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On the Extended Forms of Phenomenological Models for the Lattice Dynamics of BCC Transition Metals

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Abstract

The Sharma–Joshi and Krebs phenomenological models for the lattice dynamics of cubic metals have been extended and are applied to study the phonon dispersion curves of tungsten, a metal for which long range forces are also important. It has been found that, unlike extended forms of the de Launay model (Shukla and Cavalheiro 1973a) for certain cubic metals, the models studied here for b.c.c. transition metals are not always adequate. Possible reasons for the shortcomings of the results are pointed out and discussed. The role of angular forces, particularly when they are used along with a fairly good conduction electron–ion interaction term, has been examined and found to be unimportant.

Introduction

During the last decade a number of lattice dynamical models for cubic metals have been used to study the lattice dynamics of cubic transition metals. The model of Sharma and Joshi (1963) and that of Krebs (1965), with radial interactions extending out to second or third neighbours, have been shown to furnish a satisfactory interpretation of the lattice dynamical behaviour of these metals (Pal and Sharma 1965; Mahesh and Dayal 1966; Hautecler and van Dingenen 1967; Singh and Sharma 1968; Pal 1971). Several authors, emphasizing the importance of the angular forces, incorporated them into the above models and tried to improve the agreement between the experimental and theoretical dispersion curves (Behari and Tripathi 1972). The results obtained in all such studies, although fairly satisfactory, do not show the fine structure characteristics found in the experimental dispersion curves. Recent calculations by Shukla and Salzberg (1973) and Shukla and Cavalheiro (1973*a*, 1973*b*) on the extended forms of the Bhatia (1955) and de Launay (1956) models have renewed an interest in the study of the fine structure of the dispersion curves of cubic metals.

Owing to the simplicity and general applicability of the Sharma–Joshi model and to the overall superiority of the Krebs model, it was considered worth while to employ these models in their extended forms (i.e. including angular forces) to examine phonon dispersion in tungsten. It is well known from the Born–von Karman model analyses that, for the transition metals, the long range forces are of considerable magnitude and hence a study of these metals by means of the extended forms of successfully used phenomenological models should prove worth while.

The present work was motivated by the special characteristic features of the phonon dispersion curves of tungsten (Chen and Brockhouse 1964) and by a com-

parative study of the previously reported work (Pal and Sharma 1965; Mahesh and Dayal 1966; Behari and Tripathi 1972). A comparison of the results obtained by Mahesh and Dayal with those obtained by Behari and Tripathi shows that the Krebs model without angular forces is better than the Sharma–Joshi model with angular forces. This indicates the importance of examining the role played by the angular forces, particularly when they are coupled with more appropriate conduction electron–ion interaction models.

It is well known that, in different phenomenological models, the electron gas contribution is taken into account in various approximate ways. This implies that the extent to which a particular type of conduction electron–ion interaction model will account for long range forces will differ from model to model. In such circumstances an unlimited number of force constants coupled with a given model might lead to deterioration in the theoretical results. With these views in mind, in the present paper, we discuss the adequacy of the extended forms of the more appropriate phenomenological models in reproducing the fine structure of the dispersion curves, the importance of the angular forces and the possibility of the useful extension of a given phenomenological model. All these points are illustrated through the dispersion curves of tungsten.

Theory

The secular equation determining the angular frequencies of normal modes of vibration can be written as

$$|\mathbf{D}-m\omega^2\mathbf{I}| = 0, \tag{1}$$

where the dynamical matrix $\mathbf{D}$ is the sum of central, angular and electronic components, namely,

$$D_{ij} = D^{c}_{ij} + D^{a}_{ij} + D^{e}_{ij}, \tag{2}$$

and m is the ionic mass, ω is the angular frequency and $\mathbf{I}$ is the unit matrix. Denoting the force constants of the central interactions corresponding to the first, second, third and fourth neighbours by α_1, α_2, α_3 and α_4 respectively and those corresponding to angular interactions up to third neighbours by α', α'' and α''', we have

$$\begin{aligned} D^{c}_{11} + D^{a}_{11} = {} & \tfrac{8}{3}(\alpha_1+2\alpha')(1-C_1C_2C_3) + 4\alpha_2 S_1^2 \\ & + 4(\alpha_3+\alpha''')(2S_1^2+S_2^2+S_3^2-2S_1^2S_2^2-2S_1^2S_3^2) \\ & + \tfrac{1}{11}\alpha_4\{88+264C_1C_2C_3-32C_1C_2C_3(9C_1^2+C_2^2+C_3^2)\} \\ & + 2\alpha''\{2-\cos(2\pi ak_2)-\cos(2\pi ak_3)\}, \end{aligned} \tag{3}$$

$$\begin{aligned} D^{c}_{12} + D^{a}_{12} = {} & \tfrac{8}{3}(\alpha_1-\alpha')S_1S_2C_3 + 8(\alpha_3-\alpha''')S_1S_2C_1C_2 \\ & + \tfrac{1}{11}\alpha_4\{120S_1S_2C_3-32S_1S_2C_3(3S_1^2+3S_2^2-C_3^2)\}. \end{aligned} \tag{4}$$

Here,

$$C_i = \cos(\pi ak_i) \quad \text{and} \quad S_i = \sin(\pi ak_i), \tag{5}$$

a being the lattice constant, and k_i being the phonon wave vector component, with $i = 1, 2, 3$.

In the present study, D_{ij}^{e} is introduced through the Sharma–Joshi (1963) and Krebs (1965) models. For the Sharma–Joshi model, we have

$$D_{ij}^{e} = 2a^3\pi^2 k_i k_j V_e G^2(x), \tag{6}$$

where V_e is the bulk modulus of the electron gas, x is given by

$$x = 2\pi k r_0, \tag{7}$$

r_0 being the radius of the Wigner–Seitz sphere, and the function $G(x)$ is given by

$$G(x) = 3x^{-3}(\sin x - x\cos x). \tag{8}$$

For the Krebs model, we have

$$D_{12}^{e} = A\sum_{\boldsymbol{h}}\left(\frac{(q_1+h_1)(q_2+h_2)}{|\boldsymbol{q}+\boldsymbol{h}|^2+(a/2\pi)^2K_c^2}\,G^2(B|\boldsymbol{q}+\boldsymbol{h}|) - \frac{h_1h_2}{h^2+(a/2\pi)^2K_c^2}\,G^2(B|\boldsymbol{h}|)\right). \tag{9}$$

Here A is the electronic force constant, $\boldsymbol{h}$ is the reciprocal lattice vector, $\boldsymbol{q} = 2\pi\boldsymbol{k}$, and

$$K_c^2 = K_c^2(P)f(t), \tag{10}$$

where

$$K_c(P) = 0{\cdot}353(\gamma_e/a_0)^{\frac{1}{2}}k_F \tag{11}$$

is the Bohm–Pines screening parameter, with $\gamma_e = (3/4\pi n_e)^{\frac{1}{3}}$ being the interelectronic spacing and n_e the electronic number density, while k_F is the Fermi vector and $a_0 = 0{\cdot}529$ Å is the Bohr radius. The function $f(t)$ in equation (10) is given by

$$f(t) = \tfrac{1}{2} + \frac{1-t^2}{4t}\ln\left(\left|\frac{1+t}{1-t}\right|\right), \tag{12}$$

where

$$t = (|\boldsymbol{q}+\boldsymbol{h}|)/2k_F. \tag{13}$$

Finally, the quantity B in equation (9) is given by

$$B = 2\pi\gamma_e/a. \tag{14}$$

The force constants involved in the dynamical matrix of a b.c.c. metal are related to the measured elastic constants, the lattice constant and the zone boundary frequencies. In the present work the force constants were determined from the following experimental values (Featherston and Neighbours 1963) for tungsten at 300 K:

c_{11}	c_{12}	c_{44}		a	
$52{\cdot}327\times10^{10}$	$20{\cdot}453\times10^{10}$	$16{\cdot}072\times10^{10}$	Pa	$3{\cdot}165\times10^{-10}$	m

and the following frequencies (Chen and Brockhouse 1964):

(1 0 0)	$(\frac{1}{2}\frac{1}{2}\frac{1}{2})$	$(\frac{1}{2}\frac{1}{2}0)$	$(\frac{1}{2}\frac{1}{2}0)$	
$5{\cdot}50\times10^{12}$	$5{\cdot}50\times10^{12}$	$6{\cdot}75\times10^{12}$	$4{\cdot}40\times10^{12}$	Hz

The resulting force constants are listed in Table 1.

Fig. 1a

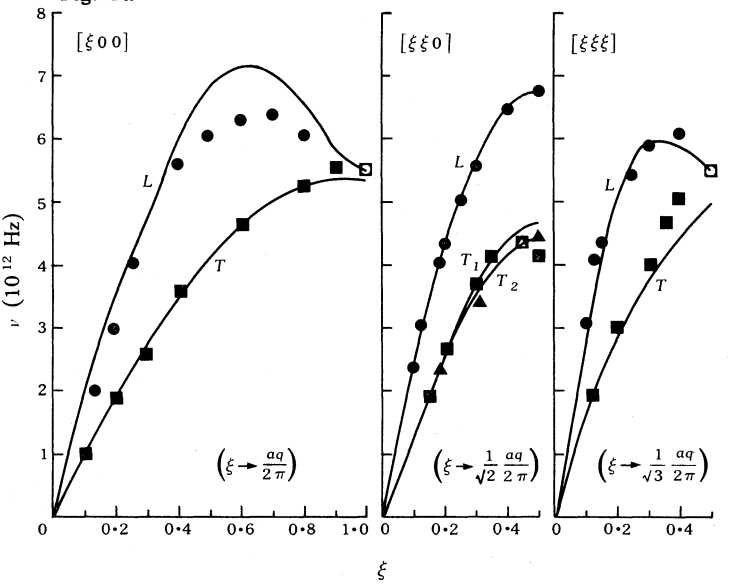
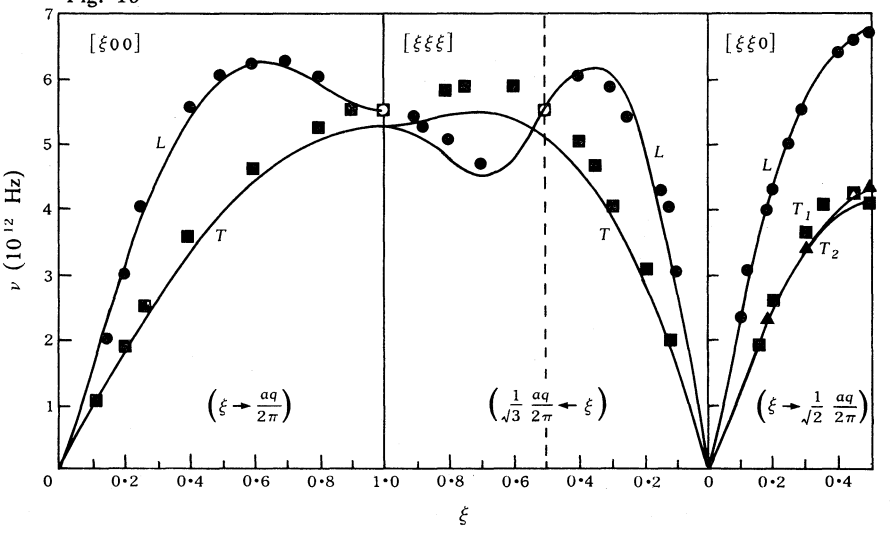


Fig. 1b



Figs 1*a*–1*d*. Phonon dispersion curves for tungsten in the indicated directions for:

(*a*) the Sharma–Joshi model incorporating radial forces out to the fourth nearest neighbours and angular forces out to the second nearest neighbours;

(*b*) the Sharma–Joshi model incorporating radial and angular forces out to the third nearest neighbours;

(*c*) the Sharma–Joshi model incorporating radial forces only out to the third nearest neighbours;

(*d*) the Krebs model incorporating radial and angular forces out to the third nearest neighbours.

Fig. 1c

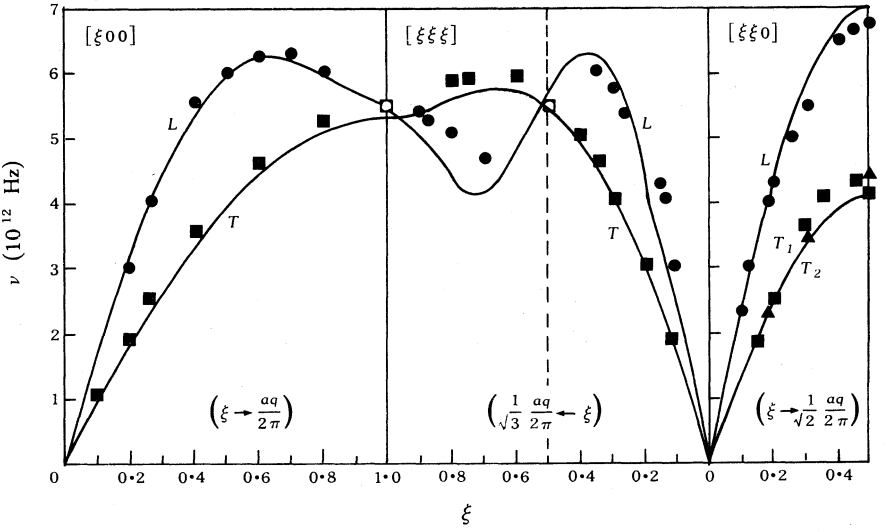
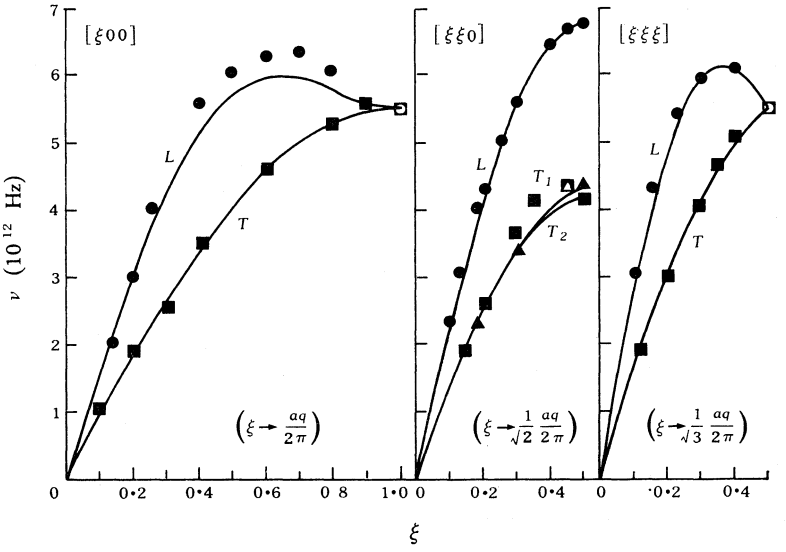


Fig. 1d



Figs 1c and 1d *(see caption opposite)*.

Table 1. Force constants for tungsten

Units are 10^2 Pa, where 1 Pa (pascal) ≡ 10 dyn cm^{-2}.

α_1	α_2	α_3	α_4	α'	α''	α'''	$V_e a$
			(a) Sharma–Joshi model				
56·483	41·344	6·717	—	2·942	−1·714	−0·358	15·914
63·128	46·049	4·392	—	—	—	—	13·866
53·532	46·119	21·752	−5·115	13·243	−14·156	—	10·213
			(b) Krebs model				
61·953	44·082	3·497	—	2·169	0·1684	−0·3294	34·827

Results and Discussion

The dispersion curves of tungsten obtained from the extended Sharma–Joshi model covering radial interactions out to the fourth nearest neighbours and angular interactions out to the second nearest neighbours are presented in Fig. 1*a*. Results of the same model coupled with three radial and three angular force constants are displayed in Fig. 1*b*. These results, when compared with those reported previously on the basis of the same model coupled with a limited number of force constants (Pal and Sharma 1965; Behari and Tripathi 1972), feature no overall improvement except for the removal of degeneracy in the transverse branch in the [110] direction at the cost of a marked deterioration in the longitudinal branch in the [100] direction (Fig. 1*a*) and the transverse branches in the [111] and [100] directions of Fig. 1*b*. Also, the frequencies for the wave vectors beyond $\xi = 0{\cdot}5$ in the [111] direction differ markedly from the experimental values and hence are not shown in Fig. 1*a*. These results clearly indicate the inadequacy of the Sharma–Joshi model in its extended form to reproduce dispersion curves which exhibit improved agreement with the experimental data. Results obtained from the Sharma–Joshi model, with three radial force constants only, are presented in Fig. 1*c*. A comparison with Fig. 1*b* and with results reported previously (Pal and Sharma 1965; Behari and Tripathi 1972) shows clearly the unimportance of the angular forces, particularly when they are coupled with the Sharma–Joshi model. Dispersion curves calculated for the Krebs model with three radial and three angular force constants (Fig. 1*d*) when compared with those reported by Mahesh and Dayal (1966) lead to the same conclusion.

It is important to comment on the deterioration apparent in certain parts of the present dispersion curves as compared with the successful application of the extended form of the de Launay model to some metals by Shukla and Salzberg (1973) and Shukla and Cavalheiro (1973*a*, 1973*b*). The behaviour of the longitudinal branch in the [100] direction (Fig. 1*a*) shows that there is an overestimation of the long range forces. Deterioration in certain parts of the theoretical dispersion curves implies that there are limitations in the conduction electron–ion interaction model, and thus the model will not reproduce the experimental frequencies at some points in the q space.

The situation is different for the de Launay (1956) model, which differs from other phenomenological models in that it does not explicitly give a separate expression for the conduction electron–ion interactions; the effect of these interactions being embodied in the modified force constants for longitudinal vibrations. In this circumstance, a gradual increase in the number of force constants and an appropriate modification of the force constants for longitudinal components would gradually account for conduction electron–ion interactions. It is for this reason that the extended forms of the de Launay model yield better results. To overcome such gratuitous agreement, it seems to us to be more important to formulate a correct account of the conduction electron–ion interactions, and not merely to increase the number of radial and angular force constants.

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