Acoustical investigation of magnesium oxide

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The higher order elastic constants of MgO starting from basic parameters viz. the nearest neighbour distance, hardness parameter using Coulomb and Börn-Mayer potentials have been obtained. These values are utilized to obtain Grüneisen parameters and non-linearity constants. Non-linearity constants ratio, ultrasonic attenuation due to phonon-phonon interaction \((\alpha/f^2)_{p-p}\) and thermo elastic loss \((\alpha/f^2)_{th}\) are calculated in a wide temperature range along <100>, <110> and <111> crystallographic directions of propagation for longitudinal and shear waves and compared with available theoretical and experimental results.

Keywords: Elastic constants, Magnesium oxide, Ultrasonics, Semiconducting crystals

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1 Introduction

Magnesium oxide is the simplest oxide, and has been a subject of intense experimental and theoretical study. Oxides and silicates make up the bulk of the Earth's mantle and crust, and thus it is important to understand and predict their behaviour. An important feature of MgO is the non-rigid behaviour of the oxygen \(\text{O}^{2-}\) ion, which makes the interactions not describable by pairwise interactions. All other more complex oxides share this feature and add additional complications as well.

The ultrasonic velocity and attenuation studies\(^1\)\(^-\)\(^10\) are widely used as a versatile tool in studying the inherent properties and internal structure of material viz. metallic, dielectric and semiconducting crystals. In different types of solids, the attenuation of acoustic waves is attributed due to different causes which are lattice imperfection, electron-phonon interaction, ferromagnetic and ferroelectric properties, phonon-viscosity, thermo elastic losses and thermal relaxation. The phonon viscosity and non-elastic phenomena are the principal causes\(^11\) of ultrasonic attenuation in all types of substances at higher temperature. The prominent mechanism for the absorption of sound wave in solids is the Akhiezer\(^12\) mechanism in which the attenuation results because, in the presence of ultrasound, the thermal phonons are not in equilibrium. In the process of coming to equilibrium, entropy is produced and energy of the sound wave is lost. In the present paper, Börn-Mayer\(^13\) and electrostatic potentials have been used to obtain second and third order elastic constants (SOECs and TOECs), taking the nearest neighbour distance and hardness parameter of MgO. These values of SOECs and TOECs have been used to obtain Grüneisen numbers hence non-linearity constants along the different directions for longitudinal and shear waves. Thermal relaxation time, non-linearity constants ratio, thermo elastic losses and ultrasonic attenuation due to p-p interaction are then calculated for magnesium oxide.

The values of \((\alpha/f^2)_{p-p}\) and \((\alpha/f^2)_{th}\) have been evaluated in the temperature range 100-1000 K for longitudinal waves along <100>, <110> and <111> directions and for shear waves, along <100>, <110> (polarized along <001> and <110>direction) and <111> (polarized along <110> and <112> directions) crystallographic directions of propagation. The results are compared with available theoretical and experimental values.

2 Theory

The theory of ultrasonic attenuation will be completed in two parts. In first part, we discuss about temperature variation of elastic constants and then ultrasonic attenuation at different temperatures in the second part.

2.1 Elastic constants

Assuming electrostatic potential \(\pm (e^2/r)\) and Börn-Mayer type potential \(Q_{\text{ov}} (r_0) = A \exp (-r_0/q); e,
\(r_0\) and \(q\) being electronic charge, nearest-neighbour distance and hardness parameter, and following Brügger's\(^{14}\) definition of elastic constants at absolute zero, second and third order elastic constants (SOECs and TOECs) are obtained. Leibfried \textit{et al.}\(^{15,16}\) have developed the theory of anharmonic properties of crystals, which has been used to obtain SOECs and TOECs at different temperatures.

2.2 Ultrasonic attenuation

As elastic constants evaluation is done, Grüneisen numbers \((\gamma_i^j)\) are calculated using Grüneisen number tables\(^{17}\). These parameters directly give Mason's\(^{18,19}\) non-linearity coupling constant \((D)\) as presented below:

\[
D = 9 <\gamma_i^j^2> - (3C_v T/ E_0) <\gamma_i^j^2>^2 \quad \ldots (1)
\]

where \(<\gamma_i^j^2>\) and \(<\gamma_i^j^2>^2\) are average Grüneisen and square average Grüneisen numbers, \(C_v\) the specific heat per unit volume, \(T\) the temperature and \(E_0\) is the energy of the crystal.

The thermo elastic losses and attenuation\(^{20}\) due to phonon-phonon interaction are given by:

\[
(\alpha/ f^2)_{th} = (2\pi)^2 <\gamma_i^j^2> K T / 2dV^s \quad \ldots (2)
\]

\[
(\alpha/ f^2)_{p-p} = (2\pi)^2 E_0 (D/3) \tau_{th} / dV^s (1+ \omega^2 \xi^2) \quad \ldots (3)
\]

In our case, \(\omega \tau_{th} \ll 1\), therefore the relation reduces to:

\[
(\alpha/ f^2)_{p-p} = (2\pi)^2 E_0 D \tau_{th} / 3 dV^s^3 \quad \ldots (4)
\]

The attenuation coefficients for longitudinal and shear waves are given by:

\[
(\alpha/ f^2)_l = (2\pi)^2 E_0 D_l \tau_l / 6dV_l^3 \quad \ldots (5)
\]

and \((\alpha/ f^2)_s = (2\pi)^2 E_0 D_s \tau_s / 3dV_s^3 \quad \ldots (6)\]

where \(d\) is crystal mass density, \(D_l\) and \(D_s\) are non-linearity coupling constants, \(\tau_l\) and \(\tau_s\) are relaxation times, and \(v_l\) and \(v_s\) are acoustic wave velocities for longitudinal(l) and shear(s) waves respectively. The two relaxation times are related as:

\[
(1/2) \tau_l = \tau_s = \tau_{th} = 3 K / <dC_v> \bar{v}^2 \quad \ldots (7)
\]

where, \(\tau_{th}\) is thermal relaxation time, \(K\) the thermal conductivity and \(\bar{v}^2\) is the Debye average velocity.

The \(v_l, v_s, v\) are given by:

\[
v_l = (C_{11}/d)^{1/2}, \quad v_s = (C_{44}/d)^{1/2}
\]

and \((3\sqrt{V^3}) = (1/v_l^3)+(2/v_s^3) \quad \ldots (8)\]

SOECs and TOECs for MgO have been calculated (taking \(r=2.014 \text{ Å}\) and \(q=0.345 \text{ Å}\)) at different temperatures (100 to 1000K) using the theory described in Refs(14-16). The variations of these constants with temperature are shown in Fig. 1(a-d). Grüneisen parameters have been evaluated using Mason’s Grüneisen tables\(^{17}\) at different temperatures and along various directions, as shown in Fig. 2(a-c).

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Fig. 1—Temperature dependence of (a)SOECs (b) \(C_{111}\) (c) \(C_{116}\) and \(C_{112}\) & (d) \(C_{123}, C_{144}\) and \(C_{256}\) (in \(10^{11} \text{ dyne / cm}^2\)) for MgO.
With the help of Eq.(1), non-linearity constants have been obtained and presented in Fig. 3(a-c). The thermal relaxation time has been obtained at different temperatures taking temperature variation of thermal conductivity as shown in Fig. (4). The temperature dependence of acoustic velocities are also presented in Fig. (5). On taking specific heat and crystal energy density at different temperatures as a function of $\theta_D/T$, where $\theta_D$ is Debye temperature, thermo elastic losses and ultrasonic attenuation at different temperatures obtained and shown in Fig. 6(a-c).

3 Results and Discussion

In the present study, SOECs are all positive, while among TOECs, three are negative and three are positive. The magnitude of SOEC and TOEC and their temperature variation {Fig. 1(a-d)} plays a crucial role in the investigation of ultrasonic attenuation in MgO. These constants may be used for the purpose of investigating anharmonic properties in MgO. Using Grüneisen tables, average Grüneisen numbers are computed for longitudinal and shear waves along <100>, <110> and <111> directions. It is observed from Fig. 2(a-c), that the average Grüneisen
numbers are decreasing with temperature for MgO. A comparison between our results and other available theoretical\textsuperscript{24} and experimental\textsuperscript{25} values have also been made in Table 1. In view of uncertainties of experimental data, our results are firmly supported by the available data.

The ratio of non–linearity constants lies in the range 1.37-1.95 for shear wave polarized along <001> direction and is less then 2 for shear wave polarized along other directions {Fig. 3(a-c)} which is also seen by other researchers for different types of substances\textsuperscript{11}. Table 2 shows the comparison with other investigations. Clearly, the value of thermal relaxation time is of the order of $10^{-12}$ s, which is expected. At 100 K, its value is very high and decreases exponentially as temperature is increased, as shown in Fig. 4. The calculated values of ultrasonic wave velocities are increasing as temperature increases for longitudinal and shear waves and are shown in Fig. 5.

It is clear that the value of $(\alpha/\beta)^{\text{th}}$ is negligible in comparison of $(\alpha/\beta)^{\text{p-p}}$ {Fig. 6(a-c)}, which divulges the fact that a major part of ultrasonic energy loss is used in achieving the equilibrium along different phonon branches and directions at various temperatures. From results shown in {Fig. 6(a-c)}; one may say that attenuation is very low at 100 K and increases as temperature increases for longitudinal and shear waves both. It has been found that the attenuation $(\alpha/\beta)^{\text{p-p}}$ in MgO assumes saturation value after 400 K. Our results are in well agreement with the experimental data\textsuperscript{26}. It is seen that attenuation for

\begin{table}[h]
\centering
\caption{Comparison data of attenuation and Grüneisen numbers for MgO at room temperature}
\begin{tabular}{|c|c|c|c|c|c|c|}
\hline
Prop. Direction & $\langle\alpha/\beta\rangle_{\text{th}}^{\text{p-p}}$ & $\langle\alpha/\beta\rangle_{\text{p-p}}^{\text{p-p}}$ & $\langle\gamma\rangle$ & $\langle\gamma\rangle$ & $\langle\gamma\rangle$ & $\langle\gamma\rangle$
\hline
<100>-long. shear & 1.27 & 1.230 & 3.94 & 3.71 & 3.39 & 3.27
\hline
<110>-long. shear & 0.55 & 0.260 & 3.12 & 0.52 & 0.18 & 0.15
\hline
<100>-long. shear & 2.81 & 0.25 & 7.89 & 1.88 & 1.93 & 1.28
\hline
<110>-long. shear & 0.943 & 0.05 & 5.32 & 0.41 & 0.18 & 0.23
\hline
<110>-long. shear & 0.944 & 0.07 & 5.32 & 1.87 & 1.39 & 1.58
\hline
<111>-long. shear & 1.74 & 1.60 & 0.78 & 1.59 & 1.34 & 0.90
\hline
<110>-long. shear & 0.59 & 0.25 & 3.33 & 3.29 & 1.90 & –
\hline
<111>-long. shear & 1.33 & 0.50 & 7.53 & 6.01 & 4.66 & –
\hline
\end{tabular}
\end{table}
longitudinal wave is more than shear wave. From the attenuation values along different directions; it is evident that the ultrasonic attenuation is different along different directions of propagation and varies with the orientation of the crystal, similar behaviour has been observed by other researchers in solids.

References

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<td>1.42</td>
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<td>1.47</td>
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<td>&lt;112&gt;</td>
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Table 3—Comparison data of thermal relaxation time for MgO

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<th>Temp(K)</th>
<th>Present</th>
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<td>16.51</td>
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<td>2.00</td>
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<td>500</td>
<td>0.92</td>
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Table 2—Comparison data of Dl/Ds (ratio of non linearity constants) for MgO at room temperature