Behaviour of ultrasonic properties on SnAs, InTe and PbSb

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Abstract

In present investigation, we studied the elastic, ultrasonic and thermal properties of SnAs, InTe and PbSb. The Coulomb and Born-Mayer potential model was utilized to compute the second and third order elastic constants up to second nearest neighbor. The direction dependent ultrasonic velocities for longitudinal and shear waves, Debye average velocity and mechanical constants such as bulk modulus, shear modulus, tetragonal modulus, Young's modulus, Poisson ratio, Pugh's indicator (shear modulus to bulk modulus ratio) and Zener anisotropy ratio were obtained with the use of the second order elastic constants and the density of the chosen materials. Since the Pugh's indicator is greater than 0.59 for all chosen materials, they have a brittle nature. Further the second and third order elastic constants with other associated acoustical parameters were used to compute the Debye temperature, thermal relaxation time, acoustic coupling constant and ultrasonic attenuation. The total ultrasonic attenuation is the smallest in the case of InTe along the <100> direction and highest for SnAs along the <111> direction. Thermo-elastic loss is insignificant in comparison to the loss due to the phonon-phonon interaction mechanism. Additionally, the thermal conductivity of these materials was found using Cahill's approach. The results of this investigation are discussed with the available findings and for other rock salt structured materials.

1. Introduction

B_1-structured superconductors have stimulated major interest among theorists and experimentalists around the scientific world since the 1960s. The chief motivation for this awareness is to recognize the source of superconductivity simply by the peculiar characteristics of B_1-structured superconductors. The physical properties of B_1-structured InTe, SnAs and PbSb superconductors have drawn considerable interest in theoretical and experimental studies [1-12]. Banus et al. [1] showed that metallic InTe (II) had a B_1-structure with $a_0 = 6.154$ Å with transition temperature of 3.5 K in a superconducting phase. Geller and Hull [2] first predicted the superconductivity properties of SnAs and InTe in a rock-salt structure. The XRD technique was applied to investigate phase transition from the tetragonal to a face centered cubic phase at 340 kBar by Chattopadhyay et al. [3]. Wang et al. [4] studied the superconductivity of SnAs in a NaCl-structure. They predicted that SnAs exhibits weakly coupled type-1 superconductivity and Sn has a single valence state such as Sn^{3+} ($5s^1$). The superconductivity of SnAs in a NaCl structure is due to this unusual chemical state. The structural, electronic, vibrational and superconducting properties of SnAs in the NaCl structure using DFT have

been investigated by Tütüncü and Srivastava [5]. They also calculated the Debye temperature of SnAs as 199 K. Kunjomana et al. [6] grew InTe crystals using a physical vapour deposition (PVD) method. Hase et al. [7] evaluated the electronic structure of PbSb using FLAPW and LDA. They found that PbSb is a soft metal. The electronic properties of SnAs, InTe, and PbSb in NaCl structure were studied by Hase et al. [8] using a tight binding analysis. PbSb, SnAs and InTe formally have typical valence states, In^{2+}, Sn^{3+} and Pb^{3+}. Pb atoms in a compound usually take a 2+ or 4+ valence state, but rarely a 3+ valence. If we put a Pb atom into a site where it should take 3+, this valence state is unstable and may have large charge fluctuations. This type of "valence-skip" charge fluctuation can induce a charge-density wave (CDW) or superconductivity [9]. Shrivastava et al. [10] studied structural phase transition, elastic and electronic properties of B_1-structured SnAs using DFT. The structural, electronic, optical and elastic properties of tin arsenide in an ambient state were reported using first principles DFT by Rahman et al. [11]. Reddy et al. [12] did a computational study of the phonon structure, electron-phonon interaction and a transition temperature at 3.08 K across the phase diagram.

To our knowledge, only few studies have been reported in the literature on the elastic and mechanical properties of these materials [10-11]. No reports have been found of thermal and ultrasonic studies of the rock-salt structure superconductors, SnAs, InTe and PbSb, in the literature. This motivated us to conduct a study of the ultrasonic and thermal properties of SnAs, InTe and PbSb. In the present work, we computed the temperature dependence of the second and third order elastic constants (SOECs and TOECs), bulk modulus (B), shear modulus (μ), tetragonal modulus (C_S), Young's modulus (Y), Poisson ratio (σ), Pugh's indicator (μ/B), Zener anisotropy ratio (Z_A), ultrasonic velocities, Debye temperature, thermal conductivity, acoustic coupling constants and ultrasonic attenuation of these materials. The results are compared with available data for the chosen material, as well as other rock-salt type materials.

2. Theoretical approach

In ultrasonic attenuation computations, the SOECs and TOECs play a crucial role. The SOECs and TOECs were computed by means of Brugger's definition of elastic constants at an absolute zero temperature (0 K) [13]:

$$C_{ijklmn} = \left(\frac{\partial^n F}{\partial \eta_{ij} \partial \eta_{kl} \partial \eta_{mn} \ldots\ldots\ldots} \right)_{T=0K} \quad (1)$$

where F is the free energy of an undeformed material and is given as:

$$F = U + F^{Vib} \quad (2)$$

Here, U is the internal energy of a unit volume of the crystal, when all ions are at rest at their lattice point. U is given as:

$$U = \frac{1}{2V_c} \sum_{v=1}^{2} \sum_{\substack{m \neq 0 \\ \chi \xi}} \phi\left(R_{\chi\xi}^{m0} \right) = \frac{1}{2V_c} \sum' \phi(R) \quad (3)$$

where V_c is the volume of an elementary cell, a^3. Here, a is the lattice parameter of a particular superconductor, $R_{\chi\xi}^{m0}$ is the distance between the ξth ion in the 0th cell and the χth ion in the mth cell. ϕ is the interaction potential between the ions.

F^{vib} is the vibrational free energy at higher temperatures and is given as:

$$F^{Vib} = \frac{k_B T}{NV_c} \sum_{i=1}^{3sN} \ln 2 \sinh\left(\frac{\hbar\omega_i}{2k_B T} \right) \quad (4)$$

and η_{ij}, η_{kl} and η_{mn} are components of the Lagrangian strain tensor (here i, j, k=1, 2, 3). η_{ij} is given as:

$$\eta_{ij} = \frac{1}{2}\left[\left(\frac{\partial y_i}{\partial x_k} \right)\left(\frac{\partial y_j}{\partial x_k} \right) - \delta_{ij} \right] \quad (5)$$

where x and y are the initial and final positions of a material point and δ_{ij} is Kronecker's delta.

In Eq. (3), we can omit the indices (ξ,0) and (χ, m) for simplicity. Then, this potential, ϕ (r), is the sum of the

Coulomb and Born -Mayer potentials.

$$\phi(r) = \phi(C) + \phi(B) \quad (6)$$

where $\phi(B)$ is the Born-Mayer potential and $\phi(C)$ is the Coulomb potential.

Their values are given as:

$$\phi(C) = \pm \frac{e^2}{r_0} \quad \text{and} \quad \phi(B) = A\exp\cdot\left(\frac{-r_0}{b} \right) \quad (7)$$

where e is the electronic charge, r_0 is the nearest neighbour distance or short range parameter, b is the hardness parameter or Born's repulsive parameter and A is the strength parameter. The expression to compute A is given as:

$$A = -3b\frac{e^2}{r_0}S_3^{(1)}\frac{1}{6\exp(-\rho_0)+12\sqrt{2}\exp(-\sqrt{2}\rho_0)} \quad (8)$$

where $S_3^{(1)}$ is the lattice sum and its value is -0.58252 [13] and $\rho_0 = \frac{r_0}{b}$.

The SOECs and TOECs at higher temperatures have been evaluated using methods developed by Mori & Hiki [14], Leibfried & Haln [15], Leibfried & Ludwig [16] and Ghate [17] for NaCl-type crystals such as the chosen superconductors, SnAs, InTe and PbSb. The lattice parameters were found for these materials are 5.81 Å for SnAs [10], 6.18 Å for InTe [2] and 6.535Å for PbSb [7]. These lattice parameters are taken at zero Kelvin (0 K) temperature by means of first principle studies [2, 10, 7]. It is also presumed that the values of the lattice parameter for the chosen materials are constants in the specific temperature regimes. Hence, SOECs and TOECs at a particular temperature have been achieved by the addition of a vibrational energy contribution and static elastic constants.

$$C_{IJ} = C_{IJ}^0 + C_{IJ}^{Vib}$$
$$C_{IJK} = C_{IJK}^0 + C_{IJK}^{Vib} \quad (9)$$

0 (zero) and Vib represent the values of the elastic constants at 0 K and at a particular temperature respectively. [18].

The detailed expressions [18] for SOECs and TOECs are given as:

Static SOECs and TOECs

$$C_{11}^0 = \frac{3e^2}{2r_0^4}S_5^{(2)} + \frac{1}{br_0}\left(\frac{1}{r_0}+\frac{1}{b} \right)\phi(r_0) + \frac{2}{br_0}\left(\frac{1}{\sqrt{2}r_0}+\frac{1}{b} \right)\phi(\sqrt{2}r_0)$$

$$C_{12}^0 = C_{44}^0 = \frac{3e^2}{2r_0^4}S_5^{(1,1)} + \frac{1}{br_0}\left(\frac{1}{\sqrt{2}r_0}+\frac{1}{b} \right)\phi(\sqrt{2}r_0)$$

$$C_{111}^0 = \frac{15e^2}{2r_0^4}S_7^{(3)} - \left(\frac{3}{r_0^2}+\frac{3}{br_0}+\frac{1}{b^2} \right)\phi(r_0) - \frac{1}{b}\left(\frac{3\sqrt{2}}{r_0^2}+\frac{6}{br_0}+\frac{2\sqrt{2}}{b^2} \right)\phi(\sqrt{2}r_0)$$

$$C_{112}^0 = C_{166}^0 = -\frac{15e^2}{2r_0^4}S_7^{(2,1)} - \frac{1}{4b}\left(\frac{3\sqrt{2}}{r_0}+\frac{6}{br_0}+\frac{2\sqrt{2}}{b^2} \right)\phi(\sqrt{2}r_0)$$

$$C_{123}^0 = C_{144}^0 = C_{456}^0 = -\frac{15e^2}{2r_0^4}S_7^{(1,1,1)}$$

$$\quad (10)$$

where:

$$\phi(r_0) = A\exp\left(-\frac{r_0}{b}\right) \text{ and } \phi(\sqrt{2}r_0) = A\exp\left(-\frac{\sqrt{2}r_0}{b}\right)$$

The values of lattice sum [14, 17] are:

$$S_3^{(1)} = -0.58252, \quad S_5^{(2)} = -1.04622, \quad S_3^{(1,1)} = -0.23185$$

$$S_7^{(3)} = -1.36852, \quad S_7^{(2,1)} = -0.16115, \quad S_7^{(2,1)} = -0.09045$$

Vibrational SOECs and TOECs

$$
\left.
\begin{aligned}
C_{11}^{Vib} &= f^{(1,1)}G_1^2 + f^{(2)}G_2 \\
C_{12}^{Vib} &= f^{(1,1)}G_1^2 + f^{(2)}G_{1,1} \\
C_{44}^{Vib} &= G_{1,1} \\
C_{111}^{Vib} &= f^{(1,1,1)}G_1^3 + 3f^{(2,1)}G_2 G_1 + f^{(3)}G_3 \\
C_{112}^{Vib} &= f^{(1,1,1)}G_1^3 + f^{(2,1)}G_1(2G_{1,1}+G_2) + f^{(3)}G_{2,1} \\
C_{123}^{Vib} &= f^{(1,1,1)}G_1^3 + 3f^{(2,1)}G_1 G_{1,1} + f^{(3)}G_{1,1,1} \\
C_{144}^{Vib} &= f^{(2,1)}G_1 G_{1,1} + f^{(3)}G_{1,1,1} \\
C_{166}^{Vib} &= 3f^{(2,1)}G_1 G_{1,1} + f^{(3)}G_{2,1} \\
C_{456}^{Vib} &= f^{(3)}G_{1,1,1}
\end{aligned}
\right\} \quad (11)
$$

where:

$$
\left.
\begin{aligned}
f^{(1,1)} = f^{(2,1)} &= -\frac{h\omega_0}{96r_0^3}\left\{\frac{h\omega_0}{2k_BT\sinh^2 x} + \coth x\right\} \\
f^{(1,1,1)} &= -\frac{h\omega_0}{384r_0^3}\left\{\frac{(h\omega_0)^2\coth x}{6(k_BT)^2\sinh^2 x} + \frac{h\omega_0\coth x}{2k_BT\sinh^2 x} + \coth x\right\}
\end{aligned}
\right\}
$$

$$(12)$$

Here, $x = h\omega_o/2k_BT$, where h is Planck's constant and k_B is the Boltzmann's constant.

ω_0 is the lattice vibrational frequency, given as:

$$\omega_0 = \left(\frac{1}{M_+} - \frac{1}{M_-}\right)\frac{1}{br_0}\left\{(\rho_0 - 2)\phi(r_0) + 2(\rho_0 - \sqrt{2})\phi(\sqrt{2}r_0)\right\}$$

$$(13)$$

M_+ and M_- are masses of positive and negative ions (Sn^{+3}, In^{+2}, Pb^{+2}, As^{-3}, Te^{-2} and Sb^{-2} in present investigation).

Expressions of G_n are given by the following relations:

$$
\left.
\begin{aligned}
G_1 &= 2\left[\left(2 + 2\rho_0 - \rho_0^2\right)\phi(r_0) + 2(\sqrt{2} + (2\rho_0 - \sqrt{2}\rho_0^2)\phi(\sqrt{2}r_0))\right]H \\
G_2 &= 2\left[\left(-6 - 6\rho_0 - \rho_0^2 + \rho_0^3\right)\phi(r_0) + (-3\sqrt{2} - 6\rho_0 - \sqrt{2}\rho_0^2 + 2\rho_0^3)\phi(\sqrt{2}r_0)\right]H \\
G_3 &= \{30 + 30\rho_0 + 9\rho_0^2 - \rho_0^3 - \rho_0^4)\phi(r_0) + (\frac{15}{2}\sqrt{2} + 15\rho_0 + \frac{9}{2}\sqrt{2}\rho_0^2 - \rho_0^3 - \sqrt{2}\rho_0^4)\phi(\sqrt{2}r_0)\}H \\
G_{1,1} &= \left[\left(-3\sqrt{2} - 6\rho_0 - \sqrt{2}\rho_0^2 + 2\rho_0^3\right)\phi(\sqrt{2}r_0)\right]H \\
G_{2,1} &= \left[\left(\frac{15}{\sqrt{2}} + 15\rho_0 + \frac{9}{\sqrt{2}}\rho_0^2 - \rho_0^3 - \sqrt{2}\rho_0^4\right)\phi(\sqrt{2}r_0)\right]H \\
G_{1,1,1} &= 0
\end{aligned}
\right\}
$$

$$(14)$$

where H is given by the following expression:

$$H = \left[\left(\rho_0 - 2\right)\phi(r_0) + 2(\rho_0 - \sqrt{2}\rho_0^2)\phi(\sqrt{2}r_0)\right]^{-1}$$

$$(15)$$

and $\rho_0 = \frac{r_0}{b}$.

The hardness parameter plays an important role in the computation the SOECs and TOECs. It is also known as the Born repulsive parameter as given in Eqs. (7-15) [14, 19-20]. It can be determined as follows:

The total free energy of a crystal in equilibrium should be minimal. In a cubic crystal for equilibrium conditions, this is:

$$-\frac{e^2}{r_0}S_3^{(1)} - \frac{2r_0}{b}\phi(r_0) - \frac{4\sqrt{2}r_0}{b}\phi(\sqrt{2}r_0) - \frac{\hbar\omega_0}{4}G_1\coth x = 0$$

$$(16)$$

$$\hbar = \frac{h}{2\pi}$$

A set of *b* which satisfied this relation and also minimizes $\sum(C_{IJ}^{Cal.} - C_{IJ}^{Exp.})^2$ is chosen as the most probable solution. $C_{IJ}^{Cal.}$ are the calculated values of the SOECs in present work and $C_{IJ}^{Exp.}$ are the experimental SOECs at room temperature. The $C_{IJ}^{Exp.}$ values have not been published in the literature. Therefore we have chosen the most probable value of *b* satisfying the above relation near about the chosen values in other families compounds from published data [14, 21]. Ghate [17] used two values of the Born parameter for NaCl-type crystals. It is further presumed that the value of *b* is independent of temperature and can empirically be expressed as *b*=0.313 Å. The values are approximately equal for SnAs, InTe and PbSb.

SOECs have been utilized to determine the values of mechanical constants such as the bulk modulus (B), shear or rigidity modulus (μ), tetragonal modulus (C_S), Young's modulus (Y), Poisson's ratio (σ), Pugh's indicator (μ/B) and Zener anisotropy factor (Z_A) for SnAs, InTe and PbSb. The values of abovementioned parameters can be determined [22] from following equations:

$$B = \frac{C_{11} + 2C_{12}}{3}; \quad \mu = \frac{\mu_V + \mu_R}{2}$$

$$\text{where } \mu_V = \frac{C_{11} - C_{12} + 3C_{44}}{5} \quad \text{and } \mu_R = \frac{5(C_{11} - C_{12})C_{44}}{4C_{44} + 3(C_{11} - C_{12})};$$

$$C_s = \frac{C_{11} - C_{12}}{2}; Y = \frac{9\mu B}{\mu + 3B}; \sigma = \frac{3B - 2\mu}{6B + 2\mu} \text{ and } Z_A = \frac{2C_{44}}{C_{11} - C_{12}}$$

$$(17)$$

The stability, strength and hardness of the materials were obtained from these parameters. In particular, in the case of cubic crystals [23-25], the conditions of stability reduce to a very simple form:

$$B = \frac{C_{11} + 2C_{12}}{3} > 0, C_s = \frac{C_{11} - C_{12}}{2} > 0, C_{44} > 0,$$

$$(18)$$

The above equations for the cubic crystal system are well known, often referred to as the "Born stability criteria".

Ultrasonic velocity plays a vital role in the characterization of materials. The propagation of ultrasonic waves through anisotropic solids depends on the strains along the <100>, <110>, <111> directions. When ultrasonic waves propagate through a medium, their velocity has three modes of propagation, one longitudinal acoustical (V_L) and two shear acoustical (V_{S1}, V_{S2}). The expressions for V_L, V_{S1} and V_{S2} are presented elsewhere [24].

Along the <100> crystallographic direction:

$$\left.\begin{array}{l} V_L = \sqrt{(C_{11}/d)}; \\ V_{S1} = V_{S2} = \sqrt{(C_{44}/d)} \quad \text{shear wave polarized} \\ \text{along} < 100 > \end{array}\right\}$$

$$(19)$$

Along the <110> crystallographic direction:

$$\left.\begin{array}{l} V_L = \sqrt{(C_{11} + C_{12} + 2C_{44})/2d}; \\ V_{S1} = \sqrt{C_{44}/d} \qquad \text{shear wave polarized along} < 001 > \\ V_{S2} = \sqrt{(C_{11} - C_{12})/d} \quad \text{shear wave polarized along} < 1\bar{1}0 > \end{array}\right\}$$

$$(20)$$

Along the <111> crystallographic direction:

$$\left.\begin{array}{l} V_L = \sqrt{(C_{11} + 2C_{12} + 4C_{44})/3d}; \\ V_{S1} = V_{S2} = \sqrt{(C_{11} - C_{12} + C_{44})/3d} \quad \text{shear wave polarized along} < \bar{1}10 > \end{array}\right\}$$

$$(21)$$

where d is the density of the chosen materials.

The Debye average velocity V_D can be determined using Debye theory [26]. V_D is the average of V_L, V_{S1} and V_{S2}. V_D is expressed as:

$$V_D = \left[\frac{1}{3}\left\{\frac{1}{V_L^3} + \frac{2}{V_s^3}\right\}\right]^{-\frac{1}{3}} \quad \text{along the <100> and <111>}$$

directions

$$(22)$$

$$V_D = \left[\frac{1}{3}\left\{\frac{1}{V_L^3} + \frac{1}{V_{s1}^3} + \frac{1}{V_{s2}^3}\right\}\right]^{-\frac{1}{3}} \text{ along the <110> direction}$$

$$(23)$$

The Debye temperature (θ_D) is obtained by substituting V_D [26] into Eq. (24).

$$\theta_D = \frac{h}{k_B}\left(\frac{3n}{4\pi}\frac{Nd}{M}\right)^{\frac{1}{3}} V_D \tag{24}$$

where n is the number of atoms in the molecule, N is Avogadro's number and M is molecular weight.

The thermal relaxation time is the period required to convey the acoustic energy into a thermal phonon and the time to balance the temperature variation of the phonons. The expressions for the thermal relaxation time (τ) to propagate along the longitudinal and shear modes are:

$$\tau_{th} = \tau_{sh} = \frac{1}{2}\tau_{long} = \frac{3\kappa}{C_V V_D^2} \tag{25}$$

where, κ is thermal conductivity and C_V is the specific heat per unit volume. The expression for the thermal conductivity is given by Cahill's approach [27]:

$$\kappa = \frac{k_B}{2.48} \times n^{2/3}(V_L + V_{S1} + V_{S2}) \tag{26}$$

There are three main causes of ultrasonic attenuation:
  (i) Electron-phonon interaction
  (ii) Phonon-phonon interaction
  (iii) Thermo-elastic relaxation mechanism

At high temperatures, ultrasonic attenuation due to phonon-phonon interaction and a thermo-elastic relaxation mechanism occurs, while ultrasonic attenuation due to electron-phonon interaction has been found absent in higher temperature regimes [28-29].

The expression to compute the ultrasonic attenuation due to thermo-elastic relaxation [26, 20] is given as:

$$\left(\frac{\alpha}{\upsilon^2}\right)_{th} = \frac{4\pi^2 <\gamma_i^j>^2 \kappa T}{2\rho V_L^5} \tag{27}$$

where α is the ultrasonic absorption coefficient, th is the thermo-elastic loss, ν is the frequency of the ultrasonic wave, γ_i^j is the Grüneisen parameter.

Ultrasonic attenuation due to the phonon-viscosity mechanism (Akhiezer loss) [26, 30] is expressed as:

Table 1 SOECs and TOECs of SnAs, InTe and PbSb at the temperature range 0 to 300 K [in 10^{10} Nm^{-2}]

Material	Temp. (K)	C_{11}	C_{12}	C_{44}	C_{111}	C_{112}	C_{123}	C_{144}	C_{166}	C_{456}
SnAs	0	4.75	1.31	1.31	-80.32	-5.29	2.20	2.20	-5.29	2.20
	100	4.66	1.26	1.32	-75.25	-5.13	1.88	2.21	-5.41	2.20
	200	4.86	1.22	1.33	-76.8	-4.95	1.71	2.22	-5.44	2.20
	300	5.07	1.19	1.33	-78.5	-4.77	1.56	2.23	-5.47	2.20
SnTe [10]	300	14.98	2.03	4.02						
SnTe [11]	300	13.80	1.60	3.30						
InTe	0	2.33	0.37	0.36	-46.10	-1.42	0.68	0.68	-1.42	6.76
	100	2.48	0.33	0.36	-47.55	-1.17	0.20	0.67	-1.42	6.76
	200	2.63	0.30	0.37	-49.20	-0.97	0.20	0.68	-1.43	6.76
	300	2.79	0.27	0.37	-50.88	-0.77	0.03	0.68	-1.44	6.76
PbSb	0	3.49	0.79	0.79	-62.10	-3.16	1.37	1.37	-3.16	1.37
	100	3.67	0.73	0.79	-63.55	-2.91	1.07	1.38	-3.17	1.37
	200	3.86	0.72	0.79	-6.522	-2.72	0.91	1.38	-3.19	1.37
	300	4.05	0.67	0.79	-66.94	-2.53	0.75	1.39	-3.21	1.37

Table 2 B, μ, Cs, Y (all in 10^{10}Nm^{-2}), σ, μ/B ratio and Z_A of SnAs, InTe, PbSb at room temperature

Material	B	μ	Cs	Y	σ	μ/B	Z_A
SnAs	2.49	1.55	1.94	3.86	0.24	0.63	0.69
SnAs [10]	6.42	4.87	-	11.6	0.20	0.76	-
SnAs [11]	5.67	4.23	-	10.16	0.21	0.75	0.54
InTe	1.11	0.62	1.26	1.57	0.26	0.56	0.29
PbSb	1.80	1.08	1.69	2.71	0.24	0.60	0.47

$$\left(\frac{\alpha}{\upsilon^2}\right)_L = \frac{4\pi^2 \tau_l E_0 D_L}{6\rho V_L^3}$$

$$\left(\frac{\alpha}{\upsilon^2}\right)_{S_1} = \frac{4\pi^2 \tau_s E_0 D_S}{6\rho V_{S_1}^3} \qquad (28)$$

$$\left(\frac{\alpha}{\upsilon^2}\right)_{S_2} = \frac{4\pi^2 \tau_s E_0 D_S}{6\rho V_{S_2}^3}$$

where E_0 is thermal energy.

The acoustic coupling constant [26], which is a measure of thermal energy conversion into acoustic energy under a relaxation process, is expressed as:

$$D = 9\left\langle \left(\gamma_i^j\right)^2 \right\rangle - \frac{3\langle \gamma_i^j \rangle^2 C_V T}{E_0} \qquad (29)$$

The total attenuation in any solid medium due to ultrasonic wave propagation can be written as the sum of attenuation due to the thermo-relaxation phenomenon and phonon-phonon interactions.

$$\left(\frac{\alpha}{\upsilon^2}\right)_{Total} = \left(\frac{\alpha}{\upsilon^2}\right)_{th} + \left(\frac{\alpha}{\upsilon^2}\right)_L + \left(\frac{\alpha}{\upsilon^2}\right)_{S_1} + \left(\frac{\alpha}{\upsilon^2}\right)_{S_2} \qquad (30)$$

3. Results and discussion

The temperature dependent higher order elastic constants (SOECs and TOECs) were found using the nearest neighbour distance and hardness parameters. The values of the higher order elastic constants (C_{11}, C_{44}, C_{12}, C_{111}, C_{112}, C_{123}, C_{144}, C_{166} and C_{456}) are given in Table 1. From Table 1, it can be seen that the C_{11}, C_{44}, C_{111}, C_{144} and C_{166} values increase with temperature, while C_{12}, C_{112} and C_{123} decrease with increasing temperature. The value of C_{456} remains invariant due to the absence of a vibrational component. The change in the higher order elastic constant is due to variation in the

inter-atomic distance with temperature. This type of trend of the higher order elastic constants was observed first in other NaCl-type materials (alkali halides) by Mori and Hiki [14]. Recently, we observed this tendency in higher order elastic constants in NaCl-type actinide carbides [29] and lutetium monopnictides [30]. Our results for second order elastic constants were compared with those of Shrivastava et al. [10] and Rahman et al. [11] for SnAs. We used the Born model potential up to the second nearest neighbour, while Shrivastava et al. [10] and Rahman et al. [11] used first principles methods up to various neighbors. Although first principles is a better approach, our method is very simple and provides an overview of the whole analysis. This type of comparative study for the higher order elastic constants has been used in case of lutetium monopnictides [30] and praseodymium monopnictides [31]. Calculations were done manually as well as in MATLAB. So, we favour our results of SOECs and TOECs in this temperature regime.

According to Ghate [17], in the central force model chosen for alkali halides, the Cauchy relations for the SOECs and TOECs, $C_{12}^0 = C_{44}^0$; $C_{112}^0 = C_{166}^0$ and $C_{123}^0 = C_{144}^0 = C_{456}^0$, are satisfied at 0 K. The failure of the Cauchy relations at a finite temperature T is due to the vibrational component of energy. This trend has previously been observed in alkali halide crystals [13-14, 29-30].

The calculated SOECs and TOECs were used to evaluate mechanical parameters such as bulk modulus (B), shear modulus (μ), tetragonal modulus (C_S), Young's modulus (Y), Poisson ratio (σ), Pugh's indicator (μ/B) and Zener anisotropy ratio (Z_A) using Eq. (17). The computed values of these mechanical parameters are given in Table 2.

We compared our results with first principles calculations [10-11]. Our results have lower magnitude compared to other published values [10-11]. This is due the lower values of the SOECs for the chosen materials. The anisotropic ratio (Z_A) is the elastic anisotropy of a solid. The value of Z_A was found to be less than one for SnAs, InTe and PbSb. So, we can conclude that these materials exhibit anisotropic behavior. From Table 2, Pugh's indicator is

Table 3 Orientation dependent V_L, V_{S1}, V_{S2} and V_D (in 10^3 ms^{-1}), θ_D (in K), κ (in Wm^{-1}K^{-1}) and τ_{th} (in ps) of SnAs, InTe and PbSb at room temperature

Material	Orientation	V_L	V_{S1}	V_{S2}	V_D	θ_D	κ	τ_{th}
SnAs	<100>	2.77	1.42	1.42	1.59	207	2.20	3.12
	<110>	2.60	1.42	2.42	1.85	240	1.24	2.32
	<111>	2.54	1.62	1.62	1.78	251	1.29	2.50
InTe	<100>	2.02	0.73	0.73	0.83	102	0.46	3.09
	<110>	1.67	0.73	1.92	1.01	124	0.32	2.81
	<111>	1.53	1.18	1.18	1.27	155	0.51	1.48
PbSb	<100>	2.27	1.00	1.00	1.13	131	1.09	0.71
	<110>	2.00	1.00	2.07	1.35	156	0.68	0.20
	<111>	1.91	1.33	1.33	1.44	166	0.84	0.65

Table 4 Orientation dependent D_L, D_{S1}, D_{S2}, $(\alpha/v^2)_L$, $(\alpha/v^2)_{S1}$, $(\alpha/v^2)_{S2}$, $(\alpha/v^2)_{th}$ and $(\alpha/v^2)_{Total}$ (all (α/v^2) in 10^{-16} Nps^{2}m^{-1}) of SnAs, InTe and PbSb at room temperature

Materials	Direction	D_L	D_{S1}	D_{S2}	$(\alpha/v^2)_L$	$(\alpha/v^2)_{S1}$	$(\alpha/v^2)_{S2}$	$(\alpha/v^2)_{th}$	$(\alpha/v^2)_{Total}$
SnAs	<100>	15.06	1.04	1.04	8.33	2.11	2.11	0.02	12.57
	<110>	17.78	0.89	26.03	8.83	1.35	7.99	0.08	18.25
	<111>	16.05	19.19	19.19	1.19	11.06	11.06	0.08	23.39
InTe	<100>	23.44	1.16	1.16	0.29	0.15	0.15	0.0002	0.59
	<110>	30.15	0.71	38.98	0.66	0.09	0.28	0.001	1.03
	<111>	23.25	31.14	31.14	0.32	0.46	0.46	0.003	1.24
PbSb	<100>	17.97	1.08	1.08	0.28	0.09	0.09	0.0002	0.46
	<110>	21.33	0.77	32.13	0.13	0.02	0.09	0.0007	0.24
	<111>	18.99	22.94	22.94	0.42	0.81	0.81	0.001	2.04
LiF	<100>	38.00	4.50	4.50					
[36]	<110>	17.00	4.50	35.00					
	<111>	11.00							
KCl [37]	<111>	13.38	36.46	36.46	10.23	249.0	249.0		
KBr [37]	<111>	16.10	30.31	30.31	25.20	380.0	380.0		
KI [37]	<111>	26.15	43.42	43.42	69.20	843.0	843.0		

greater than 0.57, which indicates the brittle nature of SnAs, InTe and PbSb. From Eq. (18) and Table 2, it can be seen that the chosen materials follow the Born stability criterion. Hence, the materials are mechanically stable. The Poisson's ratio furnishes information about the nature of bonding in a material. The values of Poisson's ratio (σ) are between 0.25 and 0.5 in the central force solid. In our case, the evaluated value is $\sigma \geq 0.25$, so interatomic forces are central in SnAs, InTe and PbSb. The values of Poisson's ratio are used to define a material's nature: (i) if $\sigma=0$, the material is ionic, (ii) if $\sigma=0.25$, the material is covalent, and (iii) if $\sigma=0.33$, the material is metallic. In our case the value of σ for the SnAs, InTe and PbSb is approximately 0.25, which verifies their covalent character at zero pressure [29]. Table 2 shows that the values of B, μ, Cs, Y, σ, μ/B and Z_A for SnAs, InTe and PbSb are lower than those of previous investigators [10-11, 23] due to our smaller second order elastic constants.

We calculated the longitudinal and shear ultrasonic velocities modes (V_L, V_{S1} and V_{S2}) for SnAs, InTe and PbSb along the <100>, <110>, <111> crystallographic directions using the SOECs and TOECs. The Debye average velocity (V_D) is calculated using Eqs. (22-23) by means of V_L and V_S along various directions. V_D was used to compute the Debye temperature (θ_D) employing Eq. (24). The thermal conductivity (κ) was computed using ultrasonic velocities with the help of Cahill's approach [27] using Eq. (20). The thermal relaxation time (τ_{th}) was computed using Eq. (25). The orientation dependence V_L, V_{S1}, V_{S2}, V_D, θ_D, κ and τ_{th} of SnAs, InTe and PbSb at room temperature are given in Table 3.

From Table 3, it can be seen that V_D is the highest for all the selected substances along the <110> direction. θ_D is lowest along the <100> direction and highest along the <111> direction. The thermal conductivity is highest for SnAs along the <100> direction. On the basis of the order of thermal relaxation times, SnAs and InTe show metallic behaviour, while PbSb demonstrates semiconducting behaviour [32]. The thermal conductivity is highest for SnAs along the <100> direction. It is comparable with other NaCl-type materials incorporating BaPo, CaPo, or PbPo [33]. The highest Debye temperature, for SnAs, means that its thermal conductivity is quite different than InTe and PbSb.

Thermal conductivity is computed using Cahill's approach [27]. In this method, the thermal conductivity is directly related to V_L, V_{S1} and V_{S2} [see Eq. (26)]. These values are different for all the chosen materials. So, the thermal conductivity of SnAs differs from those of InTe and PbSb. Equation (29) is used to compute the acoustic coupling constants (D_L, D_{S1} and D_{S2}). The ultrasonic attenuation due to the phonon-viscosity mechanism $((\alpha/v^2)_L$, $(\alpha/v^2)_{S1}$, $(\alpha/v^2)_{S2})$ has been worked out from Eqs. (27-28). Thermal elastic loss $((\alpha/v^2)_{th})$ is determined using Eq. (27). The obtained results for D_L, D_{S1}, D_{S2}, $(\alpha/v^2)_L$, $(\alpha/v^2)_{S1}$, $(\alpha/v^2)_{S2}$, $(\alpha/v^2)_{th}$ and total attenuation $(\alpha/v^2)_{Total}$ along the <100>, <110>, <111> orientations are reported at room temperature in Table 4.

Table 4 depicts that the acoustic coupling constant for the longitudinal mode, i.e., D_L is highest along <110> orientation and lowest along the <100> orientation. For the shear mode, i.e., D_S is highest along <111> direction for all the materials. We also compared our results for the acoustic

coupling constant with NaCl-type materials, LiF [34] KCl, KBr & KI [35]. We found our acoustic coupling constants satisfactory. Acoustic coupling constants values are not available in the literature, so we compared our results with the same structural material [34]. The obtained results presented in the current investigation are more or less in agreement with the experimental results for LiF [34] and KCl, KBr & KI [35].

Table 4 shows that PbSb has lowest attenuation along the <110> direction. So, it is predictable that PbSb is most suitable candidate for the future applications, especially along <110> crystallographic direction. The total ultrasonic attenuation is found highest for SnAs along the <111> orientation, while lowest for PbSb along the <110> orientation, as shown in Table 4. It can also be seen from Table 4 that the ultrasonic attenuation due to phonon-phonon interaction (Akhieser loss) is principal over the thermo-elastic loss. We also compared our results with other rock-salt structure materials, KCl, KBr and KI [35]. It can be observed from Table 4 that the values of ultrasonic absorption over frequency squared (α/v^2) for chosen materials are very much less than KCl, KBr and KI [35]. The behavior of the chosen materials is quite different from KCl, KBr and KI. It is well established that the chosen materials are superconductors, while KCl, KBr and KI are dielectric materials. Superconducting materials, such as SnAs, InTe and PbSb, have less ultrasonic attenuation than dielectrics [35].

4. Conclusions

In this investigation, the Born model potential was successfully used to compute the higher order elastic constants of SnAs, InTe and PbSb. We also calculated the mechanical constants, such as the bulk modulus, shear modulus, tetragonal modulus, Young's modulus, Poisson's ratio, Pugh's indicator and Zener's anisotropy factor of these materials for their mechanical and intrinsic behaviour. The investigation verifies that the materials are mechanically stable and possess anisotropy in elasticity and are brittle in nature. The mechanical properties of SnAs are greater in magnitude than InTe and PbSb. The ultrasonic velocities reveal that SnAs has a better crystallographic texture. The thermal properties, such as the Debye temperature, thermal conductivity and thermal relaxation time, also reveal that these materials have better performance at room temperature due to their more or less metallic character. The greater magnitude of the Debye temperature for SnAs implies that its thermal conductivity is quite large compared to InTe and PbSb. PbSb has lowest attenuation, which implies that this material is more useful in a number of industrial applications, including infrared detection and imaging, than SnAs and InTe. Overall, we observed that these superconducting materials have ultrasonic features analogous to metallic and semiconducting materials. Ultrasonic attenuation due to phonon-phonon interaction (Akhieser loss) is the major portion of total attenuation.

The results of this investigation on superconducting materials, SnAs, InTe and PbSb, are valuable for the future applications of these materials as well as for further research.

5. Acknowledgements

6. List of symbols

C_{ijklmn} = Higher order elastic constants
F = free energy of an undeformed material
U = internal energy of a unit volume of the crystal
V_c = the volume of an elementary cell
A = lattice parameter
$r_{\mu v}^{m0}$ = the distance between the vth ion in the 0th cell and μ^{th} ion in the mth cell
$\phi_{\mu v}$ = the interaction potential between ions
F^{vib} = vibrational free energy at higher temperatures
η_{ij}, η_{kl} and η_{mn} = components of Lagrangian strain tensors
x and y = initial and final positions of a point in a material
δ_{ij} = Kronecker's delta.
$\phi(B)$ = Born- Mayer potential
$\phi(C)$ = Coulomb potential
e = electronic charge,
r_0 = nearest neighbour distance or short range parameter,
b = hardness parameter or Born's repulsive parameter
A = strength parameter
$S_3^{(1)}$ = lattice sum
$\bar{h} = \frac{h}{2\pi}$ = reduced Planck's constant
h = Planck's constant,
G_1 = a term is used in the computation of the SOECs and TOECs
B = bulk modulus
μ = shear or rigidity modulus
C_S = tetragonal modulus
Y = Young's modulus
σ = Poisson's ratio
μ/B = Pugh's indicator
Z_A = Zener anisotropy factor
V_L = longitudinal velocity
V_{S1} = shear velocity
V_{S2} = shear velocity
V_D = Debye average velocity
θ_D = Debye temperature
k_B = Boltzmann constant
n = number of atoms in the molecule
N = Avogadro's number
ρ = density
M = molecular weight
τ_{th} = thermal relaxation time
τ_{long} = relaxation time for longitudinal mode propagation of wave
τ_{sh} = relaxation time for shear mode of propagation of wave
κ = thermal conductivity
C_v = specific heat per unit volume
α = ultrasonic absorption coefficient
v = frequency of an ultrasonic wave
γ_i^j = Grüneisen parameter
D = acoustic coupling constant
E_0 = thermal energy

7. References

[1] Banus MD, Hanneman RE, Strongin M, Gooen K. High-pressure transitions in $A^{(III)}B^{(VI)}$ compounds: indium telluride. Science. 1963;142:662-3.

[2] Geller S, Hull GW. Superconductivity of intermetallic

compounds with NaCl-type and related structures. Phys Rev Lett. 1964;13:127-9.

[3] Chattopadhyay T, Santandrea RP, Von Schnering HG. Temperature and pressure dependence of the crystal structure of InTe: a new high pressure phase of InTe. J Phys Chem Solids. 1985;46:351-6.

[4] Wang Y, Sato H, Toda Y, Ueda S, Hiramatsu H, Hoson H. SnAs with the NaCl-type Structure: type I superconductivity and single valence state of Sn. Chem Mater. 2014;26:7209-13.

[5] Tütüncü HM, Srivastava GP. Electron–phonon interaction and superconductivity in SnAs with the sodium chloride crystal structure. Solid State Commun. 2015;221:24-7.

[6] Kunjomana AG, Chandrasekharan KA, Teena M. Physical properties of vapour grown indium monotelluride platelets. J Cryst Growth. 2015;411: 81-7.

[7] Hase I, Yasutomi K, Yanagisawa T, Odagiri K, Nishio T. The absence of CDW order in PbSb, and its unexpected softness. Physics Procedia. 2015;65: 37-40.

[8] Hase I, Yasutomi K, Yanagisawa T, Odagiri K, Nishio T. Electronic structure of InTe, SnAs and PbSb: valence-skip compound or not ?. Physica C. 2016;527: 85-90.

[9] Varma CM. Missing valence states, diamagnetic insulators, and superconductors. Phys Rev Lett. 1988; 61:2713-20.

[10] Shrivastava D, Dhabi S, Jha PK, Sanyal SP. Structural phase transition, electrtonic and elastic properties of rocksalt structure SnAs and SnSb. Solid State Commun. 2016;243:16-22.

[11] Rahman MA, Chowdhury UK, Bhuyan TH, Rahman MA. Theoretical investigation of structural, elastic, electronic and optical properties of SnAs. Am J Condens Matt Phys. 2016;6:27-35.

[12] Reddy PVS, Kanchana V, Millichamp TE, Vaitheeswaran G, Dugdale SB. Enhanced superconductivity in the high pressure phase of SnAs studied from first principles. Physica B. 2017;505:33-40.

[13] Brugger K. Thermodynamic definition of higher order elastic coefficients. Phys Rev. 1964;133:A1611-2.

[14] Mori S, Hiki Y. Calculation of the third- and fourth order elastic constants of alkali halide crystals. J Phys Soc Jpn. 1978;45:1449-56.

[15] Liebfried G, Haln H. Zur temperaturabhangigkeit der elastischen konstantaaen von alkalihalogenididkristallen. Z Phys. 1958;150:497-525.

[16] Leibfried G, Ludwig W. Theory of anharmonic effect in crystal. In: Seitz F, Turnbull D, editors. Solid State Physics, Vol. 12. New York: Academic Press; 1961. p. 275-444.

[17] Ghate PB. Third order elastic constants of alkali halide crystals. Phys Rev. 1965;139:A1666-74.

[18] Bhalla V, Singh D, Jain SK. Mechanical and thermophysical properties of cerium monopnictides. Int J Thermophys. 2016;37(33):1-17.

[19] Tosi MP, Fumi FG. Ionic sizes and Born repulsive parameters in the NaCl-type alkali halides-II. J Phys Chem Solids. 1964;25:45-52.

[20] Fumi FG, Tosi MP. Ionic sizes and Born repulsive parameters in the NaCl-type alkali halides-I. J Phys Chem Solids. 1964;25:31-43.

[21] Yadav RR, Singh D. Ultrasonic attenuation in lanthanum monochalcogenides. J Phys Soc Jpn. 2001;70:1825-32.

[22] Tripathy C, Singh D, Paikaray R. Behaviour of elastic and ultrasonic properties of curium monopnictides. Can J Phys. 2018;96:513-8.

[23] Mir SH, Jha PC, Islam MS, Banerjee A, Luo W, Dabhi SD, et al. Static and dynamical properties of heavy actinide monopnictides of lutetium. Sci Rep. 2016;6(29309):1-7.

[24] Queheillalt DT, Wadley HNG. Temperature dependence of the elastic constants of solid and liquid $Cd_{0.96}Zn_{0.04}Te$ obtained by laser ultrasound. J Appl Phys. 1998;83:4124-33.

[25] Mouhat F, Coudert F. Necessary and sufficient elastic stability conditions in various crystal systems. Phys Rev B. 2014;90(224104):1-4.

[26] Mason WP. Effect of impurities and phonon processes on the ultrasonic attenuation in germanium crystal, quartz and silicon. In: Mason WP, editor, Physical Acoustics, Vol. III, Part B. New York: Academic Press; 1965. p. 235-86.

[27] Cahill DG, Watson SK, Pohl RO. Lower limit to the thermal conductivity of disordered crystals. Phys Rev B. 1992;46:6131-40.

[28] Singh D, Bhalla V, Kumar R, Tripathi S. Behaviour of acoustical phonons in CeAs in low temperature region. Indian J Pure Appl Phys. 2015;53:169-74.

[29] Singh D, Kumar A, Bhalla V, Thakur RK. Mechanical and thermophysical properties of actinide monocarbides. Mod Phys Lett B. 2018;32(21):1-9.

[30] Singh D, Kumar A, Thakur RK, Kumar R. Elastic and ultrasonic properties of rare-earth monopnictides. Proc Natl Acad Sci India Sect A Phys Sci. 2018; https://doi.org/10.1007/s40010-018-0529-z.

[31] Bhalla V, Kumar R, Tripathy C, Singh D. Mechanical and thermal properties of praseodymium monopnictides: an ultrasonic study. Int J Mod Phys B. 2013;27(22):1-28.

[32] Singh D, Pandey DK, Yadawa PK. Ultrasonic wave propagation in rare-earth monochalcogenides. Cent Eur J Phys. 2009;7:198-205.

[33] Singh D, Bhalla V, Bala J, Wadhwa S. Ultrasonic investigations on polonides of Ba, Ca and Pb. Z Naturforsch A. 2017;72(11):977-83.

[34] Hanson RC. Attenuation of high frequency elastic waves in LiF. J Phys Chem Solids. 1967;28:475-83.

[35] Kor SK, Yadav RR, Kailash. Ultrasonic attenuation in dielectric crystals. J Phys Soc Jpn. 1986;55:207-12.