

Ultrasonic investigations in intermetallics

DEVRAJ SINGH* and D K PANDEY

Department of Applied Physics, AMITY School of Engineering and Technology, Bijwasan,
New Delhi 110 061, India

*Corresponding author. E-mail: dsingh1@aset.amity.edu

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Abstract. Ultrasonic attenuation for the longitudinal and shear waves due to phonon-phonon interaction and thermoelastic mechanism have been evaluated in B2 structured intermetallic compounds AgMg, CuZr, AuMg, AuTi, AuMn, AuZn and AuCd along $\langle 100 \rangle$, $\langle 111 \rangle$ and $\langle 110 \rangle$ crystallographic directions at room temperature. For the same evaluations, second- and third-order elastic constants, ultrasonic velocities, Grüneisen parameters, non-linearity parameter, Debye temperature and thermal relaxation time are also computed. Although the molecular weight of these materials increases from AgMg to AuCd, the obtained results are affected with the deviation number. Attenuation of ultrasonic waves due to phonon-phonon interaction is predominant over thermoelastic loss. Results are compared with available theoretical and experimental results. The results with other well-known physical properties are useful for industrial purposes.

Keywords. Intermetallics; deviation number; ultrasonic properties.

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1. Introduction

Today ultrasound is one of the several effective methods used in the field of 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 velocities and attenuation. Ultrasonic velocities and attenuation are very important physical parameters to characterize the material, which are well related to several physical quantities like thermal conductivity, specific heat, thermal energy density, elastic constants, etc. The elastic constants play primary role for understanding the anharmonic and non-linear properties of solids [1].

Intermetallic compounds have received extensive attention in recent years because of their technical promise as high temperature structural materials [2]. The study of intermetallics has attracted the attention of the scientific world because of their anisotropic properties. It has also been found fine application in advanced power engineering. Since many intermetallic compounds of different crystal structures have been found in alloy systems, the basic reason for their stability has drawn a great

deal of attention [3]. The ultrasonic attenuation has direct/indirect relation with thermal conductivity through thermal relaxation time [4,5]. Several works have been done for the investigation of thermal conductivity of intermetallic compounds [6–10] but least for the ultrasonic study [11–13]. The study of ultrasonic attenuation in the materials has gained new dimensions with the progress in material science. Investigators have aimed their study for explaining the temperature-dependent part of the attenuation in terms of model where ultrasonic phonon interacts with thermal phonon in the lattice. All these studies indicate that major portion of attenuation is caused by direct conversion of acoustic energy into heat via phonon–phonon interaction. The present paper is focused on the ultrasonic study of intermetallic compounds.

All chosen materials AgMg, CuZr, AuMg, AuTi, AuMn, AuZn and AuCd have a CsCl-type structure (B2 structure). Ultrasonic velocities, attenuation (α), Grüneisen parameters, non-linearity parameter (acoustic coupling constant), Debye temperature and thermal relaxation time are evaluated and studied in these compounds along different crystallographic directions at room temperature. The second- and third-order elastic constants (higher-order elastic constants) are also calculated using the potential model approach for the above evaluation.

2. Theory

2.1 Higher-order elastic constants and ultrasonic velocities

The second- and third-order elastic constants are related to ultrasonic attenuation through Grüneisen parameters and acoustic coupling constant. The second- and third-order elastic constants of the chosen compounds are calculated using the simplified Ghate's formulation for CsCl-type structured crystals, that is based on the potential model and Brugger's definition of elastic constants [14,15].

There are three type of ultrasonic velocities for each direction of propagation in cubic crystals as one longitudinal (V_L) and two shear (V_{S1} and V_{S2}) [16,17]. The expressions for velocities are as follows:

Along $\langle 100 \rangle$ crystallographic direction

$$V_L = \sqrt{\frac{C_{11}}{d}}; \quad V_{S1} = V_{S2} = \sqrt{\frac{C_{44}}{d}} \quad (1)$$

Along $\langle 111 \rangle$ crystallographic direction

$$V_L = \sqrt{\frac{C_{11} + 2C_{12} + 4C_{44}}{3d}}; \quad V_{S1} = V_{S2} = \sqrt{\frac{C_{11} - C_{12} + C_{44}}{3d}}. \quad (2)$$

Along $\langle 110 \rangle$ crystallographic direction

$$V_L = \sqrt{\frac{C_{11} + C_{12} + 2C_{44}}{2d}}; \quad V_{S1} = \sqrt{\frac{C_{44}}{d}}; \quad V_{S2} = \sqrt{\frac{C_{11} - C_{12}}{d}}. \quad (3)$$

The ultrasonic velocities can be computed using calculated values of second-order elastic constants. The Debye average velocity (V_D) is useful to find Debye temperature and thermal relaxation time of the materials. The Debye temperature (T_D) is an important physical parameter of solids, which defines a division line between quantum and classical behaviour of phonons [18]. The following expressions have been used for the evaluation of Debye average velocity and Debye temperature [4,18,19].

$$\left(\frac{1}{V_D}\right)^3 = \frac{1}{3} \sum_i \frac{1}{4\pi} \int \frac{1}{V_i^3} d\Omega, \quad (4)$$

$$T_D = \frac{\hbar V_D (6\pi^2 n_a)^{1/3}}{K_B}, \quad (5)$$

where V_i is the constituent velocity along a particular direction of wave propagation, \hbar is the quantum of action and is equal to Planck's constant divided by 2π , K_B is the Boltzmann constant and n_a is the atom concentration.

2.2 Ultrasonic attenuation

The prominent reasons behind ultrasonic attenuation in a single, non-ferromagnetic, non-ferroelectric and pure crystal at high temperature are phonon viscosity mechanism (phonon-phonon interaction: Akhieser loss) and thermoelastic mechanism [4,5,11-13,20-26]. The total attenuation coefficient can be given by the following expression:

$$\left(\frac{\alpha}{f^2}\right)_{\text{Total}} = \left(\frac{\alpha}{f^2}\right)_{\text{p-p}} + \left(\frac{\alpha}{f^2}\right)_{\text{Th}} = \left(\frac{\alpha}{f^2}\right)_{\text{Akh}} + \left(\frac{\alpha}{f^2}\right)_{\text{Th}}, \quad (6)$$

$$\left(\frac{\alpha}{f^2}\right)_{\text{Akh}} = \frac{4\pi^2 \tau E_0 D}{6dV^3}, \quad (7a)$$

$$\left(\frac{\alpha}{f^2}\right)_{\text{Th}} = \frac{4\pi^2 \langle \gamma \rangle^2 kT}{2dV_L^3}, \quad (7b)$$

where

$$D = 9\langle \gamma^2 \rangle - 3\langle \gamma \rangle^2 (C_V T / E_0) \quad (8)$$

$$\tau_{\text{Th}} = \tau_s = \frac{\tau_{\text{Long}}}{2} = \frac{3k}{C_V V_D^2}. \quad (9)$$

Here, k is the thermal conductivity, C_V is the specific heat, E_0 is the thermal energy density, α is the ultrasonic attenuation coefficient, γ is the Grüneisen parameter, D is the non-linearity parameter, T_D is the Debye temperature, T is the temperature, V is the longitudinal/shear velocity and τ is the thermal relaxation time.

Grüneisen parameters are well related to the second- and third-order elastic constants [22]. C_V and E_0 can be obtained using T_D/T table of AIP Handbook [27].

Table 1. Second- and third-order elastic constants (in 10^{10} N/m²) of the compounds.

| | AgMg | CuZr | AuMg | AuTi | AuMn | AuZn | AuCd |
|-----------|---------|---------|---------|---------|---------|---------|---------|
| C_{11} | 8.471 | 7.438 | 8.642 | 6.836 | 7.046 | 9.239 | 8.006 |
| C_{12} | 4.325 | 3.637 | 4.443 | 3.234 | 3.370 | 4.860 | 4.007 |
| C_{44} | 3.802 | 3.218 | 3.901 | 2.889 | 3.003 | 4.248 | 3.537 |
| C_{111} | -79.458 | -68.393 | -81.307 | -62.041 | -64.249 | -87.779 | -74.459 |
| C_{112} | -10.001 | -9.565 | -10.066 | -9.279 | -9.379 | -10.278 | -9.814 |
| C_{123} | -6.997 | -7.087 | -6.972 | -7.087 | -7.092 | -6.865 | -7.051 |
| C_{144} | -5.669 | -5.646 | -5.662 | -5.574 | -5.604 | -5.613 | -5.674 |
| C_{166} | -9.489 | -8.851 | -9.586 | -8.430 | -8.581 | -9.904 | -9.214 |
| C_{456} | -7.577 | -7.588 | -7.564 | -7.536 | -7.559 | -7.498 | -7.596 |

Table 2. Basic input quantities of the intermetallic compounds.

| | AgMg | CuZr | AuMg | AuTi | AuMn | AuZn | AuCd |
|------------------------------------|---------|---------|---------|---------|---------|---------|---------|
| Mol. wt. (g) | 132.173 | 154.766 | 221.273 | 244.869 | 251.907 | 262.289 | 309.379 |
| d (10^3 kg/m ³) | 6.22 | 6.43 | 10.62 | 9.36 | 9.93 | 13.42 | 13.79 |
| T_D (K) | 309 | 269 | 241 | 204 | 206 | 228 | 197 |
| k (10^2 W/mK) | 1.472 | 0.124 | 0.539 | 0.135 | 0.257 | 1.103 | 0.847 |
| C_V (10^6 J/m ³ K) | 1.210 | 0.998 | 1.163 | 0.984 | 0.962 | 1.240 | 1.089 |
| E_0 (10^8 J/m ³) | 2.353 | 2.191 | 2.605 | 2.323 | 2.262 | 2.855 | 2.591 |

3. Results and discussion

The calculation of second- and third-order elastic constants has been done using lattice parameter and hardness parameter. The lattice parameters for AgMg, CuZr, AuMg, AuTi, AuMn, AuZn and AuCd compounds are 3.28 Å, 3.42 Å, 3.259 Å, 3.515 Å, 3.478 Å, 3.19 Å and 3.34 Å respectively. The calculated elastic constants are presented in table 1.

The molecular weights of the compounds are given in table 2. Densities of the materials are calculated with the help of [28] and are presented in table 2. Thermal conductivities of the chosen materials are taken from [29] and is given in table 2. The calculated T_D using eq. (5) and evaluated C_V and E_0 are also presented in table 2.

The ultrasonic velocities and Debye average velocities along different crystallographic directions of the compounds have been calculated using eqs (1)–(4) and are presented in table 3.

The ultrasonic attenuation coefficient over frequency square due to p–p interaction and thermoelastic mechanism for different directions of propagation of ultrasonic wave has been calculated using eqs (6)–(8) and are presented in table 4. The thermal conductivity, calculated thermal relaxation time and total ultrasonic attenuation coefficient over frequency square are shown in figures 1–3.

The mechanical strength and durability of the material are related to their elastic behaviour and are also used for the determination of the ultrasonic attenuation,

Table 3. Ultrasonic velocities (in 10^3 m/s) of compounds along different crystallographic directions.

| | | AgMg | CuZr | AuMg | AuTi | AuMn | AuZn | AuCd |
|-----|------------------|-------|-------|-------|-------|-------|-------|-------|
| 100 | V_L | 3.690 | 3.401 | 2.851 | 2.632 | 2.663 | 2.624 | 2.409 |
| | V_S | 2.472 | 2.237 | 1.915 | 1.711 | 1.739 | 1.779 | 1.601 |
| | V | 2.668 | 2.420 | 2.066 | 1.854 | 1.883 | 1.917 | 1.730 |
| 111 | V_L | 4.163 | 3.780 | 3.222 | 2.898 | 2.943 | 2.988 | 2.699 |
| | V_S | 2.064 | 1.909 | 1.594 | 1.481 | 1.497 | 1.464 | 1.349 |
| | V | 2.288 | 2.114 | 1.768 | 1.639 | 1.657 | 1.625 | 1.496 |
| 110 | V_L | 4.049 | 3.689 | 3.134 | 2.834 | 2.876 | 2.901 | 2.630 |
| | V_{S1}^* | 2.472 | 2.237 | 1.915 | 1.711 | 1.739 | 1.779 | 1.601 |
| | V_{S2}^\dagger | 1.826 | 1.721 | 1.406 | 1.351 | 1.361 | 1.277 | 1.204 |
| | V | 2.275 | 2.116 | 1.755 | 1.645 | 1.663 | 1.606 | 1.491 |

*Shear wave polarized along $\langle 001 \rangle$.

†Shear wave polarized along $\langle 1\bar{1}0 \rangle$.

and so elastic constants of the material give useful information. The second-order elastic constants (SOEC) of the present intermetallic compounds are given in table 1. These data can be used for the determination of bulk (B)/shear modulus of compounds. The experimental values of C_{11} , C_{12} , C_{44} and B for AgMg at 300 K are 8.46×10^{10} N/m², 5.67×10^{10} N/m², 4.85×10^{10} N/m² and 6.6×10^{10} N/m² respectively [3]. The present values for the same compound are 8.471×10^{10} N/m², 4.325×10^{10} N/m², 3.637×10^{10} N/m² and 5.71×10^{10} N/m² respectively, which are in good agreement with experimental. So the method adopted by us for the calculation of higher-order elastic constants is justified for these intermetallic compounds also. A slight variation with experimental is because the theory applied here involves atomic interaction only up to second nearest neighbour. The order of elastic constants in these compounds are the same as for CsCl, CsBr and CsI but with high C_{11} magnitude [22]. The higher values of C_{11} for these materials indicate that they have better mechanical strength than the Cs-halide compounds.

The calculated density of AgMg has been found to be 6.22×10^3 kg/m³ (see table 2) while 6.04×10^3 kg/m³ is given in [3]. Thus our calculated densities for these compounds are correct and valid. The calculated densities of the chosen compounds are less than the densities of pure noble metals (Ag, Au and Cu). Hence these materials will be lighter in weight than the pure noble metals.

The C_{11} for pure Ag, Au and Cu metals are 12.4×10^{10} N/m², 20.0×10^{10} N/m² and 16.84×10^{10} N/m² respectively [17,22]. Present values of elastic constants show that by introducing Mg, Zr, Ti, Mn, Zn, Cd in noble metals, the elastic behaviour slightly decreases due to loose interaction of impurity atoms with noble metal atoms. The calculated third-order elastic constants (TOEC) are not compared due to lack of data in literature. A comparison with literature [13,14] indicates that negative TOEC values of the compounds are justified. The calculated elastic constants have been used for the evaluation of ultrasonic velocities, Debye average velocity, Debye temperature and attenuation.

Table 4. Ultrasonic attenuation over frequency α/f^2 (in 10^{-15} Nps²/m) of compounds along $\langle 100 \rangle$, $\langle 110 \rangle$ and $\langle 111 \rangle$ directions.

| | | AgMg | CuZr | AuMg | AuTi | AuMn | AuZn | AuCd |
|-------|-------------------------------------------|---------|-------|---------|--------|--------|---------|---------|
| 1 0 0 | $(\alpha/f^2)_{\text{Th}}$ | 0.050 | 0.006 | 0.039 | 0.015 | 0.026 | 0.103 | 0.106 |
| | $(\alpha/f^2)_{\text{Akh.Long}}$ | 2.301 | 0.293 | 1.926 | 0.819 | 1.445 | 4.948 | 5.709 |
| | $(\alpha/f^2)_{\text{Akh.Shear}}$ | 0.851 | 0.141 | 0.683 | 0.465 | 0.771 | 1.548 | 2.345 |
| 1 1 1 | $(\alpha/f^2)_{\text{Th}}$ | 0.010 | 0.002 | 0.008 | 0.005 | 0.008 | 0.017 | 0.024 |
| | $(\alpha/f^2)_{\text{Akh.Long}}$ | 1.229 | 0.178 | 1.016 | 0.545 | 0.931 | 2.482 | 3.237 |
| | $(\alpha/f^2)_{\text{Akh.Shear}}$ | 12.833 | 1.378 | 10.946 | 3.384 | 6.248 | 30.335 | 29.454 |
| 1 1 0 | $(\alpha/f^2)_{\text{Th}}$ | 0.206 | 0.023 | 0.163 | 0.058 | 0.104 | 0.424 | 0.433 |
| | $(\alpha/f^2)_{\text{Akh.Long}}$ | 5.905 | 0.713 | 4.992 | 1.921 | 3.435 | 13.196 | 14.369 |
| | $(\alpha/f^2)_{\text{Akh.Shear}}^*$ | 174.285 | 6.678 | 193.155 | 11.782 | 24.057 | 276.160 | 227.428 |
| | $(\alpha/f^2)_{\text{Akh.Shear}}^\dagger$ | 23.902 | 2.397 | 20.593 | 5.632 | 10.567 | 59.185 | 53.251 |

The Debye temperature (DT) for Ag, Au, Cu, Cd, Zn and Mg are 226, 289, 224, 214, 272 and 400 K respectively [17,22]. Thus the present values of Debye temperature (table 2) for mixed compounds with Ag or Cu lie between Debye temperatures of constituent materials while Debye temperatures for mixed compounds with Au lie below the constituent materials. Debye temperatures for AgMg are 312 K [3] and 309 K in the present calculation. Thus our evaluation of T_D is justified. The relation of elastic behaviour to thermal behaviour can be made through Debye theory. According to this theory, the long-wavelength vibrational contribution to the specific heat at low temperature is proportional to $(T/T_D)^3$ [30]. Hence decrease or increase in Debye temperature indicates increase or decrease in acoustic contribution to the low temperature specific heat.

The ultrasonic velocities and average velocity of these intermetallic compounds along different crystallographic directions are given in table 3. The average sound velocities in these intermetallic compounds are larger than in the noble metals as well as in Cs/Rb halides, because of the low density of these compounds [17,22]. Ultrasonic velocity in these materials decreases with their molecular weight. On the basis of ultrasonic velocity, the differentiation among AuTi, AuMn and AuZn are difficult because they have very close values as presented in table 3. The velocity of these compounds is useful for determining their anisotropic properties.

The ultrasonic wave propagation is influenced by the microstructure of the material through which it propagates. Ultrasonic attenuation, which is the sum of absorption and scattering, is mainly depending upon the damping capacity and scattering from the grain boundary in the material. Loss due to scattering is dominant in polycrystalline solids. However, for complete characterization of the material, the attenuation requires the knowledge of a large number of thermophysical parameters [31].

Table 4 indicates that the ultrasonic attenuation in these intermetallic compounds due to phonon-phonon interaction for longitudinal waves is larger than that of shear waves along $\langle 100 \rangle$ direction due to larger acoustic coupling constants along longitudinal direction (see table 5), while attenuation of shear waves is larger than that of longitudinal waves along $\langle 110 \rangle$ and $\langle 111 \rangle$ directions due to larger acoustic

Table 5. Acoustic coupling constants of intermetallics along different directions.

| | | AgMg | CuZr | AuMg | AuTi | AuMn | AuZn | AuCd |
|-------|------------------|---------|--------|---------|--------|--------|----------|--------|
| 1 0 0 | D_L | 4.201 | 4.048 | 4.251 | 4.029 | 4.035 | 4.401 | 4.145 |
| | D_S | 0.934 | 1.106 | 0.915 | 1.258 | 1.199 | 0.859 | 0.999 |
| 1 1 1 | D_L | 2.368 | 2.573 | 2.371 | 2.796 | 2.716 | 2.342 | 2.473 |
| | D_S | 6.029 | 5.131 | 6.176 | 4.634 | 4.805 | 6.729 | 5.618 |
| 1 1 0 | D_L | 10.357 | 9.597 | 10.563 | 9.293 | 9.412 | 11.142 | 10.096 |
| | D_{S1}^* | 139.151 | 40.102 | 186.559 | 25.093 | 29.152 | 1075.371 | 72.102 |
| | D_{S2}^\dagger | 7.685 | 6.558 | 7.863 | 5.906 | 6.134 | 8.529 | 7.176 |

coupling constants for shear wave. This nature of Akhieser loss is similar to that of B1 structured (NaCl-type) materials and is different only for shear wave propagation along $\langle 110 \rangle$ (polarized along $\langle 001 \rangle$) direction [20,23].

Acoustic coupling constant is a measure of acoustic energy converted to thermal energy due to phonon-viscosity mechanism. It can be seen from table 4 that ultrasonic attenuation due to thermoelastic relaxation is less in comparison to attenuation due to phonon-viscosity mechanism. Hence the total attenuation in these intermetallic compounds are mainly governed by phonon-phonon interaction. Although experimental results for ultrasonic attenuation of these materials might not be found in literature, a comparison has been made with available experimental/theoretical results with this type of structured materials and the noble metals Cu, Ag and Au [24–26]. In general, the comparison reveals that the total attenuation in these intermetallics is greater than in the Cs/Rb-halides and is less than in the pure noble metals.

The total attenuation over frequency square and thermal relaxation time have been plotted in figures 1–3 with molecular weight of these compounds for wave propagation along different directions. Yet a large number of physical parameters are involved for evaluating the attenuation but figures 1–3 predict that the total attenuation along each direction of propagation is dominated by the thermal relaxation time along the same direction. Thermal relaxation time gives a combined effect of thermal conductivity, specific heat and average sound velocity. Attenuation over frequency square is proportional to thermal relaxation time and acoustic coupling constant, and hence attenuation in these intermetallics are affected by the combined effect of thermal conductivity, specific heat, average sound velocity and acoustic coupling constant.

The total attenuation in CuZr, AuTi and AuMn are minimum in comparison to AuMg, AgMg, AuZn or AuCd, thus they exist in the purest form at high temperature while compounds AuCd and AuZn are not in so pure form at high temperature as they have larger attenuation.

For CsCl-type structure, the deviation number ΔN exists from 1 to 3; ΔN denotes the difference of column number of noble metals and the secondary element in the helical periodic table. The thermal conductivity will be high for lower valued ΔN compounds [29]. ΔN values for AgMg, CuZr, AuMg, AuTi, AuMn, AuZn and AuCd are 1, 3, 1, 3, 2, 1 and 1 respectively. The compounds AgMg, AuMg, AuZn and AuCd for which $\Delta N = 1$, have larger conductivity.

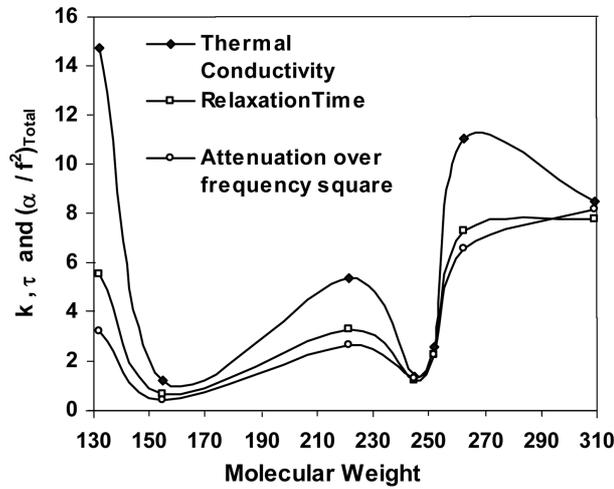


Figure 1. Thermal conductivity (in 10^2 W/mK), thermal relaxation time (in 10^{-11} s) and total ultrasonic attenuation over frequency square (in 10^{-15} Nps²/m) vs. molecular weight of the compounds for the wave propagation along $\langle 100 \rangle$ direction.

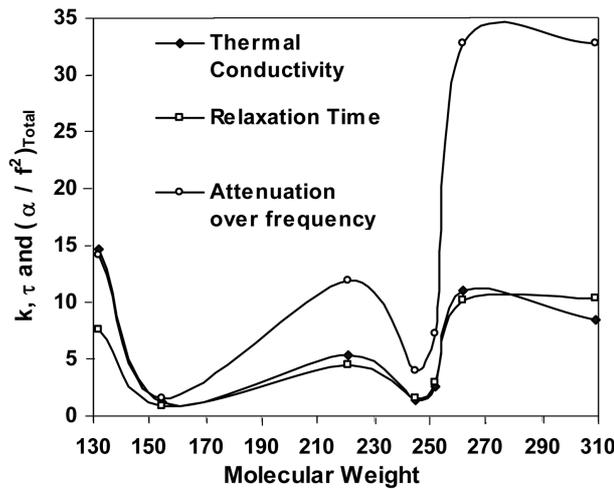


Figure 2. Thermal conductivity (in 10^2 W/mK), thermal relaxation time (in 10^{-11} s) and total ultrasonic attenuation over frequency square (in 10^{-15} Nps²/m) vs. molecular weight of the compounds for the wave propagation along $\langle 111 \rangle$ direction.

Since $(\alpha/f^2) \propto \tau \propto k$, then $(\alpha/f^2) \propto 1/\Delta N$, the trend of ultrasonic attenuation should monotonously decrease with increase in ΔN . Our results of ultrasonic attenuation in these intermetallics (figures 1–3) justify the above prediction. Thus, it may be concluded that in B2 structured intermetallic compounds the nature of

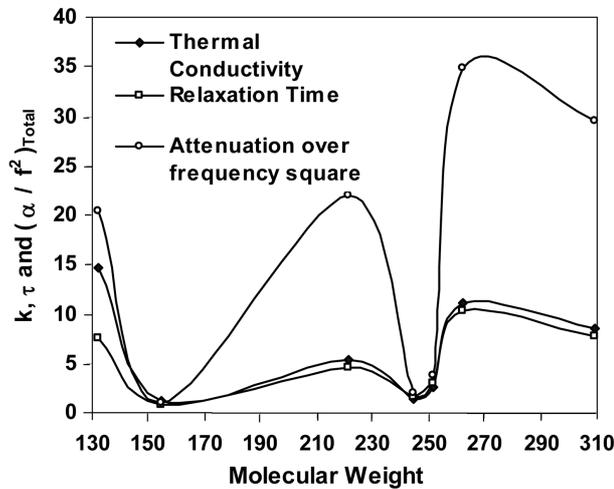


Figure 3. Thermal conductivity (in 10^2 W/mK), thermal relaxation time (in 10^{-11} s) and total ultrasonic attenuation over frequency square (in 10^{-14} Nps²/m) vs. molecular weight of the compounds for the wave propagation along $\langle 110 \rangle$ direction.

ultrasonic attenuation can be dominantly determined by the deviation number. A slight variation in attenuation for the same deviation numbered compounds is due to the corresponding values of specific heat, average sound velocity and acoustic coupling constant.

4. Conclusions

The method adopted for the theoretical evaluation of second- and third-order elastic constants is also justified for these intermetallic compounds. These materials have good mechanical strength compared to the Cs/Rb-halides. These intermetallics are light weighted materials with respect to noble metals as they have low density. The average sound velocity in the present materials is larger than in the noble metals and Cs/Rb-halides. On the basis of ultrasonic velocities, these intermetallics cannot be differentiated because they have very close values of elastic constants but are important for anisotropic characterization. The nature of thermal relaxation time follows the reciprocal trend of the deviation number. Total attenuation in these materials is mainly affected via phonon-viscosity mechanism. The larger value of longitudinal/shear attenuation due to phonon-phonon interaction is caused by their high acoustic coupling constant. The total attenuation gives direct information about the deviation number.

The obtained results in the present investigation with other well-known physical properties of these materials may expand future prospects for application/investigation of these intermetallic compounds.

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