

Interatomic interactions and the lattice statics and dynamics of TlBr crystals for both CsCl and NaCl phases

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The thallium group of halides have certain characteristic properties compared to the other alkali-halide crystals. The peculiarities are manifest in connection with the static lattice structure, the consistent description of the dielectric properties and the phonon dispersion relation, and the unified study of the lattice statics and dynamics. In this report we present a coherent account of the diverse properties of the TlBr crystal from a single model, namely, the deformable shell model with a single set of model parameters. Apart from this we present for the first time the theoretical calculation of the phonon dispersion relation for the NaCl-phase TlBr crystal. This prediction, we feel, will be helpful to the experimentalist in identifying the modes of polarization, the continuity of a particular branch in the overlapping regions and the like, if not quantitatively the frequencies themselves. Moreover, the present investigation tries to indicate the relative importance of the different interatomic interactions in relation to the typicalities. Finally it is seen that an overall description of the lattice mechanical properties of the TlBr crystal in both phases is obtained in the present model. Some of the shortcomings and possible means of improving the present calculation are also pointed out.

I. INTRODUCTION

Studies of the lattice dynamics of ionic solids have become legion. In recent years several reviews¹⁻⁴ have appeared emphasizing the different aspects of the problem. While it is gradually becoming clear that the lattice dynamics of solids cannot be studied in isolation, little attention has been paid to study the same in conjunction with other properties, like the relative stability of structure, the cohesive energy, the anharmonic properties, etc. Several models⁵⁻⁹ which have been so far developed mainly discuss the lattice dynamics of ionic crystals. They are to be suitably modified to study other properties.^{10,11} On the other hand the deformable-shell model¹²⁻¹⁴ is free from the limitations mentioned above and has already been successfully applied to calculate simultaneously the lattice statics and dynamics of a few alkali-halide crystals.¹⁵⁻¹⁷ A first-principles calculation of the lattice dynamics of ionic crystals is beset with many difficulties. Although recently several attempts¹⁸⁻²¹ at constructing microscopic theories have made some progress, they are still far from being completely satisfactory. Hence comprehensive calculations of the properties of a solid based on sensible models remain useful as a guide to both theoretical and experimental physicists. In the present work we propose to do a calculation of the lattice statics and dynamics of the TlBr crystal in both phases, from a single model, namely, the deformable shell model with

a single set of model parameters. We have selected this crystal because of the following peculiarities. Firstly the three Tl halide crystals have a time old problem²² regarding the stability of their static lattice structure. It is now well known that no reasonable two-body central interaction can explain their structure. Secondly, in this group of crystals a consistent description of the high- and low-frequency dielectric constants and the phonon frequencies has not been obtained in the calculations done so far. Cowley *et al.*²³ obtained a reasonable fit for the dispersion of phonons in a 14-parameter shell model which altered the static and high frequency dielectric constants by about 9% and 15% respectively. The 11-parameter variable-charge shell-model calculation by Srinivasan *et al.*²⁴ improves the agreement with respect to the high-frequency dielectric constant, but the discrepancy in the case of static dielectric constant is about 18%. Both calculations are, however, unable to take account of the cohesive energy, the relative stability, etc. Moreover, the physical significance of some of the parameters in the model is not clear. Thirdly, the pronounced violation of the Cauchy relation is another interesting feature of the group. Another theoretically attractive feature of the TlBr crystal is that it has a NaCl phase other than the normal CsCl phase for which some measurements are available. In Sec. II we give a brief description of the deformable-shell model and the method of determination of the parameters.

II. DEFORMABLE-SHELL MODEL AND THE DETERMINATION OF THE MODEL PARAMETERS

The details of the deformable-shell model developed by Basu and Sengupta are given in Refs. 12, 13, and 15. In order to write down the energy expression in the above model,³³ let us divide every ion into a core of charge $Z_i + Y_i$ and a shell of charge $-Y_i$. If \vec{R}_i and \vec{R}_i^s represent the position of the core and the shell, respectively, then the dipole moment and the self-energy of each ion are

given by

$$\mu_i = -Y_i(R_i^s - R_i) \quad (1)$$

and

$$\frac{1}{2}K_i(R_i^s - R_i)^2 = \mu_i^2/2\alpha_i, \quad (2)$$

where $\alpha_i = Y_i^2/K_i$, K_i being the core-shell spring constant. Assuming the overlap interaction to act through the shells only, the total energy expression in the deformable-shell model is

$$W = \frac{1}{2} \sum_{ij}' \frac{Z_i Z_j}{R_{ij}} + \frac{1}{2} \sum_{ij} b \exp(-R_{ij}/\rho) + \frac{1}{2} \sum_{ij}' -\frac{C_{ij}}{R_{ij}^6} + \frac{1}{2} \sum_{ij}' -\frac{D_{ij}}{R_{ij}^8} + \frac{1}{2} \sum_{jm} \sum_i A(K) \exp\left(-\frac{R_{ij} + R_{im}}{\rho}\right) - \sum_i \mu_i E_i^m - \frac{1}{2} \sum_i \mu_i E_i^u + \frac{1}{2} \sum_i \frac{\mu_i^2}{\alpha_i} + \frac{1}{2} \sum_{ij}' \Phi \left(\left| R_{ij} - \frac{\mu_i}{Y_i} + \frac{\mu_j}{Y_j} \right| \right); \quad (3)$$

the dipole moment μ_i being determined by the ionic coordinates through the equation

$$\frac{\partial W}{\partial \mu_i} = 0, \quad (4)$$

where the first term represents the Coulomb interaction between the ions, the second one the short-range repulsive interaction (b , ρ being the range and hardness parameters), the third and fourth the dipole-dipole and the dipole-quadrupole van der Waals interactions, respectively, the fifth the short-range three-body interaction arising out of the isotropic deformation of the charge cloud round each ion [$A(K)$ being the deformability parameter characteristic of the K ion] and the last four terms represent the energy expression corresponding to the both-ion-polarizable shell model²⁷ (Y 's and K 's being the appropriate shell charges and the core-shell spring constants).

Of the terms present in the energy expression the first four terms have a sound quantum-mechanical basis. The last four terms, namely those representing the shell model, have been justified in the works of Tolpygo and Sinha and more recently by Basu and Sengupta, but the specific forms that are in use are still questioned. This point we shall discuss in Sec. V. The justification for the fifth term stems from the works of Lowdin and Lundqvist who have demonstrated the importance of the many-body interaction in the case of ionic solids. Recently Ghosh and Basu¹⁶ have given a derivation of this term on the basis of the Thomas-Fermi-Dirac statistical model. Hence we find that the different interactions that are being considered in the present model have some at least qualitative plausibility justification.

In order to reduce the number of parameters we

have made some simplifying assumptions. As we intend to obtain a rough overall description of the lattice mechanics of the TlBr crystal in both phases with a single set of parameters rather than to obtain an exact fit for any particular property, these assumptions, we believe, will not be unjustified. The assumptions are the second-neighbor overlap is neglected, the many-body interaction is confined to the case where one of the particles is a common nearest neighbor of the other two, and while the Van der Waals interaction is retained for all neighbors in the expression for the cohesive energy and the elastic constants, it is neglected beyond nearest neighbor in the phonon frequency calculation; lastly, the ionic charge is fixed to unity.

Now, using Eqs. (3) and (4) all the lattice mechanical properties can be calculated. In this model we have altogether eight parameters, namely, b , ρ , $A(1)$, $A(2)$, $Y(1)$, $Y(2)$, $K(1)$, and $K(2)$ which are determined from the following equations. Using Eq. (3) we write the energy per unit cell for the CsCl-structure lattice

$$\Phi_{\text{CsCl}} = -\frac{\alpha_{\text{CsCl}} e^2}{\tilde{r}} + 8b e^{-\tilde{r}/\rho} + 28A e^{-2\tilde{r}/\rho} - \frac{C_{\text{CsCl}}}{\tilde{r}^6} - \frac{D_{\text{CsCl}}}{\tilde{r}^8}, \quad (5)$$

where α_{CsCl} is the Madelung constant for CsCl structure and \tilde{r} the harmonic nearest-neighbor distance.

From Eq. (3), Sarkar and Sengupta²⁵ using the homogeneous deformation theory have obtained the expression for the elastic constants. Using these equations we get for the harmonic compressibility $\tilde{\beta}$,

$$\begin{aligned} \tilde{\beta}^{-1} = & \frac{1}{\sqrt{3}} \left(\frac{\phi''}{\tilde{r}} - \frac{\phi'}{\tilde{r}^2} \right) - \frac{0.381e^2}{\tilde{r}^4} \\ & + \frac{7}{\sqrt{3}} \left(\frac{2}{\rho^2 \tilde{r}} + \frac{1}{\rho \tilde{r}^2} \right) A e^{-2\tilde{r}/\rho} \\ & - \frac{1}{3} \left(\frac{C_1}{\tilde{r}^9} + \frac{D_1}{\tilde{r}^{11}} + \frac{2C_2}{\tilde{r}^9} + \frac{2D_2}{\tilde{r}^{11}} \right), \end{aligned} \quad (6)$$

where $\phi = be^{-\tilde{r}/\rho}$, $A = A(1) + A(2)$, and the C 's and D 's are the coefficients of the Van der Waals terms for the elastic constant including the lattice sums.²⁶ The equilibrium of the static lattice structure is obtained from the following equation:

$$\frac{d\phi_{CsCl}}{d\tilde{r}} = 0. \quad (7)$$

Using the harmonic values of the compressibility, the lattice constant, and the cohesive energy the three potential parameters b , ρ , A are obtained from Eqs. (5)–(7). The parameter $A(2)$ is determined from the LA frequency for $\tilde{q} \equiv (0.5, 0, 0)$ by the following equation:

$$R_0 + \frac{e^2}{2r^3} C(1) + \frac{64A(2)e^{-2r/\rho}}{\rho^2} = (m_{T1+})\omega_{LA}^2 \quad (8)$$

where

$$\begin{aligned} R_0 = & \frac{8}{3} \left[\left(\frac{1}{\rho^2} - \frac{2}{\rho r} \right) b e^{-r/\rho} \right. \\ & \left. + \left(\frac{6}{\rho^2} - \frac{14}{\rho r} \right) A e^{-2r/\rho} - \frac{30C_{+-}}{r^8} - \frac{56d_{+-}}{r^{10}} \right] \end{aligned} \quad (9)$$

where C_{+-} , d_{+-} are the van der Waals interaction constants between the positive and negative ions.³⁵ $C(1)$ is the Coulomb coupling coefficient.

The remaining four parameters, the shell charges and the core-shell spring constants are evaluated from the values of the dielectric constants and the LO and TO frequencies for $\tilde{q} \equiv (0, 0, 0)$. The relevant equations are given by Woods *et al.*²⁷ Thus all eight parameters are obtained and given in Table I together with the input data. An idea about the uncertainties of the values of the parameters is obtained from the accuracy of the different input data. The harmonic values of the cohesive energy, the compressibility, and the lattice constant have accuracies of the order of 2%, 1%, and 0.1%, respectively. The uncertainties in the measured values of ϵ_0 and ϵ_∞ and the phonon frequencies are about 0.1%, 0.05%, and 2%. All the input data used are kept within these limits.

Further, we consider the parameters of the model to be independent of structure. This is true of the four potential parameters, b , ρ , $A(1)$, and $A(2)$, but may not be quite correct concerning the shell model parameters, i. e., the shell charges and the core-shell spring constants. But even if they are different for the NaCl phase of the crystal, the difference is certainly negligible, as is borne out by the results of the calculations that follow. With the set of parameters thus

TABLE I. Input data and values of the parameters. The quoted cohesive energy and bulk modulus refer to the harmonic values. The exact experimental values are given in parentheses when they differ from the input data.

Property		Input values	Parameters	Values
Cohesive energy ^a (kcal/mol)	$U(r)$	166.0061 (169.1)	b (10^{-8} erg)	0.3615
Bulk modulus ^b (10^{12} dyn cm ⁻²)	$\tilde{\beta}^{-1}$	0.2633	ρ (10^{-8} cm) $A(1)$ (10^{-6} erg)	0.3610 1.3034
Nearest neighbor distance (10^{-8} cm) ^c	\tilde{r} r	3.4044 3.4207(100 °K)	$A(2)$ (10^{-6} erg)	-0.4277
Dielectric constant ^d	ϵ_0 ϵ_∞	33.50(100 °K) 5.54(100 °K)	$Y(1)$ $Y(2)$	8.5600 3.1592
LA frequency ^e (10^{12} Hz [$\tilde{q} \equiv (0.5, 0, 0)$])	ν_{LA}	1.92	$K(1)$ (10^4 dyn/cm) $K(2)$ (10^4 dyn/cm)	440.0 44.0
Long-wavelength optical ^f frequencies (10^{12} Hz) [$\tilde{q} \equiv (0, 0, 0)$]	ν_{LO} ν_{TO}	3.37 1.39		

^aReference 28.

^bReference 29.

^cReference 29.

^dReference 30.

^eReference 23.

^fReference 23.

TABLE II. Relative stability of static lattice structure for the TlBr crystal.

Structure	Harmonic nn distance (10^{-8} cm)	Contribution to cohesive energy by different interaction (kcal/mol)				Cohesive energy (kcal/mol) Total	Phase transition pressure (kbar)	Phase transition volume (10^{-24} cm ³)
		Coulomb	Repulsive	van der Waals	Many body			
NaCl phase	3.3303 (3.28) ^a	-174.3168	+30.8667	-23.0589	1.8466	-164.6625	-7.1073	13.1273
CsCl phase	3.4044	-172.0000	+33.3652	-29.6371	2.2658	-166.0061		

^aRefer to the experimental nearest-neighbor (nn) distance for the NaCl phase, Ref. 31.

determined we calculate some of the principal lattice properties of the TlBr crystal.

III. LATTICE STATICS

In this section we consider first of all the relative stability of the static lattice structure. It is well known, as has been mentioned earlier, that no two-body central interaction can account for the structure of this crystal. In order to calculate the structure in the present model we write down the energy per unit cell corresponding to the NaCl structure,

$$\phi_{\text{NaCl}} = -\frac{\alpha_{\text{NaCl}}e^2}{\tilde{r}} + 6be^{-\tilde{r}/\rho} + 15Ae^{-2\tilde{r}/\rho} - \frac{C'_{\text{NaCl}}}{\tilde{r}^6} - \frac{D'_{\text{NaCl}}}{\tilde{r}^8}, \quad (10a)$$

where C' and D' are the appropriate Van der Waals constants for the new structure. The equilibrium nearest-neighbor distance for the new structure is determined by solving numerically the following equation:

$$\frac{d\phi_{\text{NaCl}}}{d\tilde{r}} = 0. \quad (10b)$$

The obtained harmonic nearest-neighbor distance

is found to compare satisfactorily with the experimental value for the NaCl phase.³¹ The phase-transition pressure at $T=0$ is obtained, by neglecting the zero-point anharmonicity effect, from the Gibbs free energy for the two phases by the following equation:

$$P = (\phi_{\text{NaCl}} - \phi_{\text{CsCl}})/(v - v'), \quad (11)$$

where, v and v' are the volumes per unit cell for the CsCl and NaCl phases, respectively. The calculated values are collected in Table II. The observed CsCl structure turns out to be stable. The contributions of the individual interactions are also given in Table II. The negative phase-transition pressure indicates that the NaCl phase is possibly stable only at high temperature or low pressure, which also agrees with observations made so far.

Next we calculate the harmonic second-order elastic constants for both phases, and these are collected in Table III. The presence of many-body interaction lifts the Cauchy relation in both cases. The agreement with experiment is also found to be quite satisfactory. For the NaCl phase, however, there is no measurement available for comparison. Treusch³¹ has made an independent estimate of the bulk modulus of TlBr in the NaCl-phase from energy-band measurements and has

TABLE III. Second-order harmonic elastic constants.

Property	Expt. ^a	Present calculation	CsCl phase		NaCl phase Present calculation
			Calculated by Cowley and Okazaki ^b	Calculated by Srinivasan <i>et al.</i> ^c	
$10^{-12}\tilde{C}_{11}$	0.4500	0.4372			0.5300
$10^{-12}\tilde{C}_{12}$	0.1700	0.1764			0.09430
$10^{-12}\tilde{C}_{44}$	0.1115	0.1179			0.04196
$10^{-12}(\tilde{C}_{12} - \tilde{C}_{44})$	0.0585	0.0585	-0.002	0.045	

^aReference 29.

^bReference 23 (refers to room-temperature values).

^cReference 24 (refers to room-temperature values).

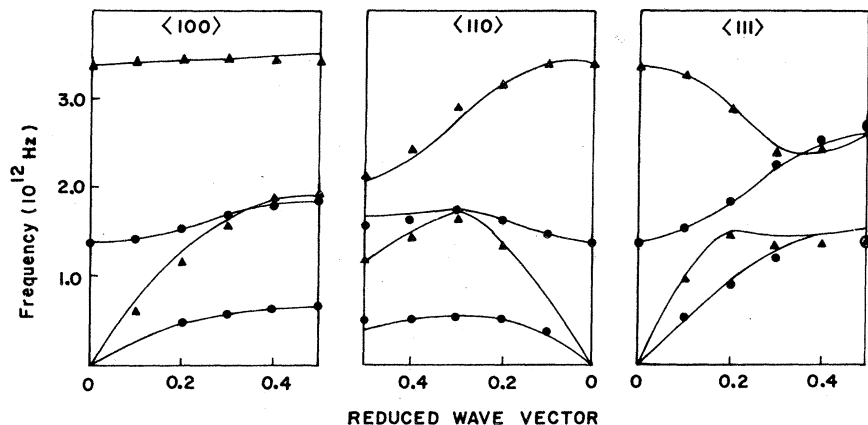


FIG. 1. Solid lines represent the deformable-shell model calculations for TlBr (CsCl phase) at 100°K. Experimental points are from Ref. 23.

found a change in bulk modulus similar to that obtained in the present investigation. This decrease in the value of the bulk modulus implies an increase in the repulsive energy as well as a decrease in the van der Waals energy (see Table II).

Thus the value of the cohesive energy which would otherwise be higher for the NaCl phase actually turns out to be lower. This also agrees with experiment, since the CsCl-phase Tl halides are the normally stable phases. Another important consequence of this is the lowering of the longitudinal phonon frequency and the increase in transverse optical frequency. This point will be discussed in the section on lattice dynamics.

IV. LATTICE DYNAMICS AND DIELECTRIC CONSTANTS

In order to calculate the dynamics, the dynamical equations are obtained by expanding \vec{R}_i in Eqs. (3) and (4) about the equilibrium configuration (see Sarkar and Sengupta³²) as follows.

$$\begin{aligned} (R + D_0 + ZCZ)U + (R + D_0 - ZCY)W &= \omega^2 m U, \\ (R^T + D_0^T - YCZ)U + (K + R + D_0 + YCY)W &= 0, \end{aligned} \quad (12)$$

where Z , Y , m are 6×6 diagonal matrices for the ionic charge, the shell charges, and the ionic mass; $U = (U_1, U_2)$ and $W = (W_1, W_2)$ are the amplitude vectors for the core-displacement and the shell-core separation vectors; R , C , K are the total overlap interaction (including Van der Waals interaction), the Coulomb interaction, and the core-shell spring interaction; and D_0 is the real 6×6 dynamical matrix corresponding to the three-body interaction [fifth term in Eq. (3)]

$$D_0 = \begin{vmatrix} M^d(33) & 0 \\ 0 & M^d(44) \end{vmatrix}. \quad (13)$$

The explicit expressions for $M^d(33)$ and $M^d(44)$ are given by Basu and Sengupta¹² and Roy *et al.*³³ for the NaCl structure and the CsCl structure, respectively. The Eqs. (12) are solved for the symmetry directions for both structures to get the eigenfrequencies shown in Figs. 1 and 2. Figure 1 shows that the frequencies (in the CsCl phase) compare quite favorably with the measured ones in all three directions. The corresponding frequencies in the NaCl phase are shown in Fig. 2. In calculating

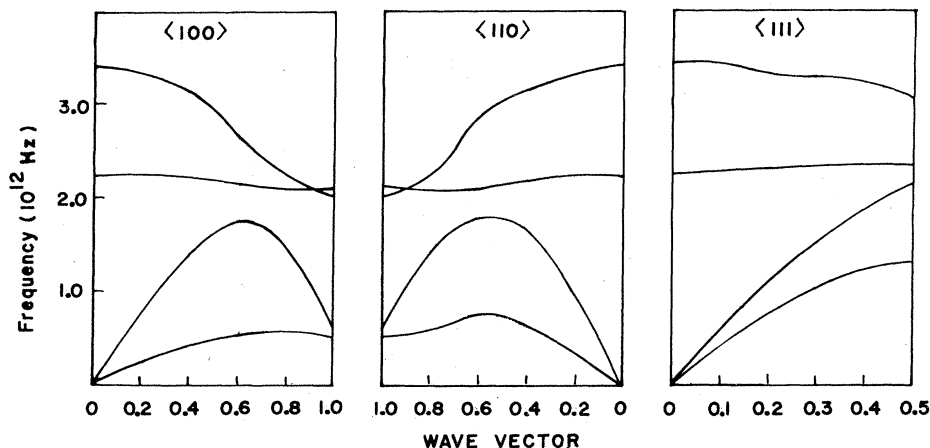


FIG. 2. Dispersion curve of TlBr in NaCl phase at room temperature.

TABLE IV. Static and high-frequency dielectric constants.

Property	Expt. ^a	CsCl phase			NaCl phase	
		Present calculation	Calculated by Cowley and Okazaki ^b	Calculated by Srinivasan <i>et al.</i> ^c	Present calculation	Estimated by Treusch ^d
Dielectric constant ϵ_0	33.50	33.59	30.50 ^e	39.40 ^e	9.30	~10.00
ϵ_∞	5.54	5.54	4.74	5.60	4.02	4.50

^aReference 30.^bReference 23.^cReference 24.^dReference 31.^eUsing the Lyddane-Sachs-Teller relation and their calculated ϵ_∞ , ω_L , ω_T .

these frequencies we have used the lattice constant as determined experimentally.³¹ All the characteristic features of the NaCl structure³⁴ are found to be present in the dispersion curves. The only other observation made in this connection by Treusch³¹ from completely separate considerations, that the ratio ω_L/ω_T should range between 1.4 and 1.8 for the NaCl phase, is found to agree with the result of the present calculation, which is 1.52. However, for any conclusive decision one must wait for a complete experimental determination of the dispersion relation in this phase.

Next we calculate the high-frequency and the static dielectric constants in the NaCl phase with the same set of parameters. The relevant equations used are the same as those of the both-ion-polarizable shell model of Woods *et al.*²⁷ with the modifications necessary to incorporate the effect of three-body and van der Waals interactions. The results are shown in Table IV together with the empirical values of the same estimated with the linear combination of atomic orbitals energy-band model by Treusch.³¹ It is interesting to note the closeness of the results obtained by two entirely different approaches.

V. DISCUSSION

The present investigation shows that the deformable-shell model with a single set of only eight parameters besides taking into account the peculiarities of the TlBr crystal provides an overall description of the lattice mechanics of the same without producing any significant discrepancy anywhere. It may be generally observed from the present calculation that the interatomic interactions affect the various lattice mechanical properties in different ways. Firstly, the significance of the van der Waals interaction which has so far been neglected is clearly demonstrated. Its contribution to the cohesive energy is >15%. The effect of this interaction on the elastic properties is quite small,

while the effect on the dielectric constant as well as on the phonon frequencies is not at all negligible. While the contribution of the three-body interaction to the cohesive energy is quite negligible (~1.5%), this is decisive in stabilizing the static lattice structure. Again, this interaction is alone responsible for the breakdown of the Cauchy relation. In addition this interaction has quite considerable effect on the zone-boundary phonon frequencies, particularly in the LO branches in all directions.

Next we consider the calculations on the NaCl phase. The agreement in the case of the lattice constant and the dielectric constants gives us some confidence about the results for the phonon frequencies in this phase. A comparison of the detailed behavior of the dispersion of phonons determined experimentally for the NaCl structure alkali halides shows some definite trends³⁴ in the [100] LO, [100] LA, and [111] LO branches at or near the zone boundaries. In the [100] LO branch there is a slope or a local minimum near the zone boundary, while there is a pronounced minimum in the [100] LA and [111] LO branches just at the zone boundary. This is particularly valid for chlorides, bromides, and iodides. The present calculation fits in well with this observation. Apart from this the present calculation will provide the structure factors for the one-phonon coherent scattering which are essential in performing thermal-neutron scattering experiments to determine the phonon frequencies.

To conclude we discuss some of the limitations of the present calculations. Although the overall agreement for the phonon frequencies in the CsCl phase is impressive and superior to that of existing calculations there are still some small localized discrepancies: the LA branch near the [111] direction zone boundary is slightly above the experimental points (~5%); the TA branch and the TO branch in the [110] direction are slightly lower and higher than the experimental ones at the zone boundary, respectively; the LO branch in the [100]

direction is about 3% higher than the experimental one at the zone boundary. It is difficult to conclude whether any adjustment in the values of the parameters can remove the discrepancies while keeping the agreement with respect to other properties intact. Rather, it appears that the effect of the quadrupolar deformation of the electron charge cloud may be a plausible means for further refinement. Another unsatisfactory feature of the present model is the rather wide variation of the value of the shell parameters (Y 's and K 's) for a small change in input data. This very fact indicates that

these parameters are to some extent artificial. The calculation of the pressure and temperature derivatives of the dielectric constants with the same set of parameters may throw some light in this direction. We hope to investigate this point in the near future.

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