MODIFIED TENSOR FORCE MODEL FOR LATTICE DYNAMICS OF B.C.C. METALS

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The point ion tensor force model has been modified to include the electron-ion interaction according to the formalism of Krebs. The proposed five parameter model has given an excellent description of the phonon dispersion relations along all the principal symmetry directions in sodium, potassium and lithium.

1. Introduction

The theoretical study of the lattice of metals has gone through several phases. In the initial phase, during the years 1900 to 1935, when only the experimental $(C_v - T)$ curves of metals were available, the intricate problem for theoreticians was to explain the exact dependence of C_v on temperature. It was found that the point ion continuum model of Debye [1] was able to explain such behaviour for higher temperatures. Only low temperature behaviours were unexplained. The second stage of studies started with the work of Fuchs [2] in 1936 when he, for the first time, divided the total interatomic interactions in metallic ions into two parts, the central ion-ion interaction part and the non-central volume dependent part. Almost all subsequent workers in the field of lattice dynamics followed Fuchs's idea. The third stage of study of lattice dynamics of metals started in 1962 when the experimental phonon dispersion relations along the principal symmetry directions of lead was determined [3]. It was realised by theoreticians that the critical test of the validity of a model was the wave vector versus frequency dispersion relations and not the C_v versus temperature curve. Because, phonon dispersion relations offers a point to point check of the phonons propagating along the symmetry directions while C_v gives only a statistical average of the entire phonon spectrum.

By now phonon dispersion relations in almost all cubic metals have been determined [4]. By endless effort theoreticians have developed models based on first principles [5] and on a phenomenological basis [6] explain the actual interatomic relations existing between

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metallic atoms. Experience shows that we are still in the initial stage. Not even a simple proposed model is free of mathematical and physical flaws. In the present note a new phenomenological model is proposed.

The phenomenological models split up the dynamical matrix of the metals into two parts, the ion-ion interaction part and the electron-ion interaction part. Let us assume that ion-ion interactions are given by short range forces and electron-ion interaction on a long ranged one. We have considered the ion-ion interaction on the basis of the tensor force model [7]. The electron-ion interaction has been considered on the formalism of Krebs [8]. We have applied the present formalism to the b. c. c. metals and that too, to three of the alkali metals i. e. sodium, potassium and lithium. Alkali metals were chosen because:

1. Not even pure tensor force model studies exist for them.

2. For all of them experimental phonon dispersion relations along the principal symmetry directions exist providing a test for the theoretical predictions.

3. Experimental constants, elastic data and lattice parameters are available to determine model parameters.

4. The bulk modulus of the electron gas for them is a small quantity compared to b. c. c. transition metals and f. c. c. metals so these metals serve as an ideal substance where the electron-ion interaction part of the dynamical matrix is like a perturbation term to the ion-ion interaction part.

The computed phonon dispersion relations of sodium, potassium and lithium form the subject matter of this paper.

2. Theory

The phonon frequencies of cubic metals are given by the solution of the secular equation

$$|D(q) - \omega^2 I| = 0, \tag{1}$$

where ω is the angular frequency, q is the propagation vector, and I is the 3×3 unit matrix.

Each element of the dynamical matrix $D(q)$ is split up into two parts, the ion-ion interaction $D^{ii}(q)$ and the electron-ion interaction part $D^{ie}(q)$ written mathematically

$$D(q) = D^{ii}(q) + D^{ie}(q). \tag{2}$$

The diagonal and non diagonal part of the $D^{ii}(q)$ are given by (see Squires [7])

$$MD_{ii}^{ii}(q) = \sum_s \left[\frac{n^s}{6} \alpha_1^s \{2 - \cos(ah_1qx)(\cos(ah_2qy)\cos(ah_3qz)\right.$$

$$+ \cos(ah_3qy)\cos(ah_2qz))\} + \alpha_2^s\{2 - \cos(ah_2qx)(\cos(ah_3qy)\cos(ah_1qz)$$

$$+ \cos(ah_1qy)\cos(ah_3qz))\} + \alpha_3^s\{2 - \cos(ah_3qx)(\cos(ah_1qy)\cos(ah_2qz)$$

$$\left. + \cos(ah_2qy)\cos(ah_1qz))\} \right], \tag{3}$$

$$MD_{ij}^{ii}(q) = \sum_s \left[\frac{n^s}{6} \beta_1^s \cos{(ah_1qz)} \{\sin{(ah_2qx)} \sin{(ah_3qy)} + \sin{(ah_3qx)} \sin{(qh_2qy)}\} \right.$$

$$+ \beta_2^s \cos{(ah_2qz)} \{\sin{(ah_3qx)} \sin{(ah_1qy)} + \sin{(ah_1qx)} \sin{(ah_3qy)}\}$$

$$+ \left. \beta_3^s \cos{(ah_3qz)} \{\sin{(ah_1qy)} \sin{(ah_2qy)} + \sin{(ah_2qx)} \sin{(ah_1qy)}\} \right]. \tag{4}$$

The diagonal and non diagonal part of $D^{ie}(q)$ are given by

$$D_{ii}^{ie}(q) = \frac{a^3\lambda^2}{4} K_e \sum_h \left[\frac{(q_i+H_i)^2 G^2(u_1)}{(q+H)^2 + \frac{a^2\lambda^2}{4\pi^2} f(H)} - \frac{H_i^2 G^2(u_2)}{H^2 + \frac{a^2\lambda^2}{4\pi^2} f(H)} \right], \tag{5}$$

$$D_{ij}^{ie}(q) = \frac{a^3\lambda^2}{4} K_e \left[\sum_h \frac{(q_i+H_i)(q_j+H_j) G^2(u_1)}{(q+H)^2 + \frac{a^2\lambda^2}{4\pi^2} f(t_1)} - \frac{H_iH_j}{H^2 + \frac{a^2\lambda^2}{4\pi^2} f(t_2)} \right]. \tag{6}$$

In the above expressions: α_i^s, β_i^s are the tensor force constants for the s-th neighbour; h_i ($i = 1, 2, 3$) are the set of non integers to specify a lattice point, $q(q_x, q_y, q_z)$ are the cartesian components of q, a is lattice parameter, K_e is the bulk modulus of the electron gas, H_i ($i = 1, 2, 3$) are the cartesian components of the reciprocal lattice vector. The functions $F(t)$ and $G(u)$ are given by

$$F(t) = \tfrac{1}{2} + \frac{1-t^2}{4t} \ln{\frac{1+t}{1-t}}, \tag{7}$$

$$G(u) = \frac{3}{u^3} (\sin{u} - u \cos{u}), \tag{8}$$

where

$$t_1 = \frac{\pi|q+H|}{aK_F}, \quad t_2 = \frac{\pi H}{aK_F} \quad u_1 = \frac{2\pi rs}{a} |q+H|, \quad u_2 = \frac{2\pi rs}{a} H, \tag{9}$$

$$\lambda = \beta \left(\frac{rs}{a_0} \right)^{1/2} K_F. \tag{10}$$

rs is the radius of the atomic sphere, K_F is the Fermi wave vector, a_0 is the Bohr radius.

By expanding the secular determinant in the long wavelength limits ($q \to 0$) the following relations are obtained between the force constants and elastic constants:

$$ac_{11} = 2\alpha_1^1 + 2\alpha_1^2 + 8\alpha_1^3 + aK_e,$$

$$a(c_{12}+c_{44}) = 4\beta_1^1 + 8\beta_3^3 + aK_e,$$

$$ac_{44} = 2\alpha_1^1 + 2\beta_2^2 + 4\alpha_1^3 + 4\beta_3^3.$$

3. *Evaluation of force constants*

We have determined two sets of force constants. The first set of force constants is for a pure tensor force model with interionic interactions extending out to third neighbours. The second set of force constants is for the modified tensor force model by restricting the interionic interactions up to the first two neighbours. The input data with proper sources are given in Table I. The output values of the force constants are given in Table II.

TABLE I

Input data used in calculations

Properties	Sodium		Potassium		Lithium	
	Values	Ref.	Values	Ref.	Values	Ref.
Elastic constants						
$(10^{11}$ dyn/cm^2) C_{11}	.808	[9]	.4167	[10]	1.484	[11]
C_{12}	.664	[10]	.3413	[10]	1.253	[11]
C_{44}	.586	[11]	.2860	[10]	1.080	[11]
Lattice constants $(10^{-8}$ cm)	4.24		5.225		3.491	
Atomic mass $(10^{-23}$ gr)	3.8163		6.4909		1.1520	
Zone frequency						
$(10^{12}$ Hz) $\nu L\xi00$	3.85	[12]	2.21	[13]	8.82	[14]
$\nu T\xi\xi\xi$	2.88	[12]	1.78	[13]	6.8	[14]
$\nu L\xi\xi0$	3.82	[12]	2.40	[13]	9.00	[14]
$\nu T\xi\xi$	2.56	[12]	1.50	[13]	5.70	[14]

TABLE II

Output values of force constants in units of 10^3 dyn cm^{-1}

Substance	α_1^1	β_1^1	α_1^2	α_2^2	α_1^3	α_3^3	β_3^3	aK_e
Lithium	2211	235	547	144	—	—	—	−336
	(2211)	(2199)	(713)	(60)	(−83)	(−109)	(−81)	—
Sodium	1207	50	440	135				130
	(1207)	(1293)	(462)	(124)	(10)	(−55)	(15)	—
Potassium	782	62	286	89				40
	(782)	(864)	(430)	(17)	(−31)	(5)	(−22)	—

5. *Results and discussion*

The phonon dispersion relations in sodium, potassium and lithium have been plotted in figures 1 to 3 after solving the secular determinant for selected wave vectors. The results for the modified tensor force model are shown by solid lines. The broken curves correspond
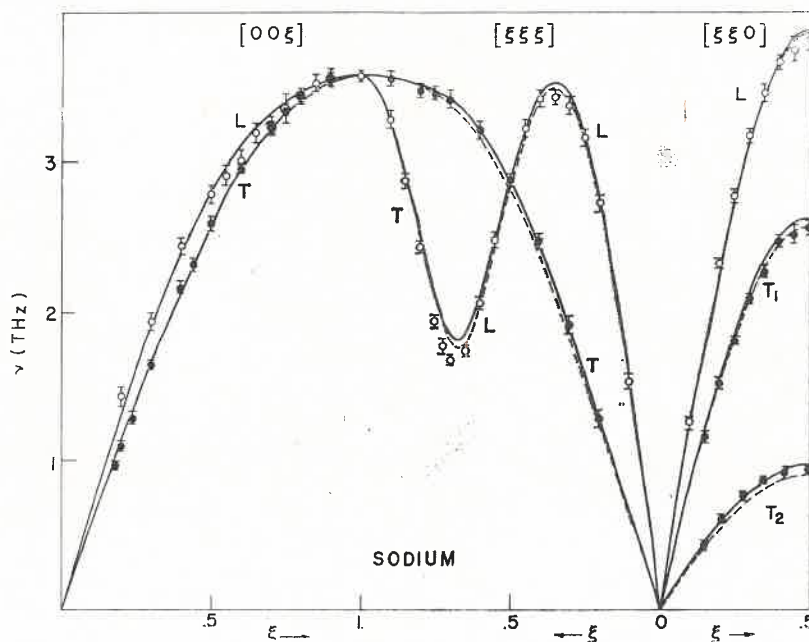

Fig. 1. Phonon dispersion relations in sodium along the $|\xi00|$, $|\xi\xi0|$ and $|\xi\xi\xi|$ directions. Computed curves are shown by solid lines. Experimental points are shown by the symbols ● and ○
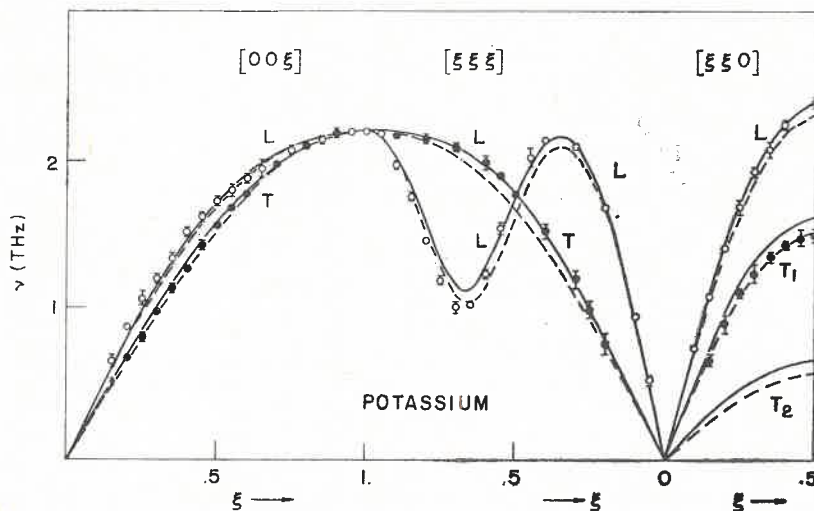

Fig. 2. Phonon dispersion relations in potassium along the potassium. Captions are the same as for fig. 1

to results of the pure tensor force model. The experimental points are shown also for comparison purposes.

A critical study of figures 1 to 3 reveals that the modified tensor force model has reproduced exceedingly well the experimental phonon dispersion relations in all the three alkali metals with equal success. At low wave vectors the calculated curves coincide with
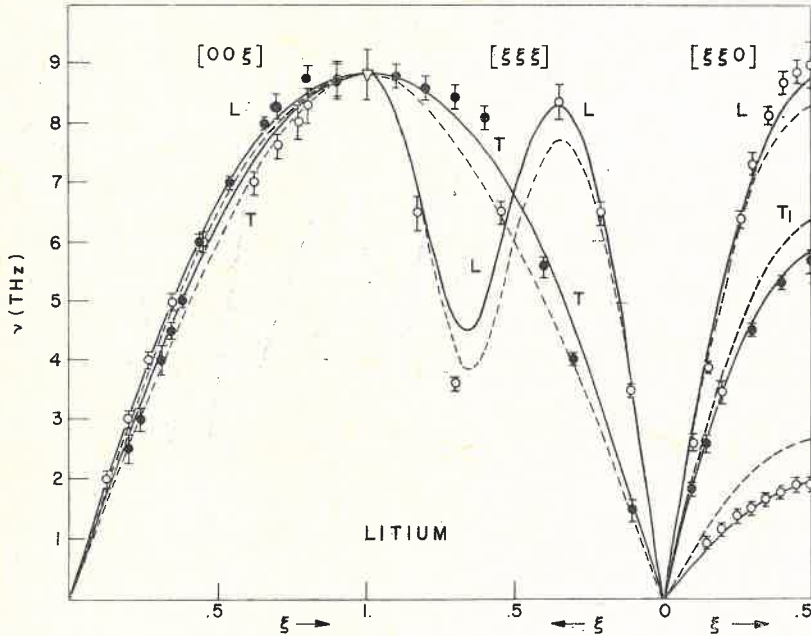

Fig. 3. Phonon dispersion relations in lithium. Captions are the same as for figure 1

the experimental results. A little discrepancy between calculated and experimental phonons have been found in all metals near the zone boundary but these deviations are small. Since the existing experimental errors in phonons were not considered while calculating the atomic force constants, this kind of results was inevitable.

The experimental phonons in lithium show a very peculiar behaviour in the [ξ 00] direction i. e. crossing of the longitudinal and transversal branches near the zone boundary. We have not obtained this effect in the theoretical results. It should be pointed out that experimentalists i.e. Woods et al. [12] for sodium, Cowley et al. [13] for potassium and Copley at al. [14] for lithium, have used as many as 13 free parameters to fit their experimental phonons. In spite of the large number of parameters they could not, as well as we could not, fit the lower portions of their experimental phonon dispersion relations.

The study of figures 1 to 3 exhibits the importance of the electron-ion interactions in the computation of the phonon frequencies. The results obtained by a five parameter model based on modified tensor forces are found to be better than those obtained by a seven parameter pure tensor force model. The present results are found to be superior to those published so far on existing phenomenological models. Such a result is the outcome of

the use of tensor forces to represent the ion-ion interaction. This fact can be very well recognized by comparing the present results for all these three alkali metals with models of Krebs [8] as well as those of Shukla et al. [15, 16] where using a similar electron-ion interaction, the ion-ion interactions are considered on simpler basis (Born Von Karman model and axially symmetry model) thus obtaining results inferior to those given by the present studies.

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