

Effect of Atoms in Different Shells on Ultrasonic Attenuation in Metallic Crystals in the Low Temperature Region

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Abstract. An attempt has been made to study the effect of neighbouring atoms on ultrasonic attenuation due to electron-phonon interaction in Ag, Cu, Au and Pb. Morse potential has been used to study the effect of atoms in different shells (from 6th shell to 12th shell). It is shown that atoms upto 6th shell are not sufficient to explain the ultrasonic attenuation. The effect of atoms in the 8th, 10th and even 12th shells are effecting the ultrasonic attenuation due to electron-phonon interaction. SOEC obtained by using Morse potential are in good agreement with the experimental values available. The variation of ultrasonic attenuation with temperature are as expected and are in good agreement with experimental values in Copper.

1. Introduction

Ultrasonic propagation parameter (velocity and absorption) studies have been made in solid, liquids and crystals. Number of book and review articles are available which gives in detail on all aspect of ultrasonic propagation parameters. Different causes of ultrasonic attenuation are attributed as due to nuclear magnetic resonance, thermal relaxation, ferroelectric and ferromagnetic losses, loss due to lattice defects, phonon-phonon interaction, electron-phonon interaction and thermoelastic loss. In pure metals^{1,2} at low temperature, ultrasonic attenuation is dominated by the interaction of the acoustical phonons and conduction electrons. This is because of the fact that at low temperature (below about 50 K), when the mean free path of the conduction electrons became large and is comparable to the wavelength of acoustic wave, the momentum given to it by vibrating lattice is not returned to the lattice at once and viscous loss occurs.

The aim of the present work is to study the effect of atoms in different shells on ultrasonic attenuation and how far the distant atoms present in different shells are affecting the ultrasonic attenuation due to electron-phonon (e-p) interaction. To the knowledge of the authors no such study has been made previously and no such result is available in literature. Ultrasonic attenuation due to e-p interaction studies have been made in some metallic crystals, making consideration of the atoms in 1st shell,

2nd shell upto 12th shell. Different potentials have been used from time to time, such as Coulomb and Born-Mayer³ adding the vibrational contribution^{4,5} for temperature variation of elastic constants, Morse potential^{6,7,8} and other potentials⁹ for determination of elastic constants. It is now well established that Morse potential is the most suitable potential for study of physical properties of metallic crystals. In the present study, Morse potential has been used to evaluate the Second Order Elastic Constants (SOEC) for interaction of atoms of different shells, then the temperature dependence of elastic constants and finally the ultrasonic attenuation due to e-p interaction.

2. Theory

(1) SOEC : According to Morse potential the interaction energy $\phi(r_{ij})$ between two atoms i and j separated by a distance r_{ij} is given by :

$$(1) \quad \phi(r_{ij}) = D [e^{-2\alpha(r_{ij}-r_0)} - 2e^{-\alpha(r_{ij}-r_0)}]$$

r_i is the distance of the i^{th} atoms from origin, D is the dissociation energy, r_0 is the separation of atoms for minimum potential and α is a parameter related to hardness of potential. The parameters D , α and r_0 used in the Morse potential can be obtained with known values of the bulk modulus, the cohesive energy and equilibrium semi-lattice constants. The expression for SOEC is as follows :

$$(2) \quad C_{11} = \frac{2Da^2\alpha^2\beta^2}{V_0} \sum \frac{m_j e^{-2\alpha a m_j}}{M_j} - \frac{2Da^2\alpha^2\beta}{V_0} \sum \frac{m_j^4 e^{-\alpha a M_j}}{M_j^2} \\ + \frac{Da\alpha\beta^2}{V_0} \sum \frac{m_j^4 e^{-2\alpha a M_j}}{M_j^3} - \frac{Da\alpha\beta}{V_0} \sum \frac{m_j^4 e^{-\alpha a M_j}}{M_j^3}$$

where $\beta = e^{\alpha r_0}$

$$r_j = [m_j^2 + n_j^2 + 1]^{\frac{1}{2}} a = M_j a$$

m_j , n_j and l_j are the position coordinates of any atom in terms of half the lattice parameter. The expression for C_{12} is obtained by replacing m_j^4 by $m_j^2 n_j^2$ in equation (2) above.

(2) **Temperature dependence of SOEC** : Applying the anharmonic theory of lattice dynamics developed by Hahn et al⁴ and Ludwig et al⁵, SOEC at different temperatures can be obtained. The vibrational energy of the crystal changes with temperature, therefore adding the vibrational energy contribution to elastic constants, we obtain :

$$(3) \quad C_{ij} = C_{ij}^0 + C_{ij}^{vib}$$

and thus the constants at the required temperature are found. The C_{ij}^{vib} are :

$$(4a) \quad C_{11}^{vib} = f^{1,1} G_1^2 + f^2 G_2$$

and

$$(4b) \quad C_{44}^{vib} = f^2 G_{1,1}$$

The f^n , s and G_n , s are given by :

$$(5) \quad f^2 = (\hbar \omega_0 / 8 r_0^3) \coth X$$

$$f^{1,1} = -(\hbar \omega_0 / 96 r_0^3) (X / \sinh^2 X) + \coth X$$

$$G_1 = 2(2 + 2q_0 - 2q_0^2) Q(r_0) + 2(\sqrt{2} + 2q_0 - \sqrt{2} q_0^2) Q(r_0 \sqrt{2}) / H$$

$$G_2 = 2(6 - 6q_0 - q_0^2 + q_0^3) Q(r_0) / H + 2G_{1,1}$$

and

$$G_{1,1} = (-3\sqrt{2} - 6q_0 - \sqrt{2} q_0^2 + 2q_0^3) Q(r_0 \sqrt{2}) / H$$

with

$$H = ((q_0 - 2) Q(r_0) + 2(q_0 - \sqrt{2}) Q(r_0 \sqrt{2})) q_0 = (r_0 / q)$$

$$X = (\hbar \omega_0 / 2kT), \omega_0^2 = (2/q r_0 H M_+)$$

and

$$A = 1.74756 (e^2 / r_0^2) q (6 \exp(-r_0 / q) + 12\sqrt{2} \exp(-r_0 \sqrt{2} / q))^{-1}$$

where r_0 is the nearest-neighbour distance, q is the hardness parameter or Born

repulsive parameter, $\hbar$ is Plank's constant divided by 2π , and M_+ is the mass of a single ion.

(3) Ultrasonic attenuation due to e-p interaction : The viscosity of electron gas is given by :

$$\eta_e = N m e \bar{l} \bar{v}$$

where N is the electron density, m is the electron's mass, $\bar{l}$ is the mean free path and $\bar{v}$ is the mean velocity of the electron. From the theory of electron gas¹⁰ using values of $\bar{l}$ and $\bar{v}$, η_e becomes :

$$(6) \quad \eta_e = (9 \times 10_{11} \times \hbar^2 (3\pi^3 N)^{2/3}) / 5e^2 R$$

where R is the electrical resistivity

In pure metals at low temperature the viscous loss occurs as the coupling of the conduction electron with a propagating ultrasonic wave causes dissipation of energy with propagating ultrasonic wave.

In equilibrium the Fermi surface of electron gas is spherical in nature. The sphere transforms into an ellipsoid¹¹ with the minor axis along the compressed part when acoustic wave propagates through it. The attenuation caused by the loss due to compressional and shear viscosities of the lattice¹² in terms of η_e given above is given by :

$$(7) \quad (\alpha_f^2)_l = 2\pi^2 \left(\frac{4\eta_e}{3} + \chi_e \right) / dv_l^3$$

$$(8) \quad (\alpha_f^2)_s = 2\pi^2 \eta_e / dv_s^3$$

where d is the density, f is the ultrasonic frequency, η_e is the transverse viscosity, χ_e is the compressional viscosity, being zero as measured by Bommel¹³

3. Evaluation

D , α and r_0 are obtained as mentioned earlier. A small computer program has been developed to obtain the consistent values of D , α and r_0 so as to satisfy the bulk modulus, cohesive energy and lattice parameter. These values are then used to evaluate SOEC and then the temperature dependance of elastic constants for Ag, Cu, Au and Pb from 10 K to 70 K. The SOEC at 300 K are also obtained so as to compare the same with experimental values available at 300 K. The SOEC along with other physical

constants (density, resistivity, etc.) are used to evaluate $(\alpha_f^2)_1$ and $(\alpha_f^2)_s$ using equations (7) and (8) as a function of temperature.

4. Results and Discussion

The SOEC's have been evaluated for the metallic crystals Ag, Cu, Au and Pb as a function of temperature from 10 K to 70 K and also for shell no. 6 (86 atoms), 8 (140 atoms), 10 (176 atoms) and 12 (224 atoms). The experimental values of SOEC's [14] available at 300 K are compared with the values evaluated by us. The results are in good agreement (Table-1). The ultrasonic attenuation due to e-p interaction for upto 6, 8, 10 and 12th shell are presented in Table-2. For Copper the experimental values are available [15] and are plotted along with the values obtained by us for interaction of atoms upto 12th shell which compares well (Fig. -1) showing the validity of the work. For other substances the values of (α_f^2) at low temperature are very high and falls sharply as the temperature increases.

From table II it is clear that at any temperature the atoms upto 6th shell are not sufficient and more shells are to be taken as still the 8th or the 10th shell are contributing to ultrasonic attenuation. For Au say, considering interaction of atoms upto 6th shell due to e-p interaction, the ultrasonic attenuation at 15 K is 9.08 (all in units as given in the table) where as for interaction of atoms upto 10th shell is 9.37, i.e. an increase of 4 percent. However between 10th shell and 12th shell absorptions are 9.37 and 9.39, showing an increase of only 0.2 percent i.e. very small. Same argument will be true for all the cases. Consideration has been made at 15 K where the fall is very sharp. However at 50 K the value is 0.17 from 6th shell to 12th shell. Similar argument can be given for shear waves also.

TABLE 1. SOEC for Ag, Cu, Au and Pb for interaction of atoms upto 6, 8, 10 and 12 shells at 300 K (in units of 10_{11} dyn/cm²).

Substance	C ₁₁				C ₁₂				C ₄₄			
	6	8	10	12	6	8	10	12	6	8	10	12
Ag(1)	14.84	14.12	13.91	13.79	8.59	8.27	8.22	8.18	9.05	8.73	8.69	8.65
Expt. #	12.22				9.07				4.54			
Cu(2)	21.69	19.45	18.65	18.21	11.63	10.66	10.4	10.3	12.6	11.6	11.4	11.2
Expt. #	16.61				9.07				7.56			
Au(3)	26.62	26.18	26.08	26.03	13.30	13.11	13.08	13.07	14.82	14.62	14.60	14.59
Expt. #	19.29				16.38				16.38			
Pb(4)	6.17	5.95	5.89	5.86	3.73	3.63	3.62	3.61	3.85	3.75	3.74	3.72

Ref. 15.

TABLE 2. Ultrasonic attenuation due to e-p interaction for interaction of atoms from 6th to 12th shell in the low temperature region (all in $10^{-17} \text{ sec}^2 \text{ cm}^{-1}$) for Ag(1), Au(3) and Pb(4).

Ag(1)	(α/f^2)							
	6 th shell		8 th shell		10 th shell		12 th shell	
	long.	sh.	long.	sh.	long.	sh.	long.	sh.
10	317	461	343	487	351	491	355	495
15	56.6	84.0	62.4	88.6	63.9	89.3	64.7	89.9
20	21.1	30.8	22.9	32.5	23.4	32.7	23.7	32.9
25	6.3	9.2	6.9	9.7	7.0	9.8	7.1	9.9
30	3.2	4.6	3.4	4.9	3.5	4.9	3.6	4.9
50	.6	.8	.6	.9	.6	.9	.6	.9
70	.4	.5	.4	.5	.4	.5	.4	.5

Au(3)	(α/f^2)							
	6 th shell		8 th shell		10 th shell		12 th shell	
	long.	sh.	long.	sh.	long.	sh.	long.	sh.
10	55.95	98.70	57.42	100.7	57.76	100.9	57.92	101.1
15	9.08	16.01	9.31	16.32	9.37	16.36	9.39	16.39
20	2.69	4.74	2.76	4.83	2.77	4.84	2.78	4.85
25	1.24	2.19	1.28	2.24	1.28	2.24	1.29	2.25
30	.67	1.19	.69	1.21	.69	1.21	.70	1.21
50	.17	.30	.17	.30	.17	.30	.17	.30
70	.09	.16	.09	.16	.09	.16	.09	.16

Pb (4)	(α/f^2)							
	6 th shell		8 th shell		10 th shell		12 th shell	
	long.	sh.	long.	sh.	long.	sh.	long.	sh.
20	.42	.62	.44	.64	.45	.65	.45	.65
50	.09	.13	.09	.13	.09	.13	.09	.13
80	.05	.07	.05	.07	.05	.07	.05	.07

Conclusions

(i) The interaction of atoms of different shells with the central atom depend on the distance of the atom from the central atom and also on the number of atoms on that particular shell.

(ii) First few shells (say upto 6th) are not sufficient to account for ultrasonic attenuation due to e-p interaction.

(iii) The atoms in the 8th, 10th and to a certain extent 12th shell are contributing to ultrasonic attenuation (Table II).

(iv) The SOEC's evaluated by us are in good agreement with the experimental values.

(v) The ultrasonic attenuation due to e-p interaction in copper as evaluated by us are of the same nature as compared to experimental values and in some region of temperature the values are in good agreement (Fig. 1).

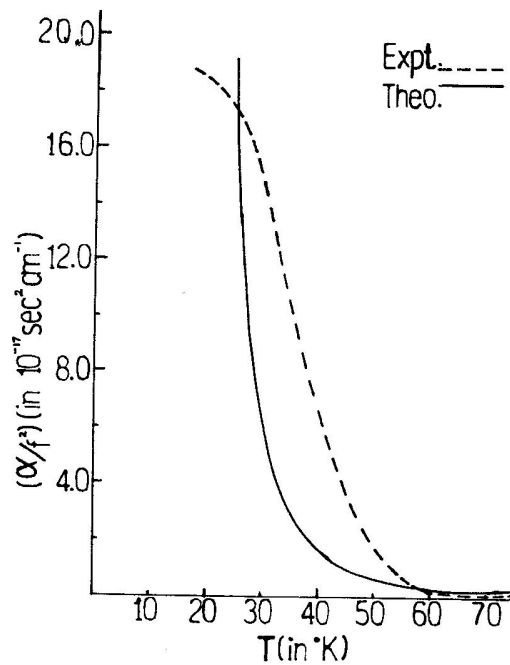


Fig. 1. Variation of Ultrasonic attenuation with temperature due to electron-photon interaction for Copper.

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