ISSN 0973-9777 Volume-6 Number-4 July-August 2012

The Indian Journal of Research

ANVIKSHIKI

Bi-monthly International Journal of all Research

Science

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Maneesha Publication

(Letter No.V-34564,Reg.533/2007-2008)

B-32/16-A-2/1,Gopalkunj,Nariya,Lanka

Varanasi,U.P.,India

Anvikshiki
The Indian Journal of Research
Volume 6 Number 4 July 2012

Science
Papers

PRINT ISSN 0973-9777,WEBSITE ISSN 0973-9777

Letter No.V-34564,Reg.533/2007-2008 INDIAN JOURNAL OF RESEARCH(2012)**6,104-111**
ANVIKSHIKI ISSN 0973-9777 Advance Access publication 20 May. 2012

STUDY OF LATTICE DYNAMICS OF DIAMOND

S.R.B.THAPA*

Declaration

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Abstract

A phenomenological model is used to study the lattice dynamics of diamond. Phonon frequencies, Debye Characteristic temperature and microscopic elastic constants are studied using Urey Bradley Valence Force Field (UVVF) approximations. The potential of the crystal involves (i) the central force due to bond-stretching (ii) the angular force due to bond bending (iii) central force between non-bonded atoms (iv) the force due to interaction of one internal co-ordinate to adjacent internal co-ordinate .Calculated results of phonon dispersion curves, Debye Characteristic temperatures and microscopic elastic constants are compared with experimental results giving fairly good agreement.

Key Words: Phenomenological model, Phonon frequencies, Debye Characteristic temperature, microscopic elastic constants, Urey Bradley Valence Force Field (UVVF) approximations.

Introduction

Diamond is an elemental semiconductor which has been studied extensively both theoretically and experimentally. Lattice dynamics of diamond has been previously investigated by Born (1914) using celebrated theory of lattice dynamics developed by Von Karman and himself. Various extensions of their work appeared in later years by Begbic and Born (1947), Smith (1948) and Herman (1959) using the model, known as Born-Von-Karman model (BKM).A two neighbour Born- Von Karman model is applied to the diamond crystal to study the lattice dynamics. The theoretical study of lattice dynamics of diamond has been made by Tubino et al.(1972) using a six parameters valence force field. First principle calculations of lattice dynamical properties of diamond have been formed using density-functional perturbation theory together with plane –wave expansion and non-local pseudopotentials by Pavone et

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al.(1993). Patel et al. (1984) reported the phonon dispersion curves along different symmetry directions. The lattice dynamics of diamond has been investigated by using central forces, angular forces of the type deLaunay and Clark, Gazis and Wallis by Bose et al. (1973).Warren et al.(1967) measured the phonon frequencies in the symmetry directions [100] and [111] by method of inelastic neutron scattering. Later on they extended their measurements to include [110] and [1ξ0] directions also. Robertson et al (1934) reported the first order Raman spectra of diamond. The one phonon infrared spectra have been studied experimentally by Smith and Hardy (1960). Hardy and Smith (1961) and Wehner et al (1967) measured the two phonon spectra of diamond. The precise data on second order Raman spectra are made available by Solin and Ramdas (1970). Various attempts have been made to study the dynamical behavior of solids with the application of valence force fields. Early attempts in this direction have been made by Herman (1959) and Pope (1965) to study the lattice dynamics of the elemental semiconductors germanium and silicon. Singh and Dayal (1970) studied extensively lattice dynamics of a number of elemental semiconductors on the basis of general valence force field. The application of valence force field to the lattice dynamics of sphalerite and wurtzite zinc sulphide has been made by Nusimovici and Birman (1970). Lattice dynamics of diamond type crystals have been studied by using Keating's valence force field by Bashenov et al.(1978). Kulda et al. (1997) explained coupling between acoustic and optic branches in longitudinal mode along [111] direction.

Phonon dispersion relation of diamond has been calculated by Pandey and Dayal (1973) on the basis of Clark-Gazis-Wallis type angular force model having only four parameters giving good agreement between theoretical and experimental data.

In this paper Urey-Bradley Valence Force Field (UVVF) is assumed to describe the forces operating inside the solids which crystallize in diamond structure. Urey-Bradley Valence Force Field is the combination of the simple valence force field and the central interaction between the non-bonded atoms. Thus forces considered are those which resist the extension or compression of valence bonds together with those which oppose the bending or torsion of bonds and central interaction between the non-bonded atoms. Urey-Bradley (1931) potential function is assumed to represent properly the forces present in the solids which are predominantly covalent. In the present work Urey Bradley (1931) valence force field (UVFF) approximations are used to express phonon dispersion relation of diamond. It is noteworthy that valence force potential is a dynamical representation of the results of a quantum mechanical treatment of a covalent crystal. Forces between atoms arise from the changes of the electronic energy due to the atomic displacement from the equilibrium configuration during the vibration. Because of the directional properties of orbital and their hybrids, forces act along the valence bond, bond angle and dihedral angles. In present formulation forces along the dihedral angle are neglected. Contribution to potential energy from other neighbours except first and second neighbours has been neglected because of the short range character of the force field. The Coulomb electrostatic interactions are not considered since atomic charges are either zero or very small.

Theoretical Model

The potential of the lattice is assumed to involve the following interactions:

(i) The central force due to bond stretching.
(ii) The angular force due to bond bending.
(iii) The central force between non-bonded atoms.
(iv) The force due to interaction of one internal co-ordinate (bond-stretching) to adjacent internal co-ordinate (bond-stretching)

Potential energy of solid having diamond structure is given by

$$2V = \sum_{j=1}^{4} K_r(\delta r_{ij})^2 + \sum_{l=1}^{12} K_{r_1}(\delta r_{il})^2 + \sum_{k=1}^{12} K_{r_2}(\delta r_{jk})^2 + \sum_{\theta_{jim}=1}^{4} K_{\theta_1}(\delta\theta_{jim})^2 + \sum_{\theta_{jil}=1}^{4} K_{\theta_2}(\delta\theta_{jil})^2 + \sum_{r_j=1}^{4} K_{rr}(\delta r_{ij})(\delta r_{im}) \quad (1)$$

Where 'i' is the reference atom of one type, 'j' and 'k' are two atoms of another type bonded to atom 'i', 'l' are atoms of type 'i' bonded to atom 'j', 'l' atoms are second nearest neighbours of 'i' type atoms and 'k' atoms are second nearest neighbours of 'j' type atoms.

k_r = bond stretching force constant

k_{r_1} = central force constant for non-bonded atoms (cation-cation)

k_{r_2} = central force constant for non-bonded atoms (anion-anion)

k_{θ_1} = bond bending force constant for the bond angle θ_{jik}

k_{θ_2} = bond bending force constant for the bond angle θ_{ijl}

k_{rr} = force constant for interaction of two adjacent bonds.

In the case of diamond lattice the two atoms are similar so that force constants $k_{r_1}=k_{r_2}$ and $k_{\theta_1} = k_{\theta_2}=\Upsilon$(say).

$ä r_{ij}$, $ä r_{il}$ and $ä r_{jk}$ are internal co-ordinate corresponding to bond stretching.

$ä\theta_{jik}$ and $ä\theta_{ijl}$ are internal co-ordinate corresponding to bond bending. Following the method of Wilson et al.(1955) and Singh and Roy(1975), Urey Bradley valence force field co-ordinates (change in bond length and bond angles) are transformed into atomic displacement co-ordinates. This transformation helps in expressing the potential energy in terms of the components of the displacements of atoms of the crystal.

The secular determinant for the normal modes of vibration of the atoms in the crystal is given by

$$\left|D_{\alpha\beta}(\boldsymbol{q},kk') - \omega^2\delta_{\alpha\beta}\delta_{kk'}\right| = 0 \quad (2)$$

$D_{\alpha\beta}(\mathbf{q},kk')$ represents the elements of the dynamical matrix. $D(\mathbf{q})$ and ω is the angular frequency of the normal modes of the vibration of the crystal. $\delta_{\alpha\beta}$ and $\delta_{kk'}$ are the usual kronecker delta functions. Dynamical matrix corresponding to the short range interaction are given by

$$D_{xx}(\mathbf{q},11) = 1/m[(4/3)k_r - (2/3)k_{rr} + 4k_{r1} + (28/3)(\Upsilon/r_0^2) + (8/3)(\Upsilon/r_0^2)(Cos\pi q_y)(Cos\pi q_z) - \{2k_{r1} + (2/3)(\Upsilon/r_0^2)\}\{(Cos\pi q_x)(Cos\pi q_z) + (Cos\pi q_x)(Cos\pi q_y)\}] \quad (3)$$

$$D_{xy}(\mathbf{q},11) = 1/m[\{2k_{r1} + (2/3)(\Upsilon/r_0^2)\}(Sin\pi q_x)(Sin\pi q_y) + (4/3)(\Upsilon/r_0^2)\, iSin\pi q_z\,(Cos\pi q_x - Cos\pi q_y)] \quad (4)$$

$$D_{xx}(\mathbf{q},22) = D_{xx}(\mathbf{q},11) \quad (5)$$

$$D_{xy}(\mathbf{q},22) = 1/m[\{2k_{r1} + (2/3)(\Upsilon/r_0^2)\}(Sin\pi q_x)(Sin\pi q_y) - (4/3)(\Upsilon/r_0^2)\, iSin\pi q_z\,(Cos\pi q_x - Cos\pi q_y)] \quad (6)$$

$$D_{xx}(\mathbf{q},12) = -1/m[\{(4/3)k_r - (2/3)k_{rr} - (32/3)(\Upsilon/r_0^2)\}\{Cos\pi(q_x/2)Cos\pi(q_y/2)Cos\pi(q_z/2) - i\,Sin\pi(q_x/2)Sin\pi(q_y/2)Sin\pi(q_z/2)\}] \quad (7)$$

$$D_{xy}(\mathbf{q},12) = 1/m[\{(4/3)k_r - (2/3)k_{rr} - (16/3)(\Upsilon/r_0^2)\}\{Sin\pi(q_x/2)\,Sin\pi(q_y/2)Cos\pi(q_z/2) - i\,Cos\,\pi(q_x/2)\,Cos\,\pi(q_y/2)Sin\pi(q_z/2)\}] \quad (8)$$

'm' is ionic mass, 'r_0' is chemical bond length and 'a' is half of the lattice parameter. Other elements of the matrix $D_{\alpha\beta}(\mathbf{q},kk')$ can be obtained by cyclic permutation of the indices x, y and z where α,β stand for x, y and z.

Solving the secular determinant (2) for long wave length, following expressions for the three elastic constants for the diamond structure crystal are obtained in terms of model parameters k_r , k_{rr} , k_{r1} and Υ/r_0^2.

$$C_{11} = 1/2a[(1/3)k_r - (1/6)k_{rr} + 4k_{r1} + 4(\Upsilon/r_0^2)] \quad (9)$$

$$C_{12} = 1/2a[(1/3)k_r - (1/6)k_{rr} + 2k_{r1} - 2(\Upsilon/r_0^2)] \quad (10)$$

$$C_{44} = 1/2a[(1/3)k_r - (1/6)k_{rr} + 2k_{r1} + (2/3)(\Upsilon/r_0^2) - A^2/B] \quad (11)$$

Where $A = -[(2/3)\,k_r - (1/3)k_{rr} - (8/3)(\Upsilon/r_0^2)]$

$B = [(4/3)\,k_r - (2/3)k_{rr} + (32/3)(\Upsilon/r_0^2)]$

Solving the secular determinant along [100] direction one gets

(i) at zone centre (Γ)

$$\omega_{LO}^2(\Gamma) = 2/m[(4/3)\,k_r - (2/3)k_{rr} + (32/3)(\Upsilon/r_0^2)] \quad (12)$$

(ii) at zone boundary (X)

$$\omega_{LO}^2(X) = 1/m[(4/3)k_r - (2/3)k_{rr} + 8k_{r1} + (40/3)(\Upsilon/r_0^2)] \quad (13)$$

$$\omega_{TO}^2(X) = 1/m[(4/3)k_r - (2/3)k_{rr} + 4k_{r1} + (20/3)(\Upsilon/r_0^2) + \{(4/3)k_r - (2/3)k_{rr} - (16/3)(\Upsilon/r_0^2)\}] \quad (14)$$

With the help of equations (12), (13) and (14) along with equilibrium condition of the lattice, model parameters are evaluated.

Input data for evaluating model parameters k_r, k_{r1}, k_{rr} and (Υ/r_0^2) are given in table 1 &2 while evaluated values of model parameters are given in table 3.

T A B L E 1 *(zone boundary phonon frequencies of Diamond)*

Crystal	Zone boundary and zone centre phonon frequencies (10^{12}Hz)			Reference
	$\nu_{LO}(X)$	$\nu_{TO}(X)$	$\nu_{LO}(\Gamma)$	
Diamond	35.52	32.16	39.975	Warren et al.(1967)

T A B L E 2 *(Lattice constant and mass)*

Crystal	Lattice constant (2a) cm	Mass(m) gm
Diamond	3.5668×10^{-8}	19.9366×10^{-24}

T A B L E 3 *(Evaluated values of Model parameters of Diamond)*

Crystal	Force Constants (10^4 dyne cm^{-1})			
	k_r	k_{r1}	k_{rr}	(Υ/r_0^2)
Diamond	-14.309212	3.577303	-76.057446	2.935913

Using the evaluated values of model parameters elastic constants C_{11}, C_{12}, C_{44} and Bulk modulus are evaluated.

T A B L E 4 *(Elastic constants and Bulk Modulus of diamond)*

	Expe-rimental values	Present Calculatedvalues	Calculated Values of others		
	10^{12} dyne cm^{-2}		Sokel and Harrison(1976)	Chadi and Martin(1976)	Harrison and Sokel(1976)
C_{11}	10.76	9.52			
C_{12}	2.75	2.58			
C_{44}	5.758	4.49	4.346	4.525	
Bulkmodulus	5.45	4.22			12.00

The calculated values of model parameters are used to obtain phonon dispersion curves of diamond along [100],[11 0] and [111] directions.

To calculate lattice specific heat the first Brillouin zone is divided into an even spaced sample of 1000 wave vectors which reduce to 48 non-equivalent points under symmetry operations. Secular determinant is solved at these non-equivalent points to calculate the frequency. An appropriate statistical

weight is attached to each of the non-equivalent points. When properly weighted the vibration spectra corresponding to these points represent the complete vibration spectra for the solid and frequency spectrum is divided into intervals of 0.1 T Hz. Frequency distribution function g(ν) is derived by making use of Blackman's sampling technique(1937).

Lattice specific heats at different temperatures is

$$C_v = \frac{3R}{6000}\sum_{\nu} g(\nu)E\left(\frac{h\nu}{k_B T}\right) \tag{15}$$

$E_x = \dfrac{x^2 e^x}{\left(e^x - 1\right)^2}$ where x = hõ/k_BT. h = Plank's constant, õ = frequency,

k_B is Botzmann constant, T = temperature of crystal, R = Molar gas constant.

The computed values of C_v are used to find Debye characteristic temperatures form standard table of (C_v - θ_D/T) from Saha and Srivastava (1965).

Results and Discussion

Elastic Constants

Observation of the table 4 shows that calculated values of elastic constants C_{11} and C_{12} in the present work are in fare agreement with their experimental values. The calculated value of C_{44} in the present work differs from the experimental one but is in excellent agreement with those of Sokel and Harrison (1976) and Chadi and Martin(1976).

The calculated values of C_{11} and C_{12} are used to calculate the bulk modulus of diamond. This value is comparable to the experimental value but the bulk modulus calculated theoretically by Harrison and Sokel (1976) is nearly three times compared to our result.

Phonon frequencies

Calculated phonon frequencies for diamond along three principal symmetry directions [100],[110] and [111] are displayed in Fig.1 along with experimental points of Warren et al.(1967). Calculated phonon frequencies for diamond along symmetry direction [1ξ0] are displayed in Fig.2 along with experimental points of Warren et al.(1967).The computed results are in excellent agreement with the experimental values except for the transverse acoustic modes. It is interesting to compare the results of the present investigation with those of the other studies. Cowley et al. (1966) calculated the dispersion curves for diamond by ten parameter shell model. Fairly good results were obtained for [100] and [111] directions. However they could not compare the results of [110] and [1ξ0] directions due to non-availability of the experimental data. Warren et al. (1967) calculated the dispersion curves on the basis of an eleven parameter shell model and compared the results with their own experimental data. The results obtained are quite satisfactory on the whole.

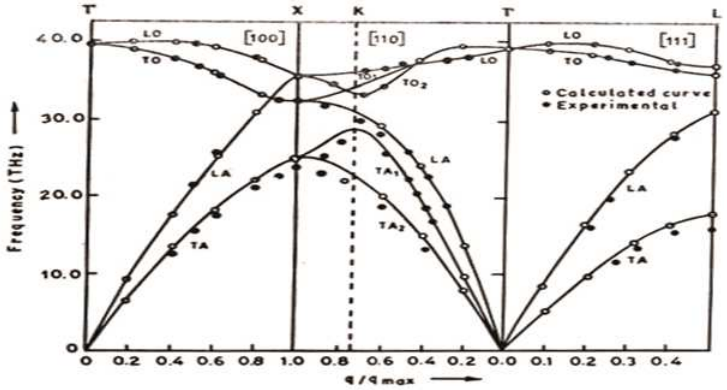

Fig.1. *Phonon dispersion curves of diamond along symmetry directions [100], [110] and [111]. Solid circles (•) represent the experimental results due to Warren et al. (1967).*

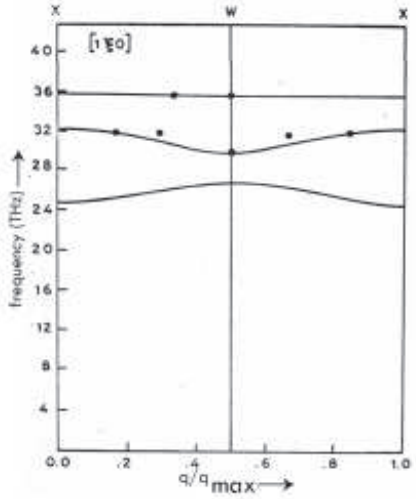

Fig.2. *Phonon dispersion curves of diamond along [1ξ0] direction. Solid circles (•) represent the experimental results due to Warren et al.(1967).*

Specific heat and Debye Temperature

Effective calorimetric and X-ray Debye temperature of diamond have been investigated on the basis of Cochran version of dipole approximation model by Blanchard and Varshni (1967). Pitzer (1938), Sorbo and Burk (1953) and Burk and Friedberg (1958) measured Debye Temperature of diamond. The Debye characteristic temperatures (θ_D) of diamond obtained from computed values of specific heat are plotted in Fig.3 against temperature (T) with experimental points of Pitzer (1938).There is a good agreement between theoretical and experimental Debye temperature at low temperatures and there is divergence at higher temperatures. Our results are comparable with those of Pandey and Dayal (1973).

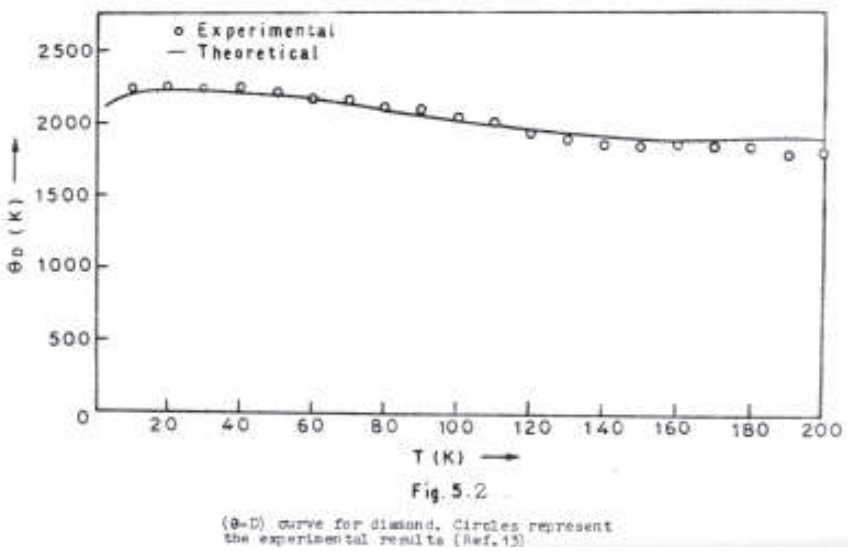

Fig. 5.2

(θ-D) curve for diamond. Circles represent the experimental results (Ref. 15)

Fig.3. *(θ_D – T) curve for diamond along with experimental points (0) due to Petzer (1938).*

Conclusion

The present lattice dynamical model having four disposable parameters is found capable of explaining the phonon dispersion relations of diamond. This model which does not require the employment of elastic constants for the evaluation of its parameters very satisfactorily reproduces the values of elastic constants and bulk modulus of diamond. The results for these quantities obtained on theoretical considerations based on pseudopotential and LCAO theories are found not superior and in some cases far inferior to our results. This gives an emphasis on the suitability of the application of valence force fields to the covalent crystals which gave highly directional bonds resulting from the well defined orbital hybridization. In view of small number of parameters used in the present work, Urey Bradley Valence Force Field Model (UBVFFM) describes satisfactorily the phonon dispersion results, elastic constants and Debye temperature of diamond.

Introduction of interaction terms for changes in bond length and bond angles in the present formulation may improve the features of the present results.

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