International Journal of Emerging Technology and Advanced Engineering
Website: www.ijetae.com (ISSN 2250-2459, ISO 9001:2008 Certified Journal, Volume 7, Issue 7, July 2017)

Theoretical Study of Dielectric Behaviour of CsH_2PO_4 Crystal

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***Abstract-* Modifying two sub lattice pseudospin lattice mode model with adding third and fourth-order interaction terms and extra spin lattice interactions terms and using double time thermal Green's funtion method, expressions for ferroelectric mode frequency, dielectric constant and loss tangent have been derived for CsH_2PO_4 crystal. Theoretical result have been compared with experimental results of Blinc et al**

***Keywords-* Anharmonic interactions , dielectric constant, antiferroelectric, Green's function**

I. INTRODUCTION

Ferroelectric materials is one of the important materials, that have vast application in different field. These materials exist spontaneous polarization which can be reversed by application of external field. Due to these properties they find potential application in fabrication of capacitors, transducer, memory devices, detectors etc. CsH_2PO_4 is one of ferroelectric materials which belong to KDP type. It undergoes ferroelectric phase transition at T_c=154K. The ferroelectric low temperature phase with completely ordered bond transform into paraelectric phase (space group P_{21}/m) with partially disordered hydrogen bonds.

At room temperature and ambient pressure, CsH_2PO_4crystal belong to monoclinic P_{21}/m space symmetry groups. Crystal structure of CsH_2PO_4 [1] is shown in fig.-1. The crystal structure contain a layer of hydrogen bonded phosphate groups. CsH_2PO_4 contain two different type of hydrogen bond .PO_4 are connected by short(2.48Å) O_3-H_2—O_4 bond into chain running along two fold b axis. PO_4 group are linked into layer by the longer (2.54Å) O_1—H_1---O_2 hydrogen bonds. H_2 atom are disorder above the ferroelectric phase transition point and ordered into one of two possible off centre sites below T_c into ferroelectric phase. The H_1 atoms are ordered on the hydrogen bond at all temperature.

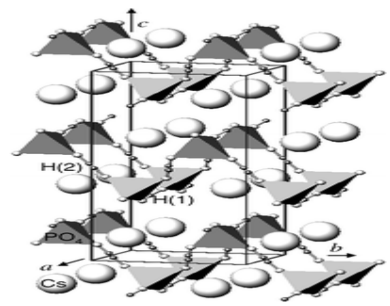

Fig. 1- Crystal Structure of CsH_2PO_4 [1]

Theoretical study on CsH_2PO_4 crystal were initiated by Ganguli et al.[2]. Considered Pseudospin model and obtain the dielectric constant, ferroelectric mode frequency and T_c etc by using Green's function method . Blinc[3] first suggest the isotopic effect in this crystal.

Many experimentalist have done extensive experimental study on this crystal. Nakamoto et al[4] have carried out dielectric constant measurement of CsH_2PO_4 at high pressure. Brononska[5] has made X-ray diffraction study on this crystal. Ahn et al[6] have grown CsH_2PO_4 crystal and made characterization study on this crystal. Kanda et al[7] have made study of measurement of specific heat using an AC calorimeter. Shchur[8] have made lattice dynamics simulation study on CsH_2PO_4 crystal. Raman scattering study is done by Magane Aoki et al[9]. Wada et al[10] have studied high temperature properties of CsH_2PO_4 single crystal by polarizing microscopic observation and thermal expension measurement. Guoliang Zhang et al[11] study the thermal behavior of this crystal and revealed that thermal behavior of this crystal is significantly influenced by Cs/P molar ratio and Cs source. Luspin et al[12] investigate the elastic properties of CsH_2PO_4 crystal by Brillouin spectroscopy throughout a temperature range which includes the supersonic transition at T_s= 233^0C and observed the discontinuities in elastic constants.

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In this paper we investigate theoretically the ferroelectric properties of CsH_2PO_4 crystal. With the help of double time Green's Function method and considering the third and fourth order anharmonic and extra spin lattice interaction terms, we find theoretically the expression for soft mode frequency, dielectric constant and loss tangent. However earlier researchers have note considered the third order anharmonic interaction terms and extra lattice interaction terms. As they decoupled the correlation at early stage so they could note produce better results. By fitting the model value of this crystal in the expression obtained, temperature dependence of soft mode frequency, tangent loss and dielectric constant is calculated for this crystal. Theoretically variation of these calculated value are compared with experimental data given by Blinc et al. Our result show good agreement with experimental data.

II. Model

For crystal CsH_2PO_4, the modified two sub-lattice pseudospin lattice coupled mode model is expressed as

$$H_1 = -2\Omega\sum_i (S_{1i}^x + S_{2i}^x) - \sum_{ij}[J_{ij}(S_{1i}^z S_{1j}^z + S_{2i}^z S_{2j}^z) + K_{ij} S_{1i}^z S_{2j}^z] - \sum_k V_{ik}(S_{1i}^z A_k + S_{2j}^z A_k^+) + \tfrac{1}{4}\sum_k \omega_k (A_k^+ A_k + B_k^+ B_k) \qquad (1)$$

where s_α^m(m=x,y,z) is m^{th} companent of Pseudospin variable S, Ω is proton tunneling frequency, J_{ij} and K_{ij} are respectively coupling constant of coupling within same lattice and different lattice.V_{ik} is spin lattice interaction constant, ω_k is phonon frequency, A_k and B_k are operator corresponding to position and momenta.

We shall add third and fourth order phonon anharmonic interactions terms

$$H_2 = \sum_{k_1k_2k_3} V^{(3)}(k_1,k_2,k_3) A_{k_1} A_{k_2} A_{k_3} + \sum_{k_1k_2k_3k_4} V^{(4)}(k_1,k_2,k_3,k_4) A_{k_1} A_{k_2} A_{k_3} A_{k_4}, \qquad (2)$$

Where $V^3(k_1 k_2 k_3)$ and $V^4(k_1,k_2,k_3,k_4)$ are third and fourth order atomic force constant.

We add

$$H_3 = -2\mu E\sum_i (S_{1i}^z + S_{2i}^z) - \sum_{ik} V_{ik}(S_{1i}^x A_k + S_{2i}^X A_k^+), \qquad (3)$$

Where μ is dipole moment of O-H—O bond and E is external electric field which we take Zero in numerical calculation. First term in above equation describe the effect of electric field on crystal and second terms describes the modulation of the distance between the two equilibrium sites in the O-H—O bonds

So total Hamiltonian become

$H = H_1 + H_2 + H_3$

III. Green's Function

Acorrding to Zubarev[13] for any pair of operators the temperature dependent double time Green function is defined by

$$G_{ij}(t-t') = \langle\langle S_{i1}^z(t); S_{j1}^z(t')\rangle\rangle = -i\theta(t-t')\langle[S_{i1}^z(t); S_{j1}^z(t')]\rangle \qquad (4)$$

In Eq (S_{i1}^z) is spin variable, θ is step function, θ=0 for t< t' and θ=1 for t> t'. Differentiating Green's function Eq. (4) two times with respect to times t and t' respectively, Fourier transforming and writing in Dyson's equation form we obtain

$$G_{ij}(\omega) = G_{IJ}^0(\omega) + G_{ij}^0(\omega)\tilde{P}(\omega)G_{ij}^0(\omega) \qquad (5)$$

$$G_{ij}^0(\omega) = \frac{\Omega\langle S_{1i}^x\rangle\delta_{ij}}{\pi[\omega^2 - 4\Omega^2]} \qquad (6)$$

$$\tilde{P}(\omega) = \pi i\frac{\langle[F_{1i}; S_{1j}^y]\rangle}{\Omega\langle S_{1i}^x\rangle^2} + \frac{\pi^2}{\Omega^2\langle S_{1i}^x\rangle^2}\langle\langle F_{1i}; F_{1j}^{'}\rangle\rangle \qquad (7)$$

Where

$$F(t) = 2\Omega V_{ik} A_k S_{1i}^z \delta_{ij} - 2\Omega J_{ij}(S_{1i}^z S_{1i}^x \delta_{ij} + S_{1i}^x S_{1j}^z) - V_{ik} A_k J_{ij}(S_{1i}^z S_{1i}^x \delta_{ij} + S_{1i}^x S_{1i}^z) - 2\Omega K_{ij} S_{1i}^x S_{2j}^z - V_{ik} A_k K_{ij} S_{1i}^x S_{2j}^z - 2\Omega V_{ik} A_k S_{1i}^x - V_{ik}^2 A_k^2 (S_{1i}^x - S_{1i}^z) - 4\Omega\mu E S_{i1}^x - 2\mu E V_{ik} A_k S_{i1}^x + 2\Omega V_{ik} A_k S_{i1}^x V_{ik} A_k S_{i1}^z, \qquad (8)$$

Second term of $\tilde{P}(\omega)$ of Eq.(7) contains higher order Green functions $\langle\langle F_{1i}; F_{1j}^{'}\rangle\rangle$, which are like <<ab,cd>>, <<abc,def>>. These are decoupled into simpler ones and then solved. In this way $\tilde{P}(\omega)$ is evaluated. Green's function finally become as

$$G_{ij}(\omega) = \frac{\Omega\langle S_{1i}^x\rangle\delta_{ij}}{\pi[\omega^2 - \hat{\Omega}^2 - 2i\Omega\Gamma(\omega)]}. \qquad (9)$$

Where

$$\tilde{\Omega}^2 = a^2 + b^2 - bc \qquad (10)$$

$$a = 2J_0\langle S_1^z\rangle + K_0\langle S_2^z\rangle + 2\mu E \qquad (11)$$
$$b = 2\Omega$$

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$$\hat{\Omega}^2 = \frac{1}{2}\left[\left(\tilde{\omega}_k^2+\tilde{\tilde{\Omega}}^2\right) \pm \left\{\left(\tilde{\omega}_k^2-\tilde{\tilde{\Omega}}\right)^2 + 4\left(\begin{array}{l} 4V_{ik}^2\Omega N_k\langle S_{1i}^x\rangle\omega_k + 4\Omega V_{ik}^2\langle S_{1i}^z\rangle a\omega_k + 2V_{ik}^2J_{ij}^2 a\langle S_{1i}^z\rangle^2\langle S_{1i}^x\rangle\omega_k + \frac{3V_{ik}^4N_k\langle S_{1i}^x\rangle}{\Omega} + \frac{3V_{ik}^4N_k a\langle S_{1i}^x\rangle\omega_k}{\Omega b} \\ + \frac{4\mu^2E^2V_{ik}^2\langle S_{1i}^x\rangle\omega_k}{\Omega} + \frac{V_{ik}^4J_{ij}^2N_k a^2\langle S_{1i}^z\rangle}{b^2} + \frac{V_{ik}^2K_{ij}^2 a\langle S_{1i}^z\rangle\langle S_{1i}^x\rangle^2\omega_k}{b\Omega}\end{array}\right)\right\}\right]^{\frac{1}{2}}. \tag{12}$$

$$c = 2J_0\langle S_1^z\rangle + K_0\langle S_2^x\rangle \tag{13}$$

utting the value of $\tilde{P}(\omega)$ into eq(8) and resolving into real $\Delta(\omega)$ and imaginary $\Gamma(\omega)$ parts we obtain

$$\begin{aligned}\Delta(\omega) = {} & \frac{a^4}{\omega^2-\tilde{\Omega}^2} + \frac{b^2c^2}{\omega^2-\tilde{\Omega}^2} + \frac{8\Omega^2V_{ik}^2N_k}{\omega^2-\tilde{\Omega}^2} + \frac{V_{ik}^2J_{ij}^2N_k\langle S_{i1}^2\rangle}{\omega^2-\tilde{\Omega}^2} + \frac{V_{ik}^2K_{ij}^2N_k\langle S_{i1}^2\rangle}{\omega^2-\tilde{\Omega}^2} + \frac{V_{ik}^2K_{ij}^2N_k\langle S_{i1}^2\rangle a^2}{b^2\left(\omega^2-\tilde{\Omega}^2\right)} + \frac{V_{ik}^4N_k a^2}{b^2\left(\omega^2-\tilde{\Omega}^2\right)} + \frac{\Omega V_{ik}^2N_k a\langle S_{i1}^z\rangle}{\omega^2-\tilde{\Omega}^2} + \frac{V_{ik}^4N_k\langle S_{i1}^x\rangle}{\omega^2-\tilde{\Omega}^2} + \frac{4\mu^2E^2V_{ik}^2N_k a^2}{b^2\left(\omega^2-\tilde{\Omega}^2\right)} \\ & + \frac{3V_{ik}^4N_k\langle S_{i1}^x\rangle}{b^2\left(\omega^2-\tilde{\Omega}^2\right)} + \frac{V_{ik}^2J_{ij}^2N_k a^2\langle S_{i1}^z\rangle^2}{b^2\left(\omega^2-\tilde{\Omega}^2\right)} + \frac{4\Omega V_{ik}^2N_k a\langle S_{1i}^z\rangle\omega_k\left(\omega^2-\tilde{\tilde{\omega}}_k^2\right)}{\left[\left(\omega^2-\tilde{\tilde{\omega}}_k^2\right)^2+4\omega_k^2\Gamma_k^2(\omega)\right]} + \frac{3V_{ik}^4N_k\langle S_{1i}^x\rangle\omega_k\left(\omega^2-\tilde{\tilde{\omega}}_k^2\right)}{\left[\left(\omega^2-\tilde{\tilde{\omega}}_k^2\right)^2+4\omega_k^2\Gamma_k^2(\omega)\right]b\Omega} + \frac{4\mu^2E^2V_{ik}^2\langle S_{1i}^x\rangle\omega_k\left(\omega^2-\tilde{\tilde{\omega}}_k^2\right)}{\left[\left(\omega^2-\tilde{\tilde{\omega}}_k^2\right)^2+4\omega_k^2\Gamma_k^2(\omega)\right]\Omega} \\ & + \frac{4\Omega V_{ik}^2N_k\langle S_{1i}^x\rangle\omega_k\left(\omega^2-\tilde{\tilde{\omega}}_k^2\right)}{\left[\left(\omega^2-\tilde{\tilde{\omega}}_k^2\right)^2+4\omega_k^2\Gamma_k^2(\omega)\right]} + \frac{2J_{ik}^2 a\langle S_{1i}^z\rangle^2\langle S_{i1}^x\rangle\omega_k\left(\omega^2-\tilde{\tilde{\omega}}_k^2\right)}{\left[\left(\omega^2-\tilde{\tilde{\omega}}_k^2\right)^2+4\omega_k^2\Gamma_k^2(\omega)\right]} + \frac{V_{ik}^2K_{ij}^2N_k a\langle S_{1i}^z\rangle\langle S_{i1}^x\rangle\omega_k\left(\omega^2-\tilde{\tilde{\omega}}_k^2\right)}{\left[\left(\omega^2-\tilde{\tilde{\omega}}_k^2\right)^2+4\omega_k^2\Gamma_k^2(\omega)\right]b\Omega}\end{aligned} \tag{14}$$

$$\begin{aligned}\Gamma(\omega) = {} & \frac{a^4}{2\tilde{\Omega}^2}\left[\delta\left(\omega-\tilde{\Omega}\right)-\delta\left(\omega+\tilde{\Omega}\right)\right] + \frac{b^2c^2}{2\tilde{\Omega}}\left[\delta\left(\omega-\tilde{\Omega}\right)-\delta\left(\omega+\tilde{\Omega}\right)\right] + \frac{8\Omega^2V_{ik}^2N_k}{2\tilde{\Omega}}\left[\delta\left(\omega-\tilde{\Omega}\right)-\delta\left(\omega+\tilde{\Omega}\right)\right] \\ & + \frac{6V_{ik}^4N_k\langle S_{1i}^x\rangle\omega_k\Gamma_k(\omega)}{2\Omega\left[\left(\omega^2-\tilde{\tilde{\omega}}_k^2\right)^2+4\omega_k^2\Gamma_k^2(\omega)\right]} + \frac{6V_{ik}^4N_k\langle S_{i1}^x\rangle\omega_k^2\Gamma_k(\omega)}{2b\Omega\left[\left(\omega^2-\tilde{\tilde{\omega}}_k^2\right)^2+4\omega_k^2\Gamma_k^2(\omega)\right]} + \frac{8\mu^2E^2V_{ik}^2\langle S_{1i}^x\rangle\omega_k^2\Gamma_k(\omega)}{2\Omega\left[\left(\omega^2-\tilde{\tilde{\omega}}_k^2\right)^2+4\omega_k^2\Gamma_k^2(\omega)\right]} \\ & + \frac{8\Omega V_{ik}^2\omega_k^2\Gamma_k(\omega)\left(\langle S_{1i}^x\rangle+\langle S_{1i}^z\rangle\right)}{2\left[\left(\omega^2-\tilde{\tilde{\omega}}_k^2\right)^2+4\omega_k^2\Gamma_k^2(\omega)\right]} + \frac{4V_{ik}^2J_{ij}^2a\langle S_{1i}^z\rangle^2\langle S_{1j}^x\rangle\omega_k^2\Gamma_k(\omega)}{2\left[\left(\omega^2-\tilde{\tilde{\omega}}_k^2\right)^2+4\omega_k^2\Gamma_k^2(\omega)\right]} + \frac{2V_{ik}^2K_{ij}^2\langle S_{1i}^z\rangle\langle S_{1j}^x\rangle^2\omega_k^2\Gamma_k(\omega)}{2b\left[\left(\omega^2-\tilde{\tilde{\omega}}_k^2\right)^2+4\omega_k^2\Gamma_k^2(\omega)\right]} + \frac{2V_{ik}^2J_{ij}^2N_k\langle S_{1i}^z\rangle^2\omega_k\Gamma_k(\omega)}{2b^2\left[\left(\omega^2-\tilde{\tilde{\omega}}_k^2\right)^2+4\omega_k^2\Gamma_k^2(\omega)\right]}\end{aligned} \tag{15}$$

Where $\Delta_k(\omega)$ is phonon shift ,$\Gamma_k(\omega)$ is phonon width due to third and fourth order phonon anharmonic interaction terms and $\tilde{\tilde{\omega}}_k$ are modified phonon frequency They are obtained by solving phonon Green's function $\langle\langle A_k; A_{k'}^+\rangle\rangle$ by using phonon Hamiltonian only.

$$\langle\langle A_k; A_{k'}^+\rangle\rangle = \frac{\omega_k\delta_{k,k'}}{\left(\omega^2-\tilde{\tilde{\omega}}_k\right)-2i\omega_k\Gamma_k(\omega)} \tag{16}$$

And

$$\tilde{\tilde{\omega}}_k = \tilde{\omega}_k + 2\omega_k\Delta_k(\omega) \tag{17}$$

According to Cochran[14], ferroelectric transition in ferroelectric and antiferroelectric crystals results from freezing of soft mode frequency at transition temperature.

Substituting the value of $\Delta(\omega)$ in Eq. (6) and simplifying we obtain the expression for soft mode frequency as

$$\hat{\Omega}^2 = \frac{1}{2}\left[\left(\tilde{\omega}_k^2+\tilde{\tilde{\Omega}}^2\right) \pm \left\{\left(\tilde{\omega}_k^2-\tilde{\tilde{\Omega}}\right)^2 + 4\left(\begin{array}{l} 4V_{ik}^2\Omega N_k\langle S_{1i}^x\rangle\omega_k + 4\Omega V_{ik}^2\langle S_{1i}^z\rangle a\omega_k + 2V_{ik}^2J_{ij}^2 a\langle S_{1i}^z\rangle^2\langle S_{1i}^x\rangle\omega_k + \frac{3V_{ik}^4N_k\langle S_{1i}^x\rangle}{\Omega} + \frac{3V_{ik}^4N_k a\langle S_{1i}^x\rangle\omega_k}{\Omega b} \\ + \frac{4\mu^2E^2V_{ik}^2\langle S_{1i}^x\rangle\omega_k}{\Omega} + \frac{V_{ik}^4J_{ij}^2N_k a^2\langle S_{1i}^z\rangle}{b^2} + \frac{V_{ik}^2K_{ij}^2 a\langle S_{1i}^z\rangle\langle S_{1i}^x\rangle^2\omega_k}{b\Omega}\end{array}\right)\right\}\right]^{\frac{1}{2}}. \tag{18}$$

The frequency $\hat{\Omega}$ is ferroelectric Soft mode frequency which decreases with temperature from below T_c and is so responsible for ferroelectric phase transition. We have neglected electric field term (E=0)

IV. DIELECTRIC CONSTANT AND LOSS TANGENT

The response of ferroelectric or dielectric crystal to electric field is expressed by susceptibility By using Zubarev's formalism this is expressed as

$$\chi = -\lim_{x\to 0} 2\pi N\mu^2 G_{ij}(\omega + ix) \tag{19}$$

Where N is no of dipoles having dipole moment μ in the unit volume. The susceptibility χ is related to dielectric constant as $\varepsilon = 1 + 4\pi\chi$. Using Eqs (9), (19) and relation $\varepsilon = 1 + 4\pi\chi$, the expression for dielectric constant is expressed as

$$\varepsilon = -\frac{8\pi N\mu^2\Omega\langle S_{1i}^x\rangle\delta_{ij}}{\left[\left(\omega^2-\hat{\Omega}^2\right)-2\Omega i\Gamma(\omega)\right]}., \tag{20}$$

Where $\varepsilon(\omega)$>>1.

Eq. (20) shows that dielectric constant depends on tunneling frequency (Ω).In dielectric or ferroelectric crystals some power is dissipated in the form of heat which is called the loss tangent. It is obtained as ratio of imaginary part and real part of dielectric constant. We therefore obtain

$$\tan\delta = \frac{\varepsilon''}{\varepsilon'}, \tag{21}$$

Using Eqs (20) and (21) we obtain

$$\tan\delta = -\frac{2\Omega\Gamma(\omega)}{\left(\omega^2-\hat{\Omega}^2\right)} \tag{22}$$

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From Eqs. (20) and (22) we observe that dielectric constant and loss tangent largely depend on modified ferroelectric mode frequency. Which in turn depends upon tunneling frequency, anharmonic interactions terms, extra spin lattice interactions terms and external electric field.

V. Numerical Calculation And Results

By using model values of physical quantities for CsH_2PO_4 crystal given in table-1, temperature dependence of antiferroelectric mode frequency, dielectric constant and loss tangent have been calculated and plotted in figs 2-4. Calculated values have been compared with experimental values of Blinc et al[3]

Table-1
Model Values of physical quantities for CDP

T_c (K)	Ω (cm^{-1})	J (cm^{-1})	J^* (cm^{-1})	V_{ik} (cm^{-1})	ω_k (cm^{-1})	Nμ x10^{18} (esu)
154	80	350	450	1.72	130	2.3

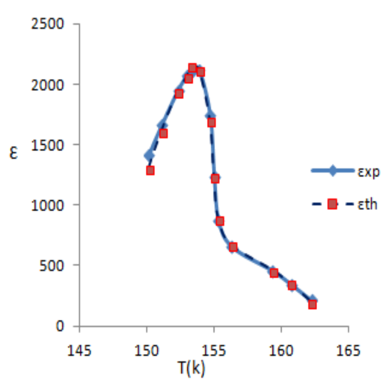

Fig 2-Temperature dependence of dielectric constant (Ɛ) in CDP crystal

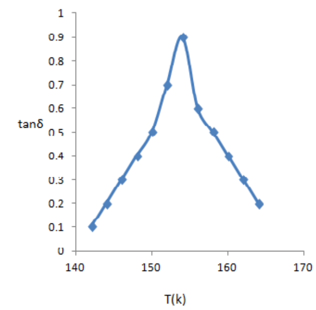

Fig 3- temperature dependence of soft mode frequency for CDP crystal(present calculation, correlated values with experimental values of Blinc et al[3])

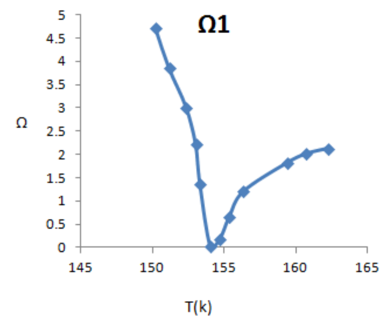

Fig 4- temperature dependence of tangent loss for CDP crystal(present calculation, correlated values with experimental values of Blinc et al[3])

The theoretical variation of dielectric constant, loss tangent and soft mode frequency with temperature are compared with values obtained by correlating the experimental result of Blinc et al[3]. The calculated dielectric constant , loss tangent versus temperature curves agree with experimental data of Blinc et al. Earlier authors decoupled the correlations functions at early stage hence some interactions disappeared from their results. We width and shift are contribution to present work.

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The soft mode frequency decreases from below Tc becoming very small near transition temperature and then increases. Dielectric constant first increases with temperature become maximum near Tc and then decreases. Similar variation are shown by tangent loss.

VI. CONCLUSION

By fitting the model value of physical parameter (Table-1) in the derived expression we calculate theoretically the dielectric constant, loss tangent and soft mode frequency for CsH_2PO_4 crystal. In present study it is shown that Pseudospin-lattice coupled mode model along with third and fourth-order phonon anharmonic interaction and extra spin lattice interaction terms explain well the temperature dependence of soft mode frequency, dielectric constant and loss tangent in CsH_2PO_4 crystals. By considering third and fourth order anharmonic interaction and extra spin lattice interaction terms our result show good agreement with experimental result of Blinc et al[3].

Acknowledgement

Authors are grateful to Eminent Physicist Prof B S Semwal (Former HOD) for his valuable suggestion and Prof. S C Bhatt (HOD), Prof N S Negi (Himanchal Pradesh University, Shimla), Prof Vinay Gupta (Delhi University Delhi), Prof K K Verma (A U Faizabad) and Dr. Manish Uniyal for their kind encouragements

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