A MODIFIED FIELEK MODEL AND PHONON DISPERSION IN BCC TRANSITION METALS

By R. N. Khanna* and R. P. S. Rathore**

Department of Physics, R. B. S. College, Agra, India

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The Fielek model is modified to account for crystal equilibrium and the Cauchy discrepancy in transition metals. The model assumes the core-core interactions to be purely central and the *d*-shell–*d*-shell interactions to be purely angular, both extending out to first neighbours only. The volume interactions are represented on the lines of the Bhatia scheme, modified to include the Umklapp processes and the effect of exchange and correlations among the electrons. The coupling between the core and *d*-shell is expressed based on the lines of the Fielek model. The simple model, employing a minimum number of input data is used to predict the dispersion relations in complicated metals like BCC Zirconium and Tantalum with a reasonable degree of success.

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1. Introduction

Recently Fielek [1] has described a model for non-simple metals, which assumes the core-core interaction to be bond streching-type as given by the axially symmetric scheme (Lehman et al. [2]) and volume interactions on the basis of Krebs [3] scheme. The model has been applied extensively (Jani and Gohel [4], Singh et al. [5–8]). All these studies require a critical review along the following lines.

(*i*) The Fielek model and its subsequent applications assume the core-conduction electron interaction of a vanishing degree. Because of the presence of intervening loosely coupled *d*-shell electrons, this interaction becomes small but its magnitude remains effective for its adequate considerations.

(*ii*) All the studies reported so far consider the lattice equilibrium separately under

* Address: Department of Physics, R. E. I. College, Dayalbagh, Agra, India.
** Address: D-132 Kamala Hagar, Agra — 262005, India.

the energies due to cores and *d*-shells. It may be mentioned that the combined effect of all the possible volume-dependent energies associated with the cores, that the *d*-shell electrons and the conduction electrons should be considered in order to arrive at the physically realistic equilibrium condition.

(*iii*) Most of the studies quoted above consider the core-core and *d*-shell–d-shell interactions to be purely of the bond stretching type. It is true that the former interactions are essentially central (Rathore and Verma [9–11], Rathore [12], Upadhyaya [13]), but the latter interactions should be related to the charge distribution of the *d*-electrons. Actually the cubic field in metals removes the five fold degeneracy and destroys the sphericity of the *d*-shell charges. This non-sphericity of overlapping *d*-shell charges clearly demands the inclusion of angular forces within the system.

(*iv*) In the Fielek model the volume interactions among the core-conduction electrons and the *d*-shell–conduction electrons are accounted for based on Krebs [3] scheme which suffers the deficiency of general inequilibrium (Cochran [14]). Moreover costly and time consuming computation is needed to sum up the series involved. In view of the recent reports on the inclusion of the equilibrium condition (Fielek [15, 16]. Shukla [17]), it is somewhat difficult and obscure to consider the equilibrium on this basis. Actually a simpler equilibrium condition, consistent with a simple scheme for the volume interactions is needed to describe the coupling between the cores and the electrons.

(*v*) In the Fielek model the Cauchy discrepancy is wholly attributed to the volume interaction among *d*-shell and conduction electrons. It may be more realistic to consider the contributions of the pressure due to conduction electrons (Rathore and Agrawal [18]) and that due to the angular interactions among *d*-shells in addition to the usual volume interaction. Moreover, a consideration of the actual amount of the discrepancy and its variation for the transition metals support this view (Hautecler and Van-Dingenen [19], Overton [20]).

In view of these remarks, it is thought worth while to modify the Fielek model for transition metals. The model considered here takes into account that:

(a) Core-core interactions are purely central and extend out to first neighbours only. The first derivative of the core-core coupling energy does not vanish because this energy is only a part of the total energy of the system. Further, the limited range of the energy is supported by the pseudopotential studies of Resolt and Taylor [21] and Degens et al. [22].

(b) *d*-shell–*d*-shell interactions are purely angular and couple only the immediate neighbours. For this purpose we have employed the best (Khanna and Rathore [23]) angular scheme of Clark et al. [24].

(c) The volume interactions on the lines of Bhatia's [25] scheme, which is modified to include (*i*) the actual form of the Bardeen [26] factor, first derived by Bross and Bohn [27] and later by Ramamurthy [28] and successfully applied by Goel et al. [29–32] and Ramamurthy and Neelkandan [33]. The inclusion of this factor makes the expression less dependent on Umklapp proceses and more symmetrical in reciprocal space. (*ii*) the screening parameter is modified properly in the light of recent reports (Singwi et al. [34]) on dielectric screening. The inclusion of $\varepsilon(q)$ needed to correct the screening for exchange and correlation effects of conduction electrons.

(d) A realistic equilibrium condition involving the volume-dependent energy due to cores and conduction electrons.

(e) The interactions among the core and d-shells as suggested by Fielek [1].

The model is adopted to calculate the dispersion relations in BCC Zirconium (β-Zr) and Tantalum (Ta).

2. Theory

The usual determinant leading to the dispersion relations may be written as

$$|D(\vec{q})-4\pi^2 M\nu^2 I| = 0, \tag{1}$$

where M is the mass of the core, ν is the dispersion frequency and I is the unit matrix of the order three. The diagonal and off-diagonal elements of the matrix $D(\vec{p})$ are expressed as

$$D_{\alpha\alpha}(\vec{q}) = \tfrac{8}{3}(\beta_1+2\alpha_1)(1-C_\alpha C_\beta C_\gamma)+K+CE(\vec{q})+\frac{K^2}{N},$$

$$D_{\alpha\beta}(\vec{q}) = \tfrac{8}{3}(\beta_1-\alpha_1)S_\alpha S_\beta S_\gamma, \tag{2}$$

where $C_\alpha = \cos(\frac{1}{2}aq_\alpha)$, $S_\alpha = \sin(\frac{1}{2}aq_\alpha)$, q_α is the α-component of the phonon wave vector q, a is the lattice constant and

$$\alpha_1 = \frac{1}{R}\left(\frac{\partial\phi_c}{\partial R}\right), \quad \beta_1 = \left(\frac{\partial^2\phi_c}{\partial R^2}\right), \tag{3}$$

where ϕ_c is the core-core potential coupling the immediate neighbours. $CE(\vec{q})$ is the matrix element representing the volume interaction among core and conduction electrons. K is the coupling constant for core and d-shells. N is given by the determinant,

$$|D'(q)-NI| = 0, \tag{4}$$

where

$$D'_{\alpha\alpha}(\vec{q}) = -16\gamma_1(1-C_\alpha C_\beta C_\gamma)+2\gamma_1(4C_{2\alpha}-C_{2\beta}-C_{2\gamma})+K-DE(\vec{q}),$$

$$D'_{\alpha\beta}(\vec{q}) = 8\gamma_1 S_\alpha S_\beta S_\gamma, \tag{5}$$

where $C_{2\alpha} = \cos(aq_\alpha)$ and γ_1 is the angular force constant of the Clark et al. [24] type. $DE(\vec{q})$ is the matrix element accounting for the volume interaction among d-shell and conduction electrons. The elements for the dynamical matrices $CE(\vec{q})$ and $DE(\vec{q})$ are written based on Bhatia's expression [25] i.e.,

$$CE_{\alpha\beta}(\vec{q}) = \frac{K_c^2\varepsilon(q)Cq_\alpha q_\beta\Omega G^2}{(K_c^2\varepsilon(q)+|q|^2)a}, \tag{6}$$

and

$$DE_{\alpha\beta}(\vec{q}) = \frac{K_c^2\varepsilon(q)Dq_\alpha q_\beta\Omega G^2}{(K_c^2\varepsilon(q)+|q|^2)a}, \tag{7}$$

where C and D are the deformation parameters for core-conduction electrons and d-shell -conduction electron potentials. K_c is the screening parameter evaluated in the Bohm–Pine [35, 36] limit, Ω is the atomic volume, G^2 is the Bardeen factor evaluated by solving the following integral over the actual atomic cell.

$$G^2 = \frac{\int_{\Omega} \exp(iq \cdot r) d\tau}{\Omega}, \tag{8}$$

and $\varepsilon(q)$ is the dielectric function given as:

$$\varepsilon(q) = \varepsilon_H(\vec{q})\,[1-G(q)]+1, \tag{9}$$

$$\varepsilon_H(\vec{q}) = 1+\frac{2K_F me^2}{\pi\hbar^2 q^2}\left\{1+\frac{4K_F^2-q^2}{4K_F q}\ln\left|\frac{2K_F+q}{2K_F-q}\right|\right\}, \tag{10}$$

$$G(q) = A[1-\exp\{-B(q/K_F)^2\}], \tag{11}$$

where

$$K_F = (3\pi^2 Z/\Omega)^{1/3}, \tag{12}$$

m is the mass of electron, e is its charge and the parameters A and B are taken from the work of Singwi et al. [34] for the inter spatial electron distant three, which is proper in view of its atomic volume and the valence of the metals under consideration.

For considering the equilibrium, the total energy ϕ_T for the system may be written as

$$\phi_T = \phi_c+\phi_d+\phi_e. \tag{13}$$

The energy of core (ϕ_c) is already included in equation (2). The energy of d-shells (ϕ_d) could not be expressed explicitly because of its obscure nature. The energy of conduction electrons (ϕ_e) comprises of Fermi, exchange and correlation parts. The Fermi and the exchange parts are explicable in definite form but the correlation part has been the subject of studies for the last forty five years. The study due to Rathore and Agrawal [18] has presented a detailed and conclusive analysis of ϕ_e in relation with the Cohesion and the Cauchy-discrepancy in metals. From this study we have inferred that the correlation part of the electron energy can suitably be given by the expression of Wigner and Seitz [37] at electron separation three.

For the general equilibrium of the lattice, the volume derivative of ϕ_T should vanish i.e.,

$$\alpha_1 = \frac{a}{2}P_e, \tag{14}$$

where P is the pressure associated with the electrons and given as

$$P_e = -\frac{\partial\phi_e}{\partial\Omega}. \tag{15}$$

Further more, the deformation parameter, C, associated with the volume interactions among core and conduction electrons may be expressed as:

$$C = -a\Omega \frac{\partial P_e}{\partial \Omega}. \tag{16}$$

3. *Calculations and results*

The model defines the dispersion relations in terms of six disposable parameters (α_1, β_1, K, γ_1, C, D). Two of these parameters are calculated using equations (14) and (16). Three of the model parameters are evaluated from elastic relations, which are obtained by comparing equation (1) with the Christoffel elastic equation in a long wave limit. The last of the model parameters is evaluated by the knowledge of an experimental frequency (ν_T,) for the transverse mode at point (100).

TABLE I

Input data for β-Zr and Ta

β-Zirconium (β-Zr)		Tantalum (Ta)	
Input data	References	Input data	References
$C_{11} = 0.783$ $C_{12} = 0.503$ } $\times 10^{12}$ dyne/cm^2 $C_{44} = 0.29$	[40]	$C_{11} = 2.609$ $C_{12} = 1.574$ } $\times 10^{12}$ dyne/cm^2 $C_{44} = 0.818$	[41]
$a = 3.64 \times 10^{-8}$ cm		$a = 3.3 \times 10^{-8}$ cm	
$m = 151.4252 \times 10^{-24}$ gm		$m = 300.377 \times 10^{-24}$ gm	
$\nu_T = 4.69$T.Hz	[38]	$\nu_T = 5.03$T.Hz	[39]

TABLE II

Computed model parameters (10^4 dyne/cm)

β-Zr	Ta
$\alpha_1 = -0.8401$	$\alpha_1 = -1.4525$
$\beta_1 = 2.5336$	$\beta_1 = 5.4166$
$K = -82.407$	$K = -1311.96$
$\gamma_1 = 0.2433$	$\gamma_1 = 0.5125$
$C = 0.2606$	$C = 0.2953$
$D = -0.8991$	$D = 0.490$

Input data and computed model parameters for β-Zirconium and Tantalum are listed in Table I and II, The calculated dispersion curves are shown by solid lines in the figure for β-Zr and Ta, respectively. The experimental data from Stassis et al. [38] and Woods

[39] for β-Zr and Ta, respectively, are also plotted with the curves for comparison. To test the utility of the present model we have shown the calculated curves of the original Fielek [1] model by dotted lines.

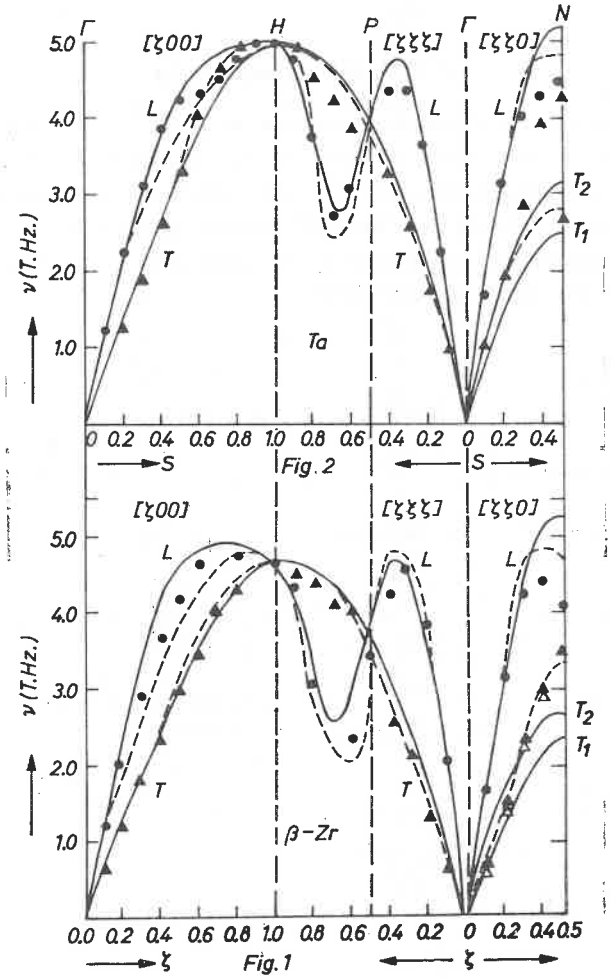

Fig. 1 and 2. Dispersion curves for β-Zr and Ta, respectively. ——— theoretical results of the present model, ····· theoretical results of the original Fielek model, and ●, ▲, △ — experimental points

4. Conclusions

A close inspection of the figure reveals that our model presents a simple successful description of phonon dispersion in complicated metals like β-Zr and Ta. The model using the minimum number of input data appears to be superior than the original Fielek model. In addition, the model incorporates the indispensable condition for lattice equili-

rium with all the effective interactions among the constituents of the transition metals. The Cauchy discrepancy is expressed as

$$aC_{12}-aC_{44} = C+D-2aP_e-8K_1. \tag{17}$$

It is therefore obvious that the electron pressure (P_e), and the angular interactions are equally effective along with the usual volume interaction in determining the discrepancy.

The slight deviations of our curves in the proximity of the zone boundary may be attributed to the deficiency of the Bhatia [25] scheme. However, these deviations loose their importance in view of the experimental errors and the different frequencies at which the input data used are measured.

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