

Acoustical investigations of uranium chalcogenides

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Ultrasonic attenuation due to phonon–phonon interaction and thermoelastic loss was evaluated in uranium chalcogenides viz. UX, X= S, Se, Te in fcc phase in the temperature range 50–600 K for longitudinal and shear waves along the $\langle 100 \rangle$, $\langle 110 \rangle$ and $\langle 111 \rangle$ directions of propagation. Electrostatic and Born–Mayer repulsive potentials were used to obtain second and third order elastic constants, taking the nearest neighbour distance and hardness parameter as the input data. Second and third order elastic constants (obtained at various temperatures) were used to obtain the Gruneisen parameters and non-linearity or anisotropy parameters, which in turn were used to evaluate the ultrasonic attenuation coefficient over the frequency square due to phonon–phonon interaction, $(\alpha f^2)_{P-P}$ in the Akhiezer regime. It has been found that at lower temperatures αf^2 increases rapidly with temperature, and at higher temperatures the rate of increase becomes small. Contribution to the total attenuation due to thermoelastic loss is negligible in comparison with that of phonon–phonon interaction, i.e. a major part of the energy from the sound wave is removed, due to interaction of acoustic phonons with thermal phonons (lattice vibrations).

Key words: *elastic constants; ultrasonic attenuation; nonlinearity coupling constants*

1. Introduction

Uranium monochalcogenides with uranium in a trivalent state under atmospheric pressure exhibit metallic conductivities. Uranium compounds, besides their important use in reactor technology, possess other interesting and also controversial properties [1–5]. Hence, they have attracted the attention of both experimental and theoretical solid state physicists. These chalcogenides (UX, X = S, Se, Te) show ferromagnetic ordering [6–9].

Ultrasonic attenuation studies have been made in different types of solids viz. dielectric, mixed dielectric, semiconducting and metallic substances at room temperature as well as at higher temperatures. There may be different reasons for the attenuation of ultrasonic waves propagating through solids (crystalline or amorphous). These

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important factors for sound attenuation are electron–phonon interaction, phonon–phonon interaction, thermoelastic loss and loss due to screw and edge dislocations. Phonon–phonon interaction is the principal reason of ultrasonic attenuation at higher temperatures viz. 100 K and above, in all types of solids, i.e. metallic, semiconducting and dielectric. In solids, a small contribution (2–5%) of the total attenuation coefficient occurs due to thermoelastic loss, i.e. due to heat transfer from compressed to rarefied parts of the materials due to strain caused by sound waves [10–12].

Sound attenuation studies offer the possibility to detect and characterize microstructural properties as well as flaws in materials, controlling material behaviour, based on physical mechanisms, to predict the future performance of the material. Structural inhomogeneities, elastic parameters, non-linearity parameters are strongly connected with frequency or temperature dependences of ultrasonic attenuation mechanisms [12–15].

The present paper deals with temperature variation of ultrasonic attenuation due to p–p interaction in the 50–600 K range, and along the $\langle 100 \rangle$, $\langle 110 \rangle$ and $\langle 111 \rangle$ crystallographic directions of propagation for longitudinal and shear waves and for shear waves polarized along various directions.

2. Theory

2.1. Theory of second and third order elastic constants

Second and third order elastic constants (SOEC and TOEC), C_{ij}^0 and C_{ijk}^0 at 0 K have been obtained using the electrostatic and Born–Mayer potentials and following Brugger’s [16] definition of elastic constants. According to Brugger’s definition, the n th order elastic constant is defined as

$$C_{ijklmn\dots} = \left(\frac{\partial u}{\partial \varepsilon_{ij} \partial \varepsilon_{kl} \partial \varepsilon_{mn} \dots} \right) \quad (1)$$

where u is the crystal free energy density and ε_{ij} is the strain tensor. In Voigt notation C_{IJK} replaces $C_{ijklmn\dots}$ in which $ij \rightarrow I\dots$, etc. For cubic crystals, (uranium chalcogenides have fcc structure) three independent SOEC (C_{11} , C_{12} , C_{44}) and six independent TOEC (C_{111} , C_{112} , C_{144} , C_{166} , C_{456} , C_{123}) occur. Using the theory discussed in [17], SOEC and TOEC at 0 K viz. C_{ij}^0 , C_{ijk}^0 obtained are given as

$$C_{11}^0 = -1.56933 \frac{z^2 e^2}{r_0^4} + \frac{1}{qr_0} \left(\frac{1}{r_0} + \frac{1}{q} \right) Q(r_1) + \frac{2}{qr_0} \left(\frac{1}{\sqrt{2}r_0} + \frac{1}{q} \right) Q(r_2)$$

$$\begin{aligned}
C_{12}^0 &= C_{44}^0 = 0.344778 \frac{z^2 e^2}{r_0^4} + \frac{1}{qr_0} \left(\frac{1}{\sqrt{2}r_0} + \frac{1}{q} \right) Q(r_2) \\
C_{111}^0 &= 10.26390 \frac{z^2 e^2}{r_0^4} - \left(\frac{3}{qr_0^2} + \frac{3}{q^2 r_0} + \frac{1}{q^3} \right) Q(r_1) - \left(\frac{3}{\sqrt{2}r_0^2 q} + \frac{3}{q^2 r_0} + \frac{\sqrt{2}}{q^3} \right) Q(r_2) \\
C_{112}^0 &= C_{166}^0 = 1.209625 \frac{z^2 e^2}{r_0^4} + \left(\frac{3}{2\sqrt{2}r_0^2 q} + \frac{3}{q^2 r_0} + \frac{\sqrt{2}}{q^3} \right) Q(r_2) \\
C_{123}^0 &= C_{144}^0 = C_{456}^0 = 1.20862 \frac{z^2 e^2}{r_0^4} - \left(\frac{3}{2\sqrt{2}r_0^2 q} + \frac{3}{2q^2 r_0} + \frac{1}{\sqrt{2}q^3} \right) Q(r_2) \\
Q(r_1) &= A \exp \left(-\frac{r_0}{q} \right), \quad Q(r_2) = A \exp \left(-\frac{r_0 \sqrt{2}}{q} \right)
\end{aligned}$$

Where z , A , r , q are the valence constant, pre-exponential constant, nearest neighbour distance and the hardness parameter, respectively

According to lattice dynamics developed by Ludwig et al. [18, 19], the temperature dependences of SOEC and TOEC can be obtained by adding a vibrational contribution to the elastic constants, SOEC and TOEC at any temperature are given by,

$$C_{ij}(T) = C_{ij}^0 + C_{ij}^{\text{vib}} \quad (2)$$

and

$$C_{ijk}(T) = C_{ijk}^0 + C_{ijk}^{\text{vib}} \quad (3)$$

where C_{ij}^{vib} and C_{ijk}^{vib} are vibrational contributions to elastic constants, where

$$C_{11}^{\text{vib}} = f^{1,1} G_1^2 + f^2 G_2, \quad C_{12}^{\text{vib}} = f^{1,1} G_1^2 + f^2 G_{1,1}, \quad C_{44}^{\text{vib}} = f^2 G_{1,1}$$

$$C_{111}^{\text{vib}} = f^{1,1,1} G_1^3 + 3f^{2,1} G_2 G_1 + f^3 G_3$$

$$C_{112}^{\text{vib}} = f^{1,1,1} G_1^3 + f^{2,1} (2G_{1,1} + G_2) G_1 + f^3 G_{2,1}$$

$$C_{123}^{\text{vib}} = f^{1,1,1} G_1^3 + 3f^{2,1} G_1 G_{1,1} + f^3 G_{1,1,1}$$

$$C_{144}^{\text{vib}} = f^{2,1} G_1 G_{1,1} + f^3 G_{1,1,1}$$

$$C_{456}^{\text{vib}} = f^3 G_{1,1}, \quad C_{166}^{\text{vib}} = f^{2,1} G_1 G_{1,1} + f^3 G_{2,1}$$

where

$$\begin{aligned}
 G_3 &= 2Q(r_1)(30 + 30q_0 + 9q_0^2 - q_0^3 - q_0^4)H + 2G_{2,1} \\
 G_{2,1} &= Q(r_2) \left(15\sqrt{2} + 15q_0 + \frac{9}{\sqrt{2}q_0^2} - q_0^3 - \sqrt{2}q_0^4 \right) H \\
 G_2 &= 2Q(r_0) (-6 - 6q_0 - q_0^2 + q_0^3)H + 2G_{1,1} \\
 G_{1,1} &= Q(r_2) (-3\sqrt{2} - 6\sqrt{2}q_0 - \sqrt{2}q_0^2 + 2q_0^3)H \\
 G_1 &= 2Q(r_0) (2 + 2q_0 - q_0^2) + Q(r_2) (\sqrt{2} + 2q_0 - \sqrt{2}q_0^2)H
 \end{aligned}$$

where

$$\begin{aligned}
 Q(r_1) &= A \exp\left(-\frac{r_0}{q}\right), \quad Q(r_2) = A \exp\left(-\frac{r_0\sqrt{2}}{q}\right) \\
 f^2 &= f^3 = \left(\frac{\hbar w_0}{8r_0^3}\right) \coth X \\
 f^{1,1} &= f^{2,1} = -\left(\frac{1}{2r_0^3}\right) \left(\frac{\hbar w_0}{48}\right) \left(\frac{X}{\sinh^2 X}\right) + \coth X \\
 f^{1,1,1} &= \left(\frac{\hbar w_0}{384r_0^3}\right) \left[\left(\frac{\hbar w_0}{kT}\right)^2 \left(\frac{\operatorname{ctgh} X}{6\sinh^2 X}\right) + \frac{X}{\sinh^2 X} + \coth X\right] \\
 X &= \left(\frac{\hbar w_0}{2kT}\right) \\
 H &= \left[\left(\frac{r_0}{q-2}\right)Q(r_1) + 2\left(\frac{r_0}{q} - \sqrt{2}\right)Q(r_2)\right]^{-1}
 \end{aligned}$$

where ω_0 is the angular frequency of the acoustic wave.

2.2. Theory of sound attenuation

In the Akhiezer regime ($\omega\tau_{th} \ll 1$), a sound wave passing through a solid can be attenuated by two processes [20]. First, if the wave is longitudinal, periodic contractions and dilations in the solid induce a temperature wave via thermal expansion. En-

ergy is dissipated by heat conduction between regions of different temperatures. This is called thermoelastic loss. Secondly, dissipation occurs as the gas of thermal phonons tries to reach an equilibrium characterized by a local (sound wave induced) strain. This is the internal friction mechanism.

The physical basis for obtaining the attenuation coefficient is that the elastic constants contributed by the thermal phonons relax [21]. The phonon contribution to the unrelaxed elastic constants is evaluated by taking into consideration the change in energy of the thermal phonons due to applied instantaneous strain. The frequency of each mode ν_i is changed by

$$\frac{\partial \nu_i}{\nu_i} = -\gamma_i^j S_j$$

where γ_i^j is the generalised Gruneisen parameter and S_j is the instantaneous strain. It is assumed that all phonons of a given direction of propagation and polarization have equal change in frequency. Then phonons of the i th branch and the j th mode undergo a change in temperature T

$$\frac{\Delta T_i}{T_0} = -\gamma_i^j S_j$$

A relaxed elastic constant is obtained after there is phonon-phonon coupling among various branches and ΔT_i relax to a common temperature change ΔT , given by

$$\frac{\Delta T}{T} = -\langle \gamma_i^j \rangle S_j$$

where $\langle \gamma_i^j \rangle$ is the average value of γ_i^j . Thermal relaxation time [22, 23] is given by the following equation

$$\tau = \tau_s = \frac{\tau_l}{2} = \frac{3K}{C_v \langle V \rangle^2} \quad (4)$$

where K is the thermal conductivity, C_v is the specific heat per unit volume and $\langle V \rangle$ is the Debye average velocity.

According to Mason and Batemann, SOEC and TOEC (which are measure of anharmonicity of the crystal) are related by the Gruneisen parameter γ_i^j and hence by the nonlinearity parameter D . Ultrasonic attenuation due to phonon-phonon interaction in the Akhiezer regime ($\omega\tau \ll 1$) is given by [22, 23]

$$\left(\frac{\alpha}{f^2} \right)_{p-p} = \frac{2\pi^2 D E_0 \tau}{3\rho V^3} \quad (5)$$

where the non-linearity coupling constant D is given by

$$D = 9 \left\langle (\gamma_i^j)^2 \right\rangle - \frac{3 \langle \gamma_i^j \rangle^2 C_v T}{E_0} \quad (6)$$

Here $\left\langle (\gamma_i^j)^2 \right\rangle$ and $\langle \gamma_i^j \rangle^2$ are the square average and the average square Gruneisen parameters, V is the sound wave velocity for longitudinal waves (V_L) and for shear waves (V_S), E_0 is the lattice energy density, and ρ is the density.

The Debye average velocity is given by [22, 23]

$$\frac{3}{\langle V \rangle^3} = \frac{1}{V_L^3} + \frac{2}{V_S^3} \quad (7)$$

Ultrasonic attenuation due to thermoelastic loss is given by [22, 23]

$$\left(\frac{\alpha}{f^2} \right)_{th} = \frac{4\pi^2 \langle \gamma_i^j \rangle^2 kT}{2\rho V_L^5} \quad (8)$$

This type of attenuation occurs for the longitudinal wave only because in shear wave propagation, the volume remains unaltered and there is no heating effect, which makes this contribution nil.

Dislocation damping due to screw and edge dislocations also produces appreciable loss, due to phonon-phonon interaction. Dislocation damping is measured in terms of viscous drag coefficients. Viscous drag coefficients due to screw and edge dislocations are given by [13]

$$B_{\text{screw}} = 0.071\eta \quad (9a)$$

$$B_{\text{edge}} = \frac{0.053\eta}{(1-\sigma^2)} + \frac{0.0079}{(1-\sigma^2)} \frac{\mu}{\phi} \theta \quad (9b)$$

where

$$\begin{aligned} \theta &= \varepsilon_L - \frac{4}{3}\varepsilon_S, & \varepsilon_L &= \frac{E_0 \Sigma_L \tau_L}{3}, & \varepsilon_S &= \frac{E_0 \Sigma_S \tau_S}{3} \\ \phi &= \frac{C_{11} + 2C_{12}}{3}, & \mu &= \frac{C_{11} - C_{12} + C_{44}}{3}, & \sigma &= \frac{C_{12}}{C_{11} + C_{12}} \end{aligned}$$

here ϕ , μ , ε , σ , θ are the bulk modulus, shear modulus, phonon viscosity, Poisson's ratio and compressional viscosity, respectively.

3. Results and discussion

The second and third order elastic constants (SOEC and TOEC) at various temperatures have been calculated by taking two basic parameters, the nearest neighbour distance using lattice constants, $r = (a / \sqrt{2})$ [24] and the hardness parameter, q (equal to 0.278 Å, 0.395 Å, 0.198 Å for US, USe and UTe, respectively). The calculated values of the second and third order elastic constants are given in Tables 1 and 2.

Table 1. C_{ij} (10^{10} N/m²) at 300 K for uranium chalcogenides

Compound	C_{11}	C_{12}	C_{44}
US	11.84	1.25	1.47
	11.9 [24]	3.9 [24]	5.5 [24]
	30.5 ± 1.5 [27]	–	1.72 ± 0.05 [27]
	24.5 ± 1.4 [25]	0.4 ± 0.7 [25]	2.1 ± 0.1 [25]
	30.17 ± 0.39 [26]	1.32 ± 0.73 [5]	1.69 ± 0.019 [2]
USe	5.96	1.24	1.35
	6.0 [24]	0.4 [24]	2.2 [24]
	19.4 ± 1.4 [4]	0.0 ± 0.7 [4]	1.6 ± 0.1 [4]
UTe	14.26	0.51	0.92
	0.20 [24]	0.002 [24]	1.90 [24]
	14.9 ± 0.3 [4]	-2.0 ± 0.4 [4]	1.13 ± 0.04 [4]
	14.34 ± 0.36 [26]	–	1.20 ± 0.02 [26]

Table 2. C_{ijk} (10^{10} N/m²) at 300 K for uranium chalcogenides

Compound	C_{111}	C_{112}	C_{123}	C_{144}	C_{166}	C_{456}
US	–245.33	–4.26	0.71	2.77	–5.52	2.76
USe	–102.03	–4.97	1.66	2.34	–5.42	2.33
UTe	–400.29	–0.12	–4.06	1.78	–3.21	1.78
LiF	–6.75	–2.60	1.01	1.01	–2.80	0.96
Ref. [17]	–5.97	–2.61	0.91	0.97	–2.64	0.88

The SOEC values were compared with experimental values obtained by other authors who performed the neutron scattering experiment [4, 25], ultrasonic experiment [26, 27] and theoretical approaches [24]. There is agreement between the evaluated and the experimental values of the second order elastic constants. However, the value of C_{11} for US agrees with the theoretical value but differs appreciably from the experimental one. There is also disagreement between the theoretical and experimental values of SOEM for some of the compounds studied, but this may be because we have derived our expressions assuming that the overlap repulsion is significant only up to the level of next nearest neighbours. Furthermore, other approaches for obtaining elastic moduli can be used at one temperature only, while our approach is simple and can

be used for obtaining the temperature dependence of the elastic constants over a wide temperature range. Our approach is very simple, involves only two basic parameters; the nearest neighbour distance (r) and the hardness parameter (q). The TOEM values have not been compared due to the unavailability of data in the literature. But, in order to justify our approach for the calculation of TOEM values, we have evaluated third order elastic constants for fcc structured LiF [17]. The values which were obtained by adopting present approach are in close agreement with the available values (Table 2).

The thermal energy density, E_0 , and the specific heat per unit volume, C_v , are calculated in function of the Debye temperature, θ_D (138.7 K, 138.7 K and 114.8 K for US, USe and UTe, respectively [28]) using a table [29, 30] of physical constants: this was the primary input data used to obtain the average square, square average Gruneisen parameters, non-linearity coupling constants (D_l and D_s) and ultrasonic attenuation along different crystallographic directions. The evaluated values of the nonlinearity coupling constants D_l and D_s for US, USe and UTe at 300 K are given in Table 3. It is clear from Table 3 that the values of D_l and D_s increase with temperature up to 400 K and then decrease at higher temperatures (i.e., anharmonicity of the crystal first increases and then decreases, since the nonlinearity coupling constant is the measure of anharmonicity of the crystal).

Table 3. Acoustic coupling constants (D_l and D_s) for longitudinal and shear waves along various directions of propagation for uranium chalcogenides at 300 K

Compound	<100>		<110>			<111>		
	D_l	D_s	D_l	D_s^*	D_s^{**}	D_l	D_s^+	D_s^{++}
US	64.32	0.782	55.83	2.32	146.32	1667.54	75.08	282.35
USE	37.73	1.294	33.86	8.14	85.37	406.85	43.61	72.22
UTE	153.98	0.779	137.51	6.34	356.52	8699.84	184.21	2036.79

* – polarization along [100], ** – polarization along [001], + polarization along [110], ++ polarization along $[\bar{1}10]$.

Temperature variation of ultrasonic attenuation due to phonon–phonon interaction for longitudinal and shear waves viz. $(\alpha f^2)_l$ and $(\alpha f^2)_s$ along different crystallographic directions of propagation and polarization are shown in Figs. 1, 2. It can be seen from the figures that the temperature variation of $(\alpha f^2)_l$ has a similar profile along all the directions of propagation for longitudinal and shear waves. $(\alpha f^2)_l$ and $(\alpha f^2)_s$ increase with temperature and the rate of their increase for longitudinal waves is higher compared with shear waves. This might be due to the higher rate of interaction of acoustical and thermal phonons. The values of $(\alpha f^2)_l$ and $(\alpha f^2)_s$ are larger along the <111> direction of propagation in comparison with the <100> and <110> direction of propagation. This may be attributed to larger anharmonicity along the <111> direction of propagation. Furthermore, as the molecular weight increases in a given chalcogenide series, $(\alpha f^2)_{th}$ is found to increase along a given direction of propagation at a particular temperature for longitudinal (or shear) waves.

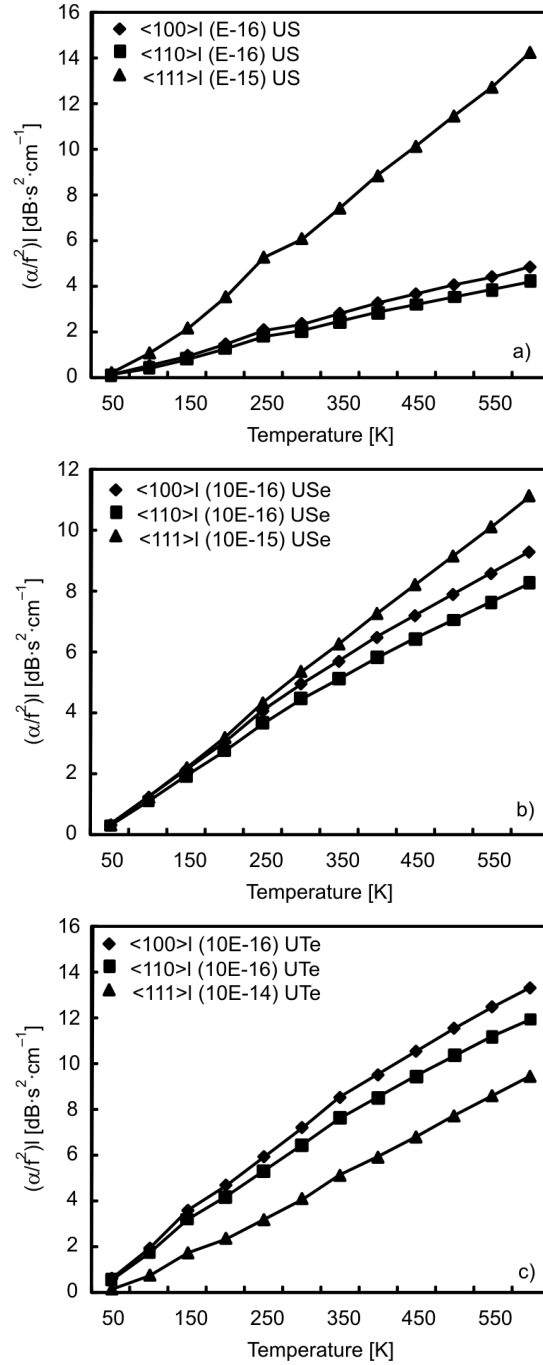


Fig. 1. Temperature dependence of ultrasonic attenuation due to phonon–phonon interaction for a longitudinal wave along various directions of propagation for: a) US, b) USe, c) UTe

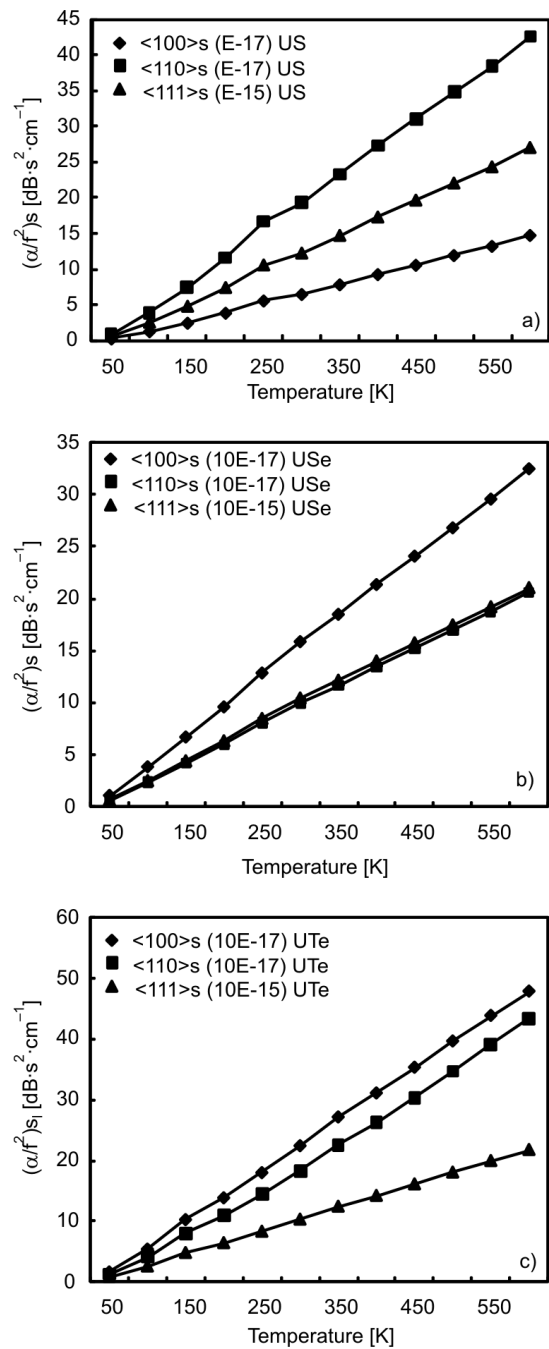


Fig. 2. Temperature dependence of the ultrasonic attenuation due to phonon–phonon interaction for a shear wave along various directions of propagation for: a) US, b) USe, c) UTe

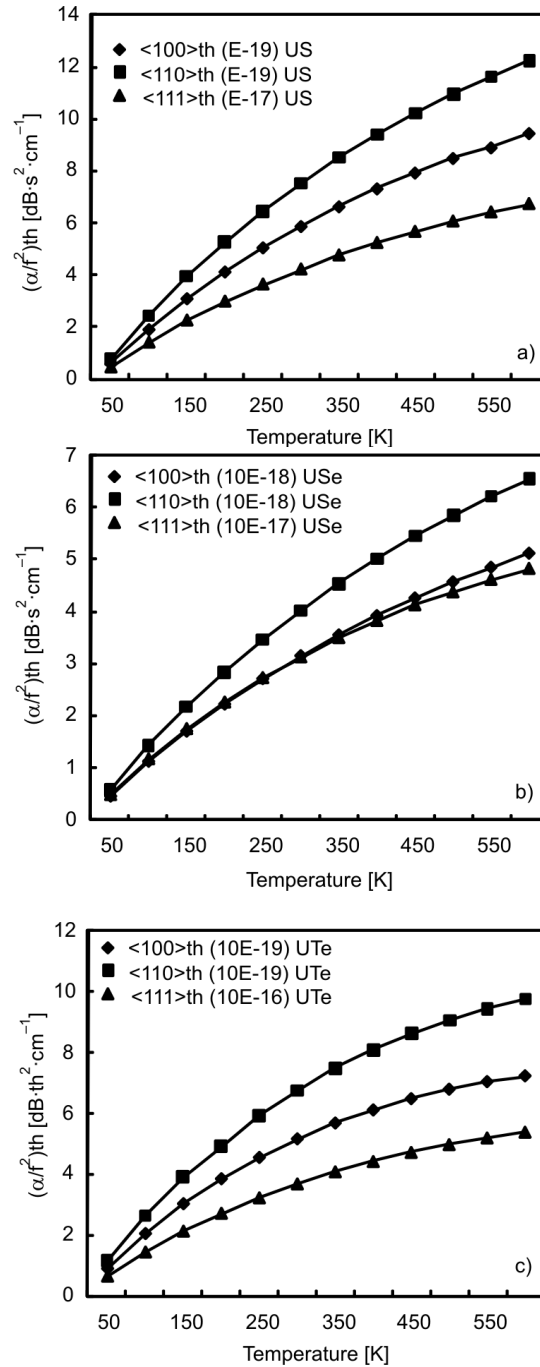


Fig. 3. Temperature dependence of the ultrasonic attenuation due to thermoelastic loss for longitudinal wave along various directions of propagation for: a) US, b) USe, c) UTe

Temperature dependences of ultrasonic attenuation due to thermoelastic loss viz. $(\alpha f)_{th}$ along various directions are shown in Fig. 3. It can be seen that $(\alpha f)_{th}$ also increases with temperature. It results from Figs. 1–3 it that ultrasonic attenuation over the frequency square due to thermoelastic loss $(\alpha f)_{th}$ is of the order of 10^{-17} – 10^{-19} dB·s²·cm⁻¹ and is negligible in comparison with attenuation due to phonon–phonon interaction, which is of the order of 10^{-15} – 10^{-17} dB·s²·cm⁻¹, i.e. attenuation due to thermoelastic loss is about 1% of the attenuation due to phonon–phonon interaction. Hence the total attenuation is predominantly due to ultrasonic attenuation .

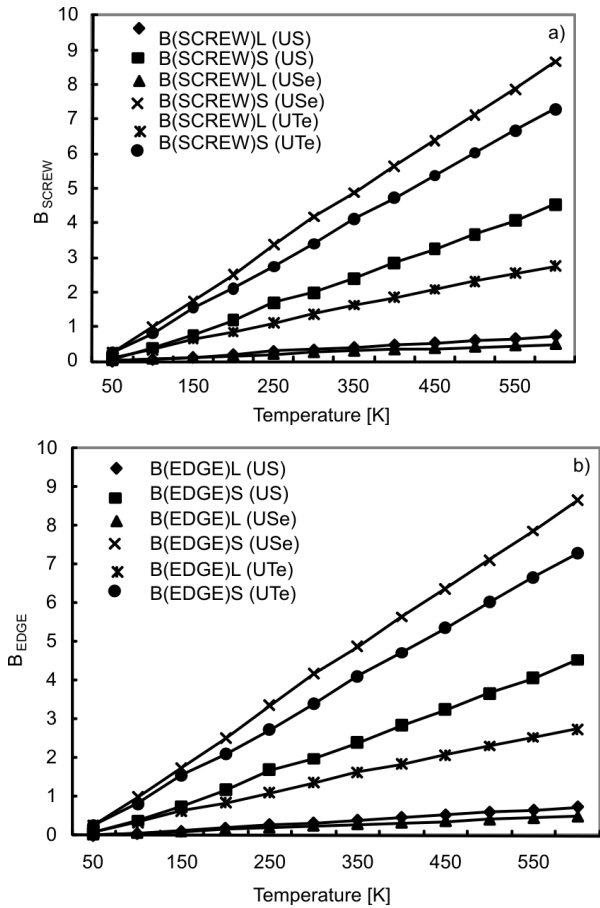


Fig. 4. Temperature dependence of the coefficient of viscosity for a longitudinal wave (cp) and shear wave (mp) due to: a) screw, b) edge dislocation along various directions of propagation

The viscous drag coefficients (B_{SCREW} and B_{EDGE}) were calculated using, Eqs. 9. The temperature dependences of viscous drag due to screw and edge dislocations for longitudinal and shear waves are shown in Figs. 4a, b, respectively. Clearly these parameters increase as the temperature increases.

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