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Temperature variation of higher-order elastic constants of MgO

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Abstract. An effort has been made for obtaining higher-order elastic constants for MgO starting from basic parameters, viz. nearest-neighbor distance and hardness parameter using Coulomb and Börn–Mayer potentials. These are calculated in a wide temperature range (100–1000 K) and compared with available theoretical and experimental results.

Keywords. Elastic energy density; higher-order elastic constants; anharmonic properties.

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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 behavior. An important feature of MgO is the non-rigid behavior of the O^{2-} ion, which makes the interactions not describable by pair-wise interactions. All other more complex oxides share this feature, and add additional complications as well.

Elastic constants are sensitive to details of the electronic interactions in a crystal, which becomes evident when one attempts to compute them. Elastic constants are very sensitive to convergence parameters and k-point sampling in self-consistent computations. There are no self-consistent studies of temperature dependencies on elasticity in MgO.

In the last and present decades considerable interest has been shown in investigating the ultrasonic properties of materials [1–5]. The present theory deals with temperature dependence of higher-order elastic constants of MgO crystal having face centered cubic crystal symmetry. In the present investigation, some efforts have been made for obtaining the second- and third-order elastic constants (SOECs and

TOECs) of MgO starting from nearest-neighbour distance and hardness parameter utilizing the Coulomb and Börn–Mayer [6] potentials.

The elastic energy density for a deformed crystal can be expanded as a power series of strains using Taylor's series expansion. The coefficients of quadratic, cubic and quartic terms are known as the second-, third- and fourth-order elastic constants (SOECs, TOECs and FOECs) respectively. When the values of these elastic constants of a crystal are known, many of the acoustic properties of the substances can be treated within the limit of the continuum approximation in a quantitative manner. Several physical properties and crystal anharmonicities such as thermal expansion, specific heat at higher temperature, temperature variation of ultrasonic velocity and attenuation, first-order pressure derivatives (FOPDs) of SOECs are directly related to SOECs and TOECs. While discussing higher order anharmonicities such as the first-order pressure derivatives of TOECS, the second-order pressure derivatives (SOPDs) of SOECs, partial contraction and deformation of crystals under large forces, the FOECs are to be considered extensively.

2. Theory

The elastic energy density for a crystal of cubic symmetry can be expanded to quartic terms as shown below [1,2]:

$$\begin{aligned} U_0 &= U_2 + U_3 + U_4 \\ &= [1/2!]C_{ijkl}x_{ij}x_{kl} + [1/3!]C_{ijklmn}x_{ij}x_{kl}x_{mn} \\ &\quad +[1/4!]C_{ijklmnpq}x_{ij}x_{kl}x_{mn}x_{pq}, \end{aligned} \tag{1}$$

where C_{ijkl}, C_{ijklmn} and $C_{ijklmnpq}$ are the SOECs, TOECs and FOECs in tensorial form and x_{ij} are the Lagrangian strain components. C_{IJ}, C_{IJK} and C_{IJKL} are the SOECs, TOECs and FOECs in Brügger's definition [7] and Voigt notations [8] which are as given below:

$$\begin{aligned} C_{ijkl} = C_{IJ} &= \left(\frac{\partial^2 U}{\partial x_{ij}\partial x_{kl}}\right)_{x=0}; \\ C_{ijklmn} = C_{IJK} &= \left(\frac{\partial^3 U}{\partial x_{ij}\partial x_{mn}\partial x_{kl}}\right)_{x=0}; \\ C_{ijklmnpq} = C_{IJKL} &= \left(\frac{\partial^4 U}{\partial x_{ij}\partial x_{kl}\partial x_{mn}\partial x_{pq}}\right)_{x=0}. \end{aligned} \tag{2}$$

An elastic constant consists of two parts as follows:

$$C_{IJ} = C^0_{IJ} + C^{\text{vib}}_{IJ};\ C_{IJK} = C^0_{IJK} + C^{\text{vib}}_{IJK};\ C_{IJKL} = C^0_{IJKL} + C^{\text{vib}}_{IJKL}. \tag{3}$$

The first part is the strain derivative of the internal energy U_0 and is known as 'static' elastic constant and the second part is the strain derivative of the vibrational free energy U^{vib} and is called 'vibrational' elastic constant. The superscript '0'

Table 1. Expressions for the SOECs and TOECs at 0 K for MgO.

$C_{11}^0 = -1.56933G + G_1 + 2G_2$
$C_{12}^0 = C_{44}^0 = 0.347775G + G_2$
$C_{111}^0 = 10.2639G - G_3 - 2G_4$
$C_{112}^0 = C_{166}^0 = 1.208625G - G_4$
$C_{123}^0 = C_{144}^0 = C_{456}^0 = 0.678375G$
where $G = e^2/r_0^4, G_1 = (1/r_0 + 1/q)Q(r_0)/qr_0$, $G_2 = (\sqrt{2}/2r_0 + 1/q)Q(r_0\sqrt{2})/qr_0$,
$G_3 = (3/r_0^2 + 3/qr_0 + 1/q^2)Q(r_0)/q$, $G_4 = (3\sqrt{2}/r_0^2 + 6/qr_0 + 2\sqrt{2}/q^2)Q(r_0\sqrt{2})/4q$

Table 2. Expressions for vibrational contribution to the SOECs and TOECs for MgO.

$C_{11}^{\text{vib}} = g_1F_1^2 + g_1F_2$
$C_{12}^{\text{vib}} = g_2F_1^2 + g_1F_5$
$C_{44}^{\text{vib}} = g_1F_5$
$C_{111}^{\text{vib}} = g_3F_1^3 + g_2F_2F_1 + g_1F_3$
$C_{112}^{\text{vib}} = g_1F_1^3 + g_2F_1(2F_5 + F_2) + g_1F_6$
$C_{123}^{\text{vib}} = g_3F_1^3 + 3g_2F_1F_5$
$C_{144}^{\text{vib}} = g_2F_1F_5$
$C_{166}^{\text{vib}} = g_2F_1F_5 + g_1F_6$
$C_{456}^{\text{vib}} = 0$

Table 3. Expression for g_n's.

$g_1 = g_0S$
$g_2 = g_0[(X/S_1) + S]/2$
$g_0 = \hbar\omega_0/8r_0^3$
$g_3 = g_0[(2X^2S/3S_1) + (X/S_1) + S]/48$
where $X = \hbar\omega_0/2KT$, $\omega_0 = (1/M^+ + 1/M^-)/qr_0F_0$, $S = \coth X, S_1 = \sinh^2 X$

has been introduced to emphasize that the static elastic constants correspond to 0 K. By adding the vibrational elastic constants to the static elastic constants, one may get SOECs and TOECs at any temperature for monovalent FCC crystals.

The equations used to calculate the static and vibrational elastic constants are presented in tables 1 and 2 respectively. These are shown as a combination of g_n's and F_n's which are evaluated conveniently by taking crystal's symmetry into account and the expressions for g_n and F_n are tabulated in tables 3 and 4.

3. Evaluation

The second- and third-order elastic constants for MgO are evaluated in the temperature range of 100–1000 K utilizing the concepts of the nearest-neighbour distance (r) and hardness parameter (q). These are calculated (taking $r = 2.014$ Å and $q = 0.345$ Å) at different temperatures (100 to 1000 K) using the theory described

Table 4. Expression for F_n's.

$F_0 = 1/[(q_0 - 2)(Q(r_0) + 2(q_0 - \sqrt{2})Q(r_0\sqrt{2})$
$F_1 = 2[(2 + 2q_0 - q_0^2)Q(r_0) + 2(\sqrt{2} + 2q_0 - \sqrt{2}q_0^2)Q(r_0\sqrt{2})]F_0$
$F_2 = 2(-6 - 6q_0 - q_0^2 + q_0^3)Q(r_0)F_0 + 2F_5$
$F_3 = 2(-30 - 30q_0 - 9q_0^2 + q_0^3 - q_0^4)Q(r_0)F_0 + 2F_6$
$F_4 = 2(-210 - 210q_0 - 75q_0^2 - 5q_0^3 + 4q_0^4 + q_0^5)Q(r_0)F_0 + 2F_7$
$F_5 = (-3\sqrt{2} - 6q_0 - \sqrt{2}q_0^2 + 2q_0^3)Q(r_0\sqrt{2})F_0$
$F_6 = [(15/\sqrt{2}) + 15q_0 - (9/\sqrt{2})q_0^2 - q_0^3 - \sqrt{2}q_0^4]Q(r_0\sqrt{2})F_0$
where $q_0 = r_0/q$

Table 5. Comparison data of SOECs and TOECs (in 10^{11} dyne/cm^2) at room temperature for MgO.

Comp.	C_{11}	C_{12}	C_{44}	C_{111}	C_{112}	C_{123}	C_{144}	C_{166}	C_{456}	Ref.
MgO	17.86	27.00	27.29	−217.41	−106.78	37.45	38.23	−107.48	38.08	Present
				−240.48	−86.20	30.06	30.84	−58.20	30.79	[9]
	29.40	9.24	15.58	−328.00	−31.80	24.20	31.80	−67.40	35.60	[10]
				−489.50	−95.00	−6.90	11.30	−65.90	14.70	[11]

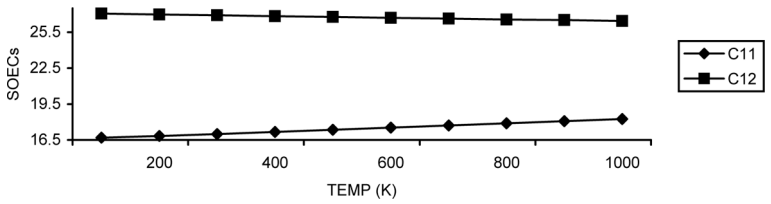

Figure 1. Temperature variation of C_{11} and C_{12} (in 10^{11} dyne/cm^2) for MgO.

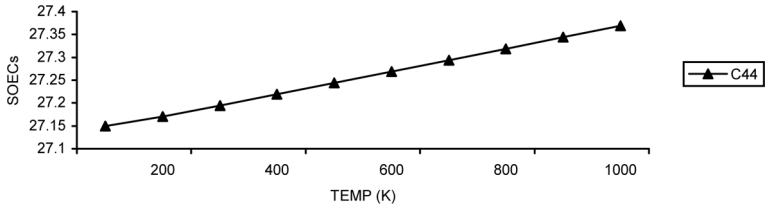

Figure 2. Temperature variation of C_{44} (in 10^{11} dyne/cm^2) for MgO.

in [6–8]. The variations of these constants with temperature are shown in figures 1–6. The theoretical [9,10] and experimental [11] data of others are presented in table 5 for comparison.

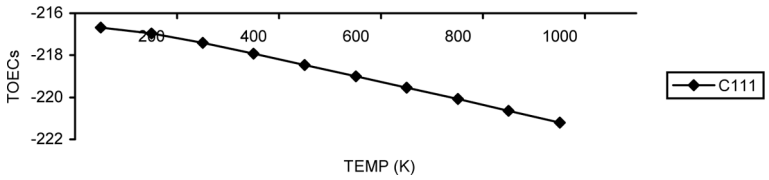

Figure 3. Temperature variation of C_{111} (in 10^{11} dyne/cm^2) for MgO.

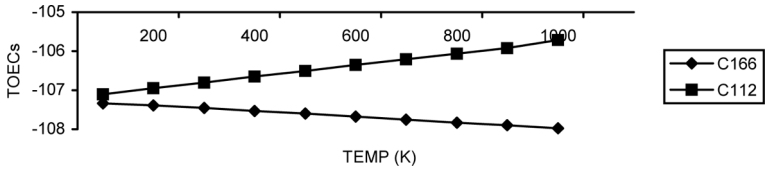

Figure 4. Temperature variation of C_{166} and C_{112} (in 10^{11} dyne/cm^2) for MgO.

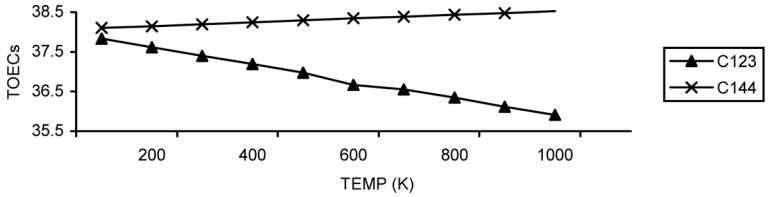

Figure 5. Temperature variation of C_{123} and C_{144} (in 10^{11} dyne/cm^2) for MgO.

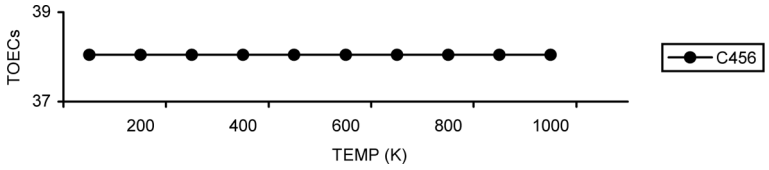

Figure 6. Temperature variation of C_{456} (in 10^{11} dyne/cm^2) for MgO.

4. Results and discussions

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 variations (figures 1–6) play 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. In general, workers [9,10] engaged in such studies have an impression that linear variation of elastic constant is true. The experimental study shows that elastic constants are not linear. In the present study, the vibrational contribution, which is anharmonic, is added to the static elastic constant. Although room temperature study is beneficial for common

use, in the present work we have tried our best to obtain the higher-order elastic constants from 100 to 1000 K for MgO. The cases discussed in the present study are in overall good agreement with theoretical and experimental results, which shows the validity of the present theory. The data obtained in the present investigation will be helpful to those workers who are engaged in such studies.

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