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Acoustic investigations on intermetallics

Devraj Singh*, D.K.Pandey

AMITY School of Engineering and Technology, Bijwasan, New Delhi-110 061 (INDIA)

Tel : 011-28062106 ext(206); Fax: 011-28062105

E-mail: devrajsingh.aset@gmail.com; pandeydrdk@rediffmail.com

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ABSTRACT

Acoustic attenuation due to phonon-phonon interaction and thermoelastic loss are studied in lanthanum chalcogenides along <111> crystallographic direction as a function of temperature. The second- and third-order elastic constants (SOEC & TOEC) of the materials are also calculated for the evaluation of ultrasonic coefficients. Shear wave attenuation is more than longitudinal wave attenuation for all three substances and total attenuation increases with temperature and predominantly affected with temperature variation of internal energy of these materials. Thermoelastic attenuation is very small as compared to phonon-viscosity loss (Akhieser loss). The behaviour of temperature dependent attenuation of these intermetallics has been discussed co-relating the important properties of metals and other NaCl-type structured crystals.

KEYWORDS

Elastic constants;
Grüneisen parameters;
Non-linearity parameters;
Acoustic velocity;
Acoustic attenuation.

INTRODUCTION

In recent years acoustic studies have been extensively used for characterization of materials[1-3]. In the present investigation we have chosen the lanthanum chalcogenides LaS, LaSe and LaTe for the evaluation of acoustic studies, which are well connected to fundamental properties of intermetallic compounds. In general intermetallics function as magnetic materials, superconductors, semiconductors, hydrogen bonding alloys, shape memory alloys and so on. The ordering of atoms and the strong interatomic bonding result many attractive properties for intermetallic compounds. The intermetallic compounds are desirable for nuclear application. Their applications are as structural materials, where strength at high temperature is required[4]. In the lanthanum chalcogenides, an appreciable fraction of total thermal conductivity is lattice thermal conductivity, hence they can be considered to be 'monovalent' metals. Lanthanum itself is typical trivalent metals[5]. These intermetallics used in carbon lighting application, especially by the motion picture industry for studio lighting and projection. To make them more useful for industry, temperature dependence of acoustic attenuation and other parameters has been studied in these materials, showing some characteristic features for them using second- and third-order elastic constants at different temperature through non-linearity parameters.

Theory

Mason's theory[6] for solids, which was initially proposed by Akhieser[7] has been established for the tem-

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TABLE 1: Higher order elastic constants in temperature range 100K to 450K

Materials	T[K]	C_{11}	C_{12}	C_{44}
LaS	100	4.700	1.261	1.346
	200	4.845	1.184	1.351
	300	5.007	1.106	1.356
	400	5.173	1.034	1.362
	450	5.266	0.985	1.365
LaSe	100	4.280	1.021	1.097
	200	4.430	0.950	1.102
	300	4.592	0.879	1.106
	400	4.757	0.808	1.111
	450	4.839	0.772	1.113
LaTe	100	3.800	0.780	0.854
	200	3.950	0.710	0.857
	300	4.106	0.640	0.861
	400	4.264	0.569	0.864
	450	4.342	0.534	0.865

Materials	T[K]	C_{111}	C_{112}	C_{123}	C_{144}	C_{166}	C_{456}
LaS	100	-75.5	-5.19	1.764	2.248	-5.49	2.231
	200	-76.1	-4.93	1.296	2.266	-5.51	2.231
	300	-76.9	-4.67	0.828	2.283	-5.53	2.231
	400	-77.7	-4.40	0.361	2.300	-5.56	2.231
	450	-78.1	-4.25	0.052	2.311	-5.60	2.231
LaSe	100	-70.6	-4.14	1.413	1.868	-4.45	1.853
	200	-71.4	-3.86	0.972	1.882	-4.47	1.853
	300	-72.2	-3.55	0.532	1.897	-4.49	1.853
	400	-73.1	-3.29	0.092	1.911	-4.51	1.853
	450	-73.5	-3.15	0.128	1.918	-4.52	1.853
LaTe	100	-64.8	-3.13	1.002	1.484	-3.43	1.472
	200	-65.6	-2.84	0.531	1.496	-3.45	1.472
	300	-66.4	-2.55	0.061	1.508	-3.46	1.472
	400	-67.3	-2.26	-0.41	1.519	-3.48	1.472
	450	-67.7	-2.12	-0.65	1.525	-3.49	1.472

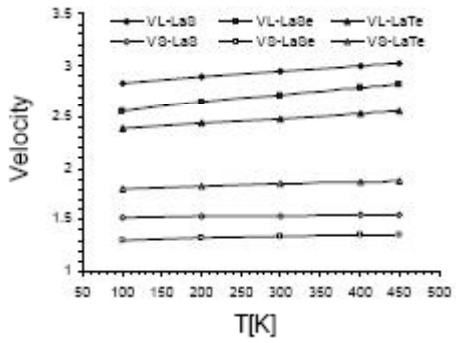

Figure 1 : Velocity(in 10^3m/s) vs temperature

perature dependence of acoustic attenuation and other related parameters for dielectrics, metals, some semiconductors along few crystallographic directions. This theory has been established for the variety of semiconductors, semimetallics and typical mixed crystal systems for different orientations and temperatures. An effort has been made to establish this theory for intermetallic compounds to evaluate acoustic attenuation coefficient and other parameters. The temperature dependence of acoustic attenuation and allied parameters has been discussed as the characteristic features of the materials taken.

Formulation for acoustic parameters ($\omega\tau_{th}<<1$) listed below:

$$\alpha_{Akh.} = \frac{\omega^2 E_0 D n \tau_{th}}{6\rho V^3} \quad (1)$$

$$\alpha_{th} = \frac{\omega^2 \langle \gamma_i^j \rangle KT}{2\rho V^5} \quad (2)$$

$$\tau_{th} = \frac{\sqrt{3}\rho K (C_{11}^{-3/2} + C_{44}^{-3/2})^{2/3}}{C_V} \quad (3)$$

where $$D = 9\langle (\gamma_i^j)^2 \rangle - \frac{3\rho C_V T}{E_0} \langle \gamma_i^j \rangle^2 \quad (4)$$

α_{Akh} = acoustic attenuation coefficient due to phonon-phonon interaction mechanism, which is valid at higher temperature, α_{th}= acoustic attenuation coefficient due to thermoelastic loss, ω= angular frequency of the wave, D=non-linearity parameters, $\langle \gamma_i^j \rangle$ =average Grüneisen number and $\langle (\gamma_i^j)^2 \rangle$ =average of square of Grüneisen number.The value of n=1 (for shear wave) and n=2 (for longitudinal wave), ρ=density of the materials. K= thermal conductivity, τ_{th}= thermal relaxation time, V=average Debye velocity, C_{11} and C_{44}=SOEC, C_V=specific heat per unit volume, E_0=internal energy density and T=temperature in Kelvin scale.

The higher order elastic constants play an important role in the evaluation of Grüneisen numbers and non-linearity parameters. Thus the higher order elastic constants are indirectly connected to acoustic attenuation. Therefore we have evaluated SOEC and TOEC following Brugger's approach and the method given by Mori and Hiki[8]. For the evaluation of SOEC and TOEC, we have taken only two basic parameters i.e. lattice parameter and hardness parameter.

RESULTS AND DISCUSSIONS

Evaluated SOEC and TOEC values (in the unit of $x10^{10}N/m^2$) are presented in TABLE 1 for La-chalcogenides.

Thermal conductivity (K) values are taken from the literature[5]. Acoustic velocities (V_L and V_S) are shown in figure 1. The Debye average velocity ($\overline{V}$)thermal

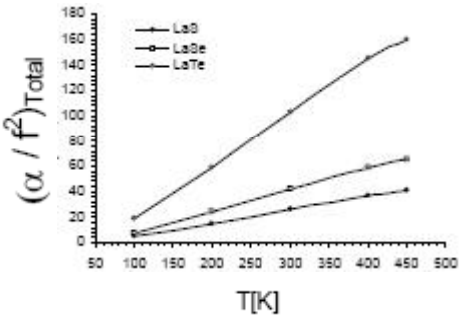

Figure 2 : α/f^2(10^{-16}Np s^2/m)vs temperature

TABLE 2: Debye average velocity (in the unit of 10^3m/s), relaxation time (in 10^{-11}sec), Grüneisen parameters and non-linearity parameters along <111> (shear wave polarized along <$\bar{1}$10>) direction in the temperature range 100-450K

Materials	T[K]	$\overline{V}$	$\tau_{th.}$	$\langle\gamma_i^j\rangle_L$	$\langle(\gamma_i^j)^2\rangle_L$	$\langle(\gamma_i^j)^2\rangle_S$	D_L	D_S
LaS	100	2.084	2.309	-0.65	1.522	2.065	10.93	18.59
	200	2.103	2.161	-0.63	1.397	1.977	10.75	17.79
	300	2.114	2.308	-0.60	1.285	1.893	10.09	17.04
	400	2.126	2.317	-0.58	1.202	1.855	9.602	16.70
	450	2.132	2.263	-0.56	1.144	1.815	9.112	16.34
LaSe	100	1.805	2.485	-0.65	1.523	2.222	11.81	20.00
	200	1.833	2.519	-0.62	1.387	2.126	10.55	19.13
	300	1.852	2.688	-0.57	1.267	2.034	10.16	18.31
	400	1.873	2.844	-0.56	1.165	1.953	9.326	17.58
	450	1.878	2.795	-0.55	1.119	1.916	8.975	17.25
LaTe	100	1.577	3.289	-0.65	1.995	2.621	15.76	23.58
	200	1.582	3.687	-0.62	1.796	2.500	14.63	22.50
	300	1.588	4.036	-0.59	1.641	2.394	13.49	21.55
	400	1.593	4.221	-0.56	1.496	2.301	12.36	20.71
	450	1.596	4.153	-0.55	1.440	2.217	11.91	19.95

TABLE 3: Acoustic attenuation {(α/f^2):in 10^{-16}Nps^2/m} in the temperature range 100-450K

Temp	$(\alpha/f^2)_{th}$			$(\alpha/f^2)_{Akh,long}$			$(\alpha/f^2)_{Akh.Shear}$		
[K]	LaS	LaSe	LaTe	LaS	LaSe	LaTe	LaS	LaSe	LaTe
100	0.077	0.106	0.190	0.724	0.508	2.371	4.019	6.838	16.642
200	0.163	0.227	0.429	2.228	2.747	6.771	12.523	20.872	51.481
300	0.250	0.312	0.661	3.622	4.817	11.018	22.787	36.731	91.678
400	0.312	0.434	0.850	5.024	6.309	12.363	31.393	52.703	131.525
450	0.325	0.459	0.907	5.251	6.722	15.272	35.653	58.547	143.579

relaxation time ($\tau_{th.}$), non-linearity parameter (D), Gruneisen parameters ($\langle\gamma_i^j\rangle$ and $\langle(\gamma_i^j)^2\rangle$) are presented in TABLE 2.

Acoustic attenuation coefficient over frequency square (α/f^2) for longitudinal and shear waves due to phonon-phonon interaction and thermoelastic loss values are presented in TABLE 3 and total attenuation $\{(\alpha/f^2)_{Total} = (\alpha/f^2)_{th} + (\alpha/f^2)_{Akh.Long} + (\alpha/f^2)_{Akh.Shear}\}$ is shown in figure 2 for La-chalcogenides along <111> direction in the temperature range 100 to 450K.

In calculation nearest neighbour distance r_0=2.894Å, 3.032 Å and 3.211 Å for LaS, LaSe and LaTe respectively were used as taken from the literature[5,10]. Computed values of SOEC and TOEC are used for evaluation of acoustic attenuation and velocity. Although positive temperature dependence of C_{11} is seen with TABLE 1, but very slight change in value of C_{44} and there is no change in value of C_{456}, because its vibration part of C_{456} is zero. Previously Kor et.al. calculated SOEC at different temperatures of KCl, KBr and KI with this theory and positive temperature dependence of C_{11} was seen in literature[11]. In the case of Pd metal there is good agreement between evaluated attenuation values using C_{11} calculated with present method and that using C_{11} values calculated by Hsu et.al[12]. Positive temperature dependence was also found in case of W in the work of Stathis et.al[13].

On the other chalcogenides system, the bulk modulus (C_{11}+2C_{44})/3 of SmS, SmSe and SmTe are 47.6 GPa[14], 40 GPa and , 40 GPa[15] respectively. These values are higher than evaluated 18 GPa, 21 GPa and 24 GPa for LaS, LaSe and LaTe respectively. There are no experiment data for LaS, LaSe and LaTe, therefore the comparison could not be made directly.

Thus with the above conclusions the theory for evaluation of SOEC and TOEC at different temperatures for LaS, LaSe and LaTe is being established. However full account of many interactions and Vander wall's interactions being ions and also consideration of non-linearity of the materials up to some extent may further improve the calculated TOEC[16]. The lanthanum chalcogenides are compounds with ionic metallic bonding possessed well developed structures of NaCl-type[5,10]. The evaluated thermal relaxation time ($\tau_{th.}$) is of the order of 10^{-11}second, which is as expected as in previous NaCl-type crystals[16].

The variation of Grüneisen parameters and non-linearity parameters with the temperature are presented in TABLE 2 for all the compounds. The non-linearity constants(acoustic coupling constant: amount of acoustical energy converted into thermal energy), D_L is decreasing with very small values as the temperature increases in all the three materials. This behaviour of 'D' is not observed previously in other NaCl-type crystals. The decreasing behaviour of acoustic coupling constant with temperature shows that they are more stable and ductile at higher temperatures in comparison to metals.

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When the shear wave is polarized along <$\bar{1}$ 10> direction, value of D_S is more than D_L for all three substances due to larger $\langle(\gamma_i^j)^2\rangle_S$ than the $\langle(\gamma_i^j)^2\rangle_L$. The ratio of D_S/D_L direction is same order as that of other NaCl-type materials[11,16]. It varies 1.69 to 1.92, 1.69 to 1.79 and 1.49 to 1.67 for LaS, LaSe and LaTe respectively.

It is obvious from the TABLE 3 that the thermal elastic loss is negligible in comparison to phonon-viscosity loss due to larger value of longitudinal velocity/C_{11} and shear wave attenuation is more than the longitudinal wave attenuation due to larger D_s. Hence total acoustic attenuation is mainly affected with shear wave attenuation. The figure 2 depicts that the $(\alpha/f^2)_{total}$ for substances along <111> direction is increasing with temperature from 100 to 450K and is predominantly affected with the temperature variation of internal energy (E_0)[9]. The larger attenuation at higher temperature is due to larger interaction of acoustic energy quanta and lattice energy quanta of material. LaS has smallest attenuation at each temperature. Thus it might be stated that the LaS has most ductility, stability and least imperfection than the other two.

Since all the computed parameters are compared with available data in literature and are justified hence it can be also concluded that Mason-Bateman theory is still good for acoustic attenuation due to phonon-phonon interaction at higher temperatures near to room temperature value for these intermetallic compounds.

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