Chapter - 5

DISCUSSION AND CONCLUSION

The present work is a systematic theoretical investigation of the cohesive properties of alkali halides. The model proposed for this purpose has been developed by incorporating the effects of van der Waals interactions (vdWI) and three-body interactions (a part of MBI) in the frame work of rigid shell model (RSM) organized by Cochran and his collaborators [43,126,132]. The introduction of three-body interactions in RSM leads to correction both in the cohesive energy and dynamical matrix. The overall fair agreement between our theoretical and experimental results established in the chapter 4 make it evident that the present model vTSM is reasonably realistic for describing the cohesive energy and optical properties of alkali halides.

It seems appropriate to point out that a vast amount of lattice dynamical computations have done in the past to predict the eigenfrequencies and associated physical properties of binary crystals and
they are still being pursued. The present model may be understood to provide a powerful and simple approach for a comprehensive study of the harmonic as well as the other properties of the crystals like MgO, BaF₂, CdWO₄, PbWO₄ and memory materials (optical ceramics of sodalites) in future. The only limitation of the model is the requirement of the knowledge of certain experimental information to be used as input data.

The major steps involved in the numerical computations of the above mentioned properties have been clearly indicated in chapter 4. The knowledge of the model parameters have been used to solve the secular equation for specified values of wave vectors in the first Brillion zone which is divided in an evenly spaced sample of 1000 wave vectors [52]. From the symmetry, these 1000- points are reduced to 56 non-equivalent points at which the vibration frequencies have been obtained by solving the secular determinant. These frequencies represent the complete vibration (or phonon) spectra. The physical properties derived from these spectra have been discussed below.

5.1 Cohesive energy of alkali halides:-

The binding in the ionic crystals has been explained by calculating their cohesive energy from a large number of interatomic
interaction potentials. The results of computations carried out in the present study are reported in the table 1&2. The values of van der Waals dipole-dipole coefficients for all alkali halides are calculated from the Slater-Kirkwood variational method [58]. It is found that the present values of van der Waals energies are very much similar the evaluated value of earlier researchers [58,171,172,173]. The reason for the differences between the results of the present work [48] and other researchers are due to the effective number of electrons in the ions. To assess the merit of the present study of van der Waals energies and zero-point energy, we have computed the values of cohesive energy of alkali halides [49]. The theoretical results thus obtained have been compared with the experimental data [84,182]. In order to show the effectiveness of various interactions involved, it is found that TBI play an important role in describing the cohesive energy of alkali halides. It is evident from the descriptions that the most realistic model for the lattice dynamics and statics of these crystals can be developed by introducing the effect of van der Waals interactions (vdWI), and three body interactions (TBI) in the framework of RSM, where the short-range interactions have been considered up to the second neighbours. The development of such a lattice
dynamical model has contributed extensively in the calculation of cohesive energies of alkali halides. The present model RSM in a way as outline above has been applied to investigate the complete cohesive energies of alkali halides. The effect of TBI and vDW and zero-point energies are quite significant and play a vital role in the description of cohesive energies of alkali halides [49]. The atomization energies of alkali halides are computed with great satisfaction with previous results [7]. The positive value of difference between cohesive energy and atomization energy gives better idea about the stability of alkali halides.

5.2 Indices of refraction of alkali halides:-

It is evident that the inclusion of three-body interactions does not lead to any deviation from the LST (Lyddane-Sachs-Teller) relation but modification in Lorentz-Lorentz and Claussius-Mosotti relations. Having taken the van der Waals interactions and three body interactions in the frame work of rigid shell model, we have computed the indices of refraction for alkali halides crystals [50]. The results are presented with the known literature values [66, 67, 68, 69, 70, and 71] of indices of refraction in the table-(4.5). The present values were found to be in a reasonable agreement with the experimentally known indices of refraction.
of alkali halides [69, 70, and 71] and also the indices of refraction, theoretically determined by Kumar et.al & Reddy et.al [66, 67, 68].

5.3 Some Concluding Remarks:

In view of the overall achievements described above, it may be concluded that the modification introduced by TBI and vdWI in the framework of RSM is very much importance in the crystals under considerations. In fact, the present model vTSM has revealed much better descriptions of dynamical properties of these compounds than those obtained by rigid shell model [52], deformation shell model [41], breathing shell model [39], extended three-body force shell model [11].

To sum up the overall achievements given in this thesis, it is concluded that the modifications introduced by vdWI and TBI in the framework of RSM with second neighbour repulsive interactions is important in alkali halides. In fact the present model vTSM has revealed better descriptions of cohesive properties of the solids under consideration. At last, we can say that the inclusion of the van der Waals interactions and three body interactions are essential for the description of cohesive properties of these crystals.