An Analysis of Lattice Dynamical Angular Force Models for Body Centered Cubic Metals

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Present angular force models for lattice dynamical behaviour of body centered cubic (bcc) metals have been analysed for the satisfaction of rotational invariance and general equilibrium conditions. It has been found that most of the angular force models (except that due to Clark et al.) are deficient and leave the lattice under stress. Further it has been inferred that either the axially symmetric or the central pair potential schemes, incorporating the concept of electron pressure, provide the realistic picture of lattice excitations in metals.

Angular force models are currently being used to study [1–3] the lattice dynamical behaviour of bcc metals. Recently Verma and the present author [4] have established that there is hardly any justification in the assumption that angular interactions couple the metallic ions. Upadhyaya [5] has also concluded that angular interactions produce no additional results and create confusion: these models need not to be retained in further lattice dynamical calculations. In view of these remarks it has been thought worthwhile to analyse the mathematical contents of the various angular force models [6–9]. The present analysis shows that (a) all angular force models for bcc metals are identical as far as their parametric representation is concerned, (b) the angular force model developed by Clark et al. [6] stands valid to the test of symmetry requirement of rigid body rotation and (c) the remaining angular force models [7–9], when subjected to a proper equilibrium condition accounting for short and long range electron response to the ionic motion, produce better results on phonon dispersion in metals. Further it has been noted that axially symmetric [10] or central pair potential [11] models, modified to contain proper equilibrium conditions, provide a more justified and the successful description of the lattice waves in the metals.

The survey of the present literature on angular force models reveals that there exist four different types of angular force models, which have been employed for lattice dynamical studies of bcc metals. They are those proposed by de Launay [7], Hendricks et al. [8], Clark et al. [6] and Rani and Gupta [9]. The elements of dynamical matrices for these models are given in the following lines:

de Launay model [7]

$$\begin{aligned} D_{11}(\boldsymbol{q}) &= \tfrac{8}{3}(\alpha_1 + 2\alpha')(1 - C_1 C_2 C_3) + 4\alpha_2 S_1^2 \\ &\quad + 2\alpha''(2 - C_2' - C_3') \\ D_{12}(\boldsymbol{q}) &= \tfrac{8}{3}(\alpha_1 - \alpha') S_1 S_2 C_3 , \end{aligned} \tag{1}$$

where $S_i = \sin(\frac{1}{2} a q_i)$, $C_i = \cos(\frac{1}{2} a q_i)$, $C_i' = \cos(a q_i)$ $(i = 1, 2, 3)$.

a is the lattice constant and q_i is the i-th component of the phonon wave vector $\boldsymbol{q}$. α_1, α_2 are the radial and α', α'' are the angular force constants for the nearest and the next nearest neighbours, respectively.

Clark et al. model [6]

$$\begin{aligned} D_{11}(\boldsymbol{q}) &= 8(\beta_1 + 2Y_1 + 3Y_2)(1 - C_1 C_2 C_3) \\ &\quad + 4\beta_2 S_1^2 - 2Y_1(4C_1' - C_2' \\ &\quad - C_3' - 2) + 3Y_2(2 - C_2' - C_3'), \\ D_{12}(\boldsymbol{q}) &= 8(\beta_1 - Y_1 + \tfrac{3}{2}Y_2) S_1 S_2 C_3 , \end{aligned} \tag{2}$$

where β_1, β_2 are the central and Y_1, Y_2 the angular force constants for the nearest and the next nearest neighbours, respectively.

Hendricks et al. model [3]

$$\begin{aligned} D_{11}(\boldsymbol{q}) &= (\sigma_1 + 2\sigma_3)(1 - C_1 C_2 C_3) + \tfrac{3}{2}\sigma_2 S_1^2 , \\ D_{12}(\boldsymbol{q}) &= (\sigma_1 - \sigma_3) S_1 S_2 C_3 , \end{aligned} \tag{3}$$

where σ_1, σ_2 are the central force constants for the nearest and the next nearest neighbours, respectively, and σ_3 is the angular force constant for the nearest neighbours only.

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Rani and Gupta model [9]

$$D_{11}(\boldsymbol{q}) = 8(\alpha + 8K_2)(1 - C_1C_2C_3) + 4(\beta + 8K_2)S_1^2 + 8K_1(S_1^2 - S_2^2 - S_3^2),$$
$$D_{12}(\boldsymbol{q}) = 8(\alpha + 4K_2)S_1S_2C_3, \quad (4)$$

where α, β are the central and K_1, K_2 are the angular force constants for the nearest and the next nearest neighbouring ions.

If we compare the elements of the dynamical matrices of these models we get the following relations expressing the perfect equivalence between the disposable parameters:

$$\alpha_1 = 3\beta_1 + 6Y_2 = \tfrac{3}{2}\sigma_1 = 3\alpha + 16K_2,$$
$$\alpha_2 = \beta_2 + 4Y_1 = \tfrac{3}{8}\sigma_2 = \beta + 8K_2 + 2K_1,$$
$$\alpha' = 3Y_1 + \tfrac{3}{2}Y_2 = \tfrac{3}{2}\sigma_3 = 4K_2,$$
$$\alpha'' = \tfrac{3}{2}Y_2 - Y_1 = \cdots = -2K_1. \quad (5)$$

The expression for the compressibility (K) may also be derived by evaluating the respective dynamical matrices in the long wave length limit and then comparing them to the Christoffel equation of elasticity. The expression thus obtained may be written as

$$K^{-1} = \frac{1}{3a}(2\alpha_1 + 2\alpha_2 - 4\alpha' - 4\alpha'')$$
$$= \frac{1}{3a}(6\beta_1 + 2\beta_2)$$
$$= \frac{1}{a}\left(\frac{\sigma_1}{4} + \frac{\sigma_2}{4} - \frac{\sigma_3}{4}\right)$$
$$= \frac{1}{3a}(6\alpha + 2\beta + 32K_2 + 12K_1). \quad (6)$$

It may be recalled that the symmetry requirement of rigid body rotation demands that the compressibility should be independent of the angular force constants. It is thus clear that all the angular force models, except the one due to Clark et al. [6], violate this symmetry requirement of rotational invariance. The condition of rotational invariance imposes a new constraint on to the angular force parameters of the three deficient models i. e. the de Launay [7], Hendricks et al. [8] and Rani and Gupta [9] models. This constraint may now be written as

$$\alpha' + \alpha'' = \sigma_3 = 8K_2 + 3K_1 = 0. \quad (7)$$

In view of Eq. (7), the contribution of the de Launay [7] and Hendricks et al. [8] angular forces towards the Cauchy discrepancy vanishes, and these forces loose their importance on this account. However, the angular forces due to Clark et al. [6] and Rani and Gupta [9] do contribute towards the Cauchy discrepancy; in the former scheme both angular force constants (Y_1 and Y_2) equally contribute, while in the later scheme only one angular force constant (K_1 or K_2) contributes effectively to the Cauchy discrepancy.

The metallic lattices may be considered as many body assemblies of the constituent ions and electrons. The effect of the conduction electrons in the angular force models is obviously limited to the screening of the inter-ionic interactions leading to the short range character of these forces, which is not wholly justified on various theoretical [12] and experimental [13] grounds in general. The electron pressure contribute significantly towards the lattice stability of metals. Some recent studies [14 – 16] have considered this stability under the combined effect of the ionic and the electron pressure. These studies have calculated the electron pressure in Hartree approximation, which is open to question. For considering the lattice stability properly we have to account for all the volume dependent electron energies i. e. Fermi, exchange and correlation energies, all of which govern the electron response to the ionic motion. The potential (Φ_e) expressing the electron-electron interaction may be written on the basis of two different approaches [17] i. e. the independent particle approach and the collective motion approach. The electron pressure (P_e) may be calculated using a correct expression of the electron-potential (Φ_e), i. e.

$$P_e = -\partial\Phi_e/\partial\Omega, \quad (8)$$

where Ω ($= \frac{4}{3}\pi r_s^3 a_0^3$) is the atomic volume.

This electron pressure P_e counter balances the ionic pressure for the maintenance of the crystal equilibrium in general. This concept of crystal equilibrium can duely be incorporated in either the central pair potential model [11] or the axially symmetric model [10]. The present author [18] has shown that the dispersion relations in metals can efficiently be explained on the basis of the central pair potential model modified for the equilibrium.

It may again be noted that the angular forces in Clark et al. [6] scheme appear to represent the electron pressure partially in quite a different manner. This is possibly the reason for the success of this scheme. In view of the rotational invariance condition [Equation (7)]. The remaining angular force models [7 – 9] do not include this effect of the electron pressure and leave the crystal under

stress, which is to be remedied. We have equated the angular force constants to the electron pressure (P_e) calculated from Equation (8). It has been found that the deficient angular force models [7 – 9] with electron pressure lead to better results on phonon dispersion as compared to those given by the Clark et al. [6] scheme. The details of these calculations shall be reported shortly. It may be mentioned that the axially symmetric model [10], the central pair potential model [11] and the angular force models (Particularly those due to de Launay [7] and Clark et al. [8]) are equivalent [19 – 21] with each other. Hence it again appears more justified to use either the axially symmetric [10] or the central pair potential [11] models for describing the lattice dynamical behaviour of the metals.

Conclusion

It can be concluded that all the existing angular force models for the bcc metals are identical in terms of their disposable parameters. The angular force model due to Clark et al. [6] obeys the rotational invariance condition, but it only partly satisfies the equilibrium condition. The remaining angular force models [7 – 9] neither satisfy the rotational invariance condition nor account for the equilibrium of the lattice. In general, the angular force models [6 – 9] do not provide a justified description of the lattice waves in bcc metals. The axially symmetric model [10] or the central pair potential model [11], modified to include the equilibrium condition may successfully be used to study the crystal dynamics of the metals.

[1] L. P. Pathak, R. C. Rai, and M. P. Hemkar, J. Phys. Soc. Japan **44,** 1834 (1978).
[2] H. L. Kharoo, O. P. Gupta, and M. P. Hemkar, Z. Naturforsch. **32 a,** 1490 (1977).
[3] R. P. S. Rathore and M. P. Verma, Ind. J. Phys. **50,** 605 (1976).
[4] R. P. S. Rathore and M. P. Verma, Ind. J. Pure and App. Phys. **15,** 1 (1977).
[5] J. C. Upadhyaya, J. Phys. **F-7,** 769 (1977).
[6] B. C. Clark, D. C. Gazis, and R. F. Wallis, Phys. Rev. **134,** 1486 (1964).
[7] J. de Launay, Solid State Phys. (Edited by F. Seitz and D. Turnbull) **2,** 276 (1956).
[8] J. B. Hendricks, H. B. Riser, and B. C. Clark, Phys. Rev. **130,** 1377 (1963).
[9] N. Rani and H. C. Gupta, Solid State Commun. **23,** 799 (1977).
[10] G. W. Lehman, T. Wolfram, and R. E. de Wames, Phys. Rev. **128,** 1593 (1962).
[11] A. A. Maradudin, E. W. Montroll, and G. H. Weiss, Solid State Phys. Suppl. **3,** 6 (1971).
[12] E. R. Cowley, Solid State Commun. **14,** 587 (1974).
[13] B. N. Brockhouse, T. Arase, G. Gaglioti, K. R. Rao, and A. O. B. Woods Phys. Rev. **128,** 1099 (1962).
[14] J. C. Upadhyaya, S. S. Sharma, and O. P. Kulshrestha, Phys. Rev. **B-12,** 2236 (1975).
[15] R. P. S. Rathore and M. P. Verma, Ind. J. Pure App. Phys. **15,** 54 (1977).
[16] R. P. S. Rathore and M. P. Verma, Ind. J. Pure App. Phys. **15,** 467 (1977).
[17] D. Pines, Solid State Phys. (edited by F. Seitz and D. Turnbull) **Vol. 1,** 368 (1955).
[18] R. P. S. Rathore, Ind. J. Phys. **52 A,** 259 (1978).
[19] J. C. Upadhyaya, J. Phys. Chem. Sol. **37,** 644 (1976).
[20] R. P. S. Rathore, Ind. J. Pure App. Phys. **15,** 730 (1977).
[21] R. P. S. Rathore, Ind. J. Pure App. Phys. **16,** 627 (1978).