

Phonon study of Gallium Antimonide (GaSb) using a theoretical approach of van der Waals three-body force shell model (VTSM)

K.K.Mishra, K.S.Upadhyaya

Material Reseach Division, Dept of Physics, Nehru Gram Bharati University, Allahabad-221505 (India)

Abstract— A theoretical approach of van der Waals three body force shell model has been used for the calculation of mechanical, elastic and anharmonic properties of zinc blende gallium antimonides (GaSb).The agreement between results for elastic constants, pressure derivative of SOEC, Cauchy discrepancy and the available experimental data is excellent. We also report Phonon dispersion curve, two-phonon IR/Raman spectra, Debye temperature variation and anharmonic elastic properties. It is concluded that VTSM is adequately capable of describing the complete crystal dynamic sduoy of zinc-blende structure crystals satisfying well.

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Index Terms— Phonon; Debye Temperature, Combined densities of states, gallium antimonide, van der Waals interaction.



1 INTRODUCTION

The vibrations of covalent crystals having the diamond structure (Ge, Si, C) and II-VI and III-V compound having zinc-blende type structure, has the characteristics feature that the transverse acoustic (TA) vibrations have a large region in which there is a very low group velocity and the difference between the frequencies of the longitudinal and transverse vibrations at boundary of the Brillouin zone is very large.

The general theory of lattice dynamics [1, 2] permits one to obtain the elastic vibration spectrum of crystals by specifying the interaction between the structural elements of the lattice. In calculating the spectra of particular crystals, however, question arises how to properly describe the interactions i.e. there is a question about the choice of model, which must take into account the important features of the given lattice.

The lattice dynamics of semiconductor compounds have been extensively studied in recent years. With the development of inelastic neutron scattering experiments, a vast amount of data has been accumulated for phonon dispersion curves for various II- VI and III-V compounds [3-6]. The phenomenological models which have been used to calculate the frequencies of zinc-blende crystals can broadly be classified into two categories, (i) rigid-ion model (RIM) [7-9] and rigid-shell model (RSM) [10-11]. The rigid ion model involves the ion rigidity hypothesis, whereas the shell model takes into account the long-range Coulomb interaction and that related to the displacement of shells of negligible mass. The ionic polarizability is also taken into consideration. The numbers of independent parameters in the case of shell model are very large. One can easily show that a wide range of parameters give the same spectrum in the symmetric q-directions. This implies that the force constants introduced are of the nature of adjustable parameters, and cannot be assigned a definite physical meaning. The same remarks hold for the theory of Kucher and coworker [12, 13] in which the previously devel-

oped polarizable atom method [14] is used to calculate the spectrum of diamond type crystals.

In constructing a model for a lattice with covalent bonding there is fundamental difficulty related to the introduction of non-central forces [15]. In the usual definition of a non-central interaction [16] the corresponding force parameters are two-centre parameters, while actuality non-central forces cannot be due to rotation of one atom about another, but must be due to a change in the configuration of the atoms or bonds. Hence these force parameters must take into account atleast two coordination spheres. The rigid ion model and its various ramifications [17-20] have been widely used to explain several lattice dynamics properties of perfect and imperfect zinc-blende type crystals. These model, although economical in force parameters, ignore completely the non-central interaction in the lattice.

The valance force field model (VFFM) as used by Price et.al. [21] incorporates bond bending, bond-stretching and point Coulombic interactions. Later on the original 14 parameter valence force field (VFFM) was modified on the lines of shell model by Vegetatos et.al [6] and Feldkamp et.al [22]. In addition to lattice dynamics Garg et.al.[28], have investigated the mechanical properties of zinc-blende semiconductor by using three body force shell model [TSM] and derive the expressions for the third order elastic constants (TOEC) and pressure derivatives of second order elastic constants (SOEC) on the line of Singh and Verma [26, 27].

Analysis of these models RIM [7-9]; RSM [10, 11]; VFFM [21]; BCM [23]; DDM [24]; ECM [25] and TSM [26-28] reveal that the crystal dynamics study of zinc-blende crystals is still not well understood. We have thought it worthwhile to develop a new model for such structures which takes into account VWI and TBI in the framework of both ion polarizable RSM with short-range interactions effective up to second neighbour. From the above description, it may be inferred that the most realistic model for the proposed purpose can be devel-

oped by introducing the effect of VWI and TBI in the framework of RSM. The present model, known as van der Waals three body force shell model (VTSM), has 14 parameter computed for each crystal with the same input data (physical properties) and VWI has been added to short-range repulsive interactions operative up to second neighbour.

Therefore, it is obvious that along with the TBIs, VWIs must be taken into consideration in any discussion of lattice dynamics of these crystals. This new model, VTSM, corporates VWIs along with long range Coulomb interactions, TBIs and short range second neighbor interactions in the framework of RSM. The effect of TBI and VWI is quite significant and plays a vital role in the description of the lattice dynamics of gallium pnictides (GaP, GaAs and GaSb). A strong support has been found with experimental results for GaAs and GaP [55 & 56] by our group. In this paper, we thought it permanent to use the corrected expression mentioned in [29] for study of elastic properties, phonon dispersion curve (PDCs) and their pressure dependence in gallium antimonides (GaSb) crystal. The proposed investigations have been carried out by adopting a simple method to determine a consistent set of 14 parameters (i.e. four TBI parameters $b, \rho, f(r_0), \rho f'(r_0)$; two nearest neighbor short-range repulsive interaction parameter parameters A_{12} and B_{12} ; four second-nearest-neighbour short-range repulsive interaction parameters $A_{11}, B_{11}, A_{22}, B_{22}$; distortion polarizabilities of negative and positive ions d_1 and d_2 and shell charges of the negative and positive ions Y_1 and Y_2 , respectively) of VTSM. These results have shown reasonably good agreement with their available experimental and theoretical data (Farr et.al. [4]; Kunc et. al.[31]; Mooradian and Wright [32]; Miller et. al.[33].

The broad outlines about the theoretical framework of present model for calculations have been classified in section 2. The calculated results and their discussions are presented in section 4.

2. THEORETICAL FRAMEWORK OF PRESENT MODEL:

A detailed description of the three body force shell model (TSM) has been given by [27]. Further we have developed a model which includes the effect of van der Waals interactions (VWI) and three body interactions (TBI) in the framework of RSM and known as van der Waals three body force shell model (VTSM). In order to explain the essential theory and method of calculations, a brief idea about the TBP has been given below:

2.1. Three Body Crystal Potential:

The present three body force shell model for zinc-blende structure (ZBS) semiconductor crystal to describe cohesion, the crystal energy for a particular lattice separation (r) has been expressed as:

$$\Phi(r) = \Phi_{LR}(r) + \Phi_{SR}(r) \quad (1)$$

where the first term $\Phi_{LR}(r)$ represents the long-range Coulomb and three body interaction (TBI) energies expressed by

$$\Phi_{LR}(r) = \sum_{i \neq j \neq k} \frac{Z_i Z_j e^2}{r_{ij}} \left\{ 1 + \sum_k f(r_{ik}) \right\} = -\frac{\alpha_M Z^2 e^2}{r} \left\{ 1 + \frac{4}{Z} f(r) \right\} \quad (2)$$

where Z is the ionic charge parameter of i^{th} ion, r_{ij} separation between i^{th} and j^{th} ion, $f(r_{ik})$ is the three body force parameter dependent on nearest-neighbour separation r_{ik} and is a measure of ion size difference [27] and α_M is Madelung constant ($=1.63805$).

The second term in (1) is short-range energy contributions from overlap repulsion and van der Waals interactions (vdWI) expressed as [30].

$$\Phi_{SR}(r) = Nb \sum_{i,j=1}^2 \beta_{ij} \exp \left[\frac{r_i + r_j + r_{ij}}{\rho} \right] - \sum_{ij} \frac{c_{ij}}{r_{ij}^6} - \sum_{ij} \frac{d_{ij}}{r_{ij}^8} \quad (3)$$

where the first term is the Hafemeister and Flygare (HF) potential [35] and used by Singh and coworkers [26, 27, 29, 30]. The second term and third term represent the energy due to vdW dipole-dipole (d-d) and dipole-quadrupole (d-q) interactions, respectively.

Using the crystal energy expression (1), the equations of motion of two cores and two shells can be written as:

$$\omega^2 \underline{M} \underline{U} = (\underline{R} + \underline{Z}_m \underline{C}' \underline{Z}_m) \underline{U} + (\underline{T} + \underline{Z}_m \underline{C}' \underline{Y}_m) \underline{W} \quad (4)$$

$$0 = (\underline{T}^T + \underline{Y}_m \underline{C}' \underline{Z}_m) \underline{U} + (\underline{S} + \underline{K} + \underline{Y}_m \underline{C}' \underline{Y}_m) \underline{W} \quad (5)$$

here \underline{U} and \underline{W} are vectors describing the ionic displacements and deformations, respectively. \underline{Z}_m and \underline{Y}_m are diagonal matrices of modified ionic charges and shell charges, respectively; M is the mass of the core; T and R are repulsive Coulombian matrices, respectively; C' and Y_m are long-range interaction matrices, that include Coulombian and TBI respectively; S and K are core-shell and shell-shell repulsive interaction matrices, respectively and T^T is the transpose of matrix T . The elements of matrix \underline{Z}_m consists of the parameter Z_m giving the modified ionic charge.

$$Z_m = \pm Z \sqrt{1 + \frac{4}{Z} f(r)} \quad (6)$$

The elimination of \underline{W} from eqns. (4) and (5) leads to the secular determinant:

$$\left| \underline{D} - \omega^2 \underline{M} \underline{I} \right| = 0 \quad (7)$$

for the elimination of the frequency determination. Here \underline{D} (q) is the (6x6) dynamical matrix given by:

$$\underline{D} = \underline{R}' + \underline{Z}_m \underline{C}' \underline{Z}_m - \underline{C}' + \underline{Z}_m \underline{C}' \underline{Y}_m \underline{Y}_m^{-1} \underline{C}' + \underline{K} + \underline{Y}_m \underline{C}' \underline{Y}_m^{-1} \underline{C}'^T + \underline{Y}_m \underline{C}' \underline{Z}_m \quad (8)$$

The numbers of adjustable parameters have been largely reduced by considering all the short-range interactions to act only through the shells.

2.2. Second and Third Order Elastic Constants:

Proceeding with the use of three body crystal potential given by (1), Sharma and Verma [36, 37] have derived the expression for the second order elastic constant and used by [29] for zinc-blende structure crystals. We are reporting them here as their corrected expressions.

The expressions for second order elastic constants (SOEC) are

$$C_{11} = L \left[\begin{array}{l} 0.2477Z_m^2 + \frac{1}{3}(A_1 + 2B_1) + \frac{1}{2}(A_2 + B_2) \\ + 5.8243Zaf'(r_0) \end{array} \right] \quad (9)$$

$$C_{12} = L \left[\begin{array}{l} -2.6458Z_m^2 + \frac{1}{3}(A_1 - 4B_1) + \frac{1}{4}(A_2 - 5B_2) \\ + 5.8243Zaf'(r_0) \end{array} \right] \quad (10)$$

$$C_{44} = L \left[\begin{array}{l} -0.123Z_m^2 + \frac{1}{3}(A_1 + 2B_1) + \frac{1}{4}(A_2 + 3B_2) \\ - \frac{1}{3}\nabla(-7.53912Z(Z + 8f(r_0)) + A_1 + B_1) \end{array} \right] \quad (11)$$

for third order elastic constants (TOEC) they are

$$C_{111} = L \left[\begin{array}{l} 0.5184Z_m^2 + \frac{1}{9}(C_1 - 6B_1 - 3A_1) + \frac{1}{8}(C_2 - B_2 - 3A_2) \\ - 2(B_1 + B_2) - 9.9325Zaf'(r_0) + 2.5220Za^2f''(r_0) \end{array} \right] \quad (12)$$

$$C_{112} = L \left[\begin{array}{l} 0.3828Z_m^2 + \frac{1}{9}(C_1 + 3B_1 - 3A_1) + \frac{1}{8}(C_2 + 3B_2 - 3A_2) \\ - 11.642Zaf'(r_0) + 2.5220Za^2f''(r_0) \end{array} \right] \quad (13)$$

$$C_{113} = L \left[\begin{array}{l} 6.1585Z_m^2 + \frac{1}{9}(C_1 + 3B_1 - 3A_1) - 12.5060Zaf'(r_0) \\ + 2.5220Za^2f''(r_0) \end{array} \right] \quad (14)$$

$$C_{144} = L \left[\begin{array}{l} 6.1585Z_m^2 + \frac{1}{9}(C_1 + 3B_1 - 3A_1) - 4.168Zaf'(r_0) + \\ 0.8407Za^2f''(r_0) + \\ \nabla \left\{ -3.3507Z_m^2 - \frac{2}{9}C_1 + 13.5486af'(r_0) \right\} + \\ \left\{ -1.681a^2f''(r_0) \right\} \\ \nabla^2 \left\{ -1.5637Z_m^2 + \frac{2}{3}(A_1 - B_1) + \frac{C_1}{9} - 5.3138Zaf'(r_0) \right\} \\ \left\{ + 2.9350Za^2f''(r_0) \right\} \end{array} \right] \quad (15)$$

$$C_{166} = L \left[\begin{array}{l} -2.1392Z_m^2 + \frac{1}{9}(C_1 - 6B_1 - 3A_1) + \frac{1}{8}(C_2 - 5B_2 - 3A_2) \\ - (B_1 + B_2) + 4.168Zaf'(r_0) + 0.8407Za^2f''(r_0) \\ \nabla \left\{ -8.3768Z_m^2 + \frac{2}{3}(A_1 - B_1) - \frac{2}{9}C_1 + \right\} + \\ \left\{ 13.5486af'(r_0) - 1.681a^2f''(r_0) \right\} \\ \nabla^2 \left\{ 2.3527Z_m^2 + \frac{C_1}{9} - 5.3138Zaf'(r_0) \right\} \\ \left\{ + 2.9350Za^2f''(r_0) \right\} \end{array} \right] \quad (16)$$

$$C_{456} = L \left[\begin{array}{l} 4.897Z_m^2 + \frac{1}{9}(C_1 - 6B_1 - 3A_1) - B_2 + \\ \nabla \left\{ -5.026Z_m^2 - \frac{1}{9}C_1 \right\} + \nabla^2 \left\{ 7.0580Z_m^2 + \frac{C_1}{3} \right\} + \\ \nabla^3 \left\{ -4.8008Z_m^2 + \frac{1}{3}(A_1 - B_1) - \frac{C_1}{9} \right\} \end{array} \right] \quad (17)$$

where Z_m is the modified ionic charge defined earlier in equation (6) with $L = e^2/4a^4$ and

$$\nabla = \left[\frac{-7.53912Z(Z + 8f(r_0)) + (A_1 - B_1)}{-3.141Z(Z + 8f(r_0)) + (A_1 + 2B_1) + 21.765Zaf'(r_0)} \right] \quad (18)$$

The values of A_i, B_i, C_i as defined by Sharma and Verma [36, 37] $V(=3.08r_0^3)$ is the volume of the unit cell.

2.3. Vibrational Properties for Zinc-blende Structure:

By solving the secular equation (4) along $[q \ 0 \ 0]$ direction and subjecting the short and long-range coupling coefficients to the long-wavelength limit $\bar{q} \rightarrow 0$, two distinct optical vibration frequencies are obtained as

$$\omega_L^2 \Big|_{q=0} = R'_0 + \frac{(e^*)^2}{Vf_L} \cdot \frac{8\pi}{3} \left(\epsilon_m^2 + 4Zr_0f'(r_0) \right) \quad (19)$$

$$\omega_T^2 \Big|_{q=0} = R'_0 - \frac{(e^*)^2}{Vf_T} \cdot \frac{4\pi}{3} Z_m^2 \quad (20)$$

where the abbreviations stand for

$$R'_0 = R_0 - e^2 \left(\frac{d_1^2}{\alpha_1} + \frac{d_2^2}{\alpha_2} \right) \quad R_0 = \frac{e^2}{V} \left[4 \frac{A + 2B}{3} \right] \quad \text{and} \quad (21)$$

$$Z' = Z_m + d_1 - d_2 \quad (21)$$

$$f_L = 1 + \left(\frac{\alpha_1 + \alpha_2}{v} \right) \cdot \frac{8\pi}{3} \left(\epsilon_m^2 + 4Zr_0f'(r_0) \right) \quad (22)$$

$$f_T = 1 - \left(\frac{\alpha_1 + \alpha_2}{v} \right) \cdot \frac{4\pi}{3} \quad (23)$$

$$\text{and } \alpha = \alpha_1 + \alpha_2 \tag{24}$$

2.4. Debye Temperature Variation:

The specific heat at constant volume (C_V), at temperature T is expressed as

$$C_V = 3Nk_B \frac{\int_0^{\nu_m} \left\{ \left(\frac{h\nu}{k_B T} \right)^2 e^{h\nu/k_B T} \right\} G(\nu) d\nu}{\left(e^{h\nu/k_B T} - 1 \right)^2} \tag{25}$$

where, ν_m is the maximum frequency, N is the Avogadro's a number, h is the Planck's constant and k_B is the Boltzmann's constant. The equation (25) can be written as a suitable form for a computational purpose as

$$C_V = 3Nk_B \frac{\sum_{\nu} \left\{ \left(\frac{h\nu}{k_B T} \right)^2 e^{h\nu/k_B T} \right\} G(\nu) d\nu}{\sum_{\nu} G(\nu) d\nu} \tag{26}$$

Where $E = \left(\frac{h\nu}{k_B T} \right)$ is the Einstein function, defined by

$$E(x) = x^2 \frac{\exp(x)}{\exp(x) - 1} \tag{27}$$

with $x = h\nu/k_B T$

Also, $\sum_{\nu} G(\nu) d\nu =$ Total number of frequencies considered.
= 6000 for zinc-blende structure.

Hence, equation (26) can be written for zinc-blende structure type crystals, as

$$C_V = \frac{3Nk_B}{6000} \sum_{\nu} E(x)G(\nu) d\nu \tag{28}$$

The contribution of each interval to the specific heat is obtained by multiplying an Einstein function corresponding to mid-point of each interval (say 0.1 THz) by its statistical weight. The statistical weight of the interval is obtained from the number of frequencies lying in that interval. The contributions of all such intervals when summed up give $\sum_{\nu} E(x)G(\nu) d\nu$. The specific heat C_V is then calculated by expression (28).

3. COMPUTATIONS:

There are three basic model parameters (b, ρ and $f(r_0)$), which have been determined by making use of the expressions for the three second order elastic constants (C_{11}, C_{12}, C_{44}), reported by Sharma and Sharma [36, 37] and the equilibrium condition,

$$\left(\frac{d\Phi(r)}{dr} \right)_{r=d\frac{\sqrt{3}}{2}} = 0$$

with inclusion of van der Waals interactions (vdWI). The values of the input data and model parameters have been listed in table 1. The values of A_i, B_i, C_i have been calculated from the knowledge of b and ρ and the values of various order of derivatives of $f(r_0)$ (like $f'(r_0), f''(r_0), f'''(r_0)$), have been obtained by using functional form $f(r_0) = f_0 e^{-r/\rho}$ as used by [29].

Table 1. Input data and model parameters for GaSb.
[C_{ij} and B (in 10^{12} dyne / cm^2), ν (in THz), r_0 (in 10^{-8} cm), α_i (in 10^{-24} cm^3), b (in 10^{-12} erg), ρ (in 10^{-8} cm)]

Input Data		Model Parameters		
Properties	Values	Ref.	Parameters	Values
C_{11}	8.84	[51]	b	1.84
C_{12}	4.03	[51]	ρ	0.358
C_{44}	4.32	[51]	$f(r_0)$	-0.237
B	5.64	[51]	$r_0 f'(r_0)$	1.7543
r_0	2.65	[51]	A_{12}	33.9777
$\nu_{LO}(\Gamma)$	7.0	[4]	B_{12}	-4.4868
$\nu_{TO}(\Gamma)$	6.87	[4]	A_{11}	95.8952
$\nu_{LO}(L)$	6.15	[4]	B_{11}	-12.9279
$\nu_{TO}(L)$	6.48	[4]	A_{22}	-16.5754
$\nu_{LA}(L)$	4.60	[4]	B_{22}	2.4817
$\nu_{TA}(L)$	1.3	[4]	d_1	0.3770
α_1	0.97	[43]	d_2	4.2379
α_2	10.65	[43]	Y_1	-0.2427
ϵ_0	15.69	[31]	Y_2	-2.1730

The values of vdW coefficients used by us in the present study have been determined using the SKV approach [38] as suggested by Singh and Singh [29]. The values of the van der Waals (vdW) coefficients are listed in table 2.

Table: 2. Van der Waals Interaction Coefficients for GaSb.
(C_{ij} in units of 10^{-60} erg cm^6 and d_{ij} in units of 10^{-76} erg cm^8)

Parameters	Numerical Values
C_{+-}	315
C_{++}	54
C_{--}	1208
d_{+-}	221
d_{++}	18
d_{--}	1085
C	1536
D	1316

Further, these model parameters and vdW coefficients have been used to compute the rest model parameters [$f(r_0), r_0 f'(r_0), A_{11}, A_{22}, A_{12}, B_{11}, B_{22}, B_{12}, d_1, d_2, Y_1$ and Y_2] of the VTSM with knowledge of experimental values of the equilibrium interatomic separation (r_0), the vibration frequencies $\nu_{LO}(\Gamma), \nu_{TO}(\Gamma), \nu_{LO}(L), \nu_{TO}(L), \nu_{LA}(L)$ and $\nu_{TA}(L)$ and with the values $\alpha_1, \alpha_2, \epsilon_0$ for gallium pnictides.

The model parameters of VTSM have been used to compute the phonon spectra for zinc-blende structured gallium antimonides (GaSb) for the allowed 48 non-equivalent wave vec-

tors in the first Brillouin zone at Computer Center, Banaras Hindu University, Varanasi. The frequencies along the symmetry directions have been plotted against the wave vector to obtain the phonon dispersion curves (PDCs) from both the models. These curves have been compared with those measured by means of the coherent inelastic neutron scattering technique. Since the neutron scattering experiments provide us only very little data for the symmetry directions, we have studied the combined density of states (CDS) for the complete description of the frequencies for the Brillouin zone.

The complete phonon spectra have been used to calculate the combined density of states (CDS), $N(\nu_i+\nu_j)$ corresponding to the sum modes $(\nu_i+\nu_j)$, following the procedure of *Smart et al.* [39]. A histogram between $N(\nu_i+\nu_j)$ and $(\nu_i+\nu_j)$ has been plotted and smoothed out to obtain the CDS curves. These curves show well defined peaks which correspond to two-phonon Raman scattering peaks. Since no observed data on two-phonon IR/Raman spectra are available, these CDS peaks have been compared with the assignments calculated by using our present theoretical data and neutron data. Furthermore, the division of the Brillouin zone in the present case is somewhat coarse; therefore, the fine structure of the infra-red and Raman shifts may not be reproduced completely. In order to interpret them, the critical point analysis has been used following the method prescribed by *Burstein et al.* [40]. Besides above properties, the third order elastic constants (TOEC) and pressure derivatives of second order elastic constants (SOEC) have also been calculated using the VTSM.

For the calculation of Debye characteristics temperature Θ_D at various temperatures, the calculated values of C_V from equation (28) at different temperature T are to be taken and corresponding values (Θ_D/T) is determined.

4. RESULT AND DISCUSSIONS:

4.1. Phonon Dispersion Curves:

The phonon dispersion frequencies of gallium antimonides (GaSb) have been computed on the basis of present formulation. Inelastic Neutron scattering results for phonon dispersion of this compound have been reported by Farr et. al.[4] and later reproduced by Rakshit et. al.[5] with improvement by addition of Raman Scattering Experiments. We have computed phonon dispersion frequencies for the allowed 48 non-equivalent wave vectors in the first Brillouin zone by using our model parameters. We present the phonon dispersion curves [PDC] only along the three principal symmetry directions $[q, 0, 0]$, $[q, q, 0]$ and $[q, q, q]$ shown Fig. 1. Our phonon dispersion curves for GaSb agree well with the measured data reported by Singh et. al. using TSM [29]. It is evident from PDCs that our predictions using present model VTSM are better than those by using other models Singh et al. [29].

Our model VTSM has successfully explained the phonon anomalies even along $(q00)$ and (qqq) directions. From figure 1 and Table 7 it is clear that VTSM, improves result of TSM by 5.19% along $LO(X)$, 10.53% along $TO(X)$, 7.41% along $LA(X)$,

1.46% along $LO(L)$, 3.24%, along $TO(L)$, 5.65% along $LA(L)$ and 14.7% along $TA(L)$ the deviation between theory and experimental due to TSM is maximum 16.17% along $TA(L)$ while due to VTSM the deviation is maximum 1.47% along $TA(X)$. Even in the present model no deviation has been observed along $LO(X)$, $LO(L)$, $LA(L)$. However, the predictions of PDC for GaSb by using VTSM may be considered more satisfactory than those from three body force shell model (TSM) [29].

4.2. Third Order Elastic Constants, Pressure Derivatives of SOECs and Values of Cauchy Discrepancy:

It is interesting to note that our results on TOEC and pressure derivatives of SOEC for gallium antimonides (GaSb) are comparable to others are closer to their experimental values. The present model computation of Third order elastic Constants (TOEC) have been reported in Table 3 and compared with the available experimental data [33]. Further pressure derivatives of SOEC for GaSb are compared with available experimental data [33] in table 4. It can also be seen from Table 5 that Cauchy discrepancies are smaller for TOE constants than for SOE constants. A possible explanation for this fact seems to be that many body and/or thermal effects are more pronounced for SOEC than for TOEC.

Table 3. Third Order Elastic Constants (in the unit of 10^{12} dyne/cm²)

Property	Present Study	Experimental [52]
C_{111}	-3.697	-4.75
C_{112}	-3.408	-3.08
C_{123}	-1.983	-0.44
C_{144}	0.148	0.50
C_{166}	-2.134	-2.16
C_{456}	-0.198	-0.25

Table 4: Values of pressure derivatives of SOEC (in dimensionless) for GaSb.

Properties	Values	
	Present Study	Exp. [53]
dK'/dP	0.91	1.00
dS'/dP	-0.54	0.15
dC'_{44}/dP	4.75	4.80

Table 5: Values of Cauchy discrepancy of TOEC (in the units 10^{12} dyne cm⁻²) for GaSb.

Properties	Values
$C_{112}-C_{166}$	-1.274
$C_{123}-C_{456}$	-2.181
$C_{144}-C_{456}$	0.346
$C_{123}-C_{144}$	-2.131

4.3. Combined Density of States:

The present model is also capable to predict the two phonon Raman/IR spectra. The results of these investigations from combined density states (CDS) approach have been shown in figure 2 which shows that the agreement between experimental data and theoretically observed data [47, 48] and our theoretical peaks is generally good for both two phonon Raman/IR spectra for GaSb. The assignment made by the critical point analysis in the present study has been listed in Table 6.

Table: 6. Assignments for the observed peak positions in Combined (Two Phonon) Density of States in terms of selected phonon frequencies at Γ , X and L critical points.

CDS Peak s (cm ⁻¹)	Raman Active			Infra-Red Active		
	Neu- tron Data Peak s (cm ⁻¹) [4 & 54]	Present Study Val- ues (cm ⁻¹)	Assign- ments	Ob- serv ed IR Peak s (cm ⁻¹) [54]	Present Study Val- ues (cm ⁻¹)	Assign- ments
126	126	126	2TA(L)
164	164	164	2TA(L)
269	269	269	LA(L)+ TA(L)
410	412	410	2LA(L)	411	410	2LA(L)
467	469	467	TO(L)+ LA(L)	468	468	TO(L)+LA (L)
524	526	524	2TO(L)	524	523	2TO(L)
544	542	544	2TO(Γ)	544	544	2TO(Γ)

The interpretation of Raman / IR spectra achieved from both CDS approach and critical point analysis is quite satisfactory.

4.4. Debye Temperature Variation:

Specific Heat C_v values calculated on the basis of present model have presented with better agreement at low temperatures also upto 1⁰K which shows predominant with the other zinc-blende semiconductor structure crystals. In the present study the variation of Debye temperatures (Θ_D) at different temperatures T have also been calculated and curve has been plotted along with available experimental data [50] in Fig. 3. If we start from the lowest temperature first decreases to a minimum, then increases to a flat maximum and finally decreases slowly. Though, the agreement is better with our model, still, there is slight discrepancy between theoretical and experimental results at higher temperatures.

5. CONCLUSION:

The computed phonon dispersion curves in Fig. 1 show that inclusions of Van der Waals interactions have improved the

agreement between experimental data[4]and our results. The inclusion of Van der Waals interaction (VWI) with (TBI) have influence both longitudinal and transverse optic modes much more than acoustic branches. The agreement between theory and experimental data at X- point is also excellent. Another striking feature of present model is noteworthy from the excellent reproduction of almost all branches hence the prediction of phonon dispersion curves (PDC) for GaSb using (VTSM) may be considered more satisfactory than from other models [49] . The combined density of states (CDS) for GaSb has plotted in Fig. 2 the basic aim of the study of two phonon IR/Raman scattering spectra is to correlate the neutron and optical experimental data of GaSb. We have also investigated the anharmonic properties of GaSb by using VTSM. Here third order elastic constants (TOEC) and pressure derivatives of second order elastic constants (SOEC) have been compared with experimental available data [33] which shows our result is excellent. We have also plotted Debye temperature curve at temperature (T). It is evident from Fig. 3 our results is good with measured experimental data [50] and generally better than of that of three body shell model [TSM].

Table 7 . Comparison of frequencies from various sources (X and L points) for GaSb

Zone (THz)	Expt. [4]	TSM [29]			VTSM (Present study)			% improvement (a~b)	
		Value	\pm deviation	% (a)	Value	\pm deviation	% (b)		
X (100)	LO	6.35	6.68	0.051	5.19	6.35	00	00	5.19
	TO	6.36	5.7	0.103	10.37	6.37	0.001	0.15	10.53
	LA	4.99	4.6	0.078	7.81	4.97	0.004	0.40	7.41
	TA	1.698	1.7	0.001	0.11	1.7	0.001	0.11	00
L(.5.5.5)	LO	6.15	6.06	0.014	1.46	6.15	00	00	1.46
	TO	6.48	6.7	0.03	3.39	6.49	0.001	0.15	3.24
	LA	4.6	4.34	0.05	5.65	4.6	00	00	5.65
TA	1.36	1.58	0.16	16.17	1.38	0.014	1.47	14.7	

In this paper, we have systemcally reported phonon dispersion curves, combined density of states, Debye temperature and a part of anharmonic property of GaSb. On the basis of overall discussion, it is concluded that the present van der waals three body force shell model (VTSM) is adequately capable of describing the crystal dynamic study of gallium antimonides (GaSb).

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AUTHOR'S PROFILE:

K.K.Mishra



is currently pursuing Doctorate degree program in Material Reseach Division, Dept of Physics at Nehru Gram Bharati University, Allahabad, India under kind supervision of Prof. K.S.Upadhyaya, Mob-+91-9451230023. E-mail: krrish1225@gmail.com

Prof. K.S.Upadhyaya



is currently Professor and Dean Faculty of Science, Nehru Gram Bharati University, Allahabad, India, He has more than 39 years experience of teaching as well as research. He has also more than two hundred publication in various international and national Journals and supervised/co-supervised 30 Ph.D.Scholars. Mob-+91-9415375648, E-mail: kripa1shankar@yahoo.com.

Appendix:

