NON CENTRAL ELECTRON FLUID MODEL FOR THORIUM

By R. M. Agrawal* and R. P. S. Rathore**

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

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The Fielek model is modified to include (a) proper equilibrium condition, (b) proper volume interaction among cores and conduction electrons and (c) proper angular interactions among the *d*-shell electrons. The model in its modified form is used to derive dispersions relations in non-simple fcc metals. The theoretical predictions using a few input data for thorium present a close agreement with the experimental ones.

1. Introduction

Calculations employing the pseudo-potential [1, 2] and the phenomenological [3–6] techniques have been reported to explain the dispersion relations in thorium. The former technique involves approximations regarding the range and the strength of the potential used while the latter technique assumes empirically the average interactions coupling the ions and the electrons. Recently Fielek [7] has developed a model which considers the interactions among various constituents separately for a non simple metal. This model has since been adopted [8–14] extensively. Most of the studies [8–13] suffer with the following shortcomings.

(a) The Fielek model [7] does not incorporate the equilibrium condition for the lattice and its inclusion in the manner outlined by Fielek [15] himself seems to be objectionable [16].

(b) The Fielek model [7] and most of its subsequent applications [8–13] ignore the volume interaction among core and conduction electrons. The said interaction is however of small magnitude but deserves consideration.

(c) The studies [8–13] based on the Fielek model [7] have assumed the interactions among *d*-shell electrons to be purely central which is hardly justified. As a matter of fact the overlapping of clouds formed by the *d*-shell electrons give rise to the non sphericity of the charge distribution which can be accounted [17, 18] for in terms of angular forces.

* Department of Physics, R. E. I. Dayalbagh, Agra-282005, India.
** Address: D-132 Kamala Nagar, Agra 282005, India.

In view of these deficiencies, the Fielek model has been modified properly. For proper inclusion of the equilibrium condition the conduction electrons behave as a fluid possessing Fermi exchange and correlation parts of the volume dependent energy. Present authors [19] have reported that the proper consideration of these energies explains both the metallic cohesion and the stability. The volume interactions among the cores and the conduction electrons as well as among the *d*-shells and the conduction electrons are assumed to follow the Bhatia [20] scheme which is modified to incorporate a proper expression for, (*i*) the inference factor G^2 to account for the convergence of the expression [20] which makes it less dependent on Umklapp terms, (*ii*) the dielectric function $\varepsilon(\vec{q})$ to account for the exchange and correlation effects among the electrons. The angular interactions among the *d*-shell electrons are considered on the lines of the most proper [21] scheme first reported by Clark et al. [22].

The Fielek model in its presently modified form is used to predict the phonon-dispersion relations in thorium. These relations are derivable in terms of a few input data and closely follow the experimental data of Reese et al. [23].

2. Theory

The determinant leading to the dispersion frequencies (ν) may be written as

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

where m is the mass of the ion, I is the unit matrix of the order three and the elements of the dynamical matrix $D(\vec{q})$ are written in terms of the first (α_1) and second (α_2) derivatives of the central pair potential coupling the first neighbours only. These interactions are so limited because of the presence of the electrons.

$$D_{ii}(\vec{q}) = -4(\alpha_2+2\alpha_1)+2(\alpha_2+\alpha_1)C_i(C_j+C_k)+4\alpha_1 C_jC_k+K+D_{ii}(E)+\frac{K^2}{N},$$

$$D_{ij}(\vec{q}) = 2(\alpha_2-\alpha_1)S_iS_j, \tag{2}$$

where $S_i = \sin(\frac{1}{2}aq_i)$, $c_i = \cos(\frac{1}{2}aq_i)$, a is the lattice constant and q_i is the i-th component of the phonon wave vector $\vec{q}$. K is the parameter representing the coupling in between cores and *d*-shells. $D_{ii}(E)$ is the dynamical matrix arising out of the volume interaction among cores and conduction electrons.

$$D_{ii}(E) = \frac{\varepsilon(q)K_c^2K_e q_i^2\Omega G^2}{\varepsilon(q)K_c^2+|q|^2}, \tag{3}$$

where Ω is the atomic volume, K_e is the bulk modulus of the electron gas and screening parameter, K_c is evaluated in Bohm–Pine [24] limit. The dielectric function $\varepsilon(\vec{q})$ may be written as

$$\varepsilon(\vec{q}) = \varepsilon_H(\vec{q})\,[1-S(\vec{q})], \tag{4}$$

where $\varepsilon_H(\vec{q})$ is the usual Hartree dielectric function and the correction term $S(\vec{q})$, accounting for the exchange and correlation effects among conduction electrons is taken from the recent work of Mandal et al. [25].

The correct form of inference factor (G^2) is obtained by evaluating the following integral over the actual polyhedron of volume Ω.

$$G^2 = \int_0^{\Omega} \frac{\exp(i\vec{q}\cdot\vec{r})d\Omega}{\Omega}. \tag{5}$$

N in equation (2) is evaluated from determinant

$$|D'(\vec{q}) - NI| = 0, \tag{6}$$

where

$$D'_{ii}(\vec{q}) = 16\chi_1[2 - C_i(C_j + C_k)] - 4\chi_1[2C'_i - C'_j - C'_k] + K + D'_{ii}(d),$$

$$D'_{ij}(\vec{q}) = -16\chi_1 S_i S_j,$$

where $C'_i = \cos(aq_i)$ and χ_1 is the angular force parameter of the type given by Clark et al. [22] and coupling the first neighbouring d-shells only because of the immediate vicinity of the conduction electrons. The dynamical matrix $D'_{ii}(d)$ expressing the binding among the d-shell electrons and the conduction electrons may be written as

$$D'_{ii}(d) = \frac{\varepsilon(q)K_c^2 K_d q_i^2 \Omega G^2}{\varepsilon(q)K_c^2 + |q|^2}, \tag{7}$$

where the screening parameter K_c is evaluated in Thomas–Fermi [26] limit, and aK_d is the deformation parameter associated with the clouds of d-electrons.

For considering the equilibrium of the whole lattice, the total energy E may be written as

$$E = E_c + E_e + E_d. \tag{8}$$

For the equilibrium

$$\frac{\partial E}{\partial \Omega} = 0. \tag{9}$$

Obviously

$$\frac{\partial E_c}{\partial \Omega} = \frac{4}{a}\alpha_1 \tag{10}$$

and

$$\frac{\partial E_e}{\partial \Omega} = -P_e, \tag{11}$$

where P_e is the pressure associated with the conduction electrons. Further

$$\Omega \frac{\partial P_e}{\partial \Omega} = -K_e. \tag{12}$$

The expression for E_e is taken from our earlier work [19]. No proper expression for E_d is however available in the literature because of the complex behaviour of the electrons occupying the *d*-levels.

Combining equations (10) and (11), we could get the equilibrium condition

$$\alpha_1 = \frac{a}{4} P_e. \tag{13}$$

For the electron separation $\gamma_s = 4$, the proper values of $P_e (= -0.9631 \times 10^{10}$ dyne/cm^2) and $K_e = (0.9838 \times 10^{10}$ dyne/cm^2) are given by the correlation scheme of Mandal et al. [25]. It may however be noted that the contribution due to E_d is not considered in Eq. (13) because of its indefinite nature due to the transitions.

3. Calculations and results

The model comprises of six parameters (α_1, α_2, K, χ_1, aK_d and aK_e). Two of the model parameters are evaluated using equations (12) and (13). Three of the remaining parameters are evaluated using usual elastic relations which are obtained by comparing the long

TABLE I

Input data and model parameters for thorium

Input data	References	Model parameters (10^4 dyne/cm)
$C_{11} = 0.753 \times 10^{12}$ dyne/cm^2		$\alpha_1 = -0.01223$
$C_{12} = 0.489 \times 10^{12}$ dyne/cm^2	[27]	$\alpha_2 = -2.392$
$C_{44} = 0.478 \times 10^{12}$ dyne/cm^2		$K = -2.147$
$a = 5.08$ Å		$\chi_1 = +0.02367$
$m = 232.12$ amu		$aK_d = -0.2749$
$\nu_{T(111)} = 1.28$ T.H_z.	[23]	$aK_e = +0.04999$

wave length form of equation (1) with the well known Christofell's equation of elasticity. The last model parameter is evaluated by the knowledge of zone boundary frequency for transverse mode at the point (111).

The input data and the computed model parameters for thorium are shown in Table I.

The computed dispersion relation along the principal symmetry directions are shown in Fig. 1. The experimental points (×, ●, ○) are shown alongside the curves for comparison purpose. The maximum deviation of the computed curves lies at the zone boundary for $\nu_{L\,[\zeta\zeta\zeta]}$ and $\nu_{T_2\,[\zeta\zeta 0]}$ each of which is about 10%. In general the agreement is good and the efficacy of the model seems to be fairly satisfactory.

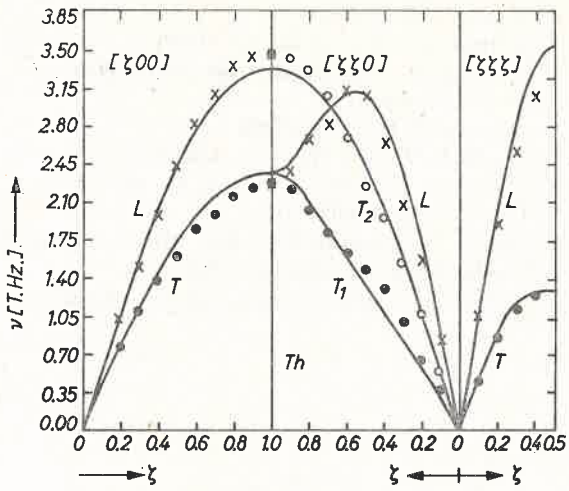

Fig. 1. Dispersion curves for thorium: (———) computed from the model, (×, ●, ○) experimental points due to Reese et al. [23]

4. Conclusion

The model, using only four input data, describes the phonon dispersion in complicated metal like thorium with fair satisfaction. Further, the model, being sound in its mathematical contents, is free from the deficiencies quoted earlier. The model is however somewhat asymmetric in reciprocal space because of the use of the Bhatia [20] expression but this shortcoming is counter balanced by the use of correct form of G^2 and $\varepsilon(\vec{q})$. A simple model involving easy computations reproduces the phonon-dispersion in complicated metal like thorium with fair degree of agreement.

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