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Study of lattice dynamics of silicon

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Abstract
Lattice dynamics of Si has been investigated by using a Urey-Bradley Valence Force Field (UVFF) model which is a phenomenological model. In this model following interactions are taken into account: (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 temperature, microscopic elastic constants and Bulk modulus of Si are compared with experimental results giving fairly good agreement.

1. Introduction

Yin and Cohen [1] investigated theoretically lattice dynamical properties of diamond crystal Si using ab initio pseudopotential theory within the local density functional formalism. Inter-planar and inter-atomic force constants in Si has been investigated using adiabatic bond-charge model by Fleszar and Resta [2].
Bose et al.[3] investigated phonon dispersion curves of Si in three symmetry directions of Brillouin zone using the angular forces of type employed by deLaunay and Clark, Gazis and Wallis. Phonon frequencies, elastic constants, specific heat and Debye temperature of diamond structure crystal Si has been investigated by Zdetsis and Wang [4] using Born Von-Karman (BVK) model. Lattice dynamical calculations of phonon scattering at ideal Si-Ge interfaces has been done by Zhao and Freund [5].
Dispersive elastic constants of Si has been determined by Jakata and Every [6].Maranganti and Sharma [7] incorporated third and fourth order spatial derivatives of the displacement field in the elastic wave equation to calculate elastic constants of Si.
Urey-Bradley Valence Force Field (UVVF) model which is successfully used to investigate the lattice dynamics of diamond by Thapa [8] is extended to study the lattice dynamics of diamond like crystals Si. 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. 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.

2. Theoretical Models

Theoretical model used in this paper is reported by Thapa [8].The secular determinant for the normal modes of vibration of the atoms in the crystal is given by

$$\left| D_{\alpha\beta}(\mathbf{q}, kk') - \omega^2 \delta_{\alpha\beta} \delta_{kk'} \right| = 0 \qquad (1)$$

$D_{\alpha\beta}$ (**q**,kk$'$) represents the elements of the dynamical matrix D(**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.
Solving the secular determinant for long wavelength limit, 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.

$$C_{11} = 1/2a[(1/3)k_r - (1/6)k_{rr} + 4k_{r1} + 4(\Upsilon/r_0^2)] \qquad (2)$$
$$C_{12} = 1/2a[(1/3)k_r - (1/6)k_{rr} + 2k_{r1} - 2(\Upsilon/r_0^2)] \qquad (3)$$
$$C_{44} = 1/2a[(1/3)k_r - (1/6)k_{rr} + 2k_{r1} + (2/3)(\Upsilon/r_0^2) - A^2/B] \qquad (4)$$

where   A = - [(2/3) k_r – (1/3)k_{rr} – (8/3)(Υ/r_0^2)],   B = [(4/3) k_r – (2/3)k_{rr} + (32/3)(Υ/r_0^2)]
Solving the secular determinant along [100] direction one gets
(i) at zone centre (Γ)

$$(\omega_{LO})^2 (\Gamma) = 2/m[(4/3)\, k_r - (2/3)k_{rr} + (32/3)(\Upsilon/r_0^2)] \qquad (5)$$

(ii) at zone boundary (X)

$$(\omega_{LO})^2 (X) = 1/m[(4/3)k_r - (2/3)k_{rr} + 8k_{r1} + (40/3)(\Upsilon/r_0^2)] \qquad (6)$$
$$(\omega_{TO})^2 (X) = 1/m[(4/3)k_r - (2/3)k_{rr} + 4k_{r1} + (20/3)(\Upsilon/r_0^2) + \{(4/3)k_r - (2/3)k_{rr} - (16/3)(\Upsilon/r_0^2)\}] \qquad (7)$$

Numerical Computation
With the help of equations (5), (6) and (7) 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.

Table-1:zone boundary phonon frequencies of Si

Crystal	Zone boundary and zone centre phonon frequencies(10^{12}Hz)			Reference
Si	$v_{LO}(X)$	$v_{TO}(X)$	$v_{LO}(\Gamma)$	Nilsson and Nelin [9]
	12.32	13.90	15.53	

Table-2: Lattice constant and mass of Si

Crystal	Lattice constant (2a)	Mass(m)
Si	5.4307×10^{-8} cm	46.629×10^{-24} gm

Table-3:Evaluated values of Model parameters of Si

Crystal	Force Constants (10^4 dyne cm^{-1})			(Υ/r_0^2)
Si	k_r	k_{r1}	k_{rr}	
	-2.141340	0.535335	-28.824330	0.548956

Using the evaluated values of model parameters elastic constants C_{11}, C_{12}, C_{44} and Bulk modulus are evaluated using equations (2),(3)&(4).

Table 4: Elastic cnstants and Bulk modulus in units of 10^{12} dyne cm^{-2}

	Experimental values Jakata and Every [6]	Present Calculated values	Calculated Values of others		
			Sokel and Harrison [10]	Chadi and Martin [11]	Baraff et al [12]
C_{11}	1.775	1.552			1.207
C_{12}	0.745	0.748			0.860
C_{44}	0.807	0.789	0.717	0.943	0.317
Bulk Mod.	1.088	2.000	2.00		

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

Lattice specific heats at different temperatures is

$$C_v = \frac{3R}{6000} \sum_v g(v) E\left(\frac{hv}{k_B T}\right) \qquad (8)$$

$E_x = \dfrac{x^2 e^x}{\left(e^x - 1\right)^2}$ where x = hυ/k$_B$T. h = Plank's constant, υ = frequency, k$_B$ is Botzmann Constant, T = temperature of crystal, R = Universal gas constant.

The computed values of C$_v$ are used to find Debye characteristic temperatures from standard table of (C$_v$ - θ_D/T) from Saha and Srivastava [13].

3. Results and Discussion

Elastic Constants

The calculated values of elastic constants C_{11},C_{12} and C_{44} and Bulk modulus of Si in the present work along with their experimental values and calculated values of by other workers are shown in the Table 4 for comparison.

Calculated result of C_{44} differs slightly with the experimental value but superior to the calculated value of Baraff et al [12]. Calculated values for C_{11} and C_{12} are in satisfactory agreement with their experimental results. The bulk modulus calculated in the present work is found to be nearly twice the experimental value. Surprisingly the calculated value of bulk modulus is almost equal to the value obtained by Harrison and Sokel [14] calculated on the basis of linear combination of atomic orbitals (LCAO).

Phonon frequencies

Propagation of acoustic phonons in Si on the basis of two lattice dynamical models: a bond-charge model and effective force constant model consisting of only short range forces have been investigated by Tamura et al. [15]. Mazur and Pollman [16] investigated lattice dynamics of Si with semi empirical approach using tight binding green functions and dynamical matrix of the system has been set up. Elastic constants and normal mode frequencies of Si has been computed with empirical Many-Body potentials

and compared with experimental results by Cowley [17]. None of the models tested is completely satisfactory.

The results of phonon dispersion curves in present work are shown in Fig.1. along three symmetry directions [100], [110] and [111].The agreement of the results with the experimental values of Dolling [18] and Nilsson and Nelin [9] shown in the figures is fairly satisfactory. Our results are comparable with those of Tubino et al. [19] and superior over the results of Martin [20], Soma and Marita [21] and Sinha et al. [22].
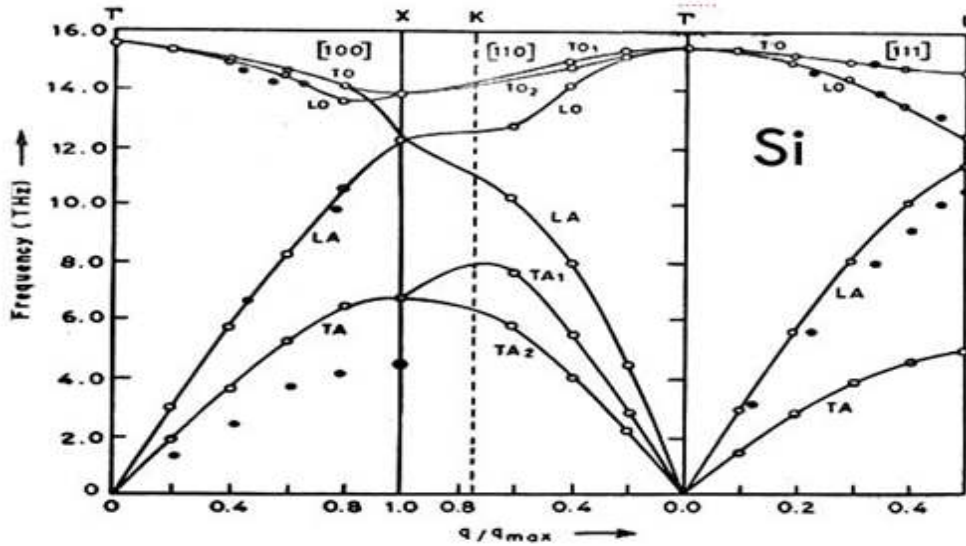

Fig.1: Phonon dispersion curves of Si along symmetry direction [100],[110] and [111]. Solid circles (•) represent the experimental results due to Dolling [18], Nilsson and Nelin [9]. Solid lines represent calculated curve.

 Debye temperature

Calculated Debye temperatures of Si are plotted against temperature in Fig.5 along with experimental points of Flubacher and Leadbetter [23] giving fairly satisfactory agreement. The calculated values deviate from experimental values at higher temperature. Divergence of the calculated result with experimental results is almost same as that of 16 parameter BVK model of Zdetsis and Wang (1979). Thus only four parameter present harmonic model provides satisfactory result of Debye temperature.
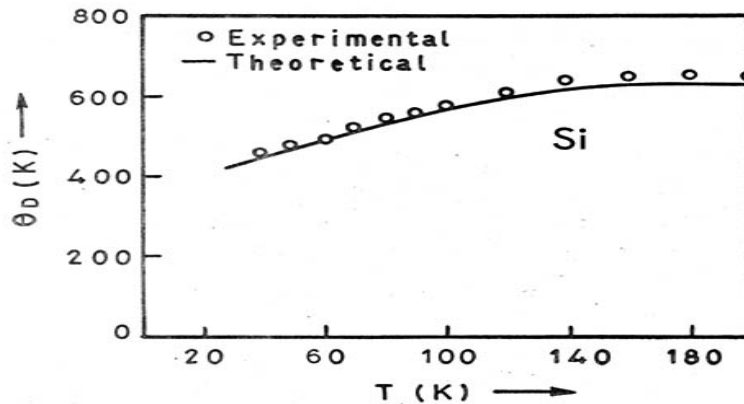

Fig.2: (θ_D – T) curve for Si. Experimental points (0) are due to Flubacher and Leadbetter [23].

4. Conclusion

The present lattice dynamical model having four disposable parameters which is successful in explaining the phonon dispersion relations, elastic constants and Debye temperature of diamond, when extended for another diamond structure crystal Si is capable to explain its lattice dynamic properties- phonon frequencies, elastic constants and Debye temperature. This model which does not require the employment of elastic constants for the evaluation of its model parameters very satisfactorily reproduces the values of elastic constants and bulk modulus of Si. 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 the present 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. Introduction of interaction terms for changes in bond length and bond angles in the present formulation may improve the features of the present results.

References

[1]   M.T. Yin and Marvin L. Cohen, Phys. Rev.B,26 (1982) 3259.
[2]   A. Fleszar and R. Resta, Phys. Rev.B, 34 (1986) 7140.
[3]   G. Bose, B.B.Tripathi and H.C. Gupta, J. Phys. Chem. Solids, 34 (1973) 1867.
[4]   A. D. Zdetsis and C. S. Wang, Phys. Rev.B, 19 (1979) 2999.
[5]   H. Zhao and J. B. Freund, J. Appl. Phys. 97 (2005) 024903.
[6]   K. Jakata and A.G. Every, Phys. Rev.B, 77 (2008) 174301.
[7]   R. Maranganti and P. Sharma, Phys. Rev. Letters ,98 (2007) 195504.
[8]   S. R. B. Thapa J. Purvanchal Acad. Sc.,17 (2011) (accepted paper).
[9]   G. Nilsson and G. Nelin, Phys. Rev.B, 6 (1972) 3777.
[10]   R. Sokel and W.A. Harrison, Phys. Rev. Letters, 36 (1976) 212.
[11]   D.J. Chadi and R. M. Martin, Solid State Commun.,19 (1976) 193.
[12]   G.A. Baraff, E.O. Kane and M. Schuluter, Phys. Rev.B, 21 (1980) 5662.
[13]   M. N. Saha and B. N. Srivastava, A Treatise on Heat,5th ed., Indian Press , Alllahabad (1965).
[14]   W.A. Harrison and R. Sokel, J.Chem. Phys.,65 (1976) 379.
[15]   S. Tamura, J.A. Shields and J.P. Wolfe, Phys. Rev.B ,44 (1991) 3011.
[16]   A. Mazur and J. Pollman, Phys. Rev.B, 39 (1989) 5261.
[17]   E. Roger Cowley, Phys. Rev.Lett. ,60 (1988) 2379.
[18]   G. Dolling, Inelastic scattering of neutrons, IAEA(Vienna) vol.II, (1965) p-249.
[19]   R. Tubino, L. Poseri and C. Zebri, J. Chem. Phys.,56 (1972) 1022.
[20]   R.M. Martin, Phys. Rev., 186 (1959) 871.
[21]   T. Soma and A. Marita, J. Phys. Soc. Japan, 32 (1972) 38.
[22]   S. K. Sinha R.P. Gupta and D.L. Price, Phys. Rev.B, 9 (1974)2564.
[23]   P. Flubacher and A.J. Leadbetter, Philos. Mag.,4 (1959) 273.