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Ultrasonic Wave Propagation in Californium Monopnictides

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Abstract

In this paper, theoretical computations have been made for the investigations of temperature dependence of the ultrasonic parameters like ultrasonic velocities and Grüneisen parameters in californium monopnictides CfY (Y: N, As and Sb) for longitudinal and shear waves along <100>, <110> and <111> crystallographic directions in the temperature range 100–500K. For the same evaluation the second- and third- order elastic constants have also been computed for these monopnictides using Coulomb and Born-Mayer potential upto second nearest neighborhood. The mechanical properties and stability of CfN is best, because of its high valued elastic constants. Ultrasonic velocity is found to be highest for CfAs along all chosen directions, so CfAs will be most suitable compound for wave propagation. The obtained results of present investigation are discussed in along with identified thermophysical properties.

Keywords: Californium monopnictides; Coulomb and Born-Mayer potential; Elastic constants; Ultrasonic velocity; Grüneisen parameters

1. Introduction

Ultrasonic non-destructive testing (NDT) is a useful technique that can be applied to a range of materials for the characterization of their microstructures, the appraisal of defects and the determination of physical properties such as density, thermal conductivity and electrical resistivity. The Grüneisen parameter is of considerable importance to Earth's scientists, because it sets limitations on the thermoelastic properties of lower mantle [1]. The study of Grüneisen parameters for a solid enables us to describe and discuss the various physical properties of a system, such as high temperature specific heats of lattice, thermal expansion, thermal conductivity and temperature variation of the elastic constants. The Grüneisen parameters play a significant role in the study of thermoelastic properties. It has its fundamental importance to the equation of state of a system and related thermodynamic properties of the solids [2]. The calculation of anharmonic effects in solids such as thermal expansion or the interaction of acoustic and thermal phonons involves Grüneisen parameters, which describe the volume and strain dependence of the lattice vibrational frequencies. In the Debye model, these vibrations are replaced by standing wave modes of a dispersionless elastic continuum. The Grüneisen parameters are then no longer frequency dependent and can be expressed in terms of second- and third- order elastic constants [3]. Wave velocity is the key parameter in ultrasonic characterization and can provide information about crystallographic texture. The ultrasonic velocity (V) is related to the elastic constant by the relation $V = \sqrt{(C/\rho)}$, where C is the relevant elastic constant and ρ is the density of that particular material. The elastic constant, in particular provides valuable information on the stability and stiffness of the materials. The elastic constants of solids also provide a link between the mechanical and dynamical behaviours of crystals and give important information concerning the nature of forces operating in solids [4]. To the best of our knowledge, the studies of californium monopnictides have not been found in detail and we hope the ultrasonic study can be of potential interest. We found few studies of californium monopnictides in literature [5-8]. Haire *et al.* [5] and Damien *et al.* [6] studied the crystal structure and lattice parameters of Cf monopnictides and also proved semimetallic nature of CfY. Magnetism of californium Monopnictides has been discussed by Nave et al. [7-8]. No experimental and theoretical result on ultra-

sonic velocity and Grüneisen parameters on these materials has been found in the literature. Moreover the temperature dependent elastic constants have not been computed and measured, which stimulates us to execute theoretical analysis of elastic constants, ultrasonic velocities and Grüneisen parameters in californium monopnictides along <100>, <110> and <111> directions at temperature range 100 K - 500 K.

2. Theory

The second and third order elastic constants (SOEC and TOEC) have been calculated following Brugger's definition of elastic constants [9-10] at absolute zero (C_{IJ}^0 and C_{IJK}^0). The SOEC and TOEC at different higher temperatures are obtained by the method developed by Leibfried and Haln [11], Ludwig [12] and Hiki [13] for NaCl-type crystals as the chosen semimetallics have well-developed structures of the NaCl-type crystals. The lattice parameters are very close to those in literature [6-8,14].

When sound wave propagates through a solid medium, there are three modes of propagation one longitudinal acoustical and two transverse acoustical. Hence, there exist three types of velocities, as one longitudinal (V_L) and two shear (V_{S1} and V_{S2}) that depend on the direction of propagation of wave [15]. The direction dependent ultrasonic velocities for a cubic crystal are expressed as given below

Along <100> crystallographic direction;

$$\left.\begin{aligned} V_L &= \sqrt{(C_{11}/\rho)}; \\ \mathrm{V_{S1}} = V_{S2} &= \sqrt{(C_{44}/\rho)} \end{aligned}\right\} \quad (1)$$

Along <111> crystallographic direction;

$$\left.\begin{aligned} V_L &= \sqrt{(C_{11}+2C_{12}+4C_{44})/3\rho}; \\ \mathrm{V_{S1}} = V_{S2} &= \sqrt{\sqrt{(C_{11}-C_{12}+C_{44})/3\rho}} \end{aligned}\right\} \quad (2)$$

Along <110> crystallographic direction;

$$\left.\begin{aligned} V_L &= \sqrt{\frac{C_{11}+C_{12}+2C_{44}}{2\rho}}\ ; \\ \mathrm{V_{S1}} &= \sqrt{\frac{C_{44}}{\rho}};\ V_{S2} = \sqrt{\frac{C_{11}-C_{12}}{\rho}} \end{aligned}\right\} \quad (3)$$

where C_{IJ} are particular elastic constant of the material and ρ is the density. The Debye average velocity (V_D) is useful for information of Debye temperature and thermal relaxation time of the materials. The following expressions have been used for evaluation of Debye average velocity [12].

$$\left.\begin{aligned} V_m &= \left[\frac{1}{3}\left\{\frac{1}{V_L^3}+\frac{2}{V_{S1}^3}\right\}\right]^{-1/3}; \\ &\quad \text{along } <100>,\ <111> \text{ directions} \\ &= \left[\frac{1}{3}\left\{\frac{1}{V_L^3}+\frac{1}{V_{S1}^3}+\frac{1}{V_{S2}^3}\right\}\right]^{-1/3}; \\ &\quad \text{along } <110> \text{ direction} \end{aligned}\right\} \quad (4)$$

A number of anharmonic properties of solids are frequently expressed in terms of Grüneisen parameters, which are expressed, in quasiharmonic approximation, as diverse weighted averages of Grüneisen tensor of the first order: $\gamma_{\alpha\beta}^j = -\omega_i^{-1}\partial\omega_i(q)/\partial\eta_{\alpha\beta}$. For example, the thermal expansivity is relative to the specific heat weighted $<\gamma_{\alpha\beta}> = \sum_{q,i} C_{q,i}\gamma_{\alpha\beta}^j \sum_{q,i} C_{q,i}$,which is thermal Grüneisen parameters γ; and the (shear) ultrasonic attenuation's Grüneisen parameter can be suitably expressed [16] by thermal conductivity weighted averages of the product $\gamma_{\alpha\beta}^j\gamma_{\gamma\delta}^j$. Brugger [17] derived expressions for the components of Grüneisen tensor in terms of SOEC and TOEC of an anisotropic elastic continuum. These relations permit the above weighted average to be reliably calculated from elastic and thermal data to give ultrasonic attenuation and non-linear parameters, which compares very well with measured results [18]. Formulae of Grüneisen parameters along different crystallographic directions are given in literature [19].

3. Results and Discussion

The elastic constants of materials are directly related to their microstructure and are used to obtain the Debye average velocity, Grüneisen parameter and other physical properties; and therefore, these are of great interest in applications where the mechanical strength and durability are important. The SOEC and TOEC have been evaluated using two basic parameters *i.e.*, lattice parameter and hardness parameter. The lattice parameters [6, 7, 14] for CfN, CfAs and CfSb are 4.95 Å, 5.809 Å, and 6.165 Å and the value of hardness parameters [20] are 0.313 Å,0.303 Å and 0.311 Å respectively. The computed results of temperature dependent SOEC and TOEC are given in **Tables 1**. No experimental/theoretical result of SOEC and TOEC of these materials was found directly in existing literature. So our achieved results have been compared with gadolinium monopnictides GdY [21]. These compared results are available in **Table 1**. It is clear from **Table 1** that out of nine elastic constants, four (*i.e.*, C_{11}, C_{44}, C_{112} and C_{144}) are increasing and four (*i.e.*, C_{12}, C_{111}, C_{166} and C_{123}) are decreasing with the temperature while C_{456} is found to be unaffected. This trend of elastic constants in chosen materials at higher

Table 1. Second and third order elastic constants of CfY at the temperature range 100 to 500K with comparable data of GdY at room temperature [in the unit of 10^{11} Dyne/cm^2].

Material	Temp (K)	C_{11}	C_{12}	C_{44}	C_{111}	C_{112}	C_{123}	C_{144}	C_{166}	C_{456}
	100	6.15	2.58	2.68	–90.42	–10.66	3.77	4.20	–11.00	4.17
	200	6.29	2.50	2.69	–90.76	–10.41	3.37	4.23	–11.03	4.17
CfN	300	6.46	2.42	2.70	–91.42	–10.15	2.98	4.26	–11.07	4.17
	400	6.65	2.33	2.71	–92.19	–9.886	2.58	4.28	–11.11	4.17
	500	6.84	2.25	2.72	–93.01	–9.623	2.18	4.31	–11.15	4.17
	100	4.96	1.22	1.30	–81.57	–4.97	1.71	2.21	–5.30	2.20
	200	5.13	1.14	1.31	–82.40	–4.66	1.23	2.23	–5.33	2.20
CfAs	300	5.37	1.06	1.31	–84.03	–4.35	0.75	2.25	–5.36	2.20
	400	5.49	0.98	1.32	–84.28	–4.04	0.26	2.26	–5.37	2..20
	500	5.68	0.90	1.32	–85.25	–3.72	–0.21	2.28	–5.40	2.20
	100	4.17	0.94	1.01	–69.96	–3.79	1.27	1.74	–4.10	1.73
	200	4.33	0.86	1.02	–70.78	–3.50	0.82	1.75	–4.12	1.73
CfSb	300	4.49	0.79	1.02	–71.65	–3.21	0.37	1.77	–4.14	1.73
	400	4.66	0.72	1.02	–72.54	–2.92	–0.07	1.78	–4.16	1.73
	500	4.82	0.06	1.03	–73.43	–2.63	0.05	1.79	–4.18	1.73
GdP[22]	300	5.54	1.16	1.43	–86.17	–4.76	0.90	2.41	–5.78	2.36
GdAs[22]	300	5.29	1.03	1.28	–83.30	–4.18	0.67	2.19	–5.19	2.14
GdSb[22]	300	4.68	0.74	0.98	–76.17	–2.92	0.17	1.17	–3.93	1.67
GdBi[22]	300	4.60	0.69	0.96	–74.78	–2.71	0.09	1.63	–3.72	1.60

temperatures is due various parameters involved in the expressions used for the evaluation of elastic constants [6-8,14]. This type of behaviour has already been found in other NaCl-type materials like gadolinium and cerium monopnictides [21-25]. Hence our approach to find out the elastic constants at different temperatures is justified.

There are no elastic data as a function of temperature for these compounds. **Table 1** depicts that CfN has highest valued SOEC and TOEC in contrast to other monopnictides. Hence mechanical properties of CfN are better than those of CfAs and CfSb. According to the Born criterion of a lattice to be stable, the elastic energy density must be positive. The values of bulk moduli (B_T), shear moduli (C_{44}) and tetragonal moduli (C_S) of the chosen materials CfN, CfAs and CfSb are $B_T = (C_{11} + 2C_{12})/3 > 0$, $C_{44} > 0$ and $Cs = (C_{11} - C_{12})/2 > 0$ respectively.

Estimated values of bulk moduli, shear moduli and tetragonal moduli are tabulated in **Table 2**. The values of bulk moduli (B_T), shear moduli (C_{44}) and tetragonal moduli (C_S) are satisfying the stability criterion for CfN, CfAs and CfSb compounds. Hence our approach to find out the elastic constants is correct and reasonable. The ultrasonic velocity is a key factor to characterize the properties of materials. It is directly related to SOEC and density of that particular material as shown in Eqs. (1)-(3) and presented in **Table 3**.

It can be seen that the velocities of the chosen materials

Table 2. Bulk moduli (B_T) and tetragonal moduli (C_S) of CfY at room temperature in the unit of 10^{11} Dyne/cm^2.

Material	B_T	C_S
CfN	3.77	1.89
CfAs	2.47	1.99
CfSb	2.01	3.23

along longitudinal and shear waves increase with increase in temperature. The Debye average velocity in these materials is highest along <100> direction and lowest along <110> direction and increases with temperature as shown in **Figures 1-3**. Due to lack of experimental data for ultrasonic velocities of CfY, we have compared our results with other B1 structured materials like semiconductor [23] and rare-earth monopnictides [21,24] and rare-earth monochalcogenides [24,25] and found that the order of ultrasonic velocities and Debye average velocity have the similar nature . It is clear from **Figures 1-3**, that the computed values of the Debye average ultrasonic velocities are the highest in case of CfAs. So we can say that the propagation of sound waves through CfAs will be better than that of other chosen materials. SOEC and TOEC are used to obtain Grüneisen parameters and average squares of the Grüneisen parameters along <110> direction for longitudinal wave over 36 modes, for shear wave polarized along <001>

Table 3. Ultrasonic velocities (in 10^5cm/s) of CfY along different crystallographic directions in the temperature range 100 K - 500 K.

Materials	Directions	Velocity	100K	200K	300K	400K	500K
CfN	<100>	V_L	2.060	2.082	2.110	2.141	2.171
		$V_{S1} = V_{S2}$	1.359	1.361	1.364	1.367	1.369
	<111>	V_L	2.250	2.251	2.253	2.257	2.260
		$V_{S1} = V_{S2}$	1.198	1.219	1.244	1.270	1.295
	<110>	V_L	2.204	2.210	2.219	2.228	2.238
		V_{S1}	1.359	1.361	1.364	1.367	1.369
		V_{S2}	1.569	1.615	1.669	1.723	1.777
CfAs	<100>	V_L	2.088	2.124	2.174	2.198	2.234
		$V_{S1} = V_{S2}$	1.072	1.074	1.076	1.078	1.080
	<111>	V_L	1.924	1.926	1.930	1.933	1.937
		$V_{S1} = V_{S2}$	1.215	1.246	1.277	1.307	1.337
	<110>	V_L	1.966	1.978	1.990	2.003	2.015
		V_{S1}	1.072	1.074	1.076	1.078	1.080
		V_{S2}	1.812	1.872	1.946	1.991	2.048
CfSb	<100>	V_L	1.988	2.026	2.064	2.101	2.138
		$V_{S1} = V_{S2}$	0.981	0.983	0.985	0.987	0.989
	<111>	V_L	1.787	1.790	1.793	1.797	1.800
		$V_{S1} = V_{S2}$	1.158	1.190	1.221	1.251	1.281
	<110>	V_L	1.840	1.852	1.865	1.877	1.890
		V_{S1}	0.981	0.983	0.985	0.987	0.989
		V_{S2}	1.750	1.811	1.872	1.930	1.987

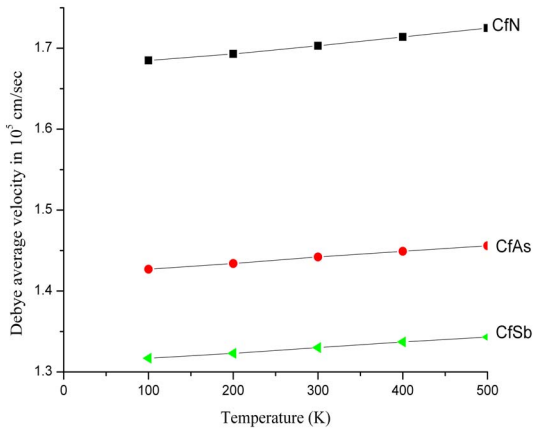

Figure 1. Debye average velocity versus temperature along <100> direction.

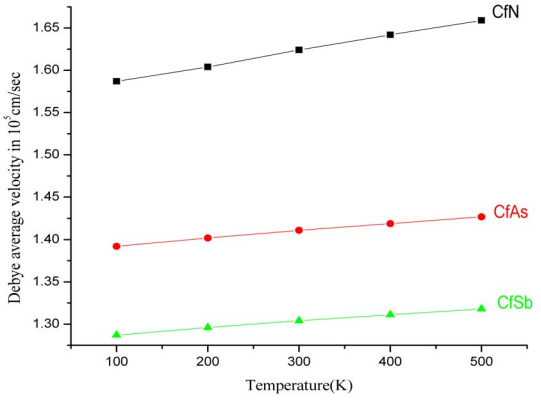

Figure 2. Debye velocity versus temperature along <110> direction.

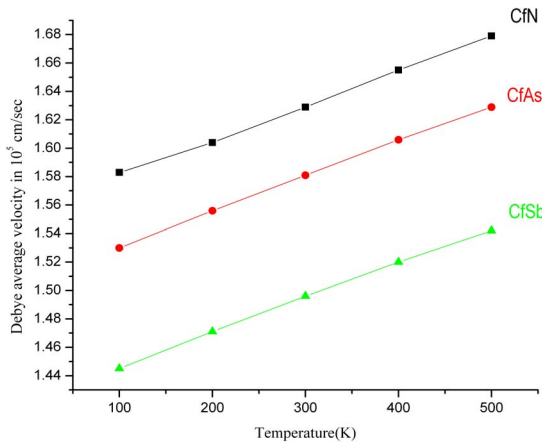

Figure 3. Debye average velocity versus temperature along <111> direction.

direction over 20 modes and for shear wave polarized along <110> direction over 18 modes and for shear wave polarized along <110> direction over 26 modes. The temperature dependent averaged ultrasonic Grüneisen parameters and averaged squares of the Grüneisen parameters are presented in **Table 4**.

The values of average Grüneisen parameters and averaged squares of the Grüneisen parameters are the highest for CfSb and lowest for CfN along <110> direction for longitudinal waves and shear as shown in **Table 4**. It is obvious from **Table 4**, that CfSb is better for thermal purposes for longitudinal wave propagation along <110> direction and CfN would be better for shear wave propagation. It is found that obtained values of Grüneisen

Table 4. Ultrasonic Grüneisen parameters of CfY along different crystallographic directions in the temperature range 100 K - 500 K.

Material	Grüneisen parameters	100K	200K	300K	400K	500K
		Ultrasonic longitudinal wave propagates along <100>				
CfN	$< \gamma_i^j >$	0.472	0.455	0.437	0.421	0.406
	$<(\gamma_i^j)^2>$	1.690	1.586	1.482	1.388	1.304
CfAs	$< \gamma_i^j >$	0.458	0.438	0.417	0.403	0.388
	$<(\gamma_i^j)^2>$	2.030	1.896	1.763	1.673	1.580
CfSb	$< \gamma_i^j >$	0.457	0.436	0.417	0.400	0.384
	$<(\gamma_i^j)^2>$	2.114	1.969	1.842	1.731	1.634
		Ultrasonic shear wave propagates along <100> and polarized along <100> direction				
CfN	$<(\gamma_i^j)^2>$	0.130	0.127	0.123	0.120	0.118
CfAs	$<(\gamma_i^j)^2>$	0.119	0.118	0.117	0.116	0.115
CfSb	$<(\gamma_i^j)^2>$	0.120	0.119	0.118	0.117	0.117
		Ultrasonic longitudinal wave propagates along <111>				
CfN	$< \gamma_i^j >$	–0.687	–0.655	–0.622	–0.592	–0.565
	$<(\gamma_i^j)^2>$	2.205	2.010	1.826	1.665	1.527
CfAs	$< \gamma_i^j >$	–0.720	–0.682	–0.644	–0.617	–0.589
	$<(\gamma_i^j)^2>$	2.333	2.100	1.881	1.730	1.584
CfSb	$< \gamma_i^j >$	–0.728	–0.688	–0.652	–0.620	–0.591
	$<(\gamma_i^j)^2>$	2.392	2.140	1.927	1.748	1.595
		Ultrasonic shear wave propagates along <110> and polarized along <110> direction				
CfN	$<(\gamma_i^j)^2>$	1.734	1.670	1.602	1.539	1.482
CfAs	$<(\gamma_i^j)^2>$	2.348	2.247	2.141	2.074	2.001
CfSb	$<(\gamma_i^j)^2>$	2.470	2.360	2.262	2.176	2.101
		Ultrasonic longitudinal wave propagates along <110>				
CfN	$< \gamma_i^j >$	–0.796	–0.767	–0.737	–0.708	–0.681
	$<(\gamma_i^j)^2>$	2.318	2.141	1.969	1.818	1.688
CfAs	$< \gamma_i^j >$	–0.768	–0.733	–0.695	–0.670	–0.642
	$<(\gamma_i^j)^2>$	2.551	2.362	2.180	2.056	1.933
CfSb	$< \gamma_i^j >$	–0.766	–0.728	–0.693	–0.661	–0.632
	$<(\gamma_i^j)^2>$	2.646	2.444	2.272	2.124	1.997
		Ultrasonic shear wave propagates along <110> and polarized along <001> direction				
CfN	$<(\gamma_i^j)^2>$	0.154	0.149	0.143	0.138	0.133
CfAs	$<(\gamma_i^j)^2>$	0.103	0.101	0.097	0.0961	0.093
CfSb	$<(\gamma_i^j)^2>$	0.099	0.097	0.094	0.092	0.090
		Ultrasonic shear wave propagates along <110> and polarized along direction<1 $\bar{1}$0>				
CfN	$<(\gamma_i^j)^2>$	2.524	2.428	2.325	2.227	2.136
CfAs	$<(\gamma_i^j)^2>$	3.471	3.306	3.132	3.020	2.899
CfSb	$<(\gamma_i^j)^2>$	3.659	3.478	3.317	3.173	3.046

parameters and average squares of the Grüneisen parameters are decreasing with increase in the temperature. This is due to adjustment of SOEC and TOEC for different modes. This type of nature is also found in other B1 structured materials like rare-earth monopnictides [21,22], semiconductors [23] and rare-earth monochalcogenides [24,25].

4. Conclusions

On the basis of analysis of the above results, we can say that

- Evaluated values of SOEC and TOEC have been compared with available same type B1 structured mate-

rials, which are in agreement; hence our approach to compute elastic constants is justified.
- SOEC and TOEC of CfN are highest so mechanical properties will be better than other CfY.
- All the chosen materials follow the Born criterion of stability. So they all are stable.
- SOEC and TOEC have been used to find out the ultrasonic velocities for longitudinal and shear waves, Debye average velocity and Grüneisen parameters in CfY.
- Ultrasonic velocity is found to be the highest for CfAs along all chosen directions, so CfAs will be the most suitable compound for wave propagation.
- CfSb is better for longitudinal wave propagation along <110> direction and CfN would be better for thermal purposes, because Grüneisen parameters are the most sensitive to temperature.

Hence, we conclude that current approach is justified and obtained results will be useful for finding various theoretical, experimental investigations like ultrasonic attenuation, non-linearity parameters, ultrasonic measurements, polarizing microscopy, solid state NMR, SEM, TEM.

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