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T. C. Upadhyay[1] and Mayank Joshi[2*]

[1&2]Department of Physics, H.N.B. Garhwal University (U.K.), INDIA

** Corresponding Author Email: mayankphysics@gamil.com*

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[1&2]Department of Physics, H.N.B. Garhwal University (U.K.), INDIA

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ABSTRACT

The third and fourth order phonon anharmonic interactions external electric field terms are added in the two -sub lattice pseudospin model for $PbHPO_4$ crystal. By using double time thermal Green's function method modified m odel, theoretical expressions for soft mode frequency, dielectric constant, shift, width tangent loss and polarizatio n are evaluated for $PbHPO_4$ crystal. Temperature and field variations of soft mode frequency, dielectric constant and loss tangent are calculated numerically. Present theoretical results agree with experimental result of Smutney and Fousek for dielectric constant of $PbHPO_4$.

***Keywords**: LHP influence, Ferroelectrics, solar-energy etc*

INTRODUCTION

Due to their promising application in the field of electronics and technology ferroelectric crystals are continuously being attracted to both physicists and material scienticts. Memory devices, infrared and pyroelectric detectors, transducers, display devices piezoelectric devices are some common uses of these materials [1].

Lead hydrogen phosphate ($PbHPO_4$) crystal and its isomorphs ($PbAsPO_4$, $CaHPO_4$, $BaHPO_4$, $CaHPO_4$ etc.) from an interesting group of quasi-one dimensional hydrogen bonded ferroelectric crystals. In $PbHPO_4$ the direction of spontaneous polarization is almost parallel to the direction of the H-bonded O-H...O projecting on the (010) plane unlike in KH_2PO_4**.** The PO_4 groups are bound to one another by the O-H...O bonds in the form of a one dimensional chain along c-axis[2] Raman spectroscopic studies showed the value of tunneling integral for $PbHPO_4$ crystal very small although very large changes of curie temperature and Curie-Weiss constant occur on deuteration [3]. According to Cochran [3] the frequency of some of the normal mode of vibration of crystal called soft mode becomes zero at the transition temperature. It is this soft mode that largely determines the dielectric and scattering properties some very interesting results are observed.

Ferroelectrics may have a bright future for solar-energy generation, following the report that the domain walls of such materials can be engineered to exhibit a photovoltaic effect with an impressively high voltage output.

Ferroelectric photovoltaic materials are recently generating much interest. Although PV effects in ferroelectrics have been known for 50 years, they have received little attention due to their initially reported low power conversion efficiency. The recent interest in PV ferroelectrics is triggered by reports that the low conversion efficiencies can be overcome by large (above-band gap) photo voltages in complex oxides, the possibility of tip-enhanced PV effects at the nanoscale or the fundamental role of domain walls which can be tuned by external fields. All this indicates that ferroelectric photovoltaic materials potentially have a bright future for solar-energy generation.

Most of the international research effort on Ferroelectric photovoltaic materials has been conducted outside Europe, despite a longstanding expertise in both ferroelectric materials and photovoltaic applications. The aim of this European workshop is to discuss both fundamental ferroelectrics-related issues and the potential of ferroelectric photovoltaic for applications.

Model Hamiltonian

Mitsui[17] and Blinc and Zeks [18] proposed a two-sublattice pseudospin model, which was applied to the case of $PbHPO_4$ and isomorphous crystals is expressed asFor $PbHPO_4$ type crystals we have, extended two-sublattice pseudospin-lattice coupled mode model[4] by adding third and fourth order phonon anharmonic interaction terms [14] as well as external electric field term which is expressed as

$$H=-2\Omega\sum_i\left(S_{1i}^x+S_{2i}^x\right)-\sum_{ij} J_{ij}\left[\left(S_{1i}^z S_{2i}^z\right)+\left(S_{2i}^z S_{2i}^z\right)\right]$$
$$-\sum_{ij} K_{ij}\left(S_{1i}^z S_{2i}^z\right)-2\mu E\sum_i(S_{1i}^z+S_{2i}^z)$$
$$+\frac{1}{4}\sum_k\omega_k\left(A_kA_k^+ + B_kB_k^+\right) \qquad \ldots(1)$$

In Eq.(1) above Ω is proton tunneling frequency between O-H…O double well potential,
Jij is exchange interaction between neighboring lattice dipoles, and k_{ij} that in same lattice μ is dipole moment of O-H…O bond, E is external electric field, is phonon frequency, A_k and B_k are position and momentum operators and S^x and S^z are components of spin variable.

Chaudhari et al[13] have modified above model by following Kobayashi [19] by adding pseudospin-lattice interaction terms

$$H_{s-p}=-\sum_{ik} V_{ik}S_{1i}^z A_k-\sum_{ik} V_{ik}S_{2i}^z A_k^+ \qquad \ldots(2)$$

In Eqs(2) above V_{ik} is spin lattice interaction constant. We add the third and the fourth-order phonon anharmonic interaction terms[14] as

$$H_{anh}=\sum_{k_1k_2k_3} V^{(3)}\left(k_1,k_2,k_3\right)A_{k_1}A_{k_2}A_{k_3}$$
$$+\sum_{k_1k_2k_3k_4} V^{(4)}\left(k_1,k_2,k_3,k_4\right)A_{k_1}A_{k_2}A_{k_3}A_{k_4}, \qquad \ldots(3)$$

$V^{(3)}$ (k_1,k_2,k_3) and $V^{(4)}$ (k_1,k_2,k_3,k_4) are third and fourth-order atomic forces constants given by Born and Huang[20].

Green's functions, Width and Shift

For the evaluation of expressions, soft mode frequency, dielectric susceptibility, dielectric constant and loss tangent we consider the evaluation of Green's function[15]

We consider the Green's function

$$G_{ij}(t-t')=\left\langle\left\langle S_{1i}^z(t);S_{1j}^z(t')\right\rangle\right\rangle$$
$$=-i\theta(t-t')\left\langle\left[S_{1i}^z(t);S_{1j}^z(t')\right]\right\rangle, \qquad \ldots(4)$$

in which θ (t- t') is unity for t<t' and zero otherwise. The angular bracket is ensemble average.

The Green's function (GF) is differentiated twice first with respect to time t and then with respect to t'

Fourier transforming the Green's function and putting in the or of Dyson's equation,

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

where $G_{ij}^0(\omega)$ is unperturbed Green's function given as

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

and $\tilde{P}(\omega)$ is polarization operator given by

$$\tilde{P}(\omega)=\pi f+\pi^2\left\langle\left\langle F_{1i}(t);F_{j1}(t')\right\rangle\right\rangle, \qquad \ldots(7)$$

where $f=\dfrac{i\left\langle\left[F,S_{1j}{}^y\right]\right\rangle}{\Omega\left\langle S^x{}_{1i}\right\rangle}$ and ...(8)

$$F_{i1}(t)=2\Omega\left(S_{1i}{}^xS_{1j}{}^z+S_{1j}{}^zS_{ij}{}^x\right)$$
$$-2\Omega K_{1j}\left(S^X{}_{1J}S^Z{}_{1J}\right)+2\Omega V_{ik}S_{1i}{}^xA_k+2\Omega V_{ik}S_{1i}{}^xA^+{}_k \qquad \ldots(9)$$

The Green's function $G(\omega)$ is then obtained as

$$G(\omega)=G^0(\omega)\left[1-G^0(\omega)\tilde{P}(\omega)\right]^{-1} \qquad \ldots(10)$$

which gives the values of Green's function (4) as

$$G_{ij}^0(\omega)=\frac{\Omega\left\langle S_{1i}^x\right\rangle\delta_{ij}}{\pi\left(\omega^2-\tilde{\Omega}^2-2i\Omega\Gamma(\omega)\right)}, \qquad \ldots(11)$$

where $\tilde{\Omega}^2=4\Omega^2+\left\langle S^x\right\rangle^{-1}f$ … ...(12)

$$\tilde{\tilde{P}}(\omega)=\frac{\pi}{\left\langle S_1^x\right\rangle}\left\langle\left\langle F_i(t);F_j'(t)\right\rangle\right\rangle,\ldots \qquad \ldots(13)$$

where <<F_i(t);F_j(t')>> are higher order Green's functions. They are evaluated by decoupling them using decoupling scheme <abcd>=<ab><cd>+<ac><bd>+<ad><bc>

$\tilde{\tilde{P}}(\omega)$ after evaluation is resolved into its real and imaginary parts using formula

$$\lim_{m\to 0}\frac{1}{x+im}=\left(\frac{1}{x}\right)\pm i\pi\delta(x)$$

…(14)

The real part is called shift $\Delta(\omega)$ and the imaginary part is called half width $\Gamma(\omega)$

We therefore obtain shift and width as

$$\Delta(\omega)=\Delta_1(\omega)+\Delta_2(\omega)+\Delta_3(\omega)+\Delta_4(\omega)\quad ,$$

…(15)

where

$$\Delta_1(\omega)=\frac{a^4}{2\Omega\left(\omega^2-\tilde{\Omega}^2\right)}\ ,\ \ldots \qquad \ldots(16)$$

$$\Delta_2(\omega)=\frac{V_{ik}^2 N_k a^2}{2\Omega\left(\omega^2-\tilde{\Omega}^2\right)}\ldots \qquad \ldots(17)$$

$$\Delta_3(\omega)=\frac{4\mu^2E^2a^2}{2\Omega\left(\omega^2-\tilde{\Omega}^2\right)}\ldots \qquad \ldots(18)$$

$$\Delta_4(\omega)=\frac{2V_{ik}^2\langle S_{1i}^x\rangle\omega_k\delta_{kk^i}\left(\omega^2-\tilde{\tilde{\omega}}_k^2\right)}{\left[\left(\omega^2-\tilde{\tilde{\omega}}_k^2\right)^2+4\omega_k^2\Gamma_k^2(\omega)\right]}\ldots \qquad \ldots(19)$$

and width $\Gamma(\omega)=\Gamma_1(\omega)+\Gamma_2(\omega)+\Gamma_3(\omega)+\Gamma_4(\omega)$,

…(20)

$$\Gamma_1(\omega)=\frac{\pi a^4}{4\Omega\tilde{\Omega}}\left[\delta\left(\omega-\tilde{\Omega}\right)-\delta\left(\omega+\tilde{\Omega}\right)\right]$$

…(21)

$$\Gamma_2(\omega)=\frac{V_{ik}^2 N_k a^2}{4\Omega\tilde{\Omega}}\left[\delta\left(\omega-\tilde{\Omega}\right)-\delta\left(\omega+\tilde{\Omega}\right)\right]$$

…(22)

$$\Gamma_3(\omega)=\frac{4V_{ik}^2\langle S_{1i}^x\rangle\omega_k\delta_{k\_k'}\left(\omega^2-\tilde{\tilde{\omega}}_k^2\right)}{\left[\left(\omega^2-\tilde{\tilde{\omega}}_k^2\right)^2+4\omega_k^2\Gamma_k^2(\omega)\right]}$$

…(23)

$$\Gamma_4(\omega)=\frac{2\pi\mu^2E^2a^2}{4\Omega\tilde{\Omega}}\left[\delta\left(\omega-\tilde{\Omega}\right)-\delta\left(\omega+\tilde{\Omega}\right)\right]$$

…(24)

In Eqs.(19) and (23) $\tilde{\tilde{\omega}}_k$ and $\Gamma_k(\omega)$ are phonon frequency and phonon half width which are obtained by solving phonon Green's Function(in a similar way)

$$G_{kk'}(t-t')=\langle\langle A_k(t),A_{k^1}(t')\rangle\rangle$$
$$=-i\theta(t-t')\langle\langle A_k(t),A_{k^1}(t)\rangle\rangle$$

…(25)

which gives

$$G_{ij'}(\omega)=\frac{\omega_k\delta_{kk'}}{\pi\left[\omega^2-\tilde{\tilde{\omega}}^2{}_k-2i\Gamma_k(\omega)\right]}\ ,$$

…(26)

where

$$\tilde{\tilde{\omega}}_k^2=\tilde{\omega}_k^2+2\omega_k\Delta_k(\omega)\ ;\ \tilde{\omega}_k^2=\omega_k+A_k$$

…(27a, b)

Phonon shift

$$\Delta_k(\omega)=\operatorname{Re}P^0(k,\omega)$$
$$=18P\sum k_1k_2\left|V^{(3)}(k_1,k_2,-k)\right|^2$$
$$\frac{\omega_{k1}\omega_{k2}}{\tilde{\omega}_{k1}\tilde{\omega}_{k2}}\left\{\left(n_{k_1}+n_{k_2}\right)\frac{\tilde{\omega}_{k1}+\tilde{\omega}_{k2}}{\omega^2-\left(\omega_{k1}+\omega_{k2}\right)^2}+\left\{\left(n_{k_2}+n_{k_1}\right)\frac{\tilde{\omega}_{k1}+\tilde{\omega}_{k2}}{\omega^2-\left(\omega_{k1}+\omega_{k2}\right)^2}\right.\right.$$

$$+48P\sum\left|V^{(4)}(k_1,k_2,k_3,-k\right|^2\frac{\omega_{k1}\omega_{k2}\omega_{k3}}{\tilde{\omega}_{k1}\tilde{\omega}_{k2}\tilde{\omega}_{k3}}$$
$$\left\{\left(1+n_{k1}n_{k2}+n_{k2}n_{k3}+n_{k3}n_{k1}\right)\frac{\tilde{\omega}_{k1}+\tilde{\omega}_{k2}+\tilde{\omega}_{k3}}{\omega^2-\left(\tilde{\omega}_{k1}+\tilde{\omega}_{k2}+\tilde{\omega}_{k3}\right)^2}\right.$$

$$+3\left(1-n_{k2}n_{k1}+n_{k2}n_{k3}-n_{k3}n_{k1}\right)$$
$$\frac{\tilde{\omega}_{k1}+\tilde{\omega}_{k2}+\tilde{\omega}_{k3}}{\omega^2-\left(\tilde{\omega}_{k1}+\tilde{\omega}_{k2}+\tilde{\omega}_{k3}\right)^2}$$

+ highter terms } …(28)

Phonon width

$$\Gamma_k(\omega)=\operatorname{Im}P(k,\omega)$$
$$=9\pi\sum\left|V^{(3)}(k_1,k_2,-k\ )\right|^2\frac{\omega_{k1}\omega_{k2}}{\tilde{\omega}_{k1}\tilde{\omega}_{k2}}$$
$$\left\{\left(n_{k2}+n_{k1}\right)\begin{bmatrix}\left[\ \delta\left(\omega+\tilde{\omega}_{k1}+\tilde{\omega}_{k1}\right)-\delta\left(\omega-\tilde{\omega}_{k1}-\tilde{\omega}_{k1}\right)+\right.\\ \left(n_{k2}-n_{k1}\right)\delta\left(\omega+\tilde{\omega}_{k1}+\tilde{\omega}_{k1}\right)-\delta\left(\omega+\tilde{\omega}_{k1}+\ \ \}\tilde{\omega}_{k1}\right)\end{bmatrix}\right\}$$

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$$+48\pi\sum\left|V^{(3)}(k_1,k_2,k_3,-k_4)\right|^2$$
$$X\{1+n_{k1}n_{k2}+n_{k2}n_{k3}+n_{k3}n_{k4}\}$$
$$X\left[\delta\left(\omega+\tilde{\omega}_{k1}+\tilde{\omega}_{k2}+\tilde{\omega}_{k3}\right)-\left[\delta\left(\omega-\tilde{\omega}_{k1}-\tilde{\omega}_{k2}-\tilde{\omega}_{k3}\right)\right]\right] \quad \ldots(29)$$

In Eq(28) and (29) $n_{ki}=Coth\left(\frac{\tilde{\omega}_{ki}}{k_BT}\right)$ and P stand for principal part.

In Eq.(8)is solved by using means field approximation for co-relation i.e. second term in Eq.(12) is evaluated using mean field approximation, i.e. correlations are finite, i.e.

$$\frac{\left\langle S_{1i}^{z}\right\rangle}{a}=\frac{\left\langle S_{1i}^{x}\right\rangle}{b}=\frac{1}{2\tilde{\Omega}}\tanh\ \beta\frac{\tilde{\Omega}}{2} \quad \ldots(30)$$

which gives

$$\tilde{\Omega}^2=a^2+b^2-bc \quad \ldots(31)$$

where $a=2J_0\left\langle S_1^z\right\rangle+K_0\left\langle S_2^z\right\rangle$, …(32)

$$b=2\Omega; \quad \ldots(33)$$

and $\boldsymbol{c}=2\boldsymbol{J}_0\left\langle \boldsymbol{S}_1^x\right\rangle+\boldsymbol{K}\left\langle \boldsymbol{S}_2^x\right\rangle$ … (34)

Therefore, the Green's function finally takes the from

$$G_{ij}(\omega)=\frac{\Omega\left\langle S_{1i}^{x}\right\rangle\delta_{ij}}{\pi\left(\omega^2-\hat{\Omega}^2-2\Omega i\Gamma(\omega)\right)} \quad \ldots(35)$$

$$\hat{\Omega}^2=\tilde{\Omega}^2+2\Omega\Delta(\omega) \quad \ldots(36)$$

In Eq.(35) and (36) $\Gamma(\omega)$ and $\Delta(\omega)$ are liven by Eq.(20) and (15) respectively. Solving Eq.(36) one gets

$$\hat{\Omega}_{\pm}^2=\frac{1}{2}\left(\tilde{\tilde{\omega}}_k^2+\tilde{\tilde{\Omega}}^2\right)\pm\frac{1}{2}\left[\left(\tilde{\tilde{\omega}}_k^2-\tilde{\tilde{\Omega}}^2\right)^2+8V_{ik}^2\left\langle S_{1i}^{x}\right\rangle\Omega\right]^{½} \quad \ldots(37)$$

The Curie temperature is given by

$$T_c=\frac{\eta}{2k_B\tanh^{-1}\left(\frac{\eta^3}{4\Omega^2J'}\right)} \quad \ldots(38a)$$

Where $\eta^2=(2J-K)^2\sigma^2+4\Omega^2$ …(38b)

$$J^*=(2J+K)+\frac{2V_{ik}^2\tilde{\tilde{\omega}}_k^2}{\left[\tilde{\tilde{\omega}}_k^4+4\omega_k\Gamma_k^2\right]} \quad \ldots(38c)$$

Dielectric constant

The response of a ferroelectric crystal to the external electric field is expressed dielectric susceptibility χ which is related to Green's function as

$$\chi=-\lim_{\in\to 0}2\pi N\mu^2G_{ij}(\omega+i\in) \quad \ldots(39)$$

The dielectric constant $\in$ is related to electrical susceptibility as

$$\in=1+4\pi\chi \quad \ldots(40)$$

By putting value of Green's function from Eq.(39) and (40) we obtain

$$\in(\omega)=\left(-8\pi N\mu^2\right)\left\langle S_1^x\right\rangle\left(\omega^2-\hat{\Omega}^2\right)\left[\left(\omega^2-\hat{\Omega}^2\right)^2+4\Omega^2\Gamma^2\right]^{-1} \quad \ldots(41)$$

The dissipation of power in dielectric material is called tangent loss which expressed as

$$\tan\ \delta=\frac{\in''}{\in'} \quad \ldots(42)$$

Where $\in''$ and $\in'$ are imaginary and real parts of dielectric constant

$$\tan\ \delta=-\frac{2\Omega\Gamma(\omega)}{\left(\omega^2-\hat{\Omega}^2\right)} \quad \ldots(43)$$

Where $\Gamma(\omega)$ and $\hat{\Omega}$ are half width and soft mode frequency given by Esq. (20) and (37) respectively.

Temperature and electric field dependence of soft mode frequency, dielectric constant and Loss tangent

By using model values of various quantities in expression for $\Delta(\omega),\Gamma(\omega),\tilde{\Omega},\hat{\Omega},\in,$ *and* $\tan\partial$ for $PbHPO_4$ crystal from literature their electric field and temperature dependences $\hat{\Omega},\in$ and $\tan\delta$ near transition temperature are calculated, which are shown in figs.1, 2 and 3

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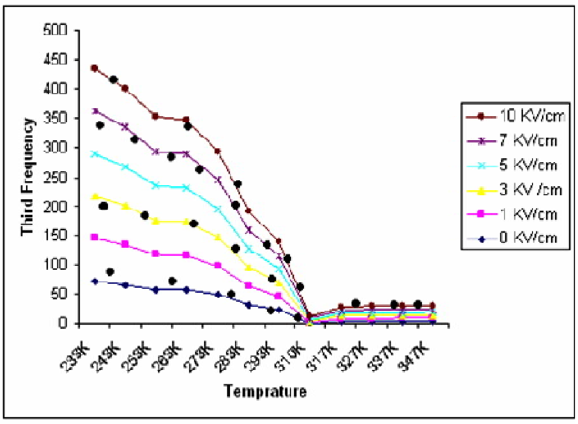
Table-1: Model values of physical parameters for $PbHPO_4$ crystal (Ref.4)

ω_0^2 (cm^{-2})	Ω (cm^{-1})	J (cm^{-1})	K (cm^{-1})	V_{ik} ($cm^{-3/2}$)	T_c(K) (cm^{-1})	C (k)	μ (10^{18}esu)	$\Omega^2 J^*$ (cm^{-3})	Ω^2 (2J+K) (cm^{-1})	ΩV_{ik} ($cm^{-5/2}$)
13.3	2.16	172.37	86.18	30.93	310	2773	0.55	2699	2024	76.75

Table-2: Value of the $2\mu E$ for different E values

E(kv/cm)	1	2	3	4	5	6	7	8	9	10
$2\mu E$	0.0204	0.0409	0.0614	0.0818	0.1023	0.1236	0.1432	0.1637	0.1842	0.2047

Calculated electric field and temperature dependences for $PbHPO_4$ crystal



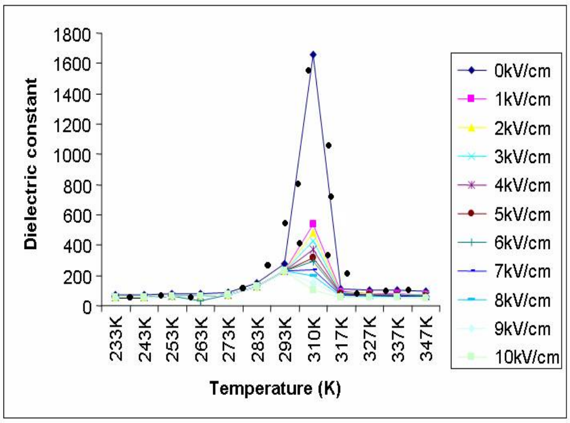


Fig 1. Soft mode frequency in $PbHPO_4$ crystal (Present calculation, ● Experimental results)
Fig.2. Dielectric constant in $PbHPO_4$ crystal (Present calculation,● Experimental results)

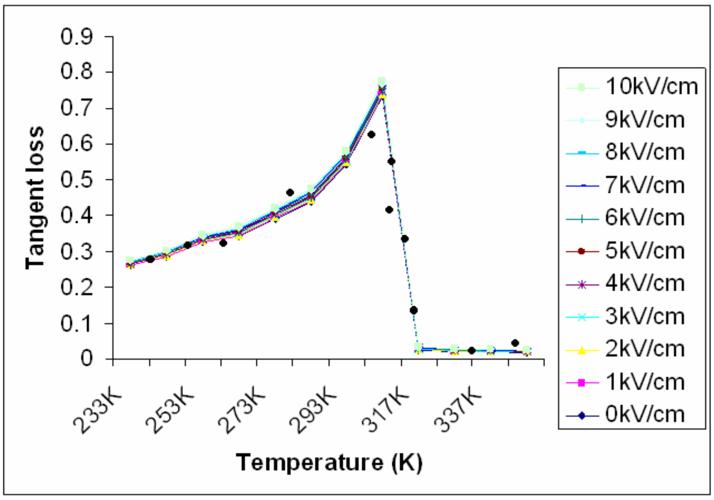


Fig 3. Tangent loss in $PbHPO_4$ crystal (Present calculation, ● Experimental results)

DISCUSSION

In the present work the effect of electric field on the dielectric properties of $PbHPO_4$ crystal has been studied .The two sub-lattice pseudo spin-lattice coupled mode model is extended by adding third and forth order phonon anharmonic interaction terms and external electric field term. With the help of double-time Green's function method, theoretically the field dependent expression for shift, width, soft mode frequency dielectric constant and loss tangent have been obtained. By fitting model values of physical quantities appearing in the expressions derived, field and temperature dependences of soft mode frequency, dielectric constant width shift and loss tangent have been calculated. Theoretical results have been compared with experimental result of Smutney & Fousek. [16] Previous workers have not considered the third order interaction an electric field terms in their calculation. Chaudhari et al [13] have not considered third-order-phonon anharmonic interaction terms in their model. This term is essential to explain linear temperature dependence (A) of soft mode frequency square $(i.e.\hat{\Omega} = AT + BT)$ since third order anharmonic term gives linear temperature dependence. Therefore, our calculation provides much better results to fit the experimental data. Secondly Chaudhuri et al [13] has decoupled the cor-relations in the early stage while we have decoupled them at a proper stage. As a result some important interactions disappeared from their calculations. If third order-phonon anharmonic terms are neglected from our expressions, these at once reduce to the expressions of previous workers. Present study shows that the electric field has pronounced effect on ferroelectric and dielectric properties of $PbHPO_4$-type crystal. The soft mode frequency increase while dielectric constant and loss tangent decrease with increase in electric field strength.

It can be seen from our expressions that our frequency $\tilde{\Omega}$ is same with the initial frequency of Chaudhuri et al [See Eqs(31)]. However, our soft mode frequency $\tilde{\Omega}$ contains extra terms $\Delta(\omega)$ [given in Eqs (15)]. Our soft mode frequency $\hat{\Omega}$ contains extra terms in $\tilde{\tilde{\omega}}_k and \Gamma_k(\omega)$ applying in $\Gamma_k(\omega)$ (Eq.27a)].These extra terms are given $\left|V^{(3)}(k_1, k_2, -k_3)\right|^2$ in $\Delta_k(\omega)$ [Eqs. (28)]and $\left|V^{(3)}(k_1, k_2, -k_3)\right|^2$ term given in $\Gamma_k(\omega)$ (Eq.29-)]. . These terms differentiate our expressions with the expressions given in the work of Chaudhuri et al[13].

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