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Temperature dependent elastic and ultrasonic properties of ytterbium monopnictides

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Ultrasonic properties of ytterbium monopnictides (YbX: X=As, P) have been evaluated along <100>, <110> and <111> directions in the temperature range 100-400 K. The second-and third-order elastic constants (SOECs and TOECs) are calculated for the computation of ultrasonic parameters. Other elastic constants like bulk modulus, shear modulus, Young's modulus, Poisson ratio, anisotropic ratio, tetragonal moduli and fracture/toughness ratio are also calculated to evaluate the performance of materials under investigation. The elastic behaviour of these materials shows their brittle nature. It is found that the ultrasonic attenuation in ytterbium monopnictides increases with temperature and have quadratic function of temperature. The nature of ultrasonic attenuation was found to be quite similar to the other observed results of rare-earth monopnictides.

Keywords: Monopnictides, Elastic constants, Ultrasonic properties

1 Introduction

The doping method is the easiest and most popular technique to design low carrier system. This method was first applied in f-electron systems particularly on the magnetic semiconductors. The semimetals and narrow gap semiconductors belong to this class, in which the carrier concentration may be controlled by pressure, magnetic field and temperature. The rareearth monopnictides, with the NaCl-type crystal structure have been found to be most potential materials under this category.

The pnictogen p state shows the dominance at the top of valence band which is a typical feature of most rare-earth monopnictides. The chosen compounds YbAs and YbP belong to low carrier heavy fermion system having same band structure and carrier concentration. It possesses Winger crystallization because the effective mass of holes is heavier than the electrons in these materials. Kasuya *et al*^{1,2}. studied such materials under different physical conditions like high pressure and high magnetic field. The temperature dependent resistivity of these materials shows the Kondo effect³. The Kondo effect is a phenomenon of divergence in resistivity at low temperatures caused by scattering of electron due to interaction between

conduction electrons and local moments. Thus, the chosen ytterbium monopnictides are, generally, known as low carrier heavy fermion Kondo system.

The ultrasonic investigations have been used as a versatile tool in studying microstructural features and inherent properties of different types of substances like metals⁴, semiconductors⁵, dielectrics⁶, semimetals⁷, intermetallics⁸, superconductors⁹, nanomaterials¹⁰, polymer¹¹, binary mixtures^{12,13}, amino acids in aqueous solution¹⁴, nitrile butyl rubber blends¹⁵, yeast enzyme¹⁶ and metallic alloys¹⁷. In the present work, ultrasonic attenuation, ultrasonic velocity and their related parameters (thermal relaxation time, Debye average velocity and acoustic coupling constant) are evaluated for YbAs and YbP semimetallics as a function of temperature and orientations.

2 Theoretical Approach

This section concentrates on the theoretical approach for the calculation of ultrasonic parameters and their correlated parameters.

2.1 Second-and third order elastic constants

The elastic energy density (U) is defined as a function of the strain components i.e.

$$U = F(e_{xx}, e_{yy}, e_{zz}, e_{yz}, e_{zx}, e_{xy})$$

= $F(e_1, e_2, e_3, e_4, e_5, e_6)$...(1)

where e_{ij} (i or j = x, y, z) is component of strain tensor. The second (C_{IJ}) and third (C_{IJK}) order elastic constants of material are defined as follows:

$$C_{IJ} = \frac{\partial^2 U}{\partial e_I \partial e_J}; \qquad \text{I or J} = 1,.....6 \qquad ...(2)$$

$$C_{IJK} = \frac{\partial^3 U}{\partial e_I \partial e_J \partial e_K}; \text{ I, J or K} = 1,.....6 \qquad ...(3)$$

The elastic energy density is related to interaction potential $\phi(r)$ between atoms. The potential used for evaluation of SOECs and TOECs is taken as sum of Coulomb and Born-Mayer potentials.

$$\phi(r) = \phi(C) + \phi(B) \qquad \dots (4)$$

where $\phi(C)$ is electrostatic or Coulomb potential and $\phi(B)$ is the repulsive or Born-Mayer potential, given as $\phi(C) = \pm (e^2 / r)$ and $\phi(B) = A_0 \exp(-r / b)$. Here 'e' is electronic charge; *r*, nearest neighbour distance; *b*, hardness parameter and A_0 is strength parameter.

The higher order elastic constants at different temperatures can be obtained with the help of anharmonic theory of lattice dynamics proposed by Leibfried and Ludwig¹⁸. According to this theory, the vibrational energy changes with temperature. Therefore, temperature dependent SOECs and TOECs can be obtained by adding vibrational energy contribution to static elastic constants at absolute zero temperature.

$$C_{IJ} = C_{IJ}^0 + C_{IJ}^{Vib}$$
 and $C_{IJK} = C_{IJK}^0 + C_{IJK}^{Vib}$...(5)

where superscript '0' has been used to denote SOECs and TOECs at 0 K (static elastic constants) and superscript '*Vib*' has been used to denote vibrational part of SOEC and TOEC at a particular temperature. The expressions for SOECs and TOECs are available in our previous studies¹⁹. The computed SOECs are used for the calculation of other mechanical parameters⁷ such as tetragonal moduli, bulk moduli, shear moduli and are very useful for the prediction of material performance.

2.2 Ultrasonic velocities

There are three modes of propagation of sound wave in a crystalline medium. The modes of propagation are longitudinal acoustical, quasi shear acoustical and shear acoustical. Hence, there exist three types of ultrasonic velocities as longitudinal (V_L) , quasi shear (V_{S1}) and shear (V_{S2}) which depend on the direction of propagation of wave²⁰⁻²². The expressions for direction dependent ultrasonic velocities in cubic crystals are:

Along <100> crystallographic direction:

$$V_L = \sqrt{\frac{C_{11}}{\rho}}$$
; $V_{S1} = V_{S2} = \sqrt{\frac{C_{44}}{\rho}}$...(6)

Along <110> crystallographic direction:

$$V_{L} = \sqrt{\frac{C_{11} + C_{12} + 2C_{44}}{2\rho}} ; V_{S1} = \sqrt{\frac{C_{44}}{\rho}} ;$$
$$V_{S2} = \sqrt{\frac{C_{11} - C_{12}}{\rho}}$$
...(7)

Along <111> crystallographic direction:

$$V_{L} = \sqrt{\frac{C_{11} + 2C_{12} + 4C_{44}}{3\rho}};$$

$$V_{51} = V_{52} = \sqrt{\frac{C_{11} - C_{12} + C_{44}}{3\rho}}$$
...(8)

The ultrasonic velocities can be computed using SOECs and density of the materials. The Debye average velocity (V_D) provides knowledge about Debye temperature and thermal relaxation time of the materials. The expression for Debye average velocity⁷ is given as:

$$V_{D} = \left[\frac{1}{3}\left\{\frac{1}{V_{L}^{3}} + \frac{2}{V_{S1}^{3}}\right\}\right]^{-1/3};$$

along <100> and <111> direction
$$= \left[\frac{1}{3}\left\{\frac{1}{V_{L}^{3}} + \frac{1}{V_{S1}^{3}} + \frac{1}{V_{S2}^{3}}\right\}\right]^{-1/3};$$

along <110> direction
$$(9)$$

The Debye temperature (θ_D) is an important physical parameter of solids, which defines a division line between quantum and classical behaviour of phonons^{7,23,24}. The Debye temperature can be obtained using following expression.

$$\theta_D = \frac{h \, V_D \, \left(6 \, \pi^2 \, n_a\right)^{1/3}}{2\pi k_B} \qquad \dots (10)$$

where k_B is Boltzmann constant, h the Planck's constant and n_a is atom concentration.

2.3 Ultrasonic attenuation and related parameters

Mason's theory for solids, proposed by Akhieser²⁵ has been well established for the temperature dependence study of ultrasonic attenuation and related ultrasonic parameters for different classes of materials along different directions. An effort has been made to establish this theory for the evaluation of ultrasonic attenuation coefficient of semimetallic YbP and YbAs and related mechanical/thermal parameters.

The principal causes of ultrasonic attenuation in a single crystal at high temperature are phonon-phonon interaction and thermo-elastic relaxation mechanism^{20,26-29}. The ultrasonic attenuation due to phonon-phonon interaction (Akhieser loss) for longitudinal and shear waves under condition $\omega \tau \leq 1$ is given by:

$$\left(\alpha/f^{2}\right)_{\text{Long}} = \frac{4\pi^{2}\tau E_{0} (D_{L}/3)}{2 \rho V_{L}^{3}} \qquad \dots (11)$$

$$\left(\alpha/f^{2}\right)_{\text{Shear}} = \frac{4\pi^{2}\tau E_{0} (D_{s}/3)}{2 \rho V_{s}^{3}} \qquad \dots (12)$$

where α : is ultrasonic attenuation coefficient, f the frequency of ultrasonic wave, ρ : density, E_0 : thermal energy density, V_L and V_S the velocity of longitudinal and shear waves, respectively, τ : thermal relaxation time, D_L and D_S are acoustic coupling constant for longitudinal and shear waves, respectively. The re-establishment time of thermal phonon after passing the ultrasonic wave is called as thermal relaxation time. The value of this time for longitudinal wave is twice that for shear wave.

$$\tau = \tau_{\text{Shear}} = \frac{\tau_{\text{Long}}}{2} = \frac{3K}{C_V V_D^2} \qquad \dots (13)$$

where C_V is the specific heat per unit volume and K is thermal conductivity.

The acoustic coupling constant is measure of ultrasonic energy converted to the thermal energy which is obtained with the expression:

$$D = 9 < \gamma^2 > -3 < \gamma >^2 (C_v T / E_0) \qquad \dots (14)$$

Grüneisen parameter³⁰ (γ) is well related to the SOECs and TOECs. In case of shear wave propagation, there is no thermoelastic loss of ultrasonic energy because there is no compression or rarefaction. The propagation of longitudinal ultrasonic wave creates compression and rarefaction throughout the lattice. The rarefied regions are colder than that of the compressed region. Thus, there is flow of heat between these two regions. The expression for ultrasonic attenuation caused by thermoelastic relaxation is given by:

$$\left(\alpha/f^{2}\right)_{\text{th}} = \frac{4\pi^{2} < \gamma >^{2} KT}{2 \rho V_{L}^{5}}$$
 ...(15)

3 Results and Discussion

3.1 Second and third order elastic constants

The lattice parameters of YbP and YbAs are 5.698 Å and 6.06 Å while the hardness parameters are 0.315 Å and 0.311 Å, respectively. The SOECs and TOECs of the chosen materials are calculated using the lattice and hardness parameters in the temperature range 100-400 K with help of Eqs (1-5). The calculated values of temperature dependent SOECs and TOECs are presented in Table 1.

Table 1 presents that out of nine elastic constants, four $(C_{11}, C_{44}, C_{112}$ and C_{144}) having positive temperature coefficient increase with temperature while other four $(C_{12}, C_{111}, C_{123} \text{ and } C_{166})$ having negative temperature coefficient decrease with increase in temperature and the value of C_{456} remains constant in this temperature range due to zero vibrational contribution. This behaviour is similar to studied rare-earth compounds like previously gadolinium praseodymium monopnictides', monopnictides²⁷, thulium monochalcogenides²⁸, lanthanum monochalcogenides³¹ and americium monopnictides³².

The bulk modulus (*B*), shear modulus (*G*), Young's modulus (*Y*), tetragonal modulus (*C_s*), anisotropic modulus (*A_m*) and Poisson's ratio (σ) of the material under investigation are calculated using following expressions:

Table 1 — Second and third order erastic constants of TDAs and TDF in the temperature range 100-400 K											
Material	<i>T</i> [K]	SOEC (10 ¹⁰ Nm ⁻²)			TOEC (10 ¹⁰ Nm ⁻²)						
		C_{11}	C_{12}	C_{44}	C_{111}	C_{112}	C_{123}	C_{144}	C_{166}	C_{456}	
YbAs	100	4.749	1.365	1.445	-75.51	-5.599	1.956	2.394	-5.899	2.375	
	200	4.909	1.291	1.451	-76.247	-5.326	1.536	2.412	-5.924	2.375	
	300	5.079	1.218	1.457	-77.074	-5.053	1.117	2.431	-5.951	2.375	
	400	5.250	1.144	1.463	-77.926	-4.781	0.697	2.45	-5.977	2.375	
YbP	100	4.358	1.020	1.097	-72.098	-4.132	1.407	1.871	-4.447	1.857	
	200	4.503	0.948	1.102	-72.751	-3.845	0.957	1.886	-4.464	1.857	
	300	4.664	0.876	1.106	-73.563	-3.556	0.507	1.900	-4.484	1.857	
	400	4.828	0.803	1.110	-74.421	-3.268	0.571	1.915	-4.504	1.857	

$$B = \frac{C_{11} + 2C_{12}}{3}; \ G = (G_V + G_R) / 2;$$

$$Y = 9GB / (G + 3B)$$

$$C_{s} = \frac{C_{11} - C_{12}}{3}; A_{m} = 2C_{44} / (C_{11} - C_{12});$$

$$\sigma = (3B - 2G) / (6B + 2G)$$

where $G_V = (C_{11} - C_{12} + 3C_{44})/5$ and $G_R = 5[(C_{11} - C_{12})C_{44}] / [4C_{44} + 3(C_{11} - C_{12})]$

The calculated parameters B, G, Y, C_s , A_m and σ are presented in Table 2. It shows that B>0, $C_{44}>0$ and $C_{12} < B < C_{11}$ for these semimetals. It reveals that the chosen materials follow the Born's mechanical stability criterion^{7,32}. Therefore, these materials are mechanically stable. The ratio of fracture and toughness (B/G ratio) is found to be 1.55 and 1.53 at room temperature for YbP and YbAs, respectively. Hence, we conclude that YbP and YbAs are brittle in nature³³. The nature of these materials is found to be same as previous studied materials^{7,27} PrX and GdX. The obtained Poisson's ratio of these materials lies below the limit of σ for central force (0.25<v<0.5).

From this result, we can say that the interatomic force for the chosen semimetals is non-central³⁴. The Young's modulus is measure of stiffness of the material. The calculated values of Y for YbAs and YbP are 4.02 GPa and 3.395 GPa, respectively. Due to small value of Young modulus for YbAs, it is much stiffer than YbP. The anisotropic modulus is unity for completely isotropic material and if less than or greater than unity it shows some degree of elastic anisotropy. The anisotropic modulus of YbP is 0.584 and is 0.755 for YbAs (Table 2). Hence, both YbP and YbAs are completely anisotropic materials. The experimental values of temperature dependent SOECs and TOECs for these low carrier heavy fermion systems are not available in literature. But if we

Table 2 — Bulk modulus (B), shear modulus (G), Poisson's ratio (σ), anisotropic modulus (A_m), Young's modulus (Y), B/G ratio and tetragonal moduli (Cs) of YbAs and YbP at room temperature

Materials	<i>B</i> (GPa)	G (GPa)	σ	A_m	Y (GPa)	B/G	Cs (GPa)
YbAs	2.505	1.631	0.233	0.755	4.020	1.536	1.931
YbP	2.139	1.374	0.235	0.584	3.395	1.557	1.894

compare the results with materials^{7,27,32} like GdX, PrX, AmX we observe the similar order and trend for the elastic constants. Hence our approach to compute higher order elastic constants is quite reasonable and justified.

3.2 Physical parameters of YbX

The density of crystalline material is the function of lattice parameter³⁵. The density of semimetals decreases with an increase in temperature. So, the change in density with change in temperature is negative. Thus, it is presumed that the temperature dependence of density of these materials pursue the following equation³⁶.

$$\frac{\rho_1}{\rho_2} = 1 + \alpha' (T_2 - T_1) \qquad \dots (16)$$

where α' is rate of change of density with temperature, ρ_1 and ρ_2 are the densities at temperatures T_1 and T_2 . The temperature dependent densities of the chosen materials are calculated using lattice parameter and Eq (16). The thermal conductivity is obtained using Weideman Franz $1aw^{37,38}$. The E_0 and C_V of chosen materials are obtained using θ_D/T tables of AIP handbook³⁹, while θ_D is determined by Eq (10). The obtained temperature dependent values of ρ , K, E_0 and C_V are presented in Table 3. We have used these values to find ultrasonic velocities, thermal relaxation time,

acoustic coupling constants and ultrasonic attenuation for longitudinal and shear modes of wave propagation.

3.3 Ultrasonic velocity and thermal relaxation time

The ultrasonic velocities for longitudinal and shear modes of propagation along <100>, <110> and <111> directions are evaluated using Eqs (6-8) and are presented in Table 4.

From Table 4, it is clear that ultrasonic velocities are increasing with temperature along different crystallographic directions. The Debye average velocities and thermal relaxation time (τ) for the chosen compounds are calculated using Eqs (9) and (13) in the temperature range 100-400 K and are shown in Figs 1 and 2.

It is clear from Figs 1 and 2 that the Debye average velocity increases with the temperature, while the thermal relaxation time decreases exponentially with temperature according to expression $\tau = \tau_0 exp(-\lambda T)$. Here τ_0 and λ are constants and have different values for different materials. This type of nature was also found in previously investigated semimetals^{7,40}. The order of thermal relaxation time of the chosen materials is of the order 10⁻¹³s. This confirms the semimetallic behaviour of material.

Table 3 — Thermal conductivity (*K*), density (ρ), specific heat (*C_V*) and energy density (*E*₀) of YbAs and YbP in the temperature range 100-400 K

$T [K] \downarrow$	$\frac{K}{(\mathrm{Wm}^{-1}\mathrm{K}^{-1})}$		ρ (10 ⁻³ kg/m ³)		C _V (J/molK)		<i>E</i> ₀ (J/mol)	
	YbAs	YbP	YbAs	YbP	YbAs	YbP	YbAs	YbP
100	0.989	2.078	9.113	6.282	0.824	6.906	505.0	425.0
200	0.934	1.695	9.011	6.189	0.886	7.402	136.3	113.8
300	0.952	1.586	8.906	6.091	0.887	7.373	223.2	185.6
400	0.945	1.553	8.801	5.992	8.840	7.315	308.3	255.1

3.4 Ultrasonic attenuation

The ultrasonic attenuation coefficient over frequency square due to phonon-phonon interaction in Akhieser regime for longitudinal and shear modes of propagation $\{(\alpha f^2)_{\text{Long}} \text{ and } (\alpha f^2)_{\text{Shear}}\}$ in the temperature range 100-400 K along <100>, <110> and <111> crystallographic directions is calculated









Table 4 — Ultrasonic velocities (10³ m/s) of YbAs and YbP along different crystallographic directions in the temperature range 100-400 K

Material		Directions							
	T[K]	<	100>	<110>			<111>		
		$V_1 = V_L$	$V_2 = V_3 = V_S$	$V_1 = V_L$	$V_2 = V_{S1}$	$V_3 = V_{S2}$	$V_1 = V_L$	$V_2 = V_3 = V_S$	
	100	2.283	1.259	2.223	1.259	1.363	2.202	1.329	
VhAa	200	2.334	1.269	2.247	1.269	1.417	2.218	1.369	
IDAS	300	2.388	1.279	2.274	1.279	1.472	2.235	1.411	
	400	2.442	1.289	2.301	1.289	1.527	2.252	1.452	
YbP	100	2.634	1.322	2.455	1.322	1.630	2.393	1.534	
	200	2.697	1.334	2.487	1.334	1.695	2.412	1.584	
	300	2.767	1.347	2.522	1.347	1.763	2.435	1.637	
	400	2.839	1.361	2.559	1.361	1.833	2.460	1.690	



Fig. 4 — $(\alpha f^2)_{\text{Total}}$ versus temperature for YbP

using Eqs (11) and (12). The ultrasonic attenuation coefficient over frequency square due to thermoelastic relaxation mechanism $(\alpha f^2)_{th}$ is evaluated using Eq. (15). The total attenuation A is obtained by taking sum of all these three types of attenuation. Then,

The total attenuation $[(\alpha f^2)_{Total}]$ for YbP and YbAs is shown in Figs 3 and 4.

The curve fitting to total attenuation suggests that the total attenuation in ytterbium monopnictides can be written as a function of temperature as $A = \sum_{n=0}^{2} A_n T^n$. The value of A_n depends on elastic

constants, ultrasonic velocity, thermal relaxation time,

thermal conductivity, specific heat per unit volume and energy density. It is observed from Figs 3 and 4 that the value of total attenuation is less in case of YbAs. Hence, we can say that the impurity will be less in YbAs in comparison to YbP. Ultrasonic energy has minimum loss for wave propagating along <100> direction in these materials. Therefore, this direction is most appropriate for the study of anisotropic nature of YbAs and YbP. The order of total attenuation is same as previously investigated materials like gadolinium monopnictides²⁷, thulium monochalcogenides^{28,41}.

4 Conclusions

On the basis of results and discussion, the following conclusions can be drawn:

- 1. Adopted approach to evaluate higher order elastic constants is justified for ytterbium monopnictides.
- 2. The materials YbP and YbAs follow the Born criterion of mechanical stability, so they are stable in temperature range 100-400 K. The interatomic forces in these materials are non-central forces as the Poisson ratio lies below the range 0.25<v<0.5. These materials are completely anisotropic as the anharmonicity deviates from unity for them.
- 3. The order of thermal relaxation time $(10^{-13}s)$ and ultrasonic attenuation $(10^{-15}Nps^2m^{-1})$ confirms the semimetallic nature of YbP and YbAs.
- 4. The minimum ultrasonic attenuation along <100> direction reveals that it is the most suitable direction for the anisoptropic study in these materials.

The obtained results are useful not only in the field of industry but also in further investigation of ytterbium monopnictides.

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