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Effect of Electric Field on Dielectric Properties of Ferroelectric Crystal Lead Hydrogen Mono Phosphate*

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Abstract: The third and fourth order phonon anharmonic interactions external electric field terms are added in the two-sublattice pseudospin model for $PbHPO_4$ crystal. By using double time thermal Green's function method modified model, theoretical expressions for soft mode frequency and dielectric constant are evaluated for $PbH(D)PO_4$ crystal. Temperature and field variations of soft mode frequency and dielectric constant are calculated numerically. Present theoretical results agree with experimental result of Smutny and Fousek[1] for dielectric constant of $PbHPO_4$.

Keywords: Anharmonic terms, Green's function, Hamiltonian, Phase transition

1. Introduction

LHP($PbHPO_4$) and LDP($PbDPO_4$) crystal undergoes ferroelectric transition at 37^0C and 179^0C respectively. It provides an interesting example of simple H-bonded ferroelectrics suitable in testing microscopic theories of ferroelectricity in H-bonded and D-bonded substances respectively. In $PbHPO_4$ crystal the direction of spontaneous polarization is almost parallel to the direction of the H-bond O-H.....O projecting on the (010) plane unlike KH_2PO_4. The PO_4 groups are bound to one another by one dimensional chain along c-axis. In LHP the low value of dielectric constant and loss combined with the relatively high SHG efficiency make LHP promising materials for LASER. The simple crystal structure of LHP has appealed many researchers for using it as a reference material to understand proton ordering in H-bonding crystal. Chunlei et al[2] have studied thermodynamic properties using three dimensional transverse Ising model. The dynamical properties were not discussed in their work. Wesselinowa[3] has studied dynamical structure factor by using Green's function approach and pseudo

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spin-lattice coupled mode model. They have not considered two-sublattice coupled model which is appropriate for LHP type crystal. The dielectric properties and phase transition have not been studied in her work. The procedure of this work is quite different to our work. Chaudhuri et al[4] have studied phase transition and dielectric properties of $PbHPO_4$ crystal. But they have not considered third-order phonon anharmonic interaction. They have decoupled the correlations in the early stage. As a result some important interactions disappeared from their results. Ohno and Lockwood[5] have carried out Raman scattering experiments on LHP crystal. Nelmes et al[6] have made neutron diffraction studies on LHP crystal. Smutny and Fousek[1] have measured dielectric constant at different temperatures and electric field strength in $PbHPO_4$ crystal. Bajpai and Chaudhary[7] have analyzed neutron structural data to study molecular distortion of polar group for explaining the phase transition in LHP. In the present work, extended two-sublattice pseudospin lattice coupled mode model[4] with third and fourth order phonon anharmonic interactions is used to derive expressions for soft mode frequency and dielectric constant of $PbHPO_4$ crystal. Earlier researchers have not considered third order phonon anharmonic interaction so could not produce better results. We compared the calculated values dielectric constant with experimental data of Smutny and Fousek[1] for LHP. We also calculated the dielectric constant value for LDP with same methods.

2. Model Hamiltonian

Earlier researchers[4] have used a two sublattice-pseudo spin lattice coupled mode model to describe the phase transition in $PbHPO_4$ crystal, which is expressed by H_s,

(2.1)
$$H_s = -2\Omega\sum_i\left(S^x_{1i}+S^x_{2i}\right)-\sum_{ij}J_{ij}\begin{bmatrix}\left(S^z_{1i}S^z_{2i}\right)\\ +\left(S^z_{2i}S^z_{2i}\right)\end{bmatrix}-\sum_{ij}K_{ij}\left(S^z_{1i}S^z_{2i}\right)-2\mu E\sum_i\left(S^z_{1i}+S^z_{2i}\right)$$
$$-\sum_{ik}V_{ik}S^z_{1i}A_k-\sum_{ik}V_{ik}S^z_{2i}A^*_k+\frac{1}{4}\sum_k\omega_k\left(A_kA_k^{+}+B_kB_k^{+}\right).$$

where Ω is proton tunnelling frequency, S_i^z and S_i^x are components of pseudospin variable, S J_{ij} is interaction between same lattice and K_{ij} is interaction between different lattices, μ is dipole moment of O-H…O bond, E is external electric field, V_{ik} is spin-lattice interaction. They have also used fourth order phonon anharmonic term.
In the present study, we use the Hamiltonian

(2.2)
$$H = H_s + H_{anh.}$$

where

(2.3) $$H_{anh} = \sum_{k_1 k_2 k_3} V^{(3)}(k_1, k_2, k_3)\, A_{k_1} A_{k_2} A_{k_3} + \sum_{k_1 k_2 k_3 k_4} V^{(4)}(k_1, k_2, k_3, k_4)\, A_{k_1} A_{k_2} A_{k_3} A_{k_4},$$

where A_k and B_k are position and momentum operators, ω_k is harmonic phonon frequency, $V^{(3)}$ and $V^{(4)}$ are third-and fourth-order atomic force constants defined by Born and Huang[8].

3. Green's Functions and Soft Mode Frequency

We shall consider the Green's function

(3.1) $$G_{ij}(t-t') = -i\theta(t-t')\left\langle\left[S^z_{1i}(t); S^z_{1j}(t') \right]\right\rangle,$$

where θ is Heaviside's function.

The Green's function (3.1) differentiated twice, first with respect to time t and then to time t' to set its Fourier transform into Dysons Equation which gives value of Green's function as

(3.2) $$G(\omega) = \frac{\Omega \left\langle s_1^x \right\rangle \delta_{ij}}{\pi\left[\omega^2 - \hat{\Omega}^2 - 2\Omega i\Gamma(\omega)\right]},$$

where

(3.3) $$\hat{\Omega}^2 = \tilde{\Omega}^2 + 2\Omega\ (\Delta(\omega)),$$

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

where

(3.5) $$a = 2J\left\langle S_1^z \right\rangle + K\left\langle S_2^z \right\rangle + 2\mu E,$$

(3.6) $$b = 2\Omega,$$

(3.7) $$c = 2J\left\langle S_1^x \right\rangle + K\left\langle S_2^x \right\rangle,$$

In Equations (3.2) and (3.3) $\Delta(\omega)$ is frequency shift and $\Gamma(\omega)$ is width defined elsewhere $\hat{\Omega}$ is obtained by solving Eq. (3.3) self consistently gives value of soft mode frequency as

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

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4. Dielectric Constant

The expression for dielectric constant ϵ is derived using relation $\epsilon = 1 + 4\pi\chi$ and $\chi = -\lim_{\epsilon \to 0} 2\pi N\mu^2 G_{ij}(\omega + i\epsilon)$, where N is number of dipoles in the unit cell and μ is dipole moment of one such dipole. By using Eq. (3.2) we obtain at once the dielectric constant as ($\epsilon >> 1$ in ferroelectric crystals)

$$(3.9) \qquad \epsilon = \left(-8\pi N\mu^2\Omega\right)\left\langle S_1^x \right\rangle \delta_{ij} \left[\omega^2 - \hat{\Omega}^2 - 2\Omega i\Gamma(\omega)\right]^{-1}.$$

5. Numerical Calculations

By using model values[3] of various quantities in expressions (Table 1) temperature and electric field dependences Dielectric constant for LHP and LDP crystal have been calculated and shown in Figs. 1, 2 and 3.

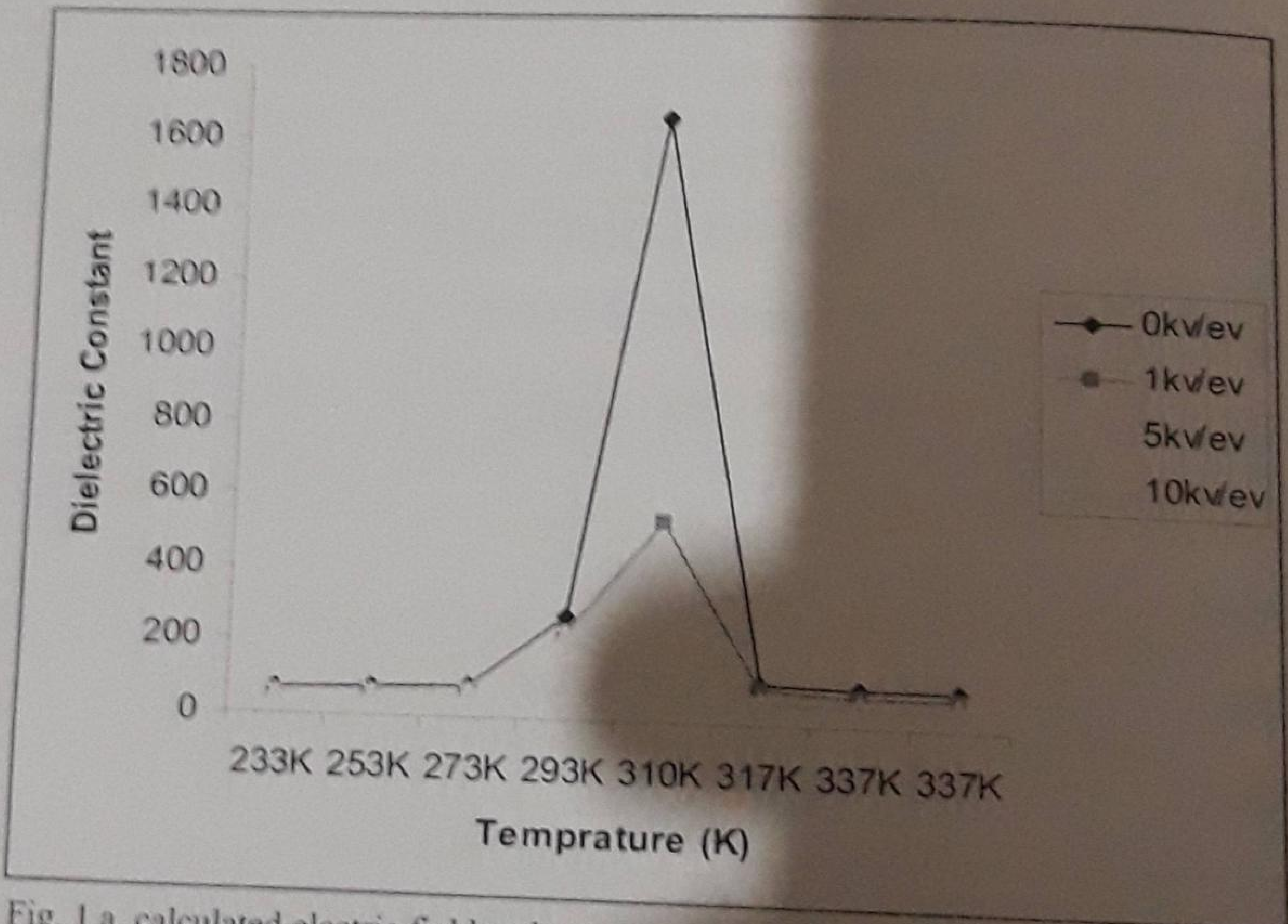

Fig. 1 a. calculated electric field and temperature dependence of dielectric constant in $PbHPO_4$ crystal

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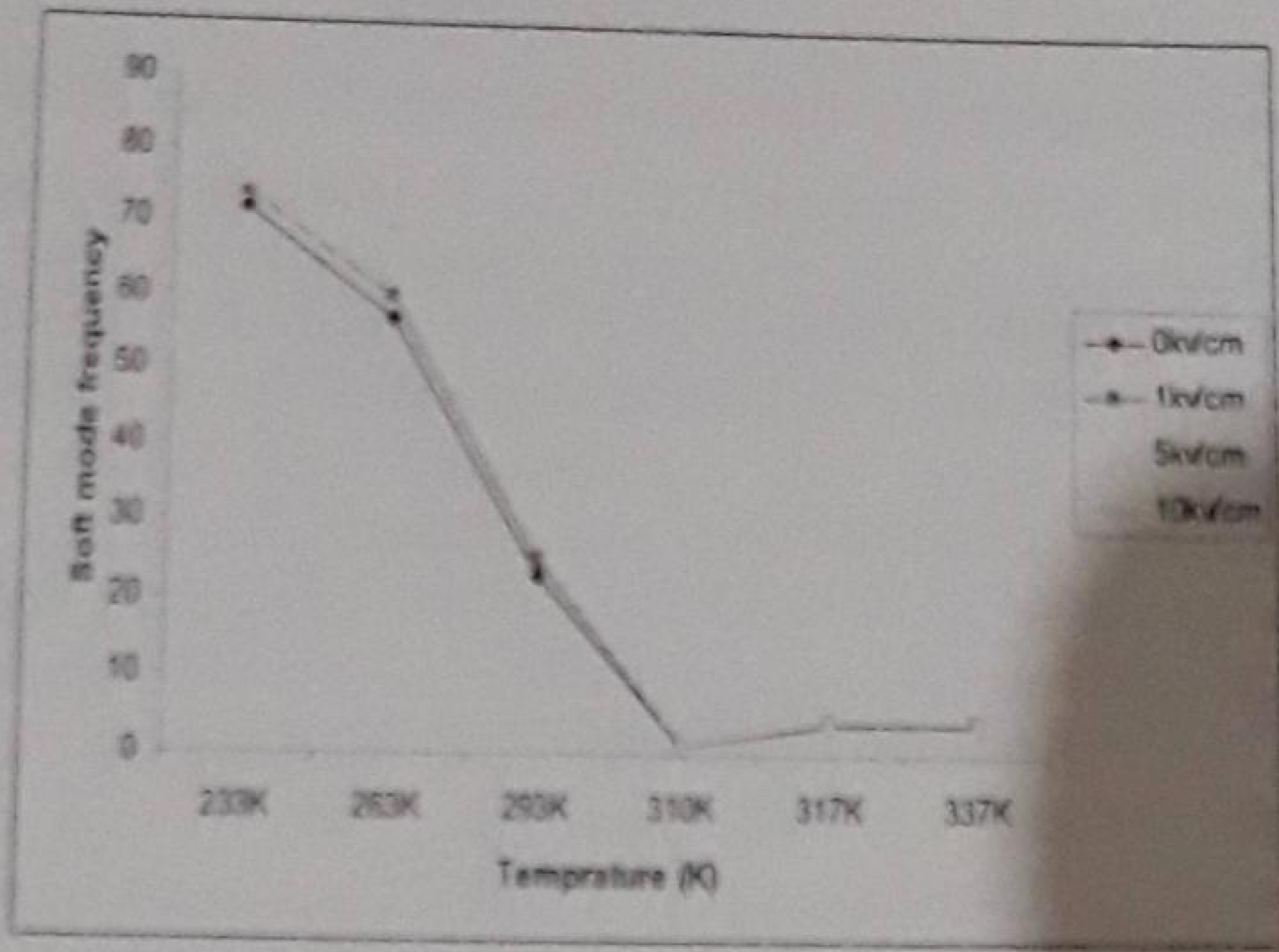

Fig.1. b. calculated electric field and temperature dependences of soft mode frequency in $PbHPO_4$ crystal

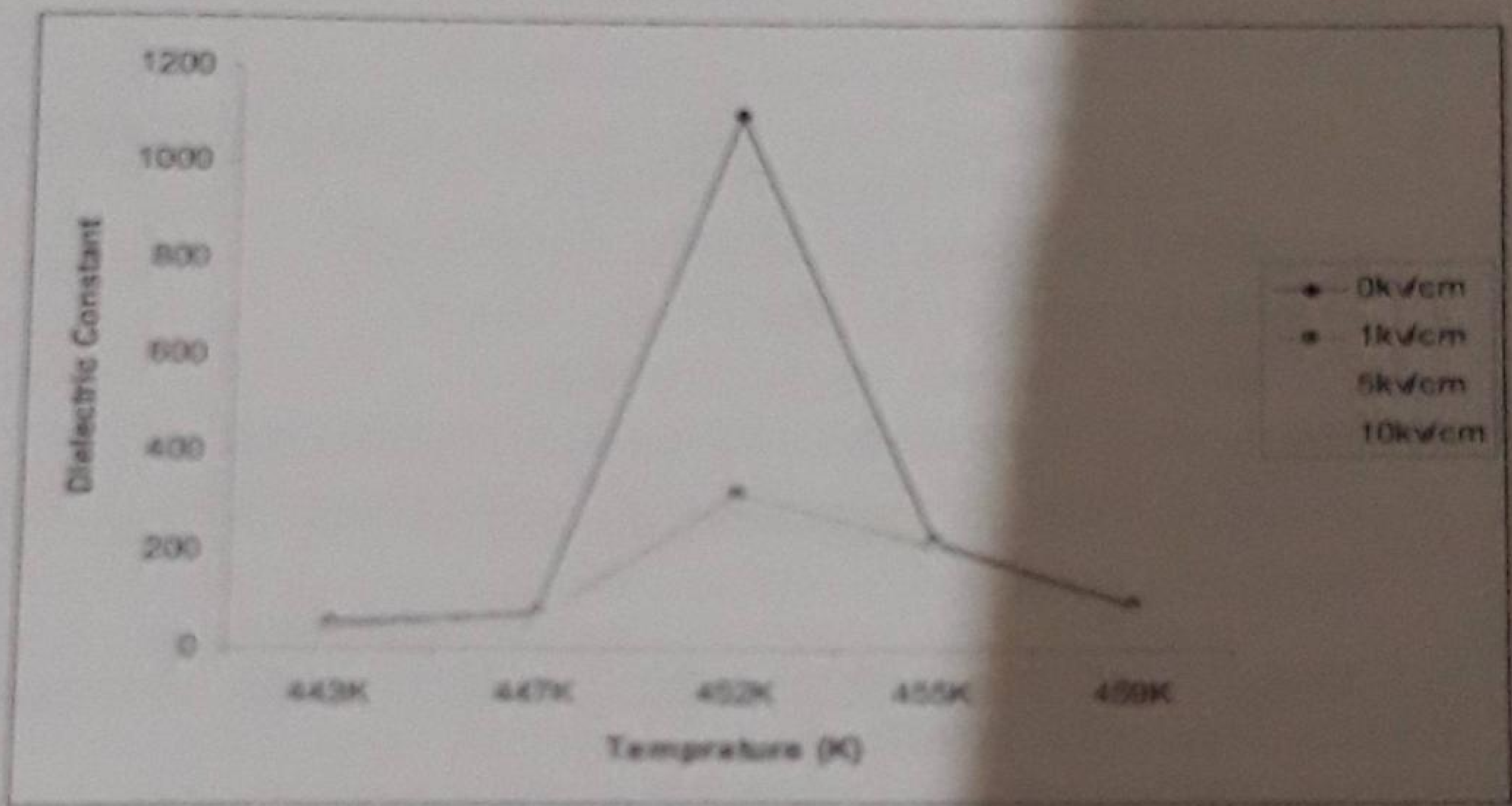

Fig.2 Calculated electric field and temperature dependences of Dielectric Constant in $PbDPO_4$ crystal

Table 1. Model values of physical quantities for LHP and LDP (Chaudhuri et al[5])

ω_k^2 (cm^{-2})	Ω (cm^{-1})	J_0 (cm^{-1})	K_0 (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^{-3})	ΩV_{ik} ($cm^{-5/2}$)
13.3	2.16	172.37	86.18	30.93	310	2773	0.55	2699	2034	76.75
30.81	0.27	251.32	125.66	63.71	452	2939	0.60	62.50	46.57	17.38

6. Conclusion

We obtain temperature and field variation of dielectric constant (ϵ) Figs (1) and (2) soft mode frequency $\hat{\Omega}$ (3). It emerges from present study that phonon anharmonic interactions significatently contribute in LHP & LDP crystal. Smutny and Fousek[1] have measured the dielectric constant at different electric field values and temperature in $PbHPO_4$ crystal our theoretical results agree with experimental data reported by Smutny and Fousek[7]. Previous authors have not studied the effect of electric field dielectric constant for LHP and LDP crystal. They have not considered third order phonon anharmonic interaction term in their calculation. High value of D.C. useful for high values capacitors. D.C. increase the electric flux density increase low dielectric makes capacitor used in high power frequency. Calculations on H-Boned crystal like $BaHPO_4$, $CaHPO_4$, and $PbHAsO_4$ etc. are progress in our laboratory.

References

1. F. Smutny and J. Fousek, *Ferroelectrics,* **21** (1978) 385.
2. B. K. Chaudhuri, S. Ganguli and D. Nath, *Phys. Rev., B.,* **23** (1981) 2308.W. Chunlei, Z. Qin and Z. Jingbo, *Ferroelectrics,* **77** (1988) 21.
3. N.-Ohno and D._J. Lockwood, *Ferroelectrics,* **72** (1977) 303.
4. R. J. Nelmes, *Ferroelectrics,* **24** (1980)237, **53** (1984) 207.
5. J. M. Wesselinowa, *Phys. Rev. B.,* **49** (1994) 3098.
6. P. K. Bajpai and R. N. P.Choudhary, *Ferroelctrics,* **29** (1999) 234.
7. M. Born and K. Huang, *Dynamical theory of Crystal Lattices*, Oxford Univ.Press, NY, 1954.
8. T. J. Negran, A. M. Glass, C. S. Brickenkamp, R. D. Rosenstein, R. K. Osterheld, R. Sussott, *Ferroelectrics,* **6** (1974) 179.
9. T. C. Upadhyay and Subodh Gairola, *Ind. J. Pure & Appl. Phys.,* **46** (2008) 727.
10. T. C. Upadhyay and Mayank Joshi, *Ind. J. Pure & Appl. Phys.,* **48** (2010) 550.
11. D. N. Zubarev, *Sov. Phys. Usp.,* **3** (1960) 320.

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