THREE-BODY FORCES AND LATTICE DYNAMICS OF ALUMINIUM

CHOPRA K. K.,[1] EL KECHAI A.,[1], Tizi-Ouzou

Modern theories based on pseudopotentials have supported the existence of three-body forces in solids. A phenomenological model incorporating these forces is used to calculate the phonon frequencies and Debye-Waller factor of aluminium. The results are found to be in good agreement with experimentally measured values.

I. INTRODUCTION

Sophisticated pseudopotential models [1—3], for the lattice dynamics of metals, although founded on a rigorous basis and very useful from a conceptual point of view, are often extremely intricate with regard to their practical use. Nevertheless these studies have provided vital information on the nature of interactions in a metal. In the present paper we have adopted a phenomenological approach [4] to incorporate some of the results of pseudopotential theories to develop a simple model for the lattice dynamics of metals and applied it to calculate the phonon frequencies and the Debye—Waller factor of aluminium.

Pseudopotential studies [5] show that the energy of a metal can be regarded as made up of three parts: (i) E_c, due to a central core-core interaction; (ii) E_r, due to the overlap of electron clouds: and (iii) E_v, the volume energy which represents the overall effects of the presence of conduction electrons and their interaction with the core. Further, Brovman et al. [6—8] and Pethick [9], also using pseudopotential methods, have shown that three-body forces play an important role in solids and are essential to obtain the required equality of static and dynamic elastic constants.

In the model presented here we have considered three types of interactions corresponding to the energies E_c, E_r and E_v and have introduced three-body forces in the calculations relating to the last two.

[1] Département de Physique, Institut National d'Enseignement Supérieur en Electrotechnique. Université de Tizi-Ouzou, TIZI—OUZOU, Algeria.

II. THEORY

II. 1. Dynamical matrix:

Following the usual theory, the phonon frequencies corresponding to a wave-vector $\boldsymbol{k}$ are given by the secular equation:

$$|D(\boldsymbol{k}) - 4\pi^2 M\nu^2| = 0$$

where M is the ionic mass and $D(\boldsymbol{k})$ the dynamical matrix of order 3×3 for a cubic metal. The dynamical matrix elements D_{ij} are, in the present case, given by:

$$D_{ij} = D_{ij}^c + D_{ij}^r + D_{ij}^v \tag{1}$$

where the superscripts c, r, and v refer respectively to the energies E_c, E_r and E_v; and i, j denote the cartesian directions x, y and z.

The energy E_c is due to core-core interactions. Pseudopotential studies of Pick et al. [10] and Resolt et al. [11] have shown that interactions among cores are essentially central. We have also, therefore, supposed this interaction to be purely central and considered it effective in the nearest neighbours only. This is then calculated in the standard manner [12]. The corresponding matrix elements are found to be

$$\begin{aligned} D_{ii}^c &= 2\alpha[2 - C_i(C_j + C_k)] \\ D_{ij}^c &= 2\alpha S_i S_j \end{aligned} \tag{2}$$

where $S_i = \sin(\pi a k_i)$; $C_i = \cos(\pi a k_i)$; "a" is the lattice constant and k_i the ith component of the phonon wave-vector.

The energy E_r is due to the overlap of electron clouds. Such an overlap is known to lead to a break-up of the sphericity of the charge distribution resulting in non-central interactions [13, 14] among electron clouds. We have considered these non-central interactions to be of a three-body character which can be phenomenologically described by the CGW type angular forces [15]. We have modified the original CGW forces to include a third type of triangles on the lines of Awasthi et al. [16]. These additional triangles are equilateral and are formed by an atom and two of its first neighbours situated on two mutually perpendicular planes. The corresponding matrix elements obtained are the following:

$$D_i^r = \frac{16(\gamma_1+\gamma_3)}{a^2}[2 - C_i(C_j + C_k)] + \frac{16\gamma_2}{a^2}[3 - (C_iC_j + C_jC_k + C_kC_i)] + \\ + \frac{8\gamma_3}{a^2}[2S_i^2 - S_j^2 - S_k^2] \tag{3}$$

$$D_{ij}^r = 8\left[\frac{\gamma_2}{a^2} - \frac{2\gamma_3}{a^2}\right] S_iS_j$$

where γ_1, γ_2, and γ_3 are the three angular force constants corresponding to the angles of 45°, 60° and 90°, respectively, of the triangles considered.

The volume energy E_v is linked to many-body interactions. We follow the idea of Sarkar et al. [17] who have developed this energy in terms of the volume strain Δ around the equilibrium volume V_0. The term of interest in the expression is $1/2\,K_eV_0\Delta^2$, where K_e is the bulk modulus of the electron gas. Interpreting Δ as a local strain one can express it in terms of the coordinates of the ions of the region. The region considered for the purpose is again confined to the nearest neighbours only. As the term in Δ is quadratic, the sum over the twelve nearest neighbours compresses the many-body interaction into an effective three-body interaction. The corresponding matrix elements are:

$$\begin{aligned} D_{ii}^v &= \frac{K_e a}{4} S_i^2(C_j + C_k)^2 \\ D_{ij}^v &= \frac{K_e a}{4} S_iS_j(C_i + C_k)(C_j + C_k). \end{aligned} \tag{4}$$

The sum of relations (2), (3) and (4) gives the total matrix elements D_{ii} and D_{ij} of the dynamical matrix.

II. 2 Debye—Waller factor

According to James [18], the exponent $2W$ of the Debye—Waller factor at a temperature T is defined by:

$$2W = \frac{\hbar}{MN}\sum_{\boldsymbol{kj}} \frac{(\Delta\boldsymbol{K}.\boldsymbol{e}_{\boldsymbol{kj}})^2}{\omega_{\boldsymbol{kj}}}\left[\langle n_{\boldsymbol{kj}}\rangle + \frac{1}{2}\right] \tag{5}$$

Here M is the mass of the atom, N is the total number of unit cells in the crystal, $\Delta\boldsymbol{K}$ is the difference between the initial and the final wave-vectors of X-rays, $\omega_{\boldsymbol{kj}}$ is the frequency of the phonon wave-vector $\boldsymbol{k}$ and polarization j, $\boldsymbol{e}_{\boldsymbol{kj}}$ is the polarization vector of the $\boldsymbol{kj}$ lattice mode and $\langle n_{\boldsymbol{kj}}\rangle$ is the average occupation number of phonons in the mode $\boldsymbol{kj}$. For monoatomic cubic crystals, the factor $(\Delta\boldsymbol{K}.\boldsymbol{e}_{\boldsymbol{kj}})$ can be replaced by its average value so that equation (5) takes the form:

$$2W = \frac{8\pi^2\hbar}{3MN}\left(\frac{2\sin^2\Theta}{\lambda^2}\right)\sum_{\boldsymbol{kj}}\frac{I}{\omega_{\boldsymbol{kj}}}\coth\left(\frac{\hbar\omega_{\boldsymbol{kj}}}{2K_BT}\right) \tag{6}$$

where K_B is the Boltzman constant, Θ the glancing angle of incidence and λ the wave-length of the incident radiation.

III. METHOD OF CALCULATION

The matrix elements given by (2) to (4) contain five parameters — α for the central forces, γ_1, γ_2 and γ_3 for the three-body angular forces and K_e for the volume forces. These have been determined from three elastic constants and two zone boundary frequencies. The input data and calculated parameters are given in Table 1.

Table 1

Input data and calculated parameters

Input Data	Model Parameters (N/m)
Elastic constants: (10^{10} Nm^2)	
C_{11}: 10.678	α: 25.310
C_{12}: 6.074	γ_1/a^2: −0.114
C_{44}: 2.821	γ_2/a^2: −0.811
Lattice constant: (10^{-10} m) 'a': 4.05	γ_3/a^2: 0.323
	$K_e a$: −2.932
Atomic mass: (amu) 'M': 26.9815	
Zone boundary frequencies (THz)	
$\nu_L(100)$: 9.63	
$\nu_T(100)$: 6.63	

For the calculation of the Debye—Waller factor the first Brillouin zone was divided into 8000 uniformly distributed points. The frequencies were calculated for the 262 nonequivalent points in the 1/48 irreducible part of the first Brillouin zone assigning proper statistical weight to each such point. The contribution of the zone centre for the zero wave-vector has been calculated as suggested by Baron et al. [19]. The results are compared with the experimental data in terms of the temperature parameter Y given by

$$Y = \log_{10} \exp \frac{\lambda^2}{\sin^2 \Theta} (2W_0 - 2W) \qquad (7)$$

where $2W_0$ is the Debye—Waller factor at the room temperature ($T_0 = 293°$). It is readily seen that Y depends only on the frequency spectrum and is independent of Θ and λ.

IV. RESULTS AND DISCUSSION

The calculated phonon dispersion curves in the principal symmetry directions are shown in Fig. 1 along with the experimental points of Stedman et al.

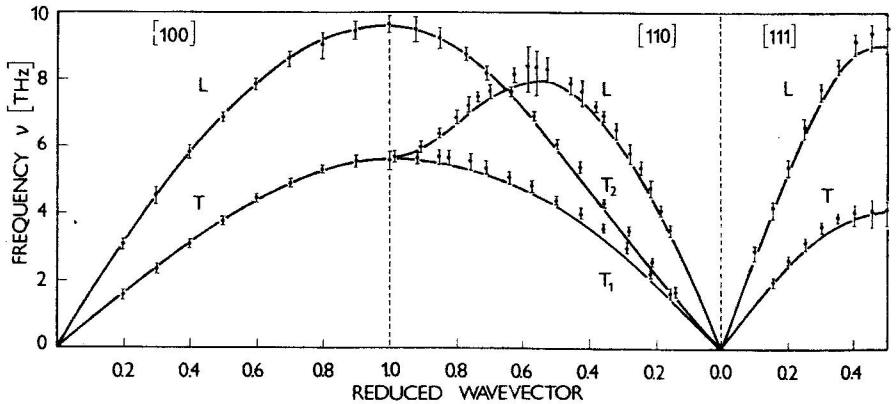

Fig. 1.: Dispersion curves of aluminium in the principal symmetry directions. Experimental points along with the corresponding uncertainties are of Stedman et al., Ref. [20].

[20]. A glance at the dispersion curves shows the good agreement obtained with the experimental values. The discrepancies are within 3%. The usual frequency distribution function $G(\nu)$ is shown in Fig. 2.

For the Debye—Waller factor, the calculated variation of the parameter Y with temperature is depicted in Fig. 3 along with experimental points (21—24).

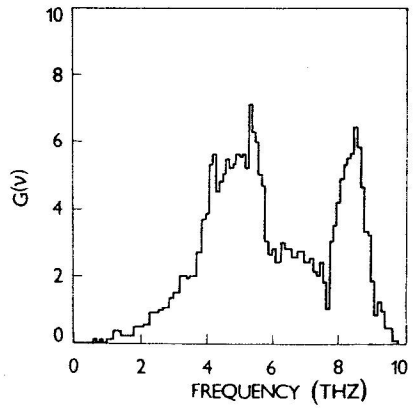

Fig. 2.: Histogram showing the frequency distribution function of Al.

Very good agreement is obtained up to 600 °K. Beyond this temperature the anharmonic effects and thermal expansion are no more negligible and are responsible for the observed discrepancies. At high temperatures, the exponent $2W$ depends on the frequency spectrum as $\sum_{kj} \nu_{kj}^{-2}$; the low frequency peak of Fig. 2 thus contributes much more at high temperatures than the high-frequency peak. A knowledge of the Gruneisen parameter corresponding to this low-frequency peak and of its variation with temperature could possibly enable one to estimate the resulting influence of thermnal expansion on the X-ray diffraction intensities and help in accounting for the observed discrepancies at high temperatures.

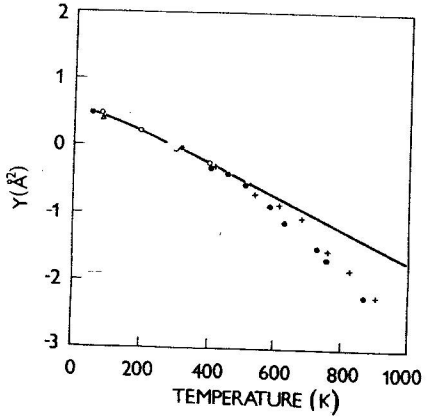

Fig. 3. Temperature dependence of the parameter Y. Experimental points are due to Chipman (●), Flinn et. al. (○), James et al. (△) and Owen et al. (+).

Nevertheless, our model gives a fairly close agreement with the experimental results and justifies the importance of various interactions considered. In particular, the many-body volume energy appears to be effectively well represented by the resulting three-body forces.

V. ACKNOWLEDGEMENTS

The authors express their gratitude to Mr. S. Guermah, Director of the Institut National d'Enseignement Supérieur en Electrotechnique de Tizi-Ouzou, for the generous facilities provided during the course of this work.

REFERENCES

[1] Oli, B. A., Animalu, A. O. E.: Phys. Rev. *B* 13 (1976), 2398.
[2] Upadhaya, J. C., Dagens, L.: J. Phys. *F8* (1978), L21.
[3] Singh, J., Singh, N., Prakash, S.: Phys. Rev. B *18* (1978), 2954.
[4] Chopra, K. K., Nait-Laziz, H.: *2nd Int. Sem. on Material Sciences.* Université de Constantine (1985).
[5] Harrison, W. A.: *Pseudopotentials in the Theory of Metals.* Benjamine, New York (1966).
[6] Brovman, E. G., Kagan, Y., Kholas, A.: Sov. Phys. JETP. *30* (1970), 883.
[7] Brovman, E. G., Kagan, Y.: Ibid, *30* (1970), 721.
[8] Brovman, E. G., Kagan, Y., Kholas, A.: Ibid, *34* (1972), 394.
[9] Pethick, C. J.: Phys. Rev. B *2* (1970), 1789.
[10] Pick, R., Sarma, G.: Phys. Rev. *135* (1964), 1363.
[11] Resolt, J. M., Taylor, R.: Phys. Rev B *11* (1975), 2717.
[12] De Launay, J.: *Solid State Physics, 2.* Academic Press, N. Y. (1956).
[13] Leigh, H. S.: Proc. Phys. Soc. London *A71* (1958), 33.
[14] Simizu, M., Niu, H.: J. Phys. Soc. Japan *27* (1962), 577.
[15] Clark, B. C., Gazis, D. C., Wallis, R. F.: Phys. Rev. *134* (1964), 1486.
[16] Awasthi, K. N., Kushwaha, S. S.: Nuovo Cimento *B54* (1979), 485.
[17] Sarkar, S. K., Das, S. K., Roy, D., Sengupta, S.: Phys. Stat. Soli. (b) *83* (1977), 615.
[18] James, R. W.: *The Optical Principles of Diffraction of X-rays.* G. Bell and Sons, London (1964)
[19] Baron, H. W. T., Smith, T.: J. Phys. Chem. Sol. *27* (1966), 1951.
[20] Stedman, R., Nilsson, G.: Phys. Rev. *145* (1966), 492.
[21] Owen, E. A., Williams, R. W.: Proc. Roy. Soc. *A188* (1947), 509.
[22] Chipman, D. R.: J. Appl. Phys. *31* (1960), 2012.
[23] James, R. W., Brindlay, G. W., Wood, R. G.: Proc. Roy. Soc. *A125* (1926), 401.
[24] Flinn, P. A., McManus, M.: Phys. Rev. *132* (1963), 2458.

Received July 18th, 1986

ТРЕХЧАСТИЧНЫЕ СИЛЫ И ДИНАМИКА РЕШЕТКИ АЛЮМИНИЯ

Современные теории, основанные на псевдопотенциалах, предполагают существование в твердых телах техчастичных сил. В работе использована феноменологическая модель, учитывающая эти силы, которая позволяет рассчитать фононные частоты и фактор Дебая—Валлера в алюминии. Обнаружено, что теоретические результаты находятся в хорошем согласии с экспериментальными значениями соответствующих величин.