

Lattice dynamical calculations for bcc caesium chloride

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Abstract

We present a lattice dynamical calculation of Caesium Chloride (CsCl) whose atoms form a bcc lattice having one type of atom at the cube centre and the other type on the corners of the cube. Dispersion curves, density of state, and lattice specific heat of bcc Caesium Chloride were computed. The code used in the computation of the densities of state and dispersion curves was obtained from the Boardman Physics program book which we modified to obtain dispersion relation in the principal symmetry directions. In general, the obtained results agree reasonably well with the experimental data of the bcc Caesium Chloride.

Keywords: Bcc caesium chloride; Lattice dynamics; Phonon dispersion; Density of state; Specific heat

1.0 Introduction

Atoms in crystalline solids vibrate about their mean equilibrium positions at all temperatures [1]. These vibrations are responsible for a number of important properties of crystalline solids, some of which are the heat capacity, mechanical, heat transport and related physical properties of the solids. The study of these vibrations whose quanta are called phonons requires the knowledge of dispersion curves and density of states.

Despite the role played by the density of states, it is very difficult to measure directly. Experimental data will normally give information on the phonon dispersion curves, and these will be used to determine the interatomic force constants, by comparing the experimental curves with those calculated from the force constants. Even when these have been determined, it is by no means simple to calculate the density of states analytically, but modern computing methods have proved an ideal tool for solving the problem numerically [2].

The space lattice of caesium chloride is body-centered cubic. The primitive basis of two identical atoms at 000 and $\frac{1}{2} \frac{1}{2} \frac{1}{2}$ is associated with each lattice point. Each atom has two nearest neighbours and eight next nearest neighbours.

In this paper, a simulation program is used and the ratio of the two force constants is set so as to generate a theoretical set of dispersion curves and examine the effect of the two constants on the dispersion curves. We also choose values for the force constant in the secular determinant and evaluate the determinant over a mesh of points in k-space. We built up a histogram of density of states from which we determine the specific heat of the solid.

2.0 Theory

2.1 Dynamical matrix for a body- centered CsCl lattice (second nearest neighbor)

In the harmonic and adiabatic approximations, the phonon frequencies of cubic systems are determined by solving the usual secular equation given by

$$|D(q) - \omega^2 I| = 0 \quad (1)$$

where ω , $D(q)$, M and I are in their usual meanings.

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The dynamical matrix element for a body-centered CsCl lattice is given by the following matrixes elements [3] given as

$$\Phi_1(1,1) = \frac{8}{3}\gamma(m_1m_2)^{-\frac{1}{2}} \begin{pmatrix} \rho^{-\frac{1}{2}}(1 + \gamma_x) & 0 & 0 \\ 0 & \rho^{-\frac{1}{2}}(1 + \gamma_y) & 0 \\ 0 & 0 & \rho^{-\frac{1}{2}}(1 + \gamma_z) \end{pmatrix} \quad (2)$$

$$\Phi_1(1,2) = \frac{8}{3}\gamma(m_1m_2)^{-\frac{1}{2}} \begin{pmatrix} -C_xC_yC_z & S_xS_yC_z & S_xC_yS_z \\ S_xS_yC_z & -C_xC_yC_z & C_xS_yS_z \\ S_xC_yS_z & C_xS_yS_z & -C_xC_yC_z \end{pmatrix} \quad (3)$$

$$\Phi_1(2,1) = \frac{8}{3}\gamma(m_1m_2)^{-\frac{1}{2}} \begin{pmatrix} -C_xC_yC_z & S_xS_yC_z & S_xC_yS_z \\ S_xS_yC_z & -S_xS_yC_z & C_xS_yS_z \\ S_xC_yS_z & C_xS_yS_z & -C_xC_yC_z \end{pmatrix} \quad (4)$$

$$\Phi_1(2,2) = \frac{8}{3}\gamma(m_1m_2)^{-\frac{1}{2}} \begin{pmatrix} \rho^{-\frac{1}{2}}(1 + \gamma_x) & 0 & 0 \\ 0 & \rho^{-\frac{1}{2}}(1 + \gamma_y) & 0 \\ 0 & 0 & \rho^{-\frac{1}{2}}(1 + \gamma_z) \end{pmatrix} \quad (5)$$

Combining Eqs. (2), (3), (4) and (5) gives

$$\mathfrak{D}(\vec{k}) = \begin{pmatrix} \Phi_1(1,1) & \Phi_1(1,2) \\ \Phi_1(2,2) & \Phi_1(2,1) \end{pmatrix} \quad (6A)$$

$$\mathfrak{D}(\vec{k}) = \frac{8}{3}\gamma(m_1m_2)^{-\frac{1}{2}} \begin{pmatrix} \rho^{-\frac{1}{2}}(1 + \gamma_x) & 0 & 0 & -C_xC_yC_z & S_xS_yC_z & S_xC_yS_z \\ 0 & \rho^{-\frac{1}{2}}(1 + \gamma_y) & 0 & S_xS_yC_z & -C_xC_yC_z & C_xS_yS_z \\ 0 & 0 & \rho^{-\frac{1}{2}}(1 + \gamma_z) & S_xC_yS_z & C_xS_yS_z & -C_xC_yC_z \\ -C_xC_yC_z & S_xS_yC_z & S_xC_yS_z & \rho^{-\frac{1}{2}}(1 + \gamma_x) & 0 & 0 \\ S_xS_yC_z & -C_xC_yC_z & C_xS_yS_z & 0 & \rho^{-\frac{1}{2}}(1 + \gamma_y) & 0 \\ S_xC_yS_z & C_xS_yS_z & -C_xC_yC_z & 0 & 0 & \rho^{-\frac{1}{2}}(1 + \gamma_z) \end{pmatrix} \quad (6)$$

where $C_x = \text{Cos}K_xa$, $S_x = \text{Sin}K_xa$, $C_{2x} = \text{Cos}2K_xa$, $C_y = \text{Cos}K_ya$, $S_y = \text{Sin}K_ya$, $C_{2y} = \text{Cos}2K_ya$, $\gamma_x = \frac{3}{4}R(1 - \text{Cos}2K_za)$ and $\rho = m_1/m_2$ (the ratio of mass of the atoms in the cell)

In order to make an analytical calculation of the density of states, it is necessary to integrate $(\nabla_k w)^{-1}$ over a constant frequency surface in k-space. Since this is not normally feasible, the usual approach is to compute the density of states numerically [5]. This is usually referred to as ‘‘root sampling method’’ since it build up the density of states by finding the roots of the secular equation at large number of points in the brillouin zone.

The usual expression for specific heat (C_v) in the Debye model with the modification made by Born is given by

$$C_v = 9nNk_B \left(\frac{T}{\theta_D} \right)^3 \int_0^{(\theta_D/T)} \frac{x^4 e^x}{(e^x - 1)^2} = f \left(\frac{\theta_D}{T} \right) \quad (7)$$

where
$$\theta_D^3 = \frac{18nN\pi^2\hbar}{k_B V \left(\frac{1}{c_l^3} + \frac{2}{c_t^3} \right)} \quad (8)$$

and $x = (\hbar\omega/k_B T)$. The quantity that reflects the properties of the material is θ_D , which depend on n , V , c_l , and c_t , where c_l and c_t elastic wave velocity for longitudinal and transverse waves, n is the number of atom, k_B is Boltzmann constants and V is the molar volume.

3.0 Results and discussion

An understanding of microscopic properties of insulating materials requires the phonon dispersion curves and density of states. In spite of the fundamental role played by density of states, it is very difficult to measure directly. The work was motivated by a recent theoretical study of the dispersion relations of bcc cerium [4] to develop a numerical method of computing not only the dispersion relations but also the densities of states and specific heat capacity.

The program used in this work originated from the “Boardman Physics Program” book, [3]. The modifications made to it, as explained in [4], include; reformatting the program to calculate in double precision in order to reduce round off errors and eliminate singularities; substituting the NAG’s subroutine with a more readily available one. We also replaced the NAG’s subroutine F02AAF with the Numerical Recipes [5] real matrix eigenvalue solver.

The normal mode frequencies obtained for [100], [110], and [111] over a cubic mesh point are weighted and stored in histograms, to form the densities of states for each mode of vibration, presented in Figure 4. Figure 5 is the total density of states for bcc caesium chloride.

4.0 Conclusions

A Fortran program developed for personal computers [2] for calculating phonon dispersion curves and density of states [Keeler 1980] was used for bcc caesium chloride structures. The efficacy of the method is seen from the agreement of the computed dispersion curves and the densities of states, bcc caesium chloride, with the theoretical and experimental results found in literatures [6]

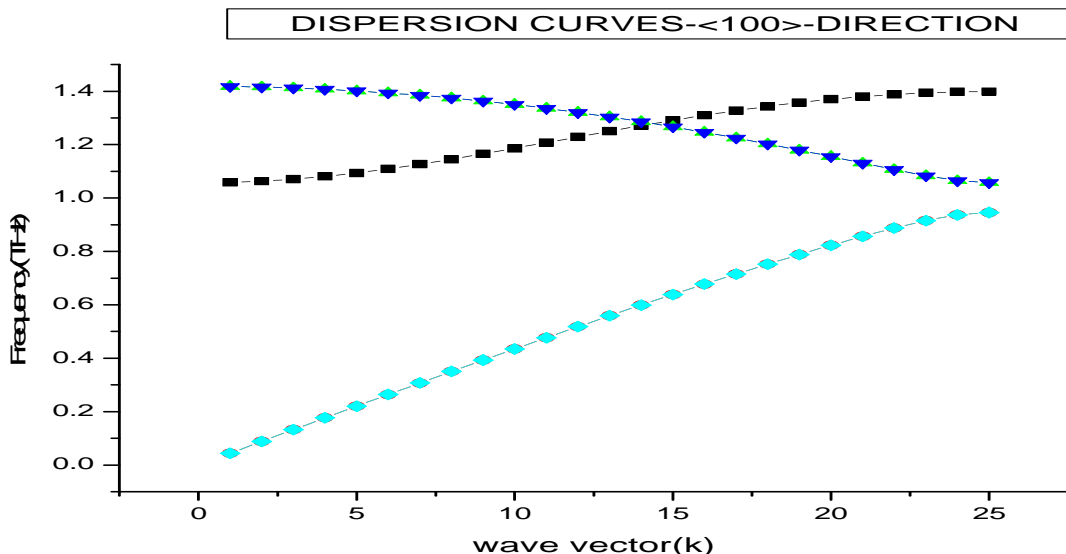


Figure 1. The [100] phonon dispersion curves for bcc CsCl, force constant ratio $R = 0.8$ where the blue line indicates Longitudinal branch and the black and the sky blue indicates the transverse branch.

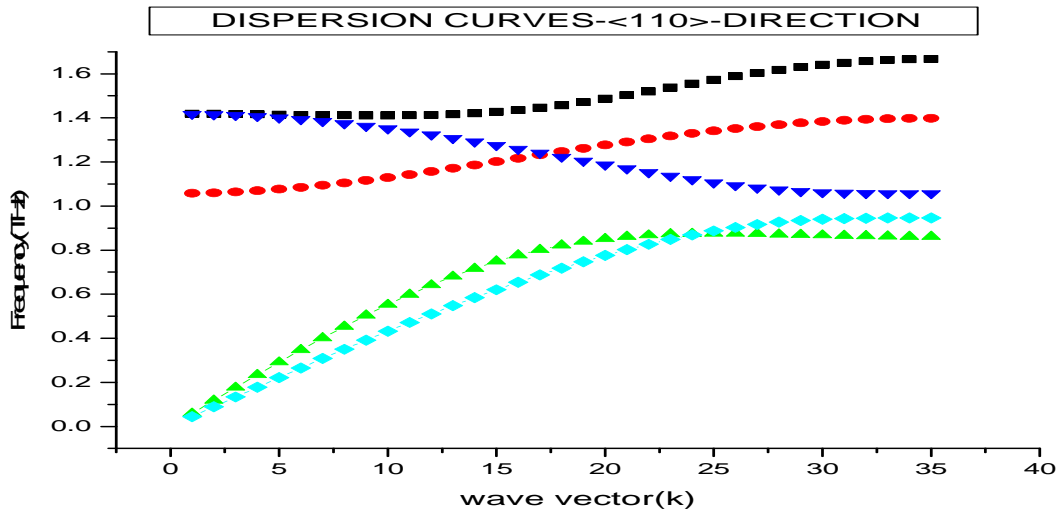


Figure 2. The [110] phonon dispersion curves for bcc CsCl, force constant ratio $R = 0.8$ where the black and blue lines indicates the longitudinal branch and the red, sky blue and the light green lines represents the transverse branch

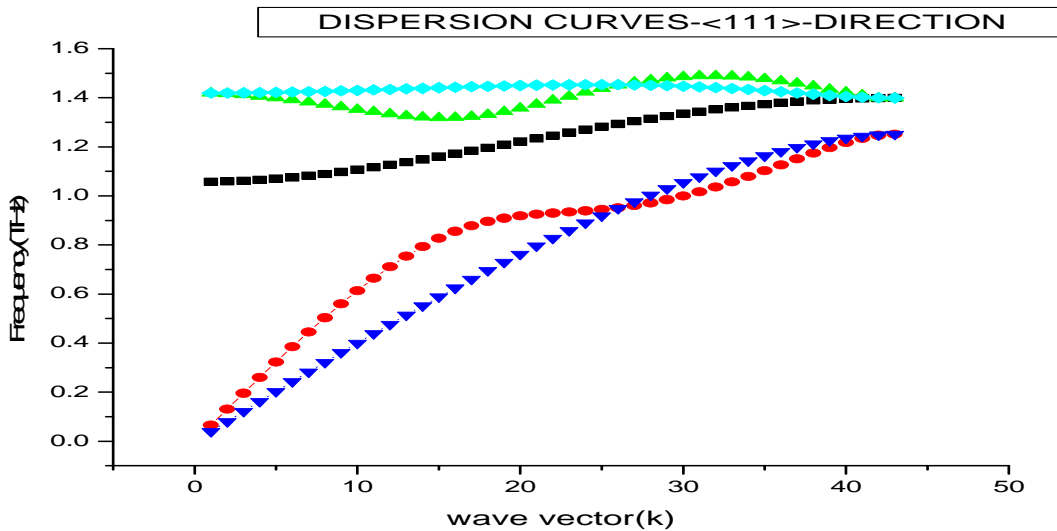


Figure 3. The [111] phonon dispersion curves for bcc CsCl, force constant ratio $R = 0.8$ where the sky blue and the light green lines represents the longitudinal branch and the black, red and the blue lines indicates the transverse branch of the dispersion curve.

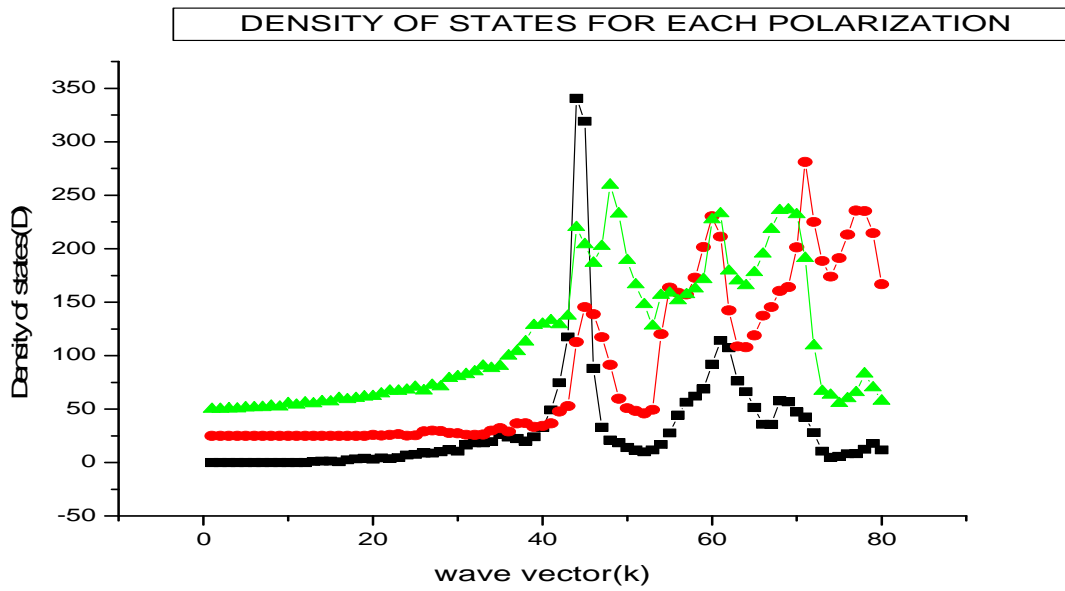


Figure 4. The phonon densities of state for the low-, medium-, and high-frequency polarization for a one-atom bcc lattice with $R = 0.8$.

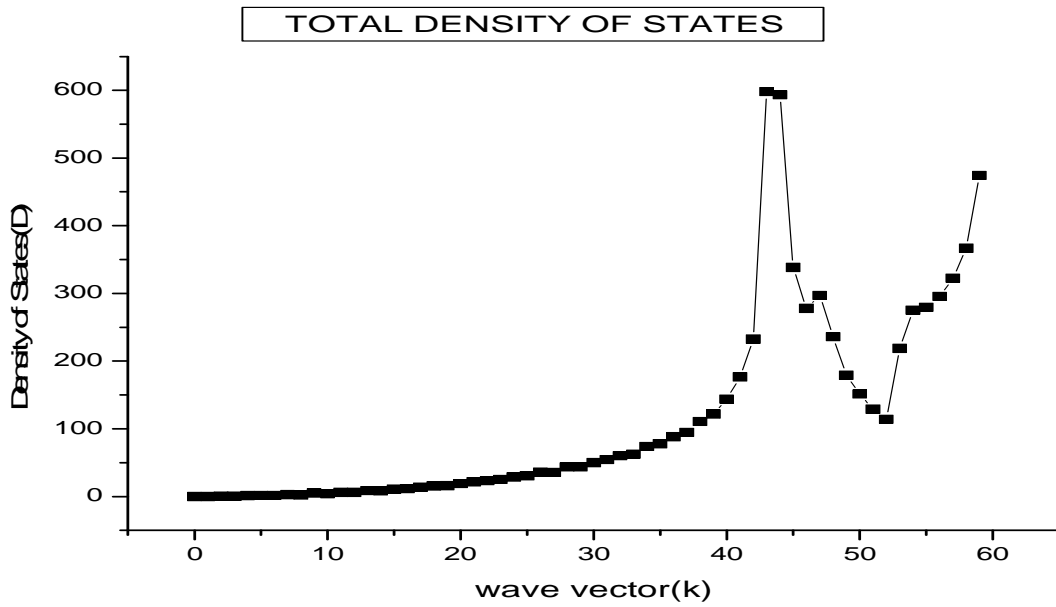


Figure 5. The total density of states of CsCl lattice with $R = 0.8$.

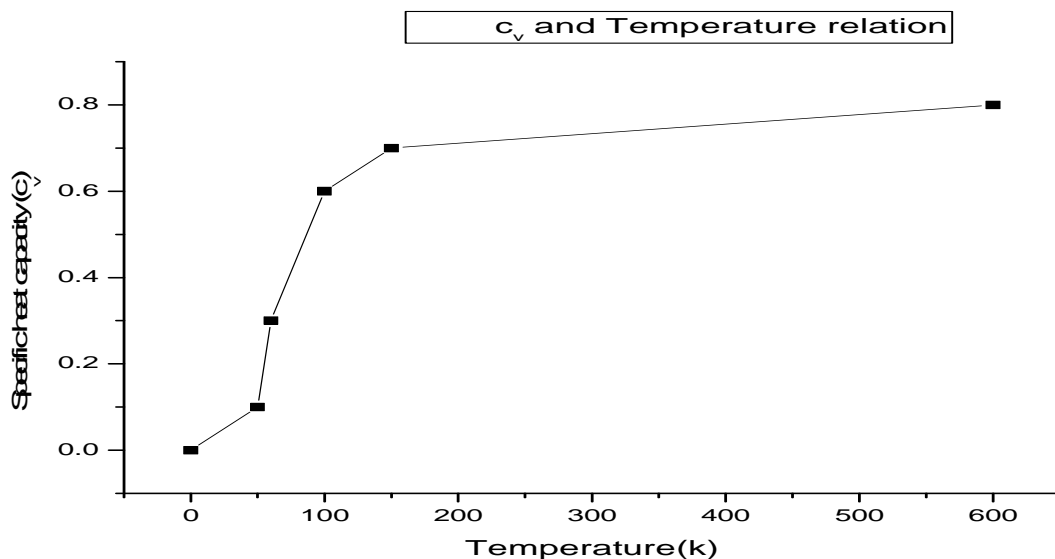


Figure 6. Relation between specific heat capacity c_v and Temperature

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