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Lattice dynamical study of body-centred tetragonal indium

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Abstract. The phonon dispersion curves, phonon frequency distribution function as well as the lattice specific heat of body-centred tetragonal indium have been deduced using a lattice dynamical model which includes central, angular and volume forces. Six elastic constants, four zone boundary frequencies and an equilibrium condition were used in the evaluation of the force constants. It is shown that this model is elastically consistent and satisfies the symmetry requirements of the lattice, the phonon frequencies of indium deduced from it are in very good agreement with the experimental values of Reichardt and Smith and the theoretical values of Garrett and Swihart, and the θ_D values compare well with the experimental values over a wide temperature range. The apparent discrepancies in the phonon dispersion curves and the θ_D-T curves obtained from deficient models, importance of umklapp processes and the significance of angular forces in the lattice dynamical models are discussed.

Keywords. Body-centred tetragonal indium; CGW angular forces; elastic consistency; translational symmetry; phonon dispersion curves; lattice specific heat; lattice dynamics.

1. Introduction

In spite of its superconductivity, the lattice dynamics of indium has not received sufficient theoretical attention so far mainly because of the increase in the number of force constants that accompany the transition from cubic to tetragonal symmetry and nonavailability of experimental data on phonon frequencies to evaluate these constants. Even the few attempts made by imposing unrealistic restrictions on the nature and range of interatomic interactions or arbitrary constraints on the force constants suffer from serious deficiencies. For instance, Slutsky and Livingston (1960) have studied the lattice dynamics of indium on the basis of the tensor force constant model (Begbie and Born 1946), restricting the range of interactions to first two neighbours whereas Sharan and Bajpai (1969, 1970) made use of angular forces or volume forces along with central forces for this purpose. However all these models are elastically inconsistent as the expression for C_{44} obtained from D_{xx} element along the [00ζ] direction differs from that obtained from D_{zz} element along the [ζ00] direction. Besides their calculations have gone wrong as they made use of elastic constants of indium referred to FCT axes (Winder and Smith 1958) to evaluate the force constants referred to BCT axes. Further, Ashokkumar (1973) and Sharan and Ashokkumar (1973) have used central forces extending up to fourth neighbours, angular forces with two force constants as well as volume forces with a theoretical

value for the bulk modulus, K_e of the electron gas (Ashokkumar *et al* 1973) in their study of indium. Neither of these models satisfies the symmetry requirements of the lattice even though the latter is elastically consistent. On the contrary Reissland and Ese (1975) and Garrett and Swihart (1976) have made use of the optimized model potential (Shaw 1968) with the effective electron mass, m^* as a free parameter to investigate the lattice dynamics of BCT and FCT indium, respectively, but there is hardly any agreement between these two sets of phonon frequencies referred to the same tetragonal axes.

It is therefore obvious that a realistic force constant model free from all inadequacies mentioned above, is required for a proper study and understanding of the lattice dynamics of indium. In this context one can exploit the claim of Garrett and Swihart (1976) that their results with $m^* = 0.8$ are in quantitative agreement with experimental phonon frequencies measured using the inelastic scattering of neutrons (supplied to them by W Reichardt and HG Smith, but unpublished), elastic constants (Chandrasekhar and Rayne 1961) as well as with the temperature dependence of specific heat (Clement and Quinnell 1953; Bryant and Keesom 1961) of indium and use their zone boundary frequencies to evaluate additional force constants. Keeping this in view, the present authors (Ramamurthy and Rajendraprasad 1981) have expressed the ion-ion interactions up to the third neighbour in terms of the modified axially symmetric forces (Dewames and Lehman 1964, Dewames *et al* 1965) and the electron-ion-interactions in terms of an electron gas model based on the deformation potential approximation (Ramamurthy and Neelakandan 1978). Besides the inclusion of umklapp processes ensures that this model is consistent with the translational symmetry of the lattice. Nevertheless, this model suffers from elastic inconsistency and the phonon frequencies deduced from it are in no way better than those deduced from earlier models (Ashokkumar 1973, Sharan and Ashokkumar 1973, Reissland and Ese 1975), in spite of the fact that the former are in very good agreement with the experimental frequencies and those deduced by Garrett and Swihart (1976). The present authors have therefore developed an elastically consistent force constant model in which the modified axially symmetric forces of their earlier model (Ramamurthy and Rajendraprasad 1981) are replaced by the central forces and angular forces incorporated in a manner suggested by Clark *et al* (1964). The present paper describes this model which satisfies the symmetry requirements of the lattice as well, and the lattice dynamical study of indium based on this new model.

2. Theory

2.1 *Secular determinant*

Proceeding in the usual way the secular determinant for lattice vibrations may be written as

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

where $\omega_{\mathbf{q}}$ is the frequency of the normal mode with wave vector $\mathbf{q}$ and $\hat{I}$ is the 3×3 unit matrix. Assuming that the atomic interactions in indium consist of central,

angular and volume forces, the dynamical matrix $D(\mathbf{q})$ is expressed as a sum of three terms, *viz.*

$$D(\mathbf{q}) = D^r + D^l + D^e, \tag{2}$$

where D^r and D^l represent the contributions arising from central forces and angular forces, respectively to the ion-ion interactions and D^e represents the contribution from volume forces to the electron-ion interactions. If the range of central force interactions is extended up to fourth neighbours to include all atoms in the unit cell, the elements of the matrix D^r are given by (Sharan and Ashokkumar 1973)

$$MD^r_{xx} = 4\,\alpha_1 S_x^2 + \frac{8\,\alpha_2}{(2+t^2)}[1 - C_x C_y C_z] + 2\,\alpha_3 [1 - C_{2x} C_{2y}], \tag{3a}$$

$$MD^r_{xy} = \frac{8\,\alpha_2}{(2+t^2)} S_x S_y C_z + 2\,\alpha_3 S_{2x} S_{2y}, \tag{3b}$$

$$MD^r_{xz} = \frac{8\,t\,\alpha_2}{(2+t^2)} S_x C_y S_z \tag{3c}$$

and

$$MD^r_{zz} = \frac{8\,t^2\,\alpha_2}{(2+t^2)}[1 - C_x C_y C_z] + 4\,\alpha_4 S_z^2, \tag{3d}$$

where $S_x = \sin(q_x a)$, $S_{2x} = \sin(2q_x a)$, $C_x = \cos(q_x a)$ and

$C_{2x} = \cos(2q_x a)$, etc.,

M is the mass of the ion, α_n is the nth neighbour central force constant and $t = c/a$, $2a$ and $2c$ being the lattice constants of BCT indium. The remaining elements of D^r are obtained by interchanging the subscripts x and y in (3).

Angular forces in the CGW model (Clark *et al* 1964) arise from the resistance to deformation of the angles of a triangle formed by joining three lattice points, but a variety of triangles can be formed by bonds between different kinds of neighbours. However the total angular energy should include the self-consistent set of triangles comprised of all combinations of interatomic distances upto the largest one considered. It is invariant under rigid body rotations only if none of the angles in these triangles is excluded. The isosceles triangles of type I, III and IV formed by joining an atom with two of its second neighbours separated by first, third and fourth neighbour distance, respectively, constitute a complete set when the range of these interactions is restricted to first four neighbours only. There are, with reference to the atom at the origin, 24 triangles of type I, 12 triangles each of types III and IV in this set. The first, third and fourth neighbour distances subtend angles θ_1, θ_3 and θ_5 in respective triangles whereas the second neighbour distances subtend angles θ_2, θ_4 and θ_6 respectively in triangles I, III and IV. The elements of the matrix D^l obtained by summing over these six angles in the set are given by

$$MD^l_{xx} = \frac{8}{(2+t^2)}\left[\frac{4\gamma_1 + 2p\gamma_2}{(1+t^2)} + t^2(\gamma_4 + 2\gamma_5 + \gamma_6)\right](1 - C_x C_y C_z) + \frac{4t^2\gamma_3}{(2+t^2)} \times$$

$$\times (1-C_{2x}\, C_{2y}) + \frac{16\,(1+t^2)\,\gamma_1}{(2+t^2)} S_x^2 - \frac{8}{(1+t^2)}\left[\frac{2\gamma_1}{(2+t^2)} - t^2\,\gamma_2\right] S_y^2$$

$$- 4\left[\frac{2t^4\,\gamma_5+(t^4-4)\,\gamma_6}{t^2\,(2+t^2)}\right] S_z^2, \tag{4a}$$

$$MD^l_{xy} = \frac{8}{(2+t^2)}[-4\gamma_1 + 2t^2\,(\gamma_2+\gamma_5) + t^2\,(\gamma_4+\gamma_6)]\; S_x\, S_y\, C_z$$

$$+ \frac{4t^2\,\gamma_3}{(2+t^2)} S_{2x}\, S_{2y}, \tag{4b}$$

$$MD^l_{xz} = -\frac{8t}{(2+t^2)}\left[\frac{2\,(t^2-1)\,\gamma_1 + (t^2-p)\,\gamma_2}{(1+t^2)} + 2\,(\gamma_3+\gamma_5)\right.$$

$$\left. - \frac{(t^2-2)\,(t^2\,\gamma_4 - 2\,\gamma_6)}{2\,t^2}\right] S_z\, C_y\, S_z, \tag{4c}$$

and

$$MD^l_{zz} = \frac{32}{(2+t^2)}\left[\frac{t^2\,(2\,\gamma_1+\gamma_2)}{(1+t^2)} + (2\,\gamma_3+\gamma_4+\gamma_6)\right](1 - C_x\, C_y\, C_z)$$

$$- \left[\frac{16\,\gamma_3 - 2\,(t^4-4)\,\gamma_4}{(2+t^2)}\right](1 - C_{2x}\, C_{2y})$$

$$- \frac{8}{(1+t^2)}\left[\frac{2t^2\,\gamma_1}{(2+t^2)} - t^4\,\gamma_2\right](S_x^2 + S_y^2) + \frac{32\,\gamma_5}{(2+t^2)} S_z^2, \tag{4d}$$

where $p = (2 + 2t^2 + t^4)$ and γ_n is the angular force constant associated with the angle θ_n. In order to facilitate the comparison of these force constants with those associated with FCT lattice and to make them dimensionally equal to the central force constants, they have been divided by a normalizing factor, $(2 + t^2)\, a^2$.

The volume forces which owe their origin to the presence of conduction electrons, are expressed in terms of the deformation potential. When the exchange and correlation effects are taken into account through an appropriate screening function, the elements of the matrix D^e which include the contributions due to umklapp processes are given by (Ramamurthy and Singh 1978)

$$MD^e_{xy} = 2\,C' \sum_{\mathbf{g}}\left[\frac{(g-q)_x\,(g-q)_y\,S\,(\mathbf{g}-\mathbf{q})\,F\,(g-q)}{4\,(g-q)^2 + K_1\,F\,(g-q)}\right], \tag{5}$$

where C' is an adjustable constant related to the bulk modulus, K_e of the electron gas $\mathbf{g}$ is a reciprocal lattice vector, $K_1 = a^{-2}$ and $S\,(\boldsymbol{\eta})$ is the interference factor defined by

$$S\,(\boldsymbol{\eta}) = \int_{\Omega} \exp\,(i\,\boldsymbol{\eta}\cdot\mathbf{r})\;\mathrm{d}\,\tau/\Omega, \tag{6}$$

and has been evaluated exactly over the actual shape of the Wigner-Seitz cell for the BCT lattice (Sharan *et al* 1972, Ramamurthy 1979). $F(\eta)$ is a screening function which is determined by the choice of the screening approximation. A number of expressions exist in the literature and the following expressions for $F(\eta)$ were used in the present investigation:

(i) the simple random-phase approximation

$$F(\eta) = F_0(\eta) = 2a^2 K_s^2 \left[1 + \frac{4K_F^2 - \eta^2}{4K_F \eta} \ln \left| \frac{2K_F + \eta}{2K_F - \eta} \right| \right], \tag{7}$$

where K_s is the Thomas-Fermi screening parameter and K_F is the Fermi wave vector;

(ii) the Hubbard-Sham approximation (Sham 1965)

$$F(\eta) = F_0(\eta) \left[1 - \frac{F_0(\eta)}{8a^2 (\eta^2 + K_F^2 + K_s^2)} \right]^{-1}; \tag{8}$$

(iii) the Vashishta-Singwi (1972) approximation

$$F(\eta) = F_0(\eta) \left[1 - \frac{F_0(\eta) A}{4a^2 \eta^2} \{1 - \exp [-B(\eta/K_F)^2]\} \right]^{-1}, \tag{9}$$

where the values of the constants A and B for indium are given by 0·9054 and 0·3202 respectively.

The first term on the right hand side of (5) corresponding to $\mathbf{g} = 0$ represents the contribution of normal processes to the elements of D^e. Since these expressions do not contribute to the transverse modes of vibration, earlier models on indium (Sharan and Bajpai 1970, Ashokkumar 1973, Sharan and Ashokkumar 1973) which ignored the contributions from umklapp processes to electron-ion interactions, do not satisfy the symmetry requirements of the lattice even when the interference factor in (6) is evaluated over an ellipsoid (instead of a sphere) of equal volume or over the actual shape of the Wigner-Seitz cell. In the present calculations, nearly 200 terms representing the contribution of umklapp processes associated with several reciprocal lattice vectors are included in the sum and the elements of D^e so obtained contribute to the longitudinal as well as transverse modes and are consistent with the translational symmetry of the lattice. Besides, these expressions reduce to a convenient form as the exact evaluation of (6) leads to

$$S(\boldsymbol{\eta}) = 0 \quad \text{if } \boldsymbol{\eta} = \mathbf{g} \neq 0. \tag{10}$$

2.2 *Force constant evaluation*

The numerical values of six elastic constants, four zone boundary frequencies along the principal symmetry directions of the crystal as well as an equilibrium condition which must be imposed on the force constant models explicitly to ensure lattice cohesion, are made use of to evaluate the eleven force constants that appear in the

dynamical matrix elements. For this purpose the secular determinant (1) is taken to the long wavelength limit and compared with the elastic determinant for a BCT lattice. This gives the following six relations between the elastic and force constants:

$$cC_{11} = a_1 + \frac{a_2}{(2+t^2)} + a_3 + \frac{2p\,\Gamma_1}{(1+t^2)(2+t^2)} + \frac{t^2\,\Gamma_2}{(2+t^2)} + \frac{C'}{2}, \quad (11a)$$

$$cC_{12} = \frac{a_2}{(2+t^2)} + a_3 - \frac{4\,\Gamma_1}{(2+t^2)} + \frac{t^2\,\Gamma_2}{(2+t^2)} + \frac{C'}{2}, \quad (11b)$$

$$cC_{13} = \frac{t^2\,a_2}{(2+t^2)} - \frac{2t^4\,\Gamma_1}{(1+t^2)(2+t^2)} - \frac{2t^2\,\Gamma_2}{(2+t^2)} + \frac{C'}{2}, \quad (11c)$$

$$cC_{33} = \frac{t^4\,a_2}{(2+t^2)} + t^2\,a_4 + \frac{4t^4\,\Gamma_1}{(1+t^2)(2+t^2)} + \frac{4t^2\,\Gamma_2}{(2+t^2)} + \frac{C'}{2}, \quad (11d)$$

$$cC_{44} = \frac{t^2\,a_2}{(2+t^2)} + \frac{2t^2\,(2\gamma_1 + p\gamma_2)}{(1+t^2)(2+t^2)} + \frac{(t^4\,\gamma_4 + 4\,\gamma_6)}{(2+t^2)}, \quad (11e)$$

and

$$cC_{66} = \frac{a_2}{(2+t^2)} + a_3 + \frac{4\,(1+t^2)\,\gamma_2}{(2+t^2)} + \frac{t^2\,\Gamma_2}{(2+t^2)}, \quad (11f)$$

where $\Gamma_1 = (2\gamma_1 + \gamma_2)$ and $\Gamma_2 = (2\gamma_3 + \gamma_4 + 2\gamma_5 + \gamma_6)$.

Further, the secular determinant is solved at the zone boundary points (100), (001) and $(\frac{1}{2}\,\frac{1}{2}\,0)$ to obtain relevant relationships between these frequencies and the force constants. These are given by

$$4\pi^2\,M\,\nu_L^2\,(100) \equiv 4\pi^2\,M\,\nu_T^2\,(001)$$

$$= \frac{16}{(2+t^2)}\left[a_2 + \frac{2(2\gamma_1 + p\gamma_2)}{(1+t^2)} + t^2\,(\gamma_4 + 2\gamma_5 + \gamma_6)\right]$$

$$+ 2C'\,SL\,(100), \quad (12a)$$

$$4\pi^2\,M\,\nu_L^2\,(001) \equiv 4\pi^2\,M\,\nu_{T_2}^2\,(100)$$

$$= \frac{16t^2}{(2+t^2)}\left[a_2 + \frac{4(2\gamma_1 + \gamma_2)}{(1+t^2)} + \frac{4(2\gamma_3 + \gamma_4 + \gamma_6)}{t^2}\right]$$

$$+ 2C'\,SL\,(001), \quad (12b)$$

$$4\pi^2\,M\,\nu_{T_1}^2\,(\tfrac{1}{2}\,\tfrac{1}{2}\,0) = 4a_1 + \frac{8(2+t^2)\,(2\gamma_1 + \gamma_2)}{(1+t^2)} + 2C'\,ST_1\,(\tfrac{1}{2}\,\tfrac{1}{2}\,0) \quad (12c)$$

and $$4\pi^2 M \nu_{T_2}^2 (\tfrac{1}{2}\,\tfrac{1}{2}\,0) = \frac{8t^2}{(2+t^2)}\left[\alpha_2 + \frac{2(2\gamma_1 + p\gamma_2)}{(1+t^2)} + \frac{4(2\gamma_3 + \gamma_4 + \gamma_6)}{t^2}\right]$$

$$+ 2C'\, ST_2 (\tfrac{1}{2}\,\tfrac{1}{2}\,0), \tag{12d}$$

where $SL(100)$, $SL(001)$, $ST_1(\frac{1}{2}\,\frac{1}{2}\,0)$ and $ST_2(\frac{1}{2}\,\frac{1}{2}\,0)$ correspond to the umklapp contributions to the appropriate modes of vibration, represented by [ ] in (5) which is summed over several reciprocal lattice vectors at the respective zone boundary points.

The lattice equilibrium condition, determined by minimizing the total potential energy of the crystal assumes the form

$$\gamma_1 + \tfrac{1}{2}\gamma_2 + 2\gamma_3 + \gamma_4 + 2\gamma_5 + \gamma_6 + C' = 0. \tag{13}$$

The experimental values for the elastic constants and the zone boundary frequencies of BCT indium were obtained from the measurements of Chandrasekhar and Rayne (1961) and Reichardt and Smith (Garrett and Swihart 1976), respectively, on FCT indium by a coordinate axes transformation (Sharan and Ashokkumar 1973) and are given in table 1. Numerical values of the axial ratio and the force constants calculated using the relevant data from table 1 are listed in table 2.

3. Results

The phonon frequencies of indium have been calculated by solving the secular determinant along the principal symmetry directions of the crystal as well as at 102 selected

Table 1. Relevant experimental data for body centred tetragonal indium.

M (10^{-27} kg)	$2a$ (10^{-10} m)	$2c$	ν_L (100)	ν_L (001) (THz)	$\nu_{T_1}(\frac{1}{2}\,\frac{1}{2}\,0)$	$\nu_{T_2}(\frac{1}{2}\,\frac{1}{2}\,0)$	Reference
190·6	3·244	4·938	1·573	3·336	2·256	1·707	Garrett and Swihart (1976)*
T (K)	C_{11}	C_{12}	C_{13} (10^{10} nm^{-2})	C_{33}	C_{44}	C_{66}	
4·2	6·315	2·947	4·513	5·162	0·797	0·760	Chandrasekhar and Rayne (1961)*

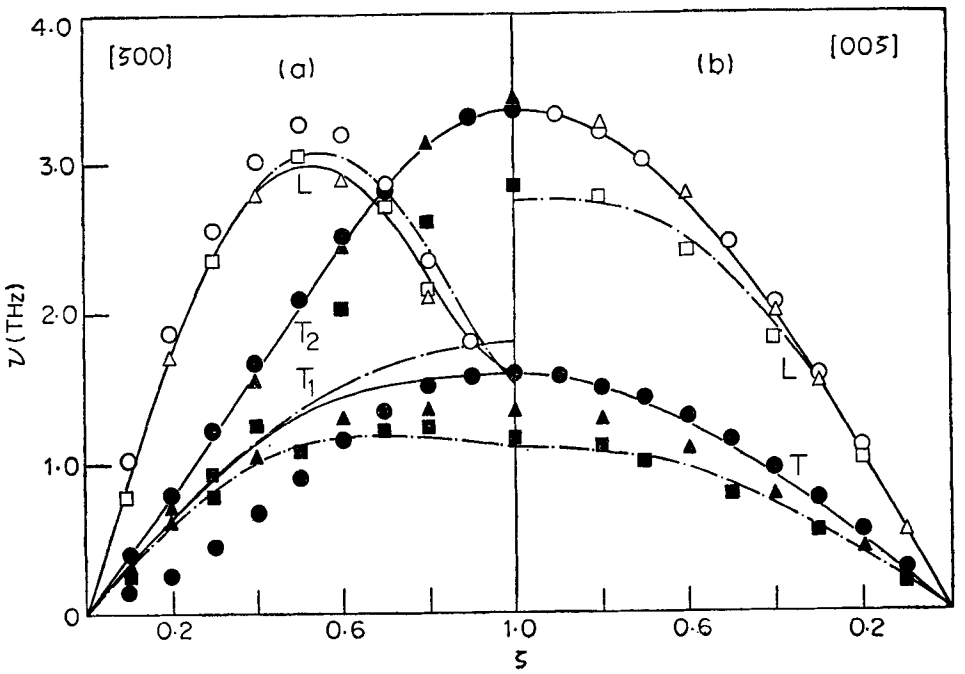
*measured on face centred tetragonal indium.

Table 2. Numerical values of the axial ratio and the force constants (in nm^{-1}).

t	α_1	α_2	α_3	α_4	C'
1·522	12·214	11·306	2·978	1·856	3·022
γ_1	γ_2	γ_3	γ_4	γ_5	γ_6
−0·342	−0·814	−0·933	−6·381	−0·907	7·789

wave vectors spread over the reduced Brillouin zone, using three different screening functions, *viz.* (7), (8) and (9) for $F(\eta)$ in the evaluation of the force constants. The increase in the number of force constants precludes the possibility of using different sets of zone boundary frequencies or elastic constants in their evaluation. The numerical values of the force constants are altered with $F(\eta)$ but it was found that the phonon frequencies deduced from three sets of force constants are not significantly different from each other. Hence only the results obtained from a typical set shown in table 2 are presented here. The dispersion curves obtained by plotting these phonon frequencies as a function of the reduced wave vector, ζ along [ζ 00], [00 ζ] and [$\zeta\zeta$ 0] directions are shown in figures 1a, b and c, respectively, whereas those along a nonsymmetry direction, [$\zeta\zeta 2\zeta$] are shown in figure 1d. In order to test the validity of present authors' force constant model, it is necessary to compare these frequencies with those measured using inelastic scattering of neutrons. For this purpose, the corresponding frequencies of BCT indium obtained, by a coordinate axes transformation from the theoretical phonon frequencies of FCT indium (Garrett and Swihart 1976) which are in good agreement with their experimental values, are plotted as a function of ζ in figure 1. Besides, phonon frequencies of indium calculated on the basis of (i) earlier force constant models (Slutsky and Livingston 1960, Sharan and Ashokkumar 1973) using correct values for the elastic constants in the former, (ii) optimized model potential (Reissland and Ese 1975) as well as (iii) the present authors' earlier model (Ramamurthy and Rajendraprasad 1981) are shown in these figures to facilitate their comparison with the present theoretical frequencies.

To determine the phonon frequency distribution function, $g(\nu)$ the Brillouin zone is divided into $10 \times 10 \times 10$ parts and each of the calculated frequencies at the 102 irreducible wave vectors is given the proper statistical weight. The entire frequency



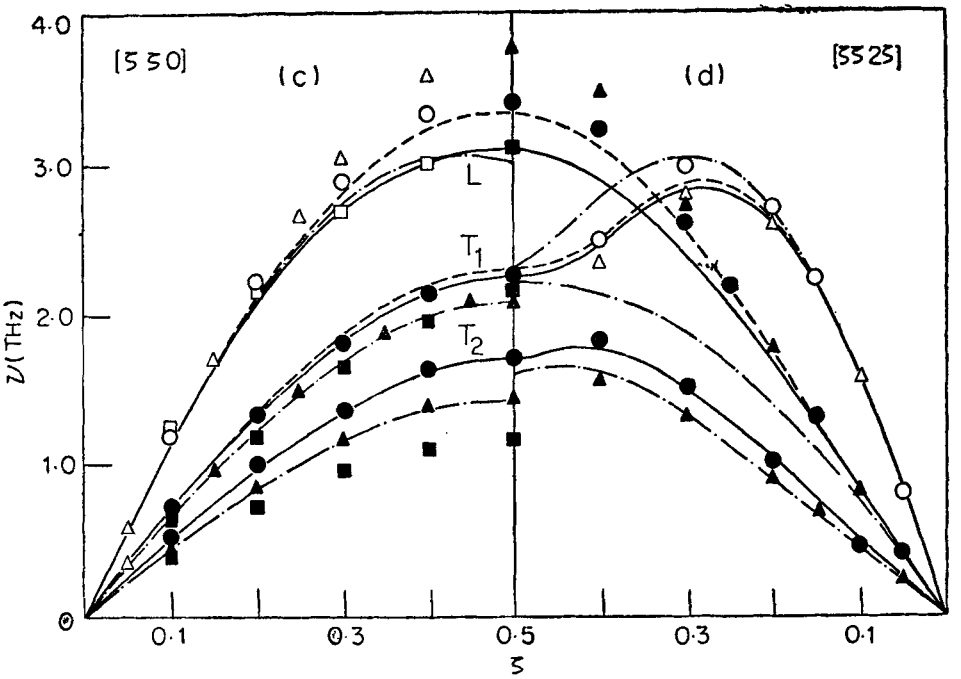


Figure 1. Dispersion curves of indium along (a) [ζ00] direction, (b) [00ζ] direction, (c) [ζζ0] direction and (d) [ζζ2ζ] direction.

Present work ——————

Ramamurthy and Rajendraprasad (1981) — — — —

Sharan and Ashokkumar (1973) — · — · — · —

Garrett and Swihart (1976) (along with experimental values) L ○ T ●;

Reissland and Ese (1975) L □ T ■;

Slutsky and Livingston (1960) (Corrected for elastic constant errors); L △ T ▲;

T_2 refers to polarization along [001] direction.

range is divided into intervals of 0·2 THz. Figure 2 shows the phonon frequency histogram, obtained by root sampling technique (Dayal and Sharan 1960, 1961), which has been made use of in the evaluation of the lattice specific heat in the temperature range of 5–250 K. The equivalent Debye temperature, θ_D calculated from these theoretical values is plotted as a function of temperature in figure 3. Besides, θ_D values calculated from the values of lattice specific heat obtained from earlier force constant models (Slutsky and Livingston 1960; Sharan and Ashokkumar 1973, Ramamurthy and Rajendraprasad 1981) using the correct values of elastic constants in the first and the correct expression for specific heat in second as well as those from the experimental values of specific heat (Clusius and Schachinger 1952; Clement and Quinnell 1953) are shown in figure 3 to facilitate their comparison with the present theoretical values.

4. Discussion

It is obvious from figures 1a to d that the phonon frequencies of indium calculated on the basis of the present authors' force constant model are in very good agreement

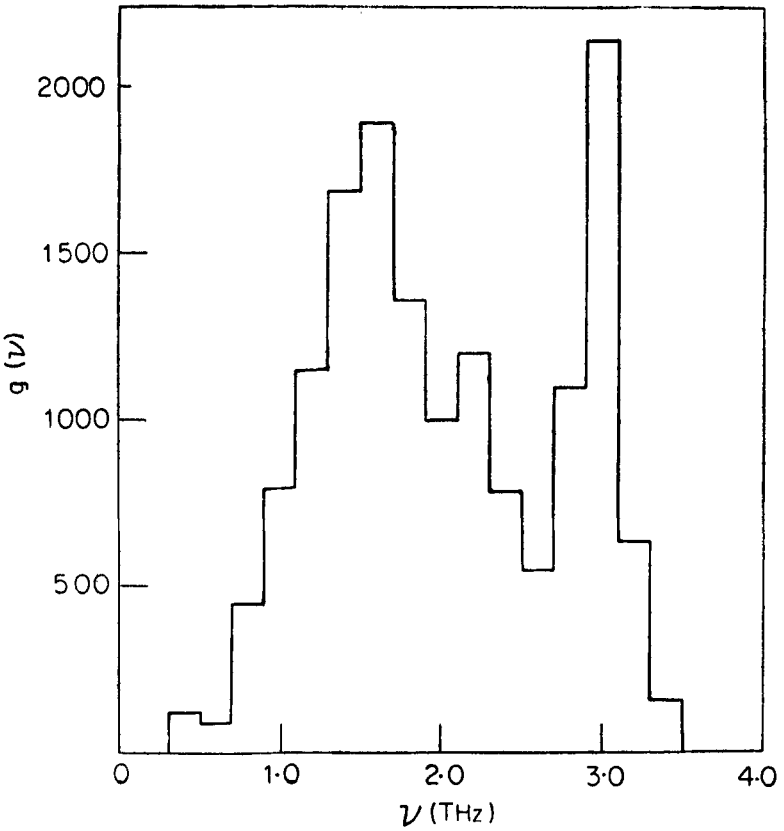


Figure 2. Phonon frequency distribution function $g(\nu)$ for the present model of indium.

with the corresponding frequencies deduced by Garrett and Swihart (1976), the only exception being the phonon frequencies of $[\zeta 00]$ T_1 branch upto $\zeta = 0{\cdot}75$. The latter are anomalously smaller than the former in this region resulting in a dispersion curve with a kink at $\zeta = 0{\cdot}2$. No other calculation on indium reported so far has predicted this anomaly. Nevertheless four zone boundary frequencies of the latter have been made use of to evaluate the force constants in the present model (as well as in the previous model) on the assumption that they are in quantitative agreement with their experimental values. It is clear from these figures that the phonon frequencies so obtained differ by not more than 9% from the corresponding experimental and theoretical frequencies reported in the latter and the maximum discrepancy between the two occurs at $[\frac{1}{2}\ \frac{1}{2}\ 0]$ L, whereas they are so close to those obtained from the present authors' previous model (Ramamurthy and Rajendraprasad 1981) that the two sets cannot be shown separately except around $\zeta = 0{\cdot}5$ in figures 1c and d where the difference between the two is $< 7\%$. Besides these frequencies are in complete agreement with those deduced from tensor force model (Slutsky and Livingston 1960) at all wave vectors along $[\zeta 00]$ T_2, $[\zeta 00]$ L and $[00\ \zeta]L$ as well as at short wave vectors along other directions provided that the force constants of the latter are evaluated using the proper values of the elastic constants. However, the longitudinal and transverse frequencies of the former are respectively higher and lower than the corresponding frequencies of the latter along other directions and the

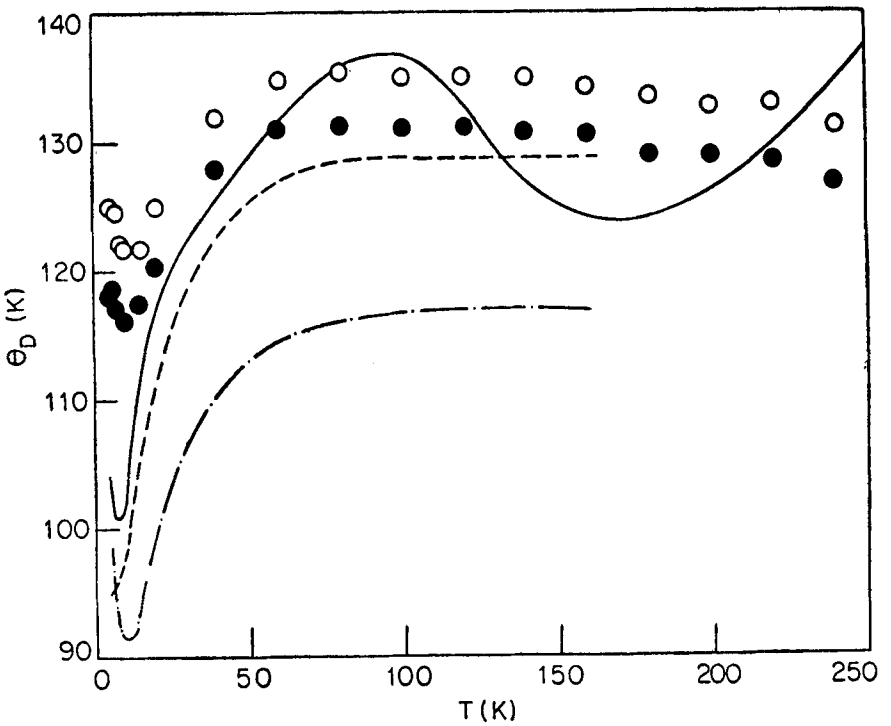


Figure 3. $\theta_D - T$ curves for indium.
Experimental ————
Slutsky and Livingston (1960) — — — —
(Corrected for elastic constant errors)
Sharan and Ashokkumar (1973) — · — · — · —
(corrected for electronic specific heat errors)
Ramamurthy and Rajendraprasad (1981) o
Present work. ●

discrepancies between the two increase with the wave vector, reaching their respective maximum values of $\sim 20\%$ and $\sim 12\%$ at the zone boundary point $(\frac{1}{2}\ \frac{1}{2}\ 0)$. On the contrary, these frequencies are invariably higher than those deduced by Sharan and Ashokkumar (1973) and Reissland and Ese (1975), the only exception being the longitudinal frequencies along $[\zeta 00]$ and $[\zeta\zeta 0]$ directions, but the latter two sets of frequencies are in excellent agreement with each other at all wave vectors and polarizations.

Figure 1 reveals that the phonon dispersion curves obtained by Sharan and Ashokkumar (1973) do not satisfy the symmetry requirements of the lattice and hence they are in poor agreement with all those which do satisfy these requirements, especially at wave vectors where two or more of these branches become degenerate. These authors evaluated the force constants associated with ion-ion interactions by making use of a theoretical value for the electron gas constant, C' (or K_e) (Ashokkumar *et al* 1973) which is approximately four times its value in the present model. Nevertheless the stronger electron-ion interactions together with the consequent weaker ion-ion interactions give rise to longitudinal frequencies which are in approximate agreement with those of other calculations, even when the umklapp processes are not considered. On the contrary the transverse frequencies of this model are very much smaller than those of others and they do not become degenerate with other

longitudinal frequencies at $\zeta=0{\cdot}5$ or $1{\cdot}0$, mainly because the former interactions do not contribute to the transverse modes when the umklapp processes are excluded. It is therefore obvious that the present model which includes contributions from umklapp processes over several reciprocal lattice vectors is superior to this model, the phonon frequencies deduced from the former are consistent with the translational symmetry of the lattice whereas those from the latter are not and the apparent discrepancies between the two have hardly any physical significance. In this context it is not at all clear how Reissland and Ese (1975) and Garrett and Swihart (1976) made use of the same pseudopotential viz. optimized model potential of Shaw (1968) and obtained phonon frequencies which do not agree with each other but are in very good agreement, respectively, with those of Sharan and Ashokkumar (1973) and with the experimental values of Reichardt and Smith (as well as those from present calculations) at several wave vectors and polarizations.

The excellent agreement between the phonon frequencies deduced from the present authors' previous and present models seem to imply that the phonon dispersion curves are not affected by the elastic inconsistency of a model. However the apparent inconsistencies between these dispersion curves and those of Slutsky and Livingston (1960) obtained by using proper values of elastic constants, should be attributed to the arbitrary constraint imposed on the force constants of the latter in order to make the two different expressions for C_{44} equal and hence have no significance. On the other hand the elastic consistency of Sharan and Ashokkumar model (1973) is of no consequence, as the phonon frequencies deduced from models with no translational symmetry have far more serious discrepancies than those from models with elastic inconsistency. It should therefore be obvious that the present force constant model with elastic consistency and translational symmetry is superior to all other force constant models considered above and to some pseudopotential models (Reissland and Ese 1975). The quantitative agreement among the experimental phonon frequencies of indium, those deduced by Garrett and Swihart (1976) and by the authors clearly indicates that the present model could be used as an alternative to pseudopotential models in the lattice dynamical study of indium and other tetragonal metals.

All force constant models based on either axially symmetric forces or general tensor forces are elastically consistent in cubic crystals but not in tetragonal crystals (Slutsky and Livingston 1960, Wolfram *et al* 1963) and close-packed hexagonal crystals (Lehman *et al* 1965, Gupta and Dayal 1966). Even the models based on modified axially symmetric forces (Dewames and Lehman 1964, Dewames *et al* 1965) or the De Launay angular forces (Ashokkumar 1973; Metzbower 1967, 1968) are not free from this deficiency. On the contrary the models based on angular forces incorporated in a manner suggested by Clark *et al* (1964) are elastically consistent and the present force constant model is an example in this context. These angular forces include some three-body interactions whereas the other type of forces between ions represent only the two-body interactions (Ramamurthy 1982). Detailed analysis of various force constant models (Ramamurthy and Rajendraprasad, to be published) reveals that the three-body interactions are important in the case of non cubic crystals. A force constant model becomes elastically inconsistent and gives rise to two different expressions for C_{44} when it includes these interactions incorrectly or excludes them altogether. On the other hand, these higher order interactions are not really significant in the case of cubic symmetry and therefore all force constant models, with or without the CGW angular forces, are elastically consistent.

The shape of the phonon frequency distribution function, $g(\nu)$ shown in figure 2 is very much similar to those obtained from other force constant models (Slutsky and Livingston 1960, Sharan and Ashokkumar 1973, Ramamurthy and Rajendraprasad 1981) except that the two maximas are further apart in the first and they occur at lower frequencies in the second. It is obvious from figure 3 that the θ_D values of indium obtained from the experimental values of lattice specific heat are in very good agreement with those from the present authors' present and previous models at all temperatures above 15K and the apparent differences between these θ_D values are not significant because of the errors involved in their calculations. Further the former θ_D values are in excellent agreement with those from Slutsky and Livingston (1960) provided the elastic constants of BCT indium are used to evaluate force constants, but are higher than those from Sharan and Ashokkumar (1973) at all temperatures. Larger values of specific heat and the consequent lower values of θ_D are due to the neglect of umklapp processes and the use of a rather large value of C' in their calculations. Since the authors deduced the θ_D values after subtracting the electronic specific heat, γT from the theoretical lattice specific heat, their $\theta_D - T$ curves (Ashokkumar and Sharan 1972; Sharan and Ashokkumar 1973) are invariably wrong. The correct θ_D values obtained from their data are shown in figure 3. On the contrary, the quantitative agreement between the experimental and theoretical values of θ_D and the corresponding lattice specific heat over a wide temperature range, clearly establishes the validity of the present force constant model.

In the present study, atomic arrangement of indium was referred to the body centred tetragonal axes on the assumption that the BCT lattice is crystallographically equivalent to the FCT lattice. However, the misinterpretation of this equivalence regarding elastic constants (Slutsky and Livingston 1960), Brillouin zone (Sharan and Bajpai 1970) and the lattice constants (Joardar and Chakraborty 1979) has resulted in many serious errors in the lattice dynamical study of indium. Further the shape of the Brillouin zone of BCT indium which resembles that of FCT lattice and the large value of t associated with this lattice seem to suggest that it is more appropriate to treat this atomic arrangement as FCT lattice (Ramamurthy 1979). The consequences of the crystallographic equivalence between BCT and FCT lattices and how they could be exploited in the lattice dynamical study of indium are discussed elsewhere (Ramamurthy and Rajendraprasad 1982).

5. Conclusions

It is obvious that the present force constant model is elastically consistent and conforms with the translational symmetry of the lattice. The phonon frequencies of BCT indium deduced from it are in very good agreement with the corresponding experimental frequencies of Reichardt and Smith and theoretical frequencies deduced by Garrett and Swihart (1976), but disagree with those obtained from inadequate models at several wave vectors and polarizations. The θ_D values deduced from this model compare well with their experimental values and theoretical values of Slutsky and Livingston (1960), Ramamurthy and Rajendraprasad (1981) but not with those of Sharan and Ashokkumar (1973), over a wide temperature range. Inclusion of angular forces and contributions from umklapp processes restores, respectively,

the elastic consistency and translational symmetry to the force constant models of non cubic crystals.

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