

Elastic and lattice dynamical behaviour of *fcc* aluminium

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Received 27 October 1995, accepted 30 January 1996

Abstract : The extended generalised exponential potential (EGEP) is employed to determine second- and third-order elastic constants of *fcc* aluminium along with its pressure derivatives of the second-order elastic constants. The cohesive energy and phonon-spectra have also been computed. The computed results show promising agreement with the experimental findings.

Keywords : Extended generalised exponential potential (EGEP), cohesive energy, phonon spectra

PACS Nos. : 63 20 Dj, 62 20 Dc

1. Introduction

Aluminium is a trivalent *p*-block metal having a much larger value for screening of conduction electrons and its dynamical behaviour has been studied earlier by many workers [1]. The lattice dynamics of aluminium have also been studied by Soma *et al* [2] using the local Heine-Abarenkov model potential with volume dependent parameters. The study due to Gupta [3] combines the short range and long range forces. The model potential due to Aschroft [4] employed by Das *et al* [5] ignores the adequate inclusion of three-body forces. Wang and Overhauser [6] have also computed phonon frequencies of aluminium using their dynamical pseudopotential theory, which involves unnecessary multiplication of interactions. A nine-parameter model has been proposed by Singh [7] to study the lattice dynamics of aluminium. Recently, Thakur and Singh [8] have employed a

phenomenological model incorporating the three-body unpaired interactions to study the phonon dispersion in aluminium.

A further review of literature reveals that the interacting potentials can be represented either as inverse or exponential function of separation or the combination of both the functions. A large number of microscopic studies [9] make use of the model potential exhibiting inverse separation dependence. Model potentials given by some authors [10] combine the inverse separation dependence with exponential dependence. The Born-Mayer function, widely used [11] to represent the repulsive interactions, is essentially an exponential function. Wang and Overhauser [6] have shown that the core and shell charge cluster assume the exponential forms. The charge density pertaining to d -shell exhibits [12] the exponential form. The couplings among d -shells and those among s - and d -shell are expressed [13] as inverse function of some power of separation. The dielectric functions [14] which either augment or subdue the microscopic interactions also implicate the direct or inverse and exponential separation dependence. These forms of interactions claim limited and selective success in explaining the static and dynamic properties of solids.

The exponential form of potential given by Morse has widely been used [15] to study the various properties of solids. Some other [16] macroscopic studies have stated the interactions as the combination of inverse and exponential separation dependent functions.

Present communication derives an empirical potential, which is an extension of the generalised exponential potential, known as extended generalised exponential potential (EGEP) and explains almost all the characteristic features of the interatomic interactions such as

- (i) The electronic exchange and correlation effects, which introduce the substantial change [17] into the width and depth of the potential, have been accounted for in an alternative form through a parameter m and therefore, properly substitute for dielectric screening functions.
- (ii) The role of the three-body forces, such as volume forces, has effectively been expressed in an indirect manner through a parameter n , because this parameter affects [18] the position as well as the depth of the potential minima (Figure 1).
- (iii) It accounts effectively for the characteristic feature of steep rise of Coulombic repulsion at small separations.
- (iv) The potential is self-convergent and therefore requires no exponential damping factor.

The potential has earlier explained [19] successfully the elastic and dynamical behaviour of *fcc* metals. The present paper aims to investigate the elastic and dynamical behaviour of *fcc* aluminium.

2. Theory

2.1. Extended generalised exponential potential :

The attractive as well as the repulsive components of the generalised exponential potential have been extended for representing their true and realistic nature. Extended generalised form of exponential potential (EGEP) so developed, assumes the form

$$\Phi_m(r_{ij}) = D/(m-1) \left[e^{-m\alpha(r_{ij}-r_0)} / (\alpha r_{ij})^n - m(\alpha r_{ij})^n e^{-\alpha(r_{ij}-r_0)} \right]. \quad (1)$$

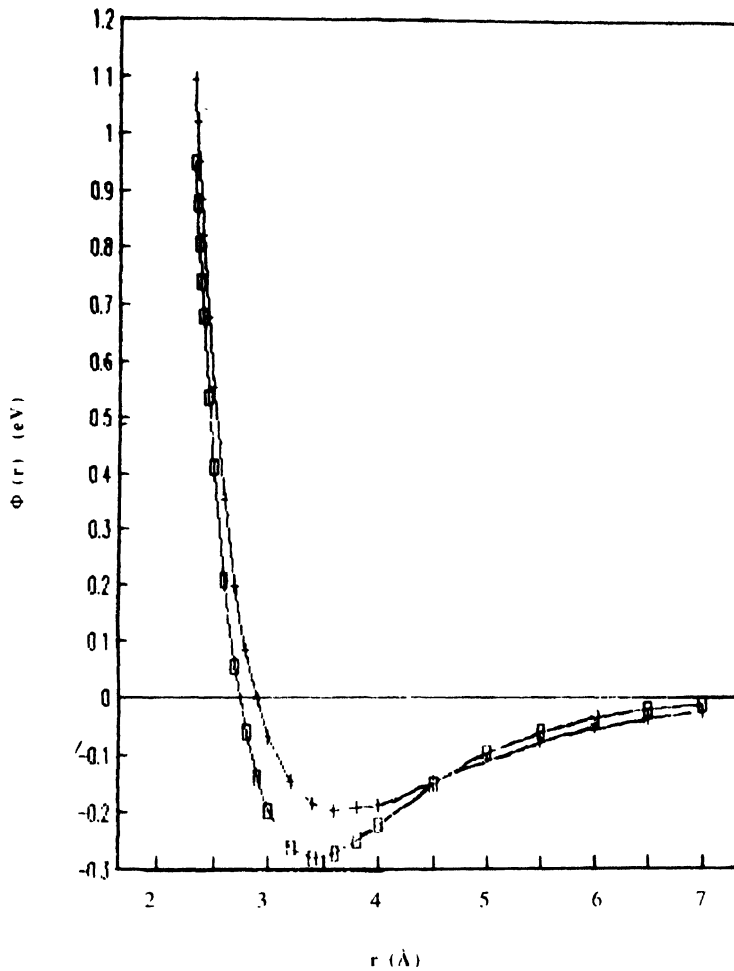


Figure 1. Variation of the potential function of Al with r for n = 0.5 (□) and 2.0 (+)

The average interaction (cohesive energy) energy per atom within the framework of EGEP is

$$\Phi_m(r_j) = D/2(m-1) \sum \left[e^{-m\alpha(r_i-r_0)} / (\alpha r_i)^n - m(\alpha r_i)^n e^{-\alpha(r_i-r_0)} \right]. \quad (2)$$

where D is the dissociation energy, α the hardness parameter and r_o the equilibrium separation parameter and r_j is the distance of the j -th atom from the origin given by

$$r_j = (l_1^2 + l_2^2 + l_3^2)^{1/2} a_o = L_j a_o. \quad (3)$$

Eq. (2) can be put in the form to represent the cohesive energy at equilibrium semi-lattice constant (a_o) as under

$$\Phi(a_o) = D/2(m-1) \left[\beta^m (\alpha a_o)^{-n} \sum_{l_1^2 l_2^2 l_3^2} e^{-m\alpha a_o l_j} L_j^{-n} - m\beta (\alpha a_o)^n \sum_{l_1^2 l_2^2 l_3^2} e^{-\alpha a_o l_j} L_j^n \right] \quad (4)$$

where $\beta = \exp(\alpha r_o)$. (5)

The three defining parameters (α , r_o and D) of the potential require for their evaluation, the precisely determined input data of equilibrium semi-lattice constant (a_o) and bulk modulus (B) of the metal only. For evaluating the three parameters α , r_o and D of the potential function, the condition

$$\sum_{l_1^2 l_2^2 l_3^2} [l_1^2 d\Phi(r) / dr^2] = 0 \quad (6)$$

for the equilibrium of the crystal in the absence of external forces is employed which gives

$$\beta^{m-1} = \frac{m(\alpha a_o)^n (U-V)}{(\alpha a_o)^{-n} (X+Y)}, \quad (7)$$

where

$$U = \left[\alpha \sum_{l_1^2 l_2^2 l_3^2} l_1^2 L_j^{n-1} \exp(-\alpha a_o L_j) \right],$$

$$V = \left[(n/a_o) \sum_{l_1^2 l_2^2 l_3^2} l_1^2 L_j^{n-2} \exp(-\alpha a_o L_j) \right],$$

$$X = \left[m\alpha \sum_{l_1^2 l_2^2 l_3^2} l_1^2 L_j^{-(n+1)} \exp(-m\alpha a_o L_j) \right],$$

$$Y = \left[(n/a_o) \sum_{l_1^2 l_2^2 l_3^2} l_1^2 L_j^{-(n+2)} \exp(-m\alpha a_o L_j) \right].$$

The bulk modulus can be expressed as

$$B = (r^2 / 9V) (d^2\Phi / dr^2)_{r=r_o}. \quad (8)$$

The parameter D can be evaluated through the expression for the bulk modulus following the condition given by eq (6) for stress-free lattice. The following expression for D is obtained

$$D = 18VB(m-1)/(P-Q), \quad (9)$$

where

$$P = \left[\beta^m (\alpha a_o)^{-n} \left\{ (m\alpha a_o)^2 \sum_{l_1 l_2 l_3} L_j^{-(n-2)} \exp(-m\alpha a_o L_j) \right. \right. \\ \left. \left. + 2n(m\alpha a_o) \sum_{l_1 l_2 l_3} L_j^{-(n-1)} \exp(-m\alpha a_o L_j) \right. \right. \\ \left. \left. + n(n+1) \sum_{l_1 l_2 l_3} L_j^{-n} \exp(-m\alpha a_o L_j) \right\} \right],$$

$$Q = \left[m\beta (\alpha a_o)^n \left\{ (\alpha a_o)^2 \sum_{l_1 l_2 l_3} L_j^{(n+2)} \exp(-\alpha a_o L_j) \right. \right. \\ \left. \left. + 2n(\alpha a_o) \sum_{l_1 l_2 l_3} L_j^{(n+1)} \exp(-\alpha a_o L_j) + n(n-1) \sum_{l_1 l_2 l_3} L_j^n \exp(-\alpha a_o L_j) \right\} \right].$$

2.2. The second-order and third order elastic constants :

The following expressions for the second-order elastic constants (SOEC) and the third-order elastic constants (TOEC) with present interatomic interactions are used

$$C_{11} = (n' a_o^4 / 2V) \sum_{l_1 l_2 l_3} l_1^4 d^2 \Phi(r) / (dr^2)^2, \quad (10)$$

$$C_{111} = (n' a_o^6 / V) \sum_{l_1 l_2 l_3} l_1^6 d^3 \Phi(r) / (dr^2)^3, \quad (11)$$

where n' is the number of atoms per unit cell (4 for fcc and 2 for bcc) and V represents the atomic volume.

The eq. (10) transforms to represent C_{12} when l_1^4 in the said equation is replaced by $l_1^2 l_2^2$. Similarly, eq. (11) transforms to represent C_{112} ($= C_{166}$) and C_{123} ($= C_{456} = C_{144}$) when l_1^6 in the said equation is replaced by $l_1^4 l_2^2$ and $l_1^2 l_2^2 l_3^2$ respectively.

In order to calculate SOEC C_{44} , we make use of the following relation existing between C_{44} and the first derivative (α_1) which describe the unpaired ionic interaction and the second derivative (β_1) of the present potential viz.,

where α_1, β_1 are the force constants for the first neighbour and a is the lattice parameter for the metal used as an input data.

The Cauchy's discrepancy in SOEC is attributed partly due to the first derivatives of the central pair-wise potential and partly due to the electron bulk modulus (K_e) viz.,

$$a (C_{12} - C_{44}) = a K_e - 8(\alpha_1 + \alpha_2), \quad (13)$$

where α_2 is the force constant for the second neighbour and the value of bulk modulus of the electron gas for Al [8] has been taken to be 0.052×10^{11} N/m². The eq. (13) clearly brings out the importance of α_1 and α_2 (which under the stress-free constraints are equal to zero) in defining C_{44} , as the electron bulk modulus K_e is very small.

2.3. The pressure derivatives of SOEC :

Birch has derived a set of effective elastic constants which determines the response of an initially stressed crystal to an additional infinitesimal strains. From these values, the pressure derivatives of the second-order elastic constants have been calculated.

2.4. Lattice dynamical behaviour :

The elements of the dynamical matrix having explicit bearing on eq. (2) may be written as

$$D_{\alpha\alpha}^{(2)}(\mathbf{q}) = 2(\alpha_1 + \beta_1) [2 - C_\alpha(C_\beta + C_\gamma)] + 4\alpha_1(1 - C_\beta C_\gamma) + 4\beta_2 S_\alpha^2 + 4\alpha_2(S_\beta^2 + S_\gamma^2), \quad (14)$$

$$D_{\alpha\beta}^{(2)}(\mathbf{q}) = 2(\beta_1 - \alpha_1) S_\alpha S_\beta, \quad (15)$$

where $S_\alpha = \sin(aq_\alpha / 2)$ and $C_\alpha = \cos(aq_\alpha / 2)$, (16)

$$\alpha_1 = [1/r (\partial\Phi / \partial r)]_N, \quad \alpha_2 = [1/r (\partial\Phi / \partial r)]_{NN}, \quad (17)$$

$$\beta_1 = [\partial^2\Phi / \partial r^2]_N, \quad \beta_2 = [\partial^2\Phi / \partial r^2]_{NN}, \quad (18)$$

q_α is the α -component of phonon wave vector \mathbf{q} , a is the lattice parameter and α_1, β_1 are the force constants for the first neighbour (N) and α_2, β_2 are those for the second nearest neighbour (NN), respectively.

The phonon frequencies (ν) are obtained by solving the usual secular equation *i.e.*

$$D_{\alpha\beta}(\mathbf{q}) - 4\pi^2\nu^2 M I = 0, \quad (19)$$

where I is the unit matrix of 3×3 order and M is the mass of the atom.

2.5. Parameter evaluation :

Eq. (7) is treated repeatedly for a chosen value of n and m to yield such a value of dimensionless quantity (αa_n) which reproduces such values of β and D [from eq. (9)] which on subsequent substitution in eq. (4) yields exactly measured value of cohesive energy. This

proper value of αa_0 is employed to evaluate α from the measured value of equilibrium semi-lattice constant (a_0). The value of r_0 is evaluated using eq. (5).

Table 1. Input data and computed parameters for fcc aluminium.

Input data	Computed parameters for $n = 0.5, m = 1.5$ and $aa_0 = 2.388$
Lattice constant	$D = 1.513965 \times 10^{-21}$ J
$a = 4.05$ Å	$\alpha = 1.179259 \times 10^{10}$ m ⁻¹
Bulk modulus	$\beta = 599.9326$
$B = 0.722 \times 10^{11}$ N/m ²	$r_0 = 5.424437 \times 10^{-10}$ m

3. Computations and results

A program was developed following the theory given in the preceding section and the same was fed to the computer to obtain the results given in Tables 2 to 6.

Table 2. Computed second-order elastic constants (in 10^{11} N/m²)

Metal	n	m	C_{11}	C_{12}	C_{44}	Temperature °K	Reference
Al	0.5	1.5	0.889	0.638	0.362		C_{44} from eq. (12)
					0.211		C_{44} from eq. (13)
Expt			1.143	0.619	0.316	0	22
			1.068	0.607	0.282	300	22

Table 3. Computed third-order elastic constants (in 10^{11} N/m²)

Metal	n	m	C_{111}	C_{112}	C_{123}	Reference
Al	0.5	1.5	-5.724	-3.546	0.530	
Expt			-10.700	-3.150	0.360	23

Table 4. Computed pressure derivatives of second-order elastic constants

Metal	n	m	$\partial C_{11}^*/\partial p$	$\partial C_{12}^*/\partial p$	$\partial C_{44}^*/\partial p$	$\partial B^*/\partial p$	$\partial C^*/\partial p$	Ref
Al	0.5	1.5	4.51	3.73	1.73	3.99	0.39	
Expt.			7.02	3.94	2.20	4.97	—	24

Table 5. Computed values of cohesive energy of Al in eV/atom.

Metal	n	m	Position of minima (Å)	Repulsive part of cohesive energy	Attractive part of cohesive energy	Magnitude of cohesive energy
Al	0.5	1.5	2.864	6.283	9.673	3.390
		6.0	2.864	1.192	4.582	3.390
	1.0	1.5	2.864	4.389	7.778	3.389
		6.0	2.864	0.924	4.314	3.390

Table 6. Computed force constants for *fcc* aluminium (in N/m).

α_1	α_2	β_1	β_2
-2.184421	0.286176	21.208030	0.124052

The needed input data and the corresponding computed parameters for the *fcc* aluminium are given in Table 1 for the most suitable combination of *n* and *m*. Table 2 displays the SOEC whereas Table 3 displays TOEC for *fcc* aluminium. The pressure derivatives of SOEC are recorded in Table 4. The computed values of cohesive energy for the said metal are recorded in Table 5. Table 6 lists the evaluated derivatives α_1 , α_2 , β_1 and β_2 for the metal. Figure 2 shows the dispersion relations for the *fcc* aluminium along with the predictions of Wang and Overhauser [6], Singh [7] and experimental findings of Stedman *et al* [20].

4. Conclusions

The extended form of the generalised exponential potential explains quantitatively the SOEC, TOEC and pressure derivatives of the SOEC in *fcc* aluminium. Besides predicting satisfactorily the elastic behaviour of *fcc* aluminium, the extended generalised exponential potential (EGEP) has produced remarkably good results particularly in case of cohesive energy, which confirms the empirical nature of the potential.

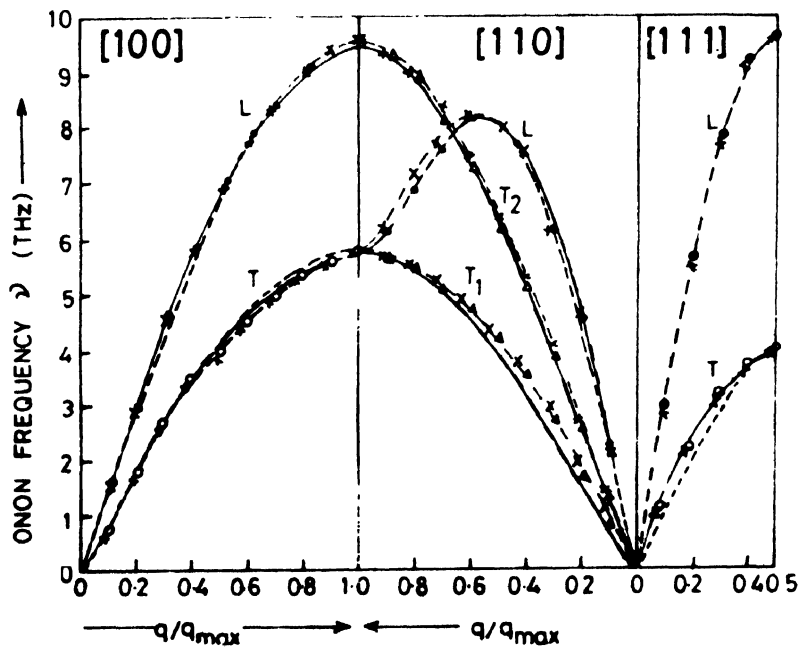


Figure 2. Phonon dispersion in *fcc* aluminium, — present study, - Wang and Overhauser [6] —x—x M Singh [7], ●, ○, ▲, △ experimental findings of Stedman *et al* [20].

The computed phonon frequencies of aluminium (Figure 2) agree satisfactorily with the measured data of Stedman *et al* [20]. The present model has successfully predicted the cohesive energy and therefore, explains all the qualitative features of phonon dispersion relations of the said metal. Our predictions are free from relative standard error and this fact enhances the reliability [21] of our model.

Acknowledgments

Two of us (MLV and AV) are thankful to N K Khurana of Universal Computer Training Point (UCTP), Palwal for valuable help in providing computation facility.

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