v_i} = -\gamma_i^j S_j$, where γ_i^j is generalised

Gruneisen parameter & 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 i th branch and j th

mode suffer a change in temperature $\frac{\Delta T_i}{T_0} = -\gamma_i^j S_j$ (T

is the 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,

$$\tau = \tau_s = \frac{\tau_l}{2} = \frac{3K}{C_v \langle V \rangle^2}, \quad (4)$$

where K is thermal conductivity, C_v is specific heat per unit volume and $\langle V \rangle$ is Debye average velocity.

According to Mason and Batemann [12, 13], SOEC and TOEC are related by Gruneisen parameter γ_i^j and hence by non-linearity parameter, D . Ultra-sonic attenuation due to phonon-phonon interaction in Akhiezer regime ($\omega\tau \ll 1$) is given by

$$\alpha = \frac{2\pi^2 f^2 D E_0 \tau}{3dV^3}, \quad (5)$$

where non-linearity coupling constant

$$\Gamma = 9 \left\langle \left(\gamma_i^j \right)^2 \right\rangle - \frac{3 \left\langle \gamma_i^j \right\rangle^2 C_v T}{E_0}, \quad (6)$$

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

Table 1. Second order elastic constants (10^{12} dyne/cm²) at 300 K

Model	C_{22}	C_{12}	C_{44}
BaS			
Present	1.13	0.14	0.03
GGA	1.05	0.11	0.10
LDA	1.31	0.13	0.11
BaSe			
Present	1.12	0.13	0.90
GGA	0.97	0.10	0.90
LDA	1.13	0.12	0.11
BaTe			
Present	1.10	0.02	0.02
GGA	0.80	0.07	0.03
LDA	0.95	0.12	0.42

Table 2. Debye temperature (Θ_D) and average Debye average velocity ($\langle V \rangle$) at 300 K and molecular weight

	Θ_D (K)	$\langle V \rangle$ (10^5 cm/s)	M (Mol. Weight)
BaS	200	4.18	169.39
BaSe	170	2.61	216.28
BaTe	143	2.39	264.92

Debye average velocity is given by

$$\frac{3}{\langle V \rangle^3} = \frac{1}{V_L^3} + \frac{2}{V_s^3}. \quad (7)$$

The Debye temperature is given by [14]

$$\Theta_D = \hbar \langle V \rangle q_d / K_B, \quad (8)$$

where is $\langle V \rangle$ Debye average velocity, K_B is Boltzmann constant and $q_d = (6\pi^2 N_a)^{1/3}$ where N_a is atom concentration.

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

$$\alpha_{th} = \frac{4\pi^2 f^2 \langle \gamma_i^j \rangle^2 KT}{2dV_L^5}, \quad (9)$$

since in nanocrystalline material the number of dislocations is large hence 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];

$$B_{screw} = 0.071\eta, \quad (10a)$$

$$E_{dge} = 0.053\eta/(1-\sigma^2) + 0.0079/(1-\sigma^2)(\mu/B)\chi, \quad (10b)$$

where

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

$$\eta_s = E_0 D_s \tau / 3,$$

$$B = (C_{11} + 2C_{12})/3,$$

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

Table 3. Square average and average square Gruneisen number for longitudinal $\langle \gamma_i^2 \rangle_1$, $\langle \gamma_i^j \rangle_1^2$ and shear $\langle \gamma_i^j \rangle_s^2$, $\langle \gamma_i^j \rangle_{s*}^2$ waves, nonlinearity coupling constants D_l , D_s and nonlinearity coupling constants ratios D_l/D_s , D_l/D_{s*} at 300 K

Compound	Direction	$\langle \gamma_i^2 \rangle_1$	$\langle \gamma_i^j \rangle_1^2$	$\langle \gamma_i^j \rangle_s^2$	$\langle \gamma_i^j \rangle_{s*}^2$	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

Note: _l for longitudinal wave.

s for shear wave, polarized along [001].

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

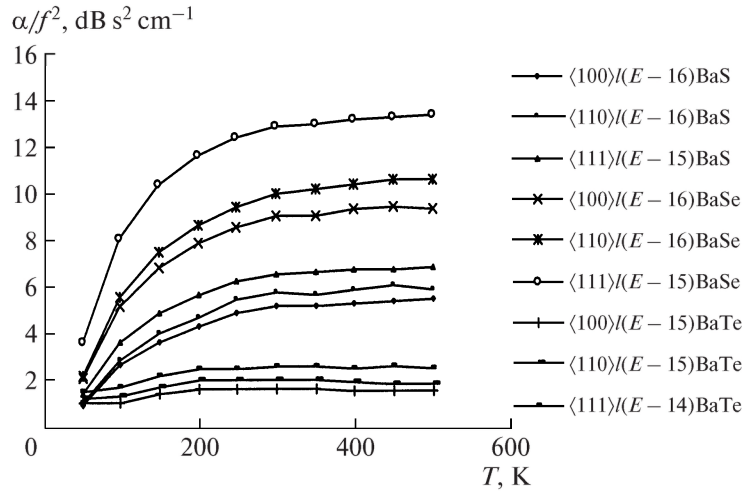


Fig. 1. Temperature variation of $(\alpha/f^2)_i$ along different direction directions.

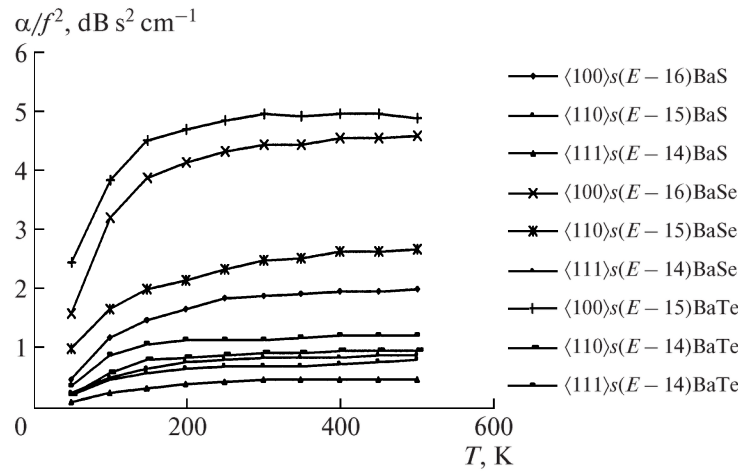


Fig. 2. Temperature variation of $(\alpha/f^2)_s$ along different direction directions.

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. RESULTS AND DISCUSSION

SOEC and TOEC have been evaluated at different temperatures with the help of lattice parameters of BaS, BaSe, BaTe, taken from [1–3] and hardness parameters 0.691 Å, 0.699 Å and 0.622 Å, respectively, using the equation (2) and (3). Evaluated SOEC values are shown in Table 1 and compared with SOEC 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 attribute to lattice and hardness parameters used by us.

Square average Gruneisen number $\langle \gamma_i^2 \rangle_1$, average square Gruneisen parameter longitudinal wave $\langle \gamma_i^2 \rangle_1^2$,

Table 4. Phonon viscosity due to screw and edge dislocation at 300 K for longitudinal (in cp) and shear (in mp) waves

Compound	B_{screw}		B_{edge}	
	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

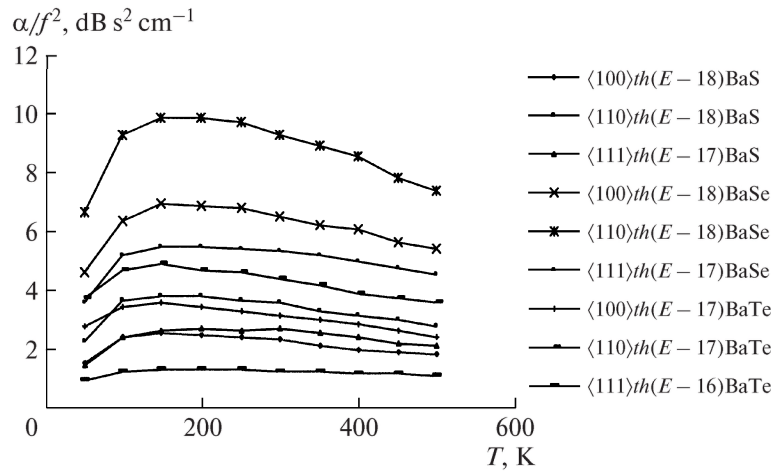


Fig. 3. Temperature variation of $(\alpha/f^2)_{th}$ along different direction.

square average Gruneisen parameter for shear wave $\langle \gamma_i^{j2} \rangle_s^2$ and $\langle \gamma_i^{j2} \rangle_{s*}^2$, nonlinearity coupling constants DI , D_s , D_s^* and their ratios DI/D_s , and DI/D_s^* along different direction of propagation are given in Table 3. Results are as expected [13, 16–18]. We have also calculated the Debye temperature using the formula (8) and Debye average velocity using (7), presented in Table 2 and viscous drag due to screw (B_{screw}) and edge dislocation (B_{edge}) using formula (10a) and (10b), as given in Table 4.

The ultrasonic attenuation due to phonon-phonon interaction for longitudinal, $(\alpha/f^2)_l$ and shear waves, $(\alpha/f^2)_s$ are evaluated using equation (5). The temperature variation of $(\alpha/f^2)_l$ and $(\alpha/f^2)_s$ along [100], [110] and [111] directions of propagation are shown in Figs. 1, 2 and ultrasonic attenuation due to thermoelastic loss $(\alpha/f^2)_{th}$ evaluated using equation (9) is

shown in Fig. 3. Thermal relaxation time is evaluated using equation (6). Temperature variation of thermal relaxation time is shown in Fig. 4 which shows exponential decay according to relation $\tau = \tau_0 \exp(-t/T)$ where τ_0 and t are constants.

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 $(\alpha/f^2)_s$ increases and for values of temperature satisfying $(\Theta_D/T) < 1$, attenuation become nearly constants, because $[(\alpha/f^2)_l \text{ or } _s]$ 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 become nearly constant for the values satisfying $(\Theta_D/T) < 1$, (Fig. 5).

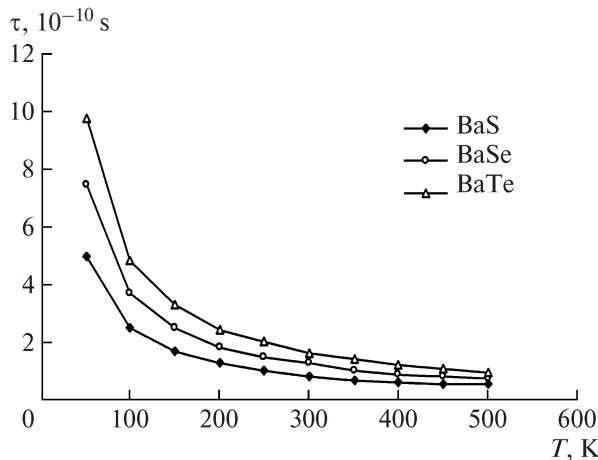


Fig. 4. Temperature variation of thermal relaxation time (τ).

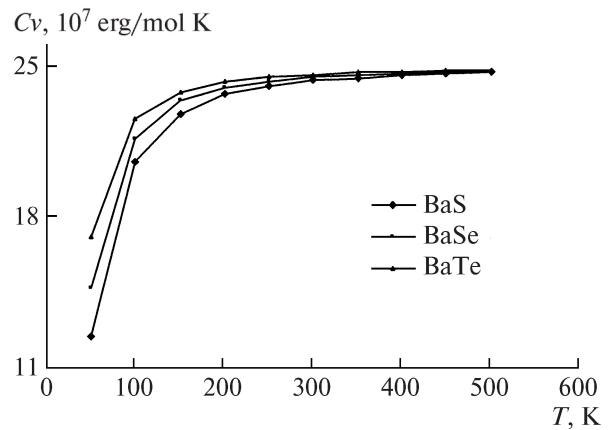


Fig. 5. Temperature variation of specific heat (C_v).

$(\alpha/f^2)_{th}$ is directly proportional to thermal conductivity (K). In low temperature range 50–200 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. The thermoelastic loss increases in small temperature range 50–200 K, and then decrease slowly in temperature range 200–500 K.

The (α/f^2) is minimum for BaS and maximum for BaTe. The (α/f^2) depends upon Debye temperature (Θ_D). The Debye temperature is maximum for BaS and minimum for BaTe (Table 2). Thus greater the Θ_D smaller the attenuation. The Θ_D depends on the Debye average velocity $\langle V \rangle$ 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. $\langle V \rangle$ is maximum for BaS and minimum for BaTe, therefore larger $\langle V \rangle$ smaller will be attenuation. The attenuation is increased in chalcogenides series with increasing the Molecular weight. The Θ_D and $\langle V \rangle$ are SOEM dependent as relation (7) and (8). Thus the increase in $(\alpha/f^2)_l$, $(\alpha/f^2)_s$ and $(\alpha/f^2)_{th}$ in a series of BaS to BaTe is mainly influenced by SOEM values and Molecular weight.

ACKNOWLEDGMENTS

One of us (R.K.S) is thankful to the University Grants Commission, New Delhi (Government of India) for financial assistance.

REFERENCES

1. Z. Charifi, H. Baoziz, F. El. Haj Hassan, and N. Bouarissa, J. Phys. Condens. Matter **17**, 4083 (2005).
2. A. Bouhemadou, R. Khenata, F. Zegrar, et al., Comput. Mater. Sci. **38**, 263 (2006).
3. F. El. Haj Hassan and H. Akbarzadeh, Comput. Mater. Sci. **38**, 362 (2006).
4. P. Cervantes, Q. Williams, M. Cote, M. Rohlfing, M. L. Cohen, and S. G. Louie, Phys. Rev. B **58**, 9793 (1998).
5. K. Brugger, Phys. Rev. A **133**, 1611 (1964).
6. P. B. Ghate, Phys. Rev. B **139** (5A), A1666 (1965).
7. W. Ludwig and G. Leibfried, *Solid State Physics* (Academic Press, New York, 1967), Vol. 12.
8. A. Akhiezer, J. Phys. (USSR) **1**, 227 (1939).
9. H. E. Bommel and K. Dransfield, Phys. Rev. **177**, 145 (1960).
10. A. B. Pippard, Phil. Mag. **45**, 1104 (1955).
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**, 645 (1964).
13. W. P. Mason, J. Acoust. Soc. Am. **42**, 253 (1967).
14. Cz. Jasiukiewicz and V. Karpus, Solid State Commun. **128**, 167 (2003).
15. R. K. Singh, R. Singh, and M. P. Singh, in *Proc. of the 19th Intern. Congress on Acoustics ICA-2007, Spain, Madrid*.
16. S. K. Kor and R. K. Singh, Acustica **79** (3), 83 (1993).
17. S. K. Kor, Sh. K. Kailash, and P. Mehrotra, J. Phys. Soc. Jpn. **56**, 2428 (1987).
18. R. K. Singh and K. K. Pandey, Acta Phys. Pol. A **109** (2), 275 (2006).