Temperature Dependence of Acoustic Attenuation in Ferrous Oxide (FeO)

Sanjay H. BAGADE *
Department of Physics, Bajaj College of Science, Wardha-442001(M S), India, sanjaybagade8@gmail.com

Parvej A. SAUDAGAR
Department of Physics, Bajaj College of Science, Wardha-442001(M S), India, saudagar.pa@gmail.com

* Author to whom correspondence should be addressed

Abstract: - The present article deals with the attenuation of acoustic waves propagating through Ferrous Oxide (FeO) based on a theory developed by W.P. Mason. The Second Order Elastic Constants (SOEC) of the material have been used to find the Gruneisen constants $\langle Y_i^j \rangle$ and its square average, using which the non-linearity parameter D has been evaluated at different temperatures. The acoustic attenuation coefficient per unit frequency square due to phonon–phonon interaction for longitudinal waves $[\propto/f^2]_L$ and shear waves $[\propto/f^2]_S$, and that due to thermoelastic losses $[\propto/f^2]_{th}$ are calculated along the <100 > crystallographic axis, within the temperature range 373-673 K. The non-linearity parameters D_L and D_S which account for the anharmonicity of the material are found to be dependent on the temperature and increase with it. The study shows that the magnitude of $[\propto/f^2]$ due to phonon-phonon interaction, for both longitudinal and shear waves is of the order 10^{-16} dbs^{2}m^{-1}, while, for thermoelastic losses, it is of the order 10^{-18} dbs^{2}m^{-1}. Thus, it can be observed that phonon-phonon interaction is the prominent cause of acoustic attenuation of the propagating waves and the contribution due to thermoelastic losses is very small. The study also shows that the longitudinal waves are more attenuated as compared to shear waves, as they propagate through the solid medium and suffer more amplitude losses. This work can be useful for the characterization and study of the anharmonic behavior of crystals as well as attenuation studies in other similar oxide crystals.

Keywords: - Acoustic Attenuation, Akhiezer losses, Phonon-Phonon Interaction, Thermoelastic losses, Ferrous Oxide.

1. INTRODUCTION

Acoustic attenuation studies have been carried out in different solids like metals [1,2], semiconductors [3,4], insulators [5] and dielectrics [6,7]. Propagation of both longitudinal and shear waves and their velocities as well as relaxation time are influenced by the temperature of the medium[8,9]. Crystals that do not obey Hooke’s law of elasticity exhibit anharmonicity, and this anharmonic nature leads to the attenuation of waves traveling through it. The attenuation coefficient of acoustic waves traveling through the medium depends on the non-linearity parameters and Gruneisen parameters, which are temperature and frequency-dependent [10,11]. Different studies have established that there are various factors like phonon-phonon interactions (Akhiezer losses), crystal defects, dislocations within the crystal body, thermoelastic losses, and interaction between electrons and phonon which contribute towards decreasing the amplitude of propagating high-frequency waves in solid. But the major cause of attenuation of ultrasonic wave is the interaction between the lattice phonon within the solids, which is popularly known as ‘phonon-phonon interaction’ and the contribution due to thermoelastic losses and other factors is very negligible [12,13]. Acoustic attenuation studies are important as various useful information regarding the ionic potentials and deviation from harmonic behavior of crystalline solids can be interpreted from it [14,15]. Besides, these studies also find applications in non-destructive testing of materials [10,16].

Attenuation studies in oxide crystals are very few and are a developing field. In the present work variation of ultrasonic attenuation with temperature are investigated for Ferrous oxide (FeO), an inorganic compound that occurs in mineral form wüstite. It is insoluble in water and alkali but soluble in acids. Oxide FeO constitutes approximately up to 10 % of the Earth's mantle. It is used to produce materials like plastics, heat-absorbing glass, ceramics, rubber, paint coatings construction materials, etc. It is relevant to study ultrasonic attenuation in FeO because of its varied industrial and technological applications. Acoustic attenuation studies have been done in a few metallic oxides like Calcium oxide, Zinc oxide, Manganese oxide, etc., but to the best of the author's knowledge, acoustic attenuation studies have not been carried out in Iron oxides.

Iron oxides occur in two different forms, where iron atom exhibits Fe^{2+} and Fe^{3+} oxidation states. This study deals with acoustic attenuations in Ferrous oxide (FeO), with the Fe^{2+} oxidation state of the iron atom, and calculations are done for both shear and longitudinal waves traveling along the direction of <100> crystallographic axis. Ultrasonic attenuations due to phonon-phonon (p-p) interactions and thermoelastic losses are evaluated for temperatures from 373 K to 623 K.

2. MATERIALS AND METHODS

Mason's theory which makes use of temperature-dependent non-linearity constant D has been employed in the present theoretical study. Initially, the Second Order Elastic Constants (SOEC) of the material FeO were used to determine the velocities of longitudinal waves $\left[\boldsymbol{V_L} = \sqrt{\boldsymbol{C_{11}/d}}\,\right]$ and shear waves $\left[\boldsymbol{V_S} = \sqrt{\boldsymbol{C_{44}/d}}\,\right]$ at different temperatures, where C_{11} and C_{44} are the SOEC and $\boldsymbol{d}$ is the density of FeO crystal. The SOEC values were further used to calculate the Gruneisen constants $\langle \boldsymbol{\Upsilon_i^j} \rangle$ and its square average, using which the non-linearity parameter D has been evaluated at different temperatures. The temperature-dependent values of DL and DS were used in the calculations of attenuation coefficients per unit frequency square $\left[\propto/\boldsymbol{f^2}\right]$ due to phonon-phonon interactions for longitudinal and shear waves respectively. Similarly, the attenuation due to thermoelastic losses was also calculated.

3. RESULTS AND DISCUSSIONS

The attenuation of waves traveling through solids is temperature-dependent and the thermal conductivity of the medium affects the attenuation process. Interactions between the quantized lattice waves, which are also known as phonons, are the most important cause of energy loss for such propagating waves through the solids[17,18]. A progressing wave causes compression of some part of the medium and the region adjacent to it gets expanded. Alternate compression and expansion within the solid produce a temperature gradient which results in the flow of thermal energy. Hence energy absorption takes place in the solid and the amplitude of the progressing wave decreases, causing its attenuation[19]. Most of the attenuation studies are related to the anharmonicity of the solids and take place due to the Akhiezer mechanism where a thermal phonon drains out energy from the propagating high-frequency wave by phonon-phonon interaction. This mechanism occurs when the condition $\boldsymbol{\omega}\tau \ll 1$ is satisfied, here τ is thermal phonon relaxation time, and $\boldsymbol{\omega} = 2\pi f$, f denotes the frequency of the acoustic wave [20,21].

The attenuation of a propagating wave due to phonon-phonon interaction is given as [4]

$$\alpha = \frac{\omega^2 \tau E_0 D}{3 d v^3 (1+\omega^2\tau^2)} \tag{1}$$

The above equation in the Akhiezer region ($\boldsymbol{\omega}\tau \ll 1$) reduces to [10]

$$\left[\frac{\alpha}{f^2}\right]_L = \frac{(2\Pi)^2\, D_L E_0\, \tau}{3 d v_L^3} \tag{2}$$

$$\left[\frac{\alpha}{f^2}\right]_S = \frac{2\Pi^2 D_S E_0\, \tau}{3 d v_S^3} \tag{3}$$

Equation (2) and (3) give attenuation coefficient per unit frequency square. D is the non-linearity coupling constant. Parameters for longitudinal waves are denoted by the suffix 'L' and those for shear waves by 'S'. The density and thermal energy density of the crystal are given by d and E_0 respectively, v_L is the velocity of the longitudinal wave and v_S the velocity of the shear wave, thermal relaxation time τ is related to thermal relaxation time for shear waves (τ_S) and thermal relaxation time for longitudinal waves (τ_L) as [10]

$$\tau = \frac{\tau_L}{2} = \tau_S = \frac{3k}{C_v \bar{v}} \tag{4}$$

where $\bar{v}$, C_v, and k denote Debye's average velocity, specific heat per unit volume, and thermal conductivity respectively. $\bar{v}$ is given by [10]

$$\bar{v} = \left[\frac{1}{3\; v_L^3} + \frac{2}{3 v_S^3}\right]^{-\frac{1}{3}} \tag{5}$$

Non-linearity parameter D is obtained as [22]

$$D = 9\,\langle {\Upsilon_i^j}^2 \rangle - \frac{3\langle \Upsilon_i^j \rangle^2 C_v T}{E_0} \tag{6}$$

$\langle \Upsilon_i^j \rangle$ and $\langle {\Upsilon_i^j}^2 \rangle$ are average and square average Gruneisen numbers.

Ultrasonic attenuation due to thermoelastic losses is given as [23]

$$\left[\frac{\alpha}{f^2}\right]_{th} = \frac{2\Pi^2 k T \langle \Upsilon_i^j \rangle^2}{d v_L^5} \tag{7}$$

$\langle \Upsilon_i^j \rangle$ is zero for shear waves, hence $\left[\frac{\alpha}{f^2}\right]_{th} = 0$, which indicates that shear waves do not show attenuation due to thermoelastic losses.

The physical parameters for FeO, like the thermal energy density of crystal E_0, the density of crystal d, thermal conductivity k, and specific heat per unit volume C_v which are required for the calculation of attenuation coefficient are taken from the references [24,25]. The longitudinal and shear wave velocities and Gruneisen parameters $\langle \Upsilon_i^j \rangle$ at different temperatures are calculated from the knowledge of second-order elastic constants (SOEC) for the oxide material. The calculated velocity values show a good agreement with the experimental values of velocity reported in [26]. The thermal relaxation time τ, non-linearity parameter D_L, and D_S at different temperatures have been computed by using equations (4) and (6) and presented in Table 1. Acoustic attenuations per unit frequency square $\left[\frac{\alpha}{f^2}\right]$ due to phonon-phonon interactions, and the thermoelastic losses were obtained for different temperatures within the range 373-623 K, using equations (2), (3), and (7) and are given in Table-2.

Table 1. Data for shear wave velocity (V_S), longitudinal wave velocity (V_L), average $\langle \Upsilon_i^j \rangle$ and square average $\langle {\Upsilon_i^j}^2 \rangle$ Gruneisen numbers, non-linearity parameter D_L and D_S, thermal relaxation time τ for Ferrous Oxide (FeO) at different temperatures

Temperature K	V_S x10^3 m s^{-1}	V_L x 10^3 m s^{-1}	$\langle \Upsilon_i^j \rangle$	$\langle {\Upsilon_i^j}^2 \rangle_L$	$\langle {\Upsilon_i^j}^2 \rangle_S$	D_L	D_S	τ x10^{-12} sec
373	6 .218	6.724	0.625	8.525	2.580	8.884	5.251	9.40
423	6.110	6.718	0.580	8.782	2.675	8.965	5.434	8.86
473	5.992	6.702	0.531	8.875	2.756	9.362	5.676	8.25
523	5.668	5.990	0.486	8.942	2.792	9.680	5.820	7.46
573	5.436	5.884	0.434	9.215	2.854	9.921	6.256	6.54
623	5.224	5.640	0.442	9.364	2.904	10.575	6.860	5.56

Table 2. Acoustic attenuations $\left[\frac{\alpha}{f^2}\right]$ in FeO due to phonon-phonon interactions and thermoelastic losses[th] at different temperatures. [L= Longitudinal wave, S= Shear wave]

Temperature K	$\left[\frac{\alpha}{f^2}\right]_L$ x 10^{-16} dbs^{2}m^{-1}	$\left[\frac{\alpha}{f^2}\right]_S$ x 10^{-16} dbs^{2}m^{-1}	$\left[\frac{\alpha}{f^2}\right]_{th}$ x 10^{-18} dbs^{2}m^{-1}
373	3.427	1.278	0.653
423	3.610	1.640	0.716
473	4.884	2.072	0.735
523	7.815	2.775	1.268
573	9.084	3.632	1.272
623	11.665	4.761	1.678

The theoretically computed values of non-linearity parameters D_L and D_S are reported in Table 1 and shown graphically in Figure 1. It is observed that the value of non-linearity parameters D_L and D_S increases with the temperature which can be attributed to the decrease in the value of the average Gruneisen parameter $\langle \Upsilon_i^j \rangle$ with increasing temperature, as mentioned in the references [27]. The increase in the values of D_L and D_S are about 19.0 % and 30.4 % respectively, within the temperature range. The ratio of D_L to D_S for FeO lies between 1 to 2, which is similar to that observed for other metals and metal oxides[28–30]. Thermal relaxation time τ decreases with temperature, but it attains saturation value at higher temperatures. The Acoustic attenuations $\left[\frac{\alpha}{f^2}\right]$ due to phonon-phonon interactions and thermoelastic losses calculated for FeO oxide crystal at different temperatures within the range 373-623 K for longitudinal as well as shear waves are given in Table 2. Variation of acoustic attenuation due to phonon-phonon interactions and thermoelastic losses for different temperatures is plotted in Figures 2-4. $\left[\frac{\alpha}{f^2}\right]$ is found to be temperature-dependent and increases with the temperature. It is observed that the magnitude of $[\propto/f^2]$ due to phonon-phonon interaction, for both longitudinal and shear waves is of the order 10^{-16} dbs^{2}m^{-1}, while, its value due to thermoelastic losses is of the order 10^{-18} dbs^{2}m^{-1}. The attenuation of the propagating waves due to phonon-phonon interaction is greater than that due to thermoelastic losses by a factor of about 10^2. Such results have been reported in previous studies for the attenuation of a few metals[2], rare-earth chalcogenides[28], semiconductors[31], and metal oxides[29].

The graphs between acoustic attenuation per unit frequency square and temperature for both longitudinal and shear waves show that the variation is a non-linear curve. Acoustic attenuation due to thermoelastic losses also exhibits the same nature of the graph. Figures 2-4 show that at lower temperatures the magnitude of attenuation of the propagating wave is small, but, as the temperature increases the attenuation of the acoustic waves also increases. It is observed from Figure 2 and Figure 3 that the magnitude of attenuation is less for shear waves as compared to that of longitudinal waves.

The attenuation coefficient has the least value in case of attenuation due to thermoelastic losses as seen in Figure 4. Two mechanisms are mainly responsible for the attenuation of acoustic waves traveling through solids. The first mechanism is phonon-phonon interactions. The second mechanism is thermoelastic losses which arise due to heat energy exchange between the adjacent regions of compression and expansion produced alternately in the solid medium, as the acoustic or ultrasonic wave propagates through it. The calculations show that the magnitude of $[\propto/f^2]$ due to phonon-phonon interaction, for both longitudinal and shear waves is of the order 10^{-16} dbs^2m^{-1}, while, for thermoelastic losses, it is of the order 10^{-18} dbs^2m^{-1}. Thus, it is seen that the contribution of thermoelastic losses to attenuation is nearly one hundred times less as compared to attenuation due to phonon-phonon interactions [32,33]. Hence, we can conclude that phonon-phonon interactions are largely responsible for high-frequency acoustic attenuations in the Ferrous oxide crystal.

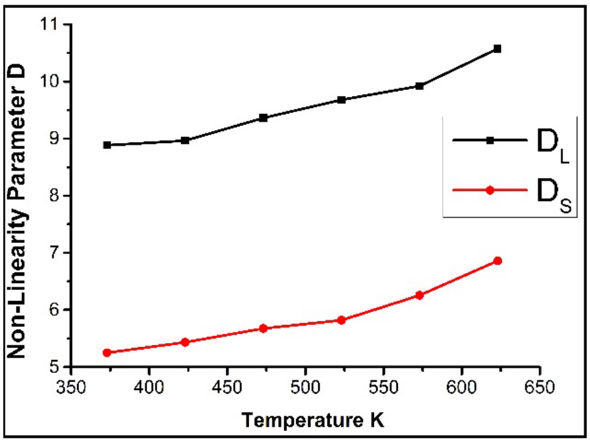

Figure 1. Variation of Non-Linearity Parameter D with Temperature.

The magnitude of the attenuation coefficient $[\propto/f^2]$ obtained for FeO crystal agrees with previously reported attenuation studies in other metal oxides like ZnO [34], MgO [29] and CaO [35], where the attenuation coefficient per unit frequency square reported was of the order of 10^{-16} dbs^2m^{-1} for longitudinal and shear waves. It is evident from Figure 2 and Figure 3 that the Longitudinal waves suffer more energy loss and hence are more attenuated as compared to shear waves as they propagate through the FeO crystal.

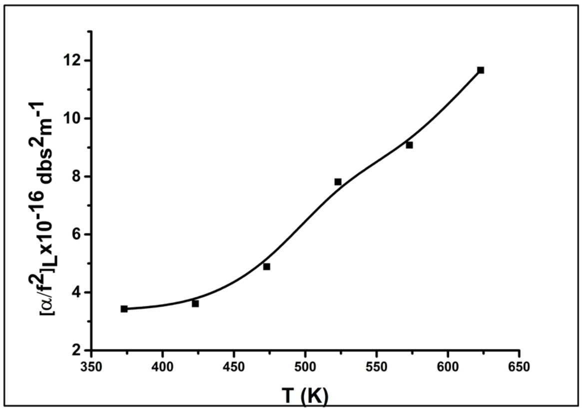

Figure 2. Attenuation of longitudinal waves propagating along (100) axis. [phonon-phonon interaction]

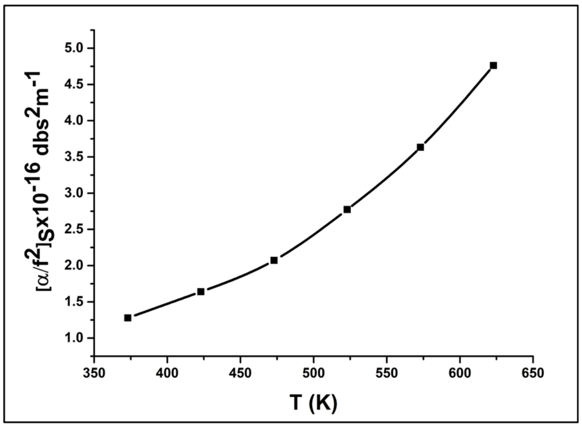

Figure 3. Attenuation of shear waves propagating along (100) axis. [phonon-phonon interaction]

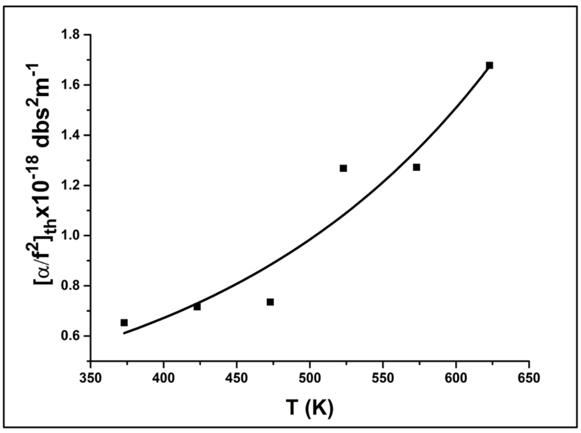

Figure 4. Attenuation of Acoustic waves due to thermoelastic losses for waves propagating along the (100) axis.

4. CONCLUSION

The behavior of acoustic attenuation in Ferrous Oxide, for both Longitudinal and shear waves is investigated within the temperature range 373-673 K, based on Mason's theory. It can be concluded that acoustic attenuation in Ferrous Oxide is temperature-dependent, and it increases non-linearly with the temperature. The increase in the values of attenuation coefficient per unit frequency square can be attributed to the increase in values of temperature-dependent non-linearity parameter D. As the non-linearity parameter takes into account the anharmonic behavior of the material, it can be concluded that attenuation of acoustic waves is mainly due to anharmonicity, which arises due to deviation from perfect elastic behavior of the crystal.

A major cause of acoustic wave attenuation is phonon-phonon interaction and contribution due to thermoelastic losses to the wave attenuation is negligible. Longitudinal waves are more attenuated than shear waves as they propagate through the solid. Work presented here may find application for wave attenuation studies in metals, insulators, semiconductors, dielectrics, and metal oxides of similar nature. Results obtained can be useful for various industrial and technological purposes, particularly in the transmission and absorption of acoustic waves and where paint coatings, surface polishing, and acoustic insulations are employed. Future work may include attenuation studies in other Iron Oxides and similar metal oxides useful for various industrial and scientific applications like surface coatings, thermal and electrical insulations, and thin film applications.

CONFLICT OF INTEREST

The author declares that no conflict of interest is associated with the present work.

ACKNOWLEDGEMENT

The author is sincerely grateful to the unknown referees for their valuable suggestions for improving the quality of the research paper.

REFERENCES

[1] Mason W. P. and Rosenberg A, Phonon and Electron Drag Coefficients in Single-Crystal Aluminum, *Physical Review*, Vol. 151, 1996, 434–41.

[2] A K Upadhyay, Study of Attenuation Coefficient of Cu, Ag & Au, *International Journal of Composite Material and Matrices,* Vol. 17, No. 1, 2021, 1-7.

[3] Sakurai H. and Suzuki K., Ultrasonic Attenuation in n-Type Ge. II: Effects of Uniaxial Stress, *J Physical Soc. Japan,* Vol.52, 1983, 4199–205.

[4] Bagade S H., High frequency acoustic attenuation in pure semiconductors: A comparative study, *AIP Conference. Proceedings, National Conference on Physics and Chemistry of Materials, Indore,* 2021, 020097.

[5] Woodruff T. O. and Ehrenreich H., Absorption of Sound in Insulators, *Physical Review,* Vol. 123, 1961, 1553–1559.

[6] Ožvold M., On Acoustic Attenuation in Dielectric Solids, *physica status solidi (b),* Vol. 37. 1970, 79–90.

[7] Raju K M, Srivastava R K, Arun Kumar and Suneeta Devi, Acoustical investigation of lead chalcogenides, Physica B Condense Matter, Vol.405, 2010, 4855–4867.

[8] Kurashkin K V., Gonchar A V., Klyushnikov V A and Mishakin V. V., Effect of Microstructure on Temperature Dependence of Ultrasonic Velocity in Aluminum, Acoustical Physics, Vol. 69, 2023, 335–340

[9] Koltsova I S and Khomutova A S., Acousto- electronic Method for Measuring the Velocity of Ultrasonic Waves in Liquids Acoustical Physics, Vol. 69, 2023, 330–334.

[10] Jyoti B, Singh S P, Gupta M, Tripathi S, Verma A K, Singh D and Yadav R R., Ultrasonic and Thermophysical Properties of Cobalt Nanowires, Acoustical Physics, Vol. 67, 2021, 584–589.

[11] Amara S and Kanouni F., Investigation of Frequency Response of Yttrium Doped AIN-Based Saw Devices Acoustical Physics, Vol. 69, 2023, 48–52.

[12] Tawfeeq W. Mohammed, The Combined Effects of Thermal and Acoustic Parameters on the Sound Absorption of Aluminum Foam Panels, Romanian Journal of Acoustics and Vibration, Vol. 20, 2023, 78–84.

[13] Upamanyu A., Dhiman M., Gupta D., and Kumar M., Frequency Dependence Studies of Acoustical and Thermo-Dynamical Parameters of The Binary Mixtures of Isopropyl Sulphide and Acetic Acid, Romanian Journal of Acoustics and Vibration, Vol. 17, 2020,128–134.

[14] Gupta M, Dhawan P K, Verma S K and Yadav R., Diameter Dependent Ultrasonic Characterization of InAs Semiconductor Nanowires, Open Journal of Acoustics, Vol. 05, 2015, 218–225.

[15] Gokhale V. J. and Rais-Zadeh M., Phonon-Electron Interactions in Piezoelectric Semiconductor Bulk Acoustic Wave Resonators Scientific Reports, Vol. 4, 2015, 5617.

[16] Kumar S and Lee H P., Recent Advances in Acoustic Metamaterials for Simultaneous Sound Attenuation and Air Ventilation Performances, Crystals (Basel), Vol. 10, 2020, 686.

[17] Rajagopalan S and Joharapurkar D. N., Temperature dependence of the nonlinearity constant and ultrasonic attenuation in pure silicon and germanium, J. Appl. Phys., Vol. 54, 1983, 3166–3171.

[18] Joharapurkar D N., Study of acoustic and elastic behavior of some solids, J. Acoust. Soc. Am., Vol. 81, 1987, 1990–2001.

[19] Verma A K., Yadav N., Singh S P., Dey K., Singh D. and Yadav R., Study of Ultrasonic Attenuation and Thermal Conduction in Bimetallic Gold/Platinum Nanofluids, Johnson Matthey Technology Review, Vol. 65, 2021, 556–567.

[20] Bhalla V., Singh D., Jain S K. and Kumar R., Ultrasonic attenuation in rare-earth monoarsenides, Pramana, Vol. 86, 2016, 1355–1367.

[21] Mason W P and Bateman T B., Relation between Third-Order Elastic Moduli and the Thermal Attenuation of Ultrasonic Waves in Nonconducting and Metallic Crystals, J. Acoust. Soc. Am. Vol. 40, 1966, 852–862.

[22] Rajagopalan S., Joharapurkar D. N. and Shende P. R., Ultrasonic attenuation and nonlinearity constant in silicon in the temperature range 73–293 K, J. Appl. Phys. Vol. 55, 1984, 275–287.

[23] Kor S. K., Mishra P. K. and Tandon U. S., Ultrasonic attenuation in aluminum, Solid State Communications, Vol. 15, 1974, 499–501.
[24] Dwight E Gray, American Institute of Physics Handbook (New York: McGraw Hill), 1963.
[25] Lide D. R., CRC Handbook of Chemistry and Physics (New York: CRC Press) 1998.
[26] Anastasia K., Elasticity measurements at extreme conditions: Application to FeO and FeNi.(Ph. D Thesis) 2008. urn: nbn: de: bvb:703-opus-3929
[27] Joharapurkar D., Gerlich D. and Breazeale M. A., Temperature dependence of elastic nonlinearities in single-crystal gallium arsenide, J. Appl. Phys. Vol. 72, 1992, 2202–2208.
[28] Singh D., Behaviour of acoustic attenuation in rare earth chalcogenides, Mater. Chem. Phys., Vol. 115, 2009, 65–80.
[29] Lewis M. F. and Patterson E., Microwave Phonon Attenuation in MgO, J. Appl. Phys., Vol. 39, 1968, 2469–75.
[30] Winterhager P. and Lücke K., Quantitative investigation of ultrasonic attenuation in copper single crystals, J. Appl. Phys., Vol. 44, 1973, 4855–4862.
[31] Rajagopalan S. and Joharapurkar D. N., Temperature variation of acoustic attenuation and nonlinearity constant in Ge and GaAs, J. Appl. Phys. Col. 56,1984, 1333–1337.
[32] Joharapurkar D. N., Rajagopalan S. and Basu B. K., Ultrasonic velocity, attenuation, and nonlinearity constant in pure and Cd-doped KCl, Physical Review B, Vol. 37, 1988, 3101–3104.
[33] Joharapurkar D, N., Temperature variation of silicon's nonlinearity constant and acoustic attenuation, J. Appl. Phys., Vol. 64, 1988, 1726–1729.
[34] Claiborne L T., Hemphill R. B. and Einspruch N. G., Ultrasonic Attenuation in ZnO, J. Acoust. Soc. Am. Vol. 45, 1969, 1352–55.
[35] Kumar J., Kailash K., Shrivastava S., Singh D. and Virendra K., Ultrasonic Attenuation in Calcium Oxide, Advances in Materials Physics and Ch*emistry,* Vol. 1, 2011, 44-49.