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# Computations of ultrasonic parameters of lanthanide metals Ti, Zr and Hf

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## Abstract

The ultrasonic properties in the hexagonal structured lanthanide metals Ti, Zr and Hf have been studied at room temperature. The higher order elastic constants have been calculated for these metals using Lennard-Jones Potential for evaluating orientation-dependent ultrasonic properties. An anomalous behaviour in orientation dependent velocity is obtained, which is due to the combined effect of second order elastic constants and density. The achieved ultrasonic properties are compared with known physical parameters to differentiate them from third group nitrides/laves phase compounds.

**Key Words:** Lanthanide metals, elastic properties, thermal conductivity, ultrasonic properties.

**PACS number:** 43.35.cg; 62.20.Dc; 63.20.Kr

## 1. Introduction

At room temperature and ambient pressure, the stable crystalline state of the early transition metals Ti, Zr and Hf is a hexagonal closed packed (hcp) structure. The lanthanides are used as catalyst to speed up chemical reactions, refining agent for the conversion of crude oil into gasoline, kerosene and diesel. They are also used as phosphors, such as in colour television sets. Other lanthanide compounds are used in streetlights, searchlights, and in the high-intensity lighting present in sports stadiums.

At high temperature, all these three metals undergo a phase transformation into a body-centered cubic (bcc) structure. The crystal structures of these metals and their phase transformation under effect of pressure have been studied by Xia et.al. [1]. The electronic structure and properties of these transition metals have been

studied by some methods of energy band theory [2]. You-qing et al. have described the electronic structure and properties of these metals via one-atom theory [2]. The XRD and TEM study explore that the alloy of these transition metals with platinum forms the solid solution at nanoscale [3]. The hydrogen storing property of Ti/Zr-Hf quasi-crystal has been explained by Kelton and co-workers [4]. The elastic behaviour of Ti, Zr and Hf has been examined by the calculation through first principle. In ground state, the total energy of these metals has minimum value for hcp structure in comparison to their fcc/bcc structure, thus they possess hcp structure in ground state [5].

Ultrasound is one of several effective methods used in the field of non-destructive testing (NDT) and inspection. Non-destructive ultrasonic techniques are used to study physical properties of the materials through the process of evaluating and measuring the ultrasonic attenuation. Ultrasonic attenuation is very important physical parameter to characterize the material, which is well related to several physical quantities, such as thermal conductivity, specific heat, thermal energy density and higher order elastic constants. Ultrasonic characterization of  $XCr_2$  ( $X = \{\text{Ti, Zr and Hf}\}$ ) can be found in the literature [6]. In the present investigation, the ultrasonic properties of hexagonal structured lanthanide metals have been characterized at room temperature. The ultrasonic attenuation coefficient, acoustic coupling constants, higher order elastic constants, thermal relaxation time and ultrasonic wave velocities for these metals for each direction of propagation of wave are calculated temperature at 300 K. The obtained results are analyzed in comparison to other hexagonal structured materials.

## 2. Theory

### 2.1. Higher order elastic constants

The elastic energy density  $U$  is a function of the strain components

$$U = F(\varepsilon_{xx}; \varepsilon_{yy}; \varepsilon_{zz}; \varepsilon_{yz}; \varepsilon_{zx}; \varepsilon_{xy}) = F(\varepsilon_1; \varepsilon_2; \varepsilon_3; \varepsilon_4; \varepsilon_5; \varepsilon_6) \quad (1)$$

where  $\varepsilon_{ij}$  ( $i$  or  $j = x, y, z$ ) are components of the strain tensor. The second ( $C_{IJ}$ ) and third ( $C_{IJK}$ ) order elastic constants of material are defined by expressions

$$C_{IJ} = \frac{\partial^2 U}{\partial \varepsilon_I \partial \varepsilon_J}; \quad I \text{ or } J = 1, \dots, 6 \quad (2)$$

$$C_{IJK} = \frac{\partial^3 U}{\partial \varepsilon_I \partial \varepsilon_J \partial \varepsilon_K}; \quad I \text{ or } J \text{ or } K = 1, \dots, 6 \quad (3)$$

The elastic energy density is well related to interaction potential  $\phi(r)$  between atoms. Let the interaction potential be the Lennard-Jones Potential or many body interaction potential, which is formulated as

$$\Phi(r) = -\frac{a_0}{r^m} + \frac{b_0}{r^n}, \quad (4)$$

where  $a_0, b_0$  are constants and  $m, n$  are integers. The definition of higher order elastic constants, as expressed in equations (2) and (3), with potential (4) under equilibrium and symmetric condition, leads six second and

ten third order elastic constants (SOEC and TOEC) for the hexagonal closed packed structured materials [7, 8]:

$$\left. \begin{aligned} C_{11} &= 24.1p^4C' & C_{12} &= 5.918p^4C' \\ C_{13} &= 1.925p^6C' & C_{33} &= 3.464p^8C' \\ C_{44} &= 2.309p^4C' & C_{66} &= 9.851p^4C' \end{aligned} \right\} \quad (5a)$$

$$\left. \begin{aligned} C_{111} &= 126.9p^2B + 8.853p^4C' & C_{112} &= 19.168p^2B - 1.61p^4C' \\ C_{113} &= 1.924p^4B + 1.155p^6C' & C_{123} &= 1.617p^4B - 1.155p^6C' \\ C_{133} &= 3.695p^6B & C_{155} &= 1.539p^4B \\ C_{144} &= 2.309p^4B & C_{344} &= 3.464p^6B \\ C_{222} &= 101.039p^2B + 9.007p^4C' & C_{333} &= 5.196p^8B, \end{aligned} \right\} \quad (5b)$$

where  $p = c/a$  is the axial ratio;  $C' = \chi a/p^5$ ;  $B = \psi a^3/p^3$ ; and the rest second and third order elastic constants have zero value, because under  $180^\circ$  rotation they have equal and opposite value for the same stress. The harmonic and anharmonic parameters ( $\chi$  and  $\psi$ ) can be calculated using one experimental SOEC [7, 8]. In the present study, we have expanded the theory for theoretical evaluation of parameters  $\chi$  and  $\psi$ . The potential energy can be expanded in the powers of changes in the squares of distances. The expansion up to cubic term can be written as

$$\Phi = \Phi_0 + \chi \sum_{i=1}^2 [\Delta r_i^2]^2 + \psi \sum_{i=1}^2 [\Delta r_i^2]^3 \quad (6)$$

According to the equation (6),  $\chi$  and  $\psi$  can be written as

$$\chi = \frac{1}{2!} \left[ \frac{d^2\Phi(r)}{d(r^2)^2} \right] \quad (7)$$

$$\psi = \frac{1}{3!} \left[ \frac{d^3\Phi(r)}{d(r^2)^3} \right] \quad (8)$$

In solving equations (7) and (8) for hexagonal closed packed structured materials, we have the relations

$$\chi = (1/8)[\{nb_0(n-m)\}/\{a^{n+4}\}] \quad (9)$$

$$\psi = -\chi/\{6a^2(m+n+6)\}. \quad (10)$$

Parameters  $\chi$  and  $\psi$  can be calculated using equations (9) and (10) with appropriate values of  $m$ ,  $n$  and  $b_0$  so that the calculated values of elastic constants justify the experimental data.

## 2.2. Ultrasonic attenuation and allied parameters

Predominant cause for ultrasonic attenuation in a solid at room temperature is phonon-phonon interaction (Akhieser loss) and thermoelastic relaxation mechanisms. The ultrasonic attenuation coefficient  $(A)_{Akh}$  due to phonon-phonon interaction and thermoelastic relaxation mechanisms is given by the expressions [7, 8]

$$(A/f^2)_{Akh} = 4\pi^2 \left( 3E_0 \langle (\gamma_i^j)^2 \rangle - \langle \gamma_i^j \rangle^2 C_V T \right) \tau / 2\rho V^3 \quad (11)$$

$$(A/f^2)_{Th} = 4\pi^2 \langle \gamma_i^j \rangle^2 kT/2\rho V_L^5, \quad (12)$$

where  $f$  denotes frequency of the ultrasonic wave;  $V$  is the ultrasonic velocity for longitudinal and shear waves;  $V_L$  is longitudinal ultrasonic velocity;  $E_0$  is thermal energy density; and  $\gamma_i^j$  denotes Grüneisen number ( $i, j$  are the mode and direction of propagation, respectively).

The Grüneisen number for hexagonal structured crystal along  $\langle 001 \rangle$  orientation or  $\theta = 0^\circ$  is a direct consequence of second and third order elastic constants. The relation  $D = 3 \left( 3E_0 \langle (\gamma_i^j)^2 \rangle - \langle \gamma_i^j \rangle^2 C_V T \right) / E_0$  is known as acoustic coupling constant, which is the measure of acoustic energy converted to thermal energy. When the ultrasonic wave propagates through crystalline material, the equilibrium of phonon distribution is disturbed. The time for re-establishment of equilibrium of the thermal phonon distribution is called the thermal relaxation time  $\tau$  and is given by the expression

$$\tau = \tau_S = \tau_L/2 = 3k/C_V V_D^2 \quad (13)$$

Here  $\tau_L$  and  $\tau_S$  are the thermal relaxation time for longitudinal and shear wave.  $k$  and  $C_V$  denote the thermal conductivity and specific heat per unit volume of the material, respectively. The Debye average velocity  $V_D$  is well-related to longitudinal ( $V_L$ ) and shear wave ( $V_{S1}, V_{S2}$ ) velocities. The expressions for ultrasonic velocities are [7, 8]

$$V_L^2 = \{C_{33}\cos^2\theta + C_{11}\sin^2\theta + C_{44} + \{[C_{11}\sin^2\theta - C_{33}\cos^2\theta + C_{44}(\cos^2\theta - \sin^2\theta)]^2 + 4\cos^2\theta\sin^2\theta(C_{13} + C_{44})^2\}^{1/2}\}/2\rho \quad (14)$$

$$V_{S1}^2 = \{C_{33}\cos^2\theta + C_{11}\sin^2\theta + C_{44} - \{[C_{11}\sin^2\theta - C_{33}\cos^2\theta + C_{44}(\cos^2\theta - \sin^2\theta)]^2 + 4\cos^2\theta\sin^2\theta(C_{13} + C_{44})^2\}^{1/2}\}/2\rho \quad (15)$$

$$V_{S2}^2 = \{C_{44}\cos^2\theta + C_{66}\sin^2\theta\}/\rho \quad (16)$$

$$V_D = \left\{ \frac{1}{3} \left( \frac{1}{V_L^3} + \frac{1}{V_{S1}^3} + \frac{1}{V_{S2}^3} \right) \right\}^{-1/3}, \quad (17)$$

where  $\rho$  and  $\theta$  are the density of the material and angle with the unique axis of the crystal, respectively. The above formulations have been used for the calculation of ultrasonic attenuation and allied parameters in the chosen lanthanide metals.

### 3. Results and discussion

#### 3.1. Higher order elastic constants

The unit cell parameters  $a$  (basal plane parameter) and  $p$  (axial ratio) for Ti, Zr and Hf are 2.95 Å, 3.23 Å, 3.19 Å and 1.586, 1.594, 1.583, respectively. The value of  $m$  and  $n$  for chosen metals are 6 and 7. The values of  $b_0$  are  $0.875 \times 10^{-64}$  erg cm<sup>7</sup>,  $1.79 \times 10^{-64}$  and  $1.93 \times 10^{-64}$  for Ti, Zr and Hf respectively. The second and third order elastic constants have been calculated for lanthanide metals using equation (5) and are presented in Table 1.

**Table 1.** Second and third order elastic constants of lanthanide metals at 300 K, in units of  $\times 10^{10}$  Nm<sup>-2</sup>.

SOEC	Ti	Zr	Hf	TOEC	Ti	Zr	Hf
C <sub>11</sub>	23.308	19.159	23.561	C <sub>111</sub>	-380.086	-312.419	-384.208
C <sub>12</sub>	5.724	4.705	5.786	C <sub>112</sub>	-60.262	-49.533	-60.915
C <sub>13</sub>	4.683	3.888	4.716	C <sub>113</sub>	-12.012	-9.973	-12.097
C <sub>33</sub>	21.197	17.778	21.266	C <sub>123</sub>	-15.267	-12.676	-15.374
C <sub>44</sub>	5.617	4.664	5.657	C <sub>133</sub>	-71.602	-60.050	-71.832
C <sub>66</sub>	9.141	7.513	9.240	C <sub>344</sub>	-67.125	-56.296	-67.341
B	10.85	8.98	10.93	C <sub>144</sub>	-17.788	-14.769	-17.913
[5]	11.20	9.50	10.90	C <sub>155</sub>	-11.856	-9.844	-11.939
[9]	10.84	8.98	10.90	C <sub>222</sub>	-330.734	-247.194	-303.996
[10]	10.60	9.50	10.80	C <sub>333</sub>	-253.270	-214.560	-253.125
[11]	10.51	8.33	10.90				

The elastic constants are important, since they are related to hardness and are used for the determination of the ultrasonic attenuation. The bulk moduli of chosen metals are evaluated using the second order elastic constants. It is obvious from Table 1 that there is good agreement between the present and reported theoretical/experimental bulk moduli [5, 9–11]. Hence applied theory for the evaluation of higher order elastic constants at room temperature is justified. The second order elastic constants for Zr lies in between Ti and Hf because the total lattice energy of Zr lies between them [5]. It is observed that the higher order elastic constants of these metals are comparable to Laves phase compounds [6].

### 3.2. Ultrasonic velocity and allied parameters

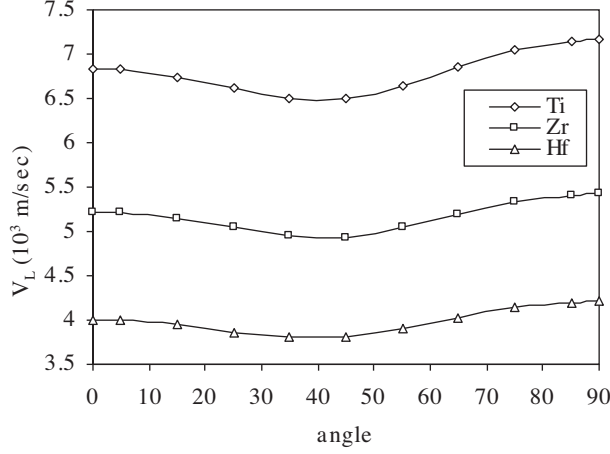
The density  $\rho$  and thermal conductivity  $k$  at room temperature have been taken from the literature [9]. The value of specific heat per unit volume  $C_V$  and thermal energy density  $E_0$  are evaluated using tables of physical constants and Debye temperature. The quantities  $k$ ,  $\rho$ ,  $C_v$  and  $E_0$  and calculated acoustic coupling constants  $D_L$  and  $D_S$  are shown in Table 2.

**Table 2.** Density  $\rho$ , specific heat per unit volume  $C_V$ , thermal energy density  $E_0$ , thermal conductivity  $k$  and acoustic coupling constants  $D_L$  and  $D_S$  of the covered lanthanide metals.

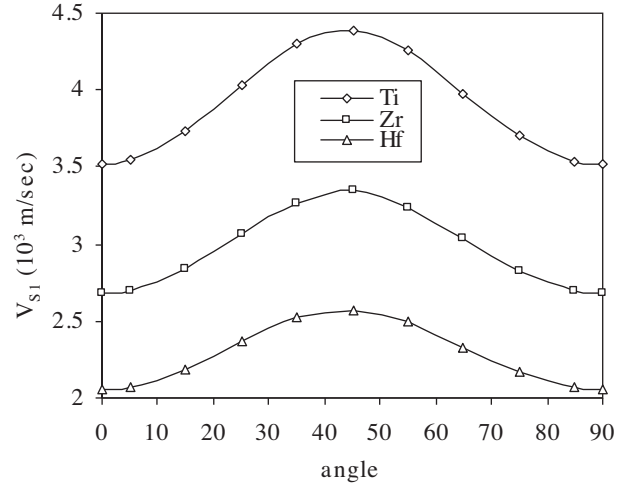
	Ti	Zr	Hf
$\rho$ ( $\times 10^3$ kgm <sup>-3</sup> )	4.54	6.51	13.31
$C_V$ ( $\times 10^6$ Jm <sup>-3</sup> K <sup>-1</sup> )	1.995	1.642	1.776
$E_0$ ( $\times 10^8$ Jm <sup>-3</sup> )	3.272	3.174	3.373
$k$ (Wm <sup>-1</sup> K <sup>-1</sup> )	21.9	22.7	23.0
$D_L$	55.965	56.314	56.521
$D_S$	1.406	1.249	1.418

The calculated orientation dependent ultrasonic wave velocities and Debye average velocities at 300 K are shown in Figures 1–4. Figures 1–3 show that the velocities  $V_L$  and  $V_{S1}$  have minima and maxima respectively at 45° with the unique axis of the crystal while  $V_{S2}$  increases with the angle from the unique axis.

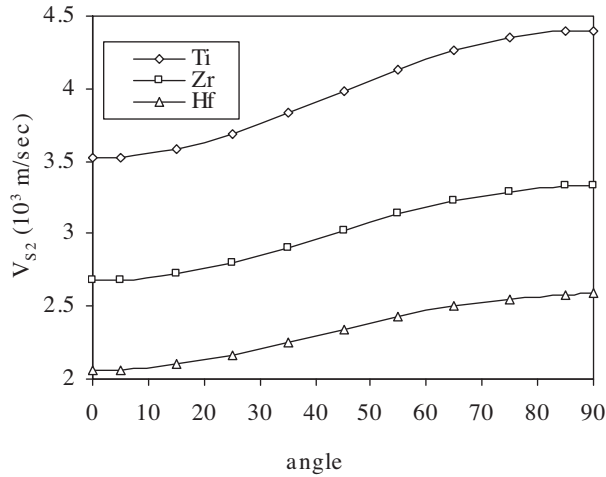
The combined effect of second order elastic constants and density is reason for abnormal behaviour of angle dependent velocities.



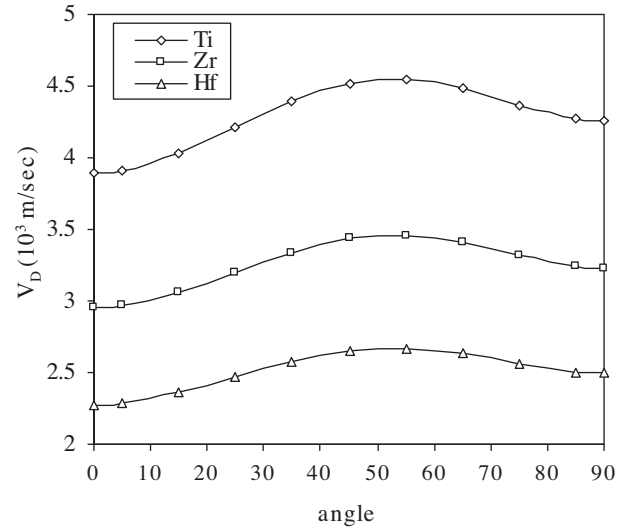
**Figure 1.**  $V_L$  versus angle (in degrees) with respect to the unique axis of the crystal.



**Figure 2.**  $V_{S1}$  versus angle (in degrees) with respect to the unique axis of the crystal.



**Figure 3.**  $V_{S2}$  versus angle (in degrees) with respect to the unique axis of the crystal.

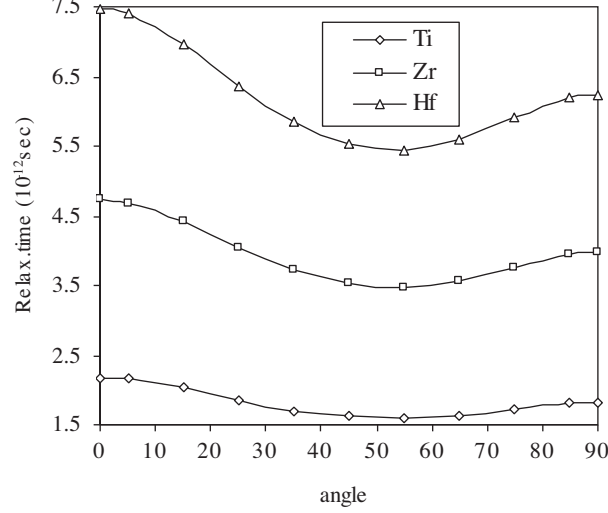


**Figure 4.**  $V_D$  versus angle (in degrees) with respect to the unique axis of the crystal.

The nature of the angle dependent velocity curves in the present work is found similar to that for Laves phase compounds and hexagonal wurtzite structured materials (GaN, AlN, InN, CdS and CdSe) [6, 12–14]. Thus the computed velocities for these lanthanide metals are justified.

Debye average velocities  $V_D$  of these metals are increasing with the angle and have maxima at  $55^\circ$  at 300 K (Figure. 4). Since  $V_D$  is calculated using  $V_L$ ,  $V_{S1}$  and  $V_{S2}$  [7, 8], the angle variation of  $V_D$  is influenced by the constituent ultrasonic velocities. The maximum  $V_D$  at  $55^\circ$  is due to a significant increase in longitudinal and

pure shear ( $V_{S2}$ ) wave velocities and a decrease in quasi-shear ( $V_{S1}$ ) wave velocity. Thus it can be concluded that the average sound wave velocity is maximum for a sound wave traveling at  $55^\circ$  with respect to the unique axis of these crystals.



**Figure 5.** Relaxation time versus angle (in degrees) with respect to the unique axis of the crystal.

The calculated thermal relaxation time is visualized in Figure 5. The angle dependent thermal relaxation time curves follow the reciprocal nature of  $V_D$  as  $\tau \propto V_D^{-2}$ .  $\tau$  has maximum values 2.18 ps, 4.73 ps and 7.48 ps for the wave propagation along unique axis ( $\theta = 0^\circ$ ) of metals Ti, Zr and Hf, respectively. The minimum values of  $\tau$  for Ti, Zr and Hf are found to be 1.59 ps, 3.5 ps and 5.45 ps, respectively, along  $\theta = 55^\circ$ . The order of  $\tau$  for hexagonal structured materials is in picoseconds [12]. Hence the calculated  $\tau$  justifies the hcp structure of chosen metals at room temperature. The minimum thermal relaxation time for wave propagation along  $\theta = 55^\circ$  implies that the re-establishment time for the equilibrium distribution of thermal phonons will be minimum for propagation of wave along this direction.

### 3.3. Ultrasonic attenuation

In the evaluation of ultrasonic attenuation, it is supposed wave propagation occurs along the unique axis ( $\langle 001 \rangle$  direction) of the lanthanide metals. The attenuation coefficient over frequency square  $(A/f^2)_{Akh}$ , for longitudinal  $(A/f^2)_L$  and shear wave  $(A/f^2)_S$  are calculated using equation (11) under the condition  $\omega\tau \ll 1$ , at room temperature. The thermoelastic loss over frequency square,  $(A/f^2)_{Th}$ , is calculated with equation (5). The values of  $(A/f^2)_L$ ,  $(A/f^2)_S$ ,  $(A/f^2)_{Th}$  and total attenuation  $(A/f^2)_{Total}$  are given in Table 3.

**Table 3.** Ultrasonic attenuation coefficient (in  $\times 10^{-17}$  Nps $^2$ m $^{-1}$ ) of lanthanide metals.

	Ti	Zr	Hf
$(A/f^2)_{Th}$	0.091	0.253	0.475
$(A/f^2)_L$	72.400	239.339	488.050
$(A/f^2)_S$	6.666	19.746	44.617
$(A/f^2)_{Total}$	79.157	259.338	533.142



Table 3 indicates that the thermoelastic loss is very small in comparison to Akhieser loss and ultrasonic attenuation for longitudinal wave  $(A/f^2)_L$  is greater than that of shear wave  $(A/f^2)_S$ . This reveals that ultrasonic attenuation due to phonon-phonon interaction along longitudinal wave is the factor governing total attenuation  $((A/f^2)_{Total} = (A/f^2)_{Th} + (A/f^2)_L + (A/f^2)_S)$ . The total attenuation is mainly affected by thermal conductivity and acoustic coupling constants. The total attenuation of these transition metals are larger than group III nitrides (AlN:  $4.441 \times 10^{-17}$  Nps<sup>2</sup>m<sup>-1</sup>; GaN:  $14.930 \times 10^{-17}$  Nps<sup>2</sup>m<sup>-1</sup> and InN:  $20.539 \times 10^{-17}$  Nps<sup>2</sup>m<sup>-1</sup>) due to their large thermal conductivity and acoustic coupling constants [6, 12]. This implies that the interaction between acoustical phonon and quanta of lattice vibration for these metals is large in comparison to group III nitrides. The density of states at the Fermi energy (in states eV<sup>-1</sup> atom<sup>-1</sup>) for Ti, Zr and Hf are 0.90, 0.87 and 0.68, respectively [5]. The values of total ultrasonic attenuation for the present investigated metals Ti, Zr and Hf are found  $79.157 \times 10^{-17}$  Nps<sup>2</sup>m<sup>-1</sup>,  $259.338 \times 10^{-17}$  Nps<sup>2</sup>m<sup>-1</sup> and  $533.142 \times 10^{-17}$  Nps<sup>2</sup>m<sup>-1</sup> respectively (Table 3). Hence, the ultrasonic attenuation has reciprocal nature to the density of states for these metals. At room temperature and zero pressure, the Ti metal is quite stable in hcp structure [5]. The minimum ultrasonic attenuation for Ti justifies its quite stable hcp structure state.

## 4. Conclusion

The above discussion reveals that:

- Our theory of higher order elastic constants is justified for hexagonal structured lanthanide metals.
- The higher order elastic constants, bulk modulus and velocity have low values for these metals in comparison to group III nitrides.
- The order of thermal relaxation time for these metals is found on the order of picoseconds, which justifies their hcp structure at 300 K. The re-establishment time for the equilibrium distribution of thermal phonons will be minimum for the wave propagation along  $\theta = 55^\circ$  due to being smallest value of  $\tau$  along this direction.
- The acoustic coupling constants of these metals for longitudinal wave are found five times larger than GaN /AlN /InN. Hence the conversion of acoustic energy into thermal energy will be large for these metals.
- The dominant mechanism for ultrasonic attenuation in these metals is phonon-phonon interaction. The thermal conductivity and acoustic coupling constant are the governing factors to the total attenuation. The density of states at Fermi energy of these metals can be understood with the knowledge of total ultrasonic attenuation.
- The mechanical properties of these metals will not be better than the group III nitrides due to their low ultrasonic velocity, second order elastic constants and high ultrasonic attenuation.

These results, together with other well-known physical properties, may expand future prospects for the application and study of lanthanides and their compounds.

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