

Dielectric Constant and Tangent Loss in Paraelectric Phase for KH_2PO_4

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Abstract- By adding third and fourth order phonon anharmonic interaction terms in the four particle cluster model Hamiltonian for the stochastic motion of H_2PO_4^- groups and using double-time temperature dependent Green's function method and Dyson's equation techniques, expressions for the dielectric constant and dielectric tangent loss have been derived and discussed for KH_2PO_4 . Using model parameters given by Ganguli S, Nath D and Chaudhary B K (Phy. Rev., 1980 B21, 2937) the transverse dielectric constant and observed dielectric constant have been calculated for KH_2PO_4 in its paraelectric phase. The temperature dependence of this dielectric constant for KH_2PO_4 have been calculated and compared with experimental results of Raman Intensity (Ferroelectrics, 1983, 52, 91), Bush G (Phys. Acta., 1938,11, 265) and Kaminow I P and Harding G O (Phys. Rev., 1963,129, 1562), and Deguchi K. and Nakamura E (J. Phys. Soc. Jpn., 1980,49, 1887). The observed dielectric constant explains the Curie-Weiss behaviour of dielectric constant along the c-axis of KH_2PO_4 crystal in the paraelectric phase. Also the temperature dependence of tangent loss in paraelectric phase for KH_2PO_4 at 9.2 GHz for field along the a-axis, and c-axis have been calculated and compared with experimental results of Kaminow I P and Harding G O (Phys. Rev., 1963,129, 1562). A good agreement has been found.

At higher temperature the loss deviates from the Curie-Weiss type behaviour and increases linearly with temperature. This behaviour suggests that at higher temperature the phonon anharmonicity contributes significantly in the observed loss.

Keywords- Phonon Anharmonicity, Transverse dielectric constant, observed dielectric constant, tangent loss.

I. INTRODUCTION

The tunneling proton model has been long believed to be an established model of the phase transition in KH_2PO_4 and other hydrogen-bonded crystals. Experimental results of dielectric dispersion [1,2], Brillouin spectroscopy [3,4] and low frequency Raman spectroscopy [5] have shown, however, that the dynamical spectra of polarization fluctuations in pure and mixed crystals $\text{KDP}_{1-x}\text{DKDP}(\text{KD}_2\text{PO}_4)_x$ can be well analyzed in terms of the Debye type susceptibility [6], which is characterized by a relaxation time.

On the other hand, the origin of spontaneous polarization in KDP was established to be shift of P and O ions relative to K ions in diffraction experiments [7], so that the site symmetry of a PO_4 tetrahedron was determined to be C_2 below the transition temperature T_c . Consequently, the Debye susceptibility suggests that the order-disorder of PO_4 dipoles with C_2 site symmetry may be the transition mechanism of KDP. Raman spectroscopic studies confirm that the ferroelectric phase transition in KH_2PO_4 , KD_2PO_4 and their mixed crystals is due to the "order-disorder dynamics" of PO_4 dipoles [8].

The frequency and temperature dependence of dielectric constant near the Curie point of several ferroelectric crystals have investigated by [9,10]. Yamada *et al* [11] have suggested that one can have a fairly simple picture of dielectric relaxation process in the vicinity of the Curie point of order-disorder type ferroelectrics and the complex dielectric constant of these substances in the paraelectric (hereafter referred to as PE) region can be well explained by a mono disperse theory together with the critical slowing down of the relaxation time at the Curie point. The first non-disperse theory of this kind has been presented by Mason [12] in order to explain the observed behaviour in Rochelle salt. The Mason theory could give a qualitative account for the experimental results obtained by Akao and Sasaki [13], but still left some gulf between theory and experiment. For instance, the cole-cole plot given by Akao *et al* [13] clearly shows a deviation from the Debye circle to which the Mason theory should also be led. This suggests that a distribution of the relaxation time plays an important role. The theoretical studies of temperature dependence of microwave loss in Rochelle salt crystal have discussed [14] by considering PLCM model Hamiltonian with phonon anharmonicity up to fourth order and using double time thermal Green's function.

Yoshimitsu and Matsubara [15] have shown that most of the anomalous dielectric behaviours of order-disorder type crystals can be explained in terms of a special distribution with one parameter of the relaxation times.

Levitsky *et al* [16], on the basis of the model of relaxational dynamics, have shown that the two particle cluster theory treating the ferroelectric phase transition as a result of an instability in a deuteron (proton) subsystem gives a fair quantitative description of the relaxation soft mode dynamics of quasi-one-dimensional hydrogen bonded crystals.

An alternating field heats the dielectrics. This heat is known as dielectric loss. The dielectric loss consists of conduction losses corresponding to a constant voltage and loss due to the active component of the displacement current. The dielectric loss is associated with the slow establishment of polarization accompanied by absorption currents. In the high fields, the dielectric loss in these ferroelectrics is due to hysteresis. Microwave losses in the KDP have studied by Kaminow *et al* [9] and other [17, 18]. The temperature dependence of microwave loss tangent in KDP is empirically represented by $(T - T_C) \tan \delta = \alpha + \beta T + \gamma T^2$, this dependence can be explained in terms of slowing down of a relaxational mode. Experiments using the Brillouin scattering [4, 19] resonance vibration [20, 21] and pulse-echo [22] techniques for attenuation studies in these materials reflect the substantial asymmetry in the temperature dependence of the polarization relaxation rate in the PE phases analogous to that of susceptibility [22, 24]. Also the attenuation is found strongly frequency dependent and becomes so large to measure close to T_C [23, 24].

In our previous works [25] (hereafter referred to as I) and [26] (hereafter referred to as II), we have designed the four particle cluster model Hamiltonian for the stochastic motion of H_2PO_4^- groups. Applying Green's function method and Dyson's equation technique, the higher order correlations have been evaluated using the renormalized Hamiltonian. The collective frequencies, corresponding width and shift have been evaluated for KDP-type ferroelectrics in I. In the present study, we use same Hamiltonian as in I. Expressions for the dielectric constant and dielectric tangent loss have been derived and discussed for KH_2PO_4 . Using model parameters given by Ganguli *et al* [27] the transverse dielectric constant and observed dielectric constant has been calculated for KH_2PO_4 . The temperature dependence of the transverse dielectric constant for KH_2PO_4 have been calculated and compared with experimental results of Raman Intensity [27], Bush [29] and Kaminow *et al* [9].

The temperature dependence of the observed dielectric constant for KH_2PO_4 have been calculated and compared with experimental results of Bush [28], Deguchi *et al* [30], and Kaminow *et al* [9]. The observed dielectric constant explains the Curie-Weiss behaviour of dielectric constant along the c-axis of KH_2PO_4 crystal in the PE phase. The temperature dependence of tangent loss of KH_2PO_4 at 9.2 GHz for field along the a-axis ($\tan \delta_a$) along the c-axis ($\tan \delta_c$) have been calculated and compared with experimental results of Kaminow *et al* [9] in PE phase. At higher temperature the loss deviates from the Curie-Weiss type behaviour and increases linearly with temperature. This behaviour suggests that at higher temperatures the phonon anharmonicity contributes significantly in the observed loss.

II. DIELECTRIC PROPERTIES

The response of a dielectric field is conveniently described by the dielectric susceptibility. Following Kuo [31] and Zubarev [32], the general expression for complex dielectric susceptibility tensor $\chi_{mn}(\omega)$ can be expressed as:

$$\chi_{mn}(\omega) = \lim_{\varepsilon \rightarrow 0} -2\pi G_{mn}(\omega + j\varepsilon), \quad (1)$$

where $G_{mn}(\omega)$ the Fourier transform of the retarded double-time thermal Green's function between the m^{th} and n^{th} components of the crystal dipole moment operators $\vec{M}(t)$ in the Heisenberg representation and is defined as

$$\begin{aligned} G_{mn}(t-t') &= \langle\langle M_m(t); M_n(t') \rangle\rangle \\ &= -j\theta(t-t') \langle [M_m(t); M_n(t')] \rangle \end{aligned} \quad (2)$$

where $\theta(t-t')$ is the Heaviside step function and the angular brackets $\langle \dots \rangle$ denote the thermal ensemble average. The crystal dipole moment $\vec{M}(t)$ depends on the ionic co-ordinates, like potential energy, i.e., on the lattice configurations and can be expanded in a Taylor's series in terms of ionic displacements. Because of the periodic boundary conditions, i.e., symmetry considerations, imposed on the ionic motions, only the low lying relaxational modes have non-zero polarization associated with them.

Thus only the expansion coefficients which correspond to lowest frequency mode, i.e., $\bar{M}(q, j)$ {where $q = 0$ for ferroelectrics and j relates the modes of spectrum} contribute to the dielectric susceptibility, significantly. Thus we can write the dielectric susceptibility as:

$$\chi_{mn}(\omega) = \lim_{\varepsilon \rightarrow 0} -2\pi N\mu^2 G_{mn}(\omega + j\varepsilon), \quad (3)$$

where N is the number of unit cells in the sample and μ is the effective dipole moment per unit cell, and

$$G_{mn}(\omega + j\varepsilon) = \langle\langle A_q(t); A_q(t') \rangle\rangle = G'(\omega) - jG''(\omega) \quad (4)$$

and $G'(\omega)$ and $G''(\omega)$ are real and imaginary parts of the Green's function defined by equation (4) in I. The dielectric constant can be evaluated using the relation:

$$\varepsilon(\omega) = 1 + 4\pi\chi = \varepsilon'(\omega) - j\varepsilon''(\omega) \quad (5)$$

and $\varepsilon'(\omega)$ and $\varepsilon''(\omega)$ are real and imaginary parts of the dielectric constant. The real part of the dielectric constant can be expressed as

$$\varepsilon'(\omega) - 1 = -8\pi N\mu^2 G'(\omega), \quad (6)$$

and the imaginary part

$$\varepsilon''(\omega) = -8\pi N\mu^2 G''(\omega), \quad (7)$$

The dielectric loss ($\tan \delta$), for the dissipation of power in the dielectric crystal is defined as the ratio of imaginary and real parts of the dielectric constant, i.e.,

$$\tan \delta = \frac{\varepsilon''(\omega)}{\varepsilon'(\omega)} = \frac{G''(\omega)}{G'(\omega)}, \quad (8)$$

The dielectric susceptibility, equation (3); dielectric constant equation (6); and dielectric loss equation (8); can thus be calculated by using the Green's function method, Dyson's equation techniques and the same model Hamiltonian as in I.

Using above equation (3), and equation (4) in I, the dielectric susceptibility for KDP - system can be obtained as

$$\chi(\omega) = \frac{-2N\mu^2 \tilde{\omega}}{[\omega^2 - \tilde{\omega}^2 + 2j\omega \Gamma_P(\omega)]}, \quad (9)$$

where $\tilde{\omega}$, the collective mode frequency, represented in equation (5) in I, and the damping constant $\Gamma_P(\omega)$ is related as [32]. The range of frequencies used in ultrasound [31], Brillouin [4] and susceptibility [31] measurement experiments are such that $\omega \ll \tilde{\omega}$. Thus equation (9) reduces to:

$$\chi(\omega) = \frac{2N\mu^2 \tilde{\omega}}{\tilde{\omega}^2 [1 - j\omega\tau_P]}, \quad (10)$$

where the polarization relaxation time ($\tau_P = 2\Gamma_P / \tilde{\omega}^2$) is given by [33] for resonant interaction. This approximation of equation (9) is equivalent to Debye relaxation susceptibility. Furthermore, if $\omega\tau_P \ll 1$, which is true for KDP-system [34], equation (10) can be further reduced to:

$$\chi(\omega) = \frac{2N\mu^2 \tilde{\omega}}{\tilde{\omega}^2} (1 + j\omega\tau_P), \quad (11)$$

Using equation (5), the expression for dielectric constant from equation (9), can be obtained as

$$\varepsilon(\omega) - 1 = \frac{-8\pi N\mu^2 \tilde{\omega} \left[(\omega^2 - \tilde{\omega}^2) - 2j\omega\Gamma_P(\omega) \right]}{\left[(\omega^2 - \tilde{\omega}^2)^2 + 4\omega^2 \Gamma_P^2(\omega) \right]}, \quad (12)$$

The imaginary part of which can be written as:

$$\varepsilon''(\omega) = -\frac{8\pi N\mu^2 \tilde{\omega} 2\omega\Gamma_P(\omega)}{(\omega^2 - \tilde{\omega}^2)^2 + 4\omega^2 \Gamma_P^2(\omega)}, \quad (13)$$

and the real part as

$$\varepsilon'(\omega) - 1 = -\frac{8\pi N\mu^2 \tilde{\omega} (\omega^2 - \tilde{\omega}^2)}{(\omega^2 - \tilde{\omega}^2)^2 + 4\omega^2 \Gamma_P^2(\omega)}, \quad (14)$$

For the experimental range of frequencies, $\omega \ll \tilde{\omega}$ and ($\omega\tau_P \ll 1$ for KDP), equation (14) reduced to ($\varepsilon' \gg 1$)

$$\varepsilon'(\omega) = \frac{8\pi N\mu^2 \tilde{\omega}}{\tilde{\omega}^2 + \omega^2 \tau_p^2} = \frac{8\pi N\mu^2 \tilde{\omega}}{\tilde{\omega}^2} \quad (15)$$

where $\tilde{\omega}$ is given by equation (6) in I, and $\tilde{\omega}$ by equation (5) in I. The $\tilde{\omega}_+$ mode corresponds to E (x, y) mode, which is responsible for the observed transverse dielectric properties of KDP. In the simplest approximation $\tilde{\omega}_+$ can be written as, (from eq. (5) in I)

$$\tilde{\omega}_+ = K_1 + K_2 T, \quad (16)$$

where K_1 and K_2 are temperature independent parameters.

The dielectric tangent loss ($\tan \delta$) for the dissipation of power in a dielectric sample is given by equation (8), which can be written as:

$$\tan \delta = \frac{-\omega \Gamma_p}{(\omega^2 - \tilde{\omega}^2)}. \quad (17)$$

For the experimental values of the applied field frequency ω , one has $\omega \tau_p \ll 1$ for KDP system, and then the above equation can be approximated as

$$\tan \delta = \frac{\omega \tau_p}{2}, \quad (18)$$

By using Blinc-de Gennes model parameter values for KH_2PO_4 crystal as given by Ganguli *et al* [27] (represented in table 1 of II), the transverse dielectric constant $\varepsilon_a(0)$, and observed dielectric constant $\varepsilon_c(0)$ in PE phase for KH_2PO_4 , and tangents loss ($\tan \delta$) of KH_2PO_4 at 9.2 GHz for fields along the a-axis ($\tan \delta_a$), and c-axis ($\tan \delta_c$) are calculated represented in table 3 in of II.

III. COMPARISON WITH EXPERIMENTS AND DISCUSSION

Dielectric Constant: The result for transverse dielectric constant $\varepsilon_a(0)$ obtained from integrated intensity of Raman spectroscopy [31] and those measured by Bush [29] and Kaminow *et al* [9] are shown in figure 1, together with the theoretical result of Havlin, Litov and Uehling [35].

The temperature dependence of the transverse dielectric crystal, shown in figure 1, can be explained well by equation (31) in I and equation (32) in II, for KH_2PO_4 . This indicates that the low frequency $\tilde{\omega}_+$ {E(x, y) mode} is

closely related to the macroscopic dielectric constant ε_a' . It should be mentioned here that the low frequency E - mode continuous appears also in a deuterated KDP (DKDP), although the intensity is about one-third of that of KDP [36], which indicates the possibility that the spectrum is due to the hydrogen collective motion. Using equation (5) of I, for $\tilde{\omega}_+$ mode, it can be seen that the E-mode collective hydrogen motion has a characteristics damping factor which slowly increases as the temperature approaches T_c , while that of $\tilde{\omega}_-$ { $B_2(z)$ } soft mode the damping factor slowly decreases down to a finite value, which agrees with the observations of Kaminow *et al* [10]. The present results agree with the behaviour of the observed E - mode Raman spectrum in the following aspects

- (i) $\tilde{\omega}_+$ does not change appreciably as $T \rightarrow T_c$ in PE phase,
- (ii) $\Gamma_p(\omega_+)$ is weakly dependent on temperature, and
- (iii) because of the factor $(\omega^2 - \tilde{\omega}^2)$ in the numerator of equation (14), the susceptibility derived changes the corresponding spectrum from a simple overdamped form to a more flat one, like the E-mode Raman spectrum of KH_2PO_4 [36].

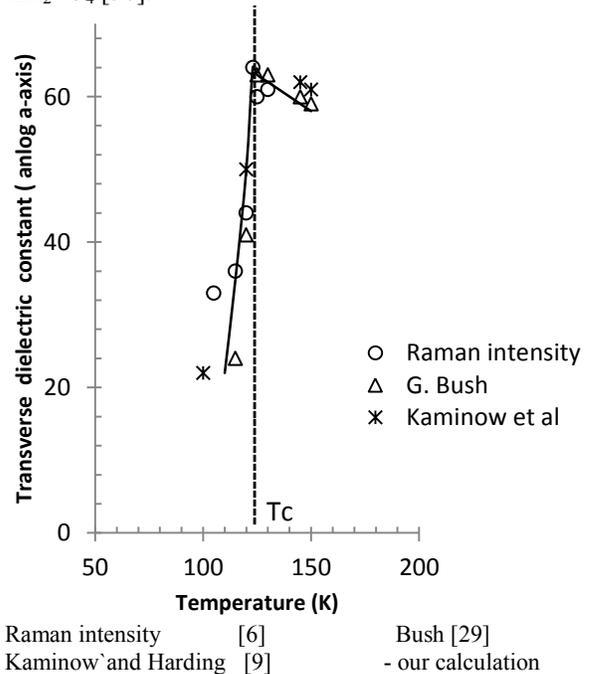
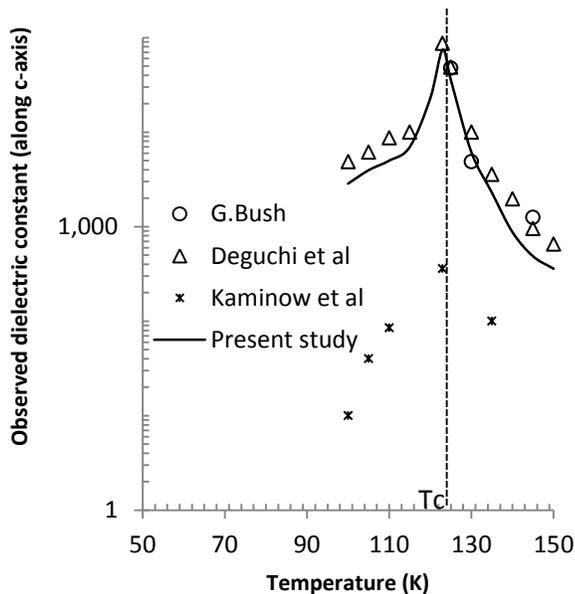


Figure 1. Temperature dependence of transverse dielectric constant $\varepsilon_a(0)$ in PE phase for KH_2PO_4 .

The observed dielectric constant (ϵ_c) of KH_2PO_4 along c-axis are shown in figure 2. The $\tilde{\omega}_q^- \{B_2(z)\}$ mode may be assigned for the observed temperature dependence of ϵ_c . Equation, $\tilde{\omega}_q^- \propto (T - T_C)$, the real part of the dielectric constant associated with this mode, from equation (41) in II, which explains the Curie-Weiss behaviour of dielectric constant along the c-axis of KH_2PO_4 crystal in the PE phase observed by Deguchi *et al* [30], Bush [29] and Kaminow *et al* [9], shown in figure 2. For temperature $T \rightarrow T_C$, ϵ_c tends to maximum value, which is consistent with the theory of Hill and Ichiki for TGS and KDP crystals. While Mason mono disperse theory [13] gives $\epsilon_c \rightarrow 0$ as $T \rightarrow T_C$. The origin of this difference in the temperature dependence of ϵ_c is easily traced back in mono disperse theory, the critical slowing down of the relaxation time has a dominant effect over the Curie-Weiss law of static dielectric constant, while the Hill-Ichiki theory of distribution function of relaxation time makes contribution to finite $\tau \neq 0$ to ϵ_c more dominant.



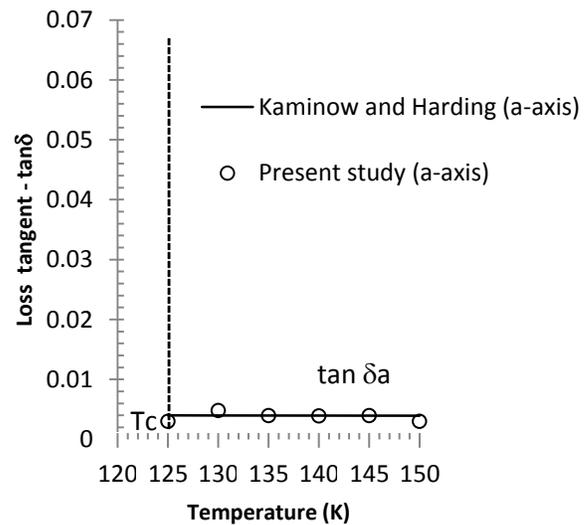
Bush [28]
Deguchi and Nakamura [30] Kaminow *et al* [9]

Figure 2. Temperature dependence of observed dielectric constant $\epsilon_c(0)$ in PE phase for KH_2PO_4 .

There are actually, however, many cases in which ϵ_c takes a minimum of finite value at $T = T_C$ being neither zero as in Mason's theory nor maximum as in Hill and Ichiki theory. In order to meet these experimental observations, the four-particle cluster collective motion model with anharmonicity, manifests plenty of varieties in the anomalous dielectric behaviour of KDP - type system near the critical points. The distribution of relaxation times and their slowing down was considered phenomenologically by Kubo - formula [30] for the dielectric susceptibility.

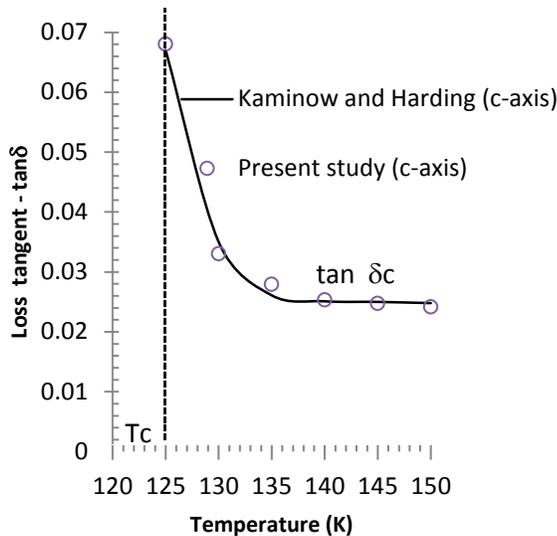
The possible origin of a distribution of the relaxation time, the effect of the short range correction between dipoles was examined for a model of KH_2PO_4 by Tokunaga and Matsubara [36]. They considered KD_2PO_4 as a kind of Ising spin system with two kinds of nearest neighbor interactions.

Dielectric Tangent loss: Calculated values of loss tangent of KH_2PO_4 at 9.2 GHz for fields along the a - axis ($\tan \delta_a$), and c - axis ($\tan \delta_c$) are given in table 3 in [26], and their temperature dependence are shown in figure 3 and 4 respectively.



Kaminow and Harding [9] O our calculation

Figure 3. Temperature dependence of Tangent loss in PE phase for KH_2PO_4 at 9.2 GHz. - for fields along the a-axis ($\tan \delta_a$).



Kaminow and Harding [9]

○ our calculation

Figure 4. Temperature dependence of Tangent loss in PE phase for KH_2PO_4 at 9.2 GHz. - for fields along the c-axis ($\tan \delta_c$).

The tangent loss is associated with damping parameter Γ {equation (25) in II}, damping can be understood as the creation of a virtual polarization mode excited by the transverse electromagnetic radiation and its subsequent decay into the real phonons by scattering from crystal defects, higher order phonon anharmonicities, etc. At higher temperature the loss deviates from the Curie-Weiss type behaviour and increases linearly with temperature. This behaviour suggests that at higher temperatures the phonon anharmonicity contributes significantly in the observed loss.

IV. CONCLUSIONS

From this paper, it is concluded that the consideration of four cluster Hamiltonian along with the third and fourth-order anharmonicities for KDP-type ferroelectrics leads to the renormalization and stabilization of the relaxational soft mode and the renormalization of the pseudo-spin exchange interaction constant. The decoupling of the correlations appearing in the dynamical equation after applying Dyson's equation, result in shift in frequency and facilitate the calculation of damping parameter, which is related to the tangent loss and relaxation time. The width and shift lead to the renormalization of the relaxational soft mode.

The present results reduce to the results of others [33, 38, 39] if the width and shift are neglected. The method of double time temperature dependent Green's function and Dyson's equation formalism have been found convenient and systematic to give the static and dynamical properties on a single framework of KDP-type system, using four-particle cluster model Hamiltonian along with phonon anharmonicities up to fourth order. The anomalous behaviour in order-disorder KDP-type ferroelectrics finds explanation by the consideration of collective proton-phonon interaction and third and fourth-order phonon anharmonicities in the four-particle cluster Hamiltonian. The dielectric properties and ultrasonic attenuation strongly depend on the relaxational mode behaviour of stochastic motion of H_2PO_4^- group in KDP-type ferroelectrics

Many workers [38, 39-41] used four proton cluster model but could not explain most of the features of KDP-system except the difference between the Curie and Curie-Weiss temperature. Vaks and Zinenko [42] Yoshimitsu and Matsubara [43] and Havlin and Sompolinsky [44] performed extensive calculations for the static thermodynamics behaviour in the four-particle cluster approximation and found satisfactory agreement with the experimental data, but they could not explain the observed relaxational behaviour of dielectric properties and ultrasonic attenuation explicitly. Ganguli *et al* [26] modified Ramakrishnan and Tanaka [38] theory by considering anharmonic interaction. Their treatment explains many features of order-disorder ferroelectrics. However, due to insufficient treatment of anharmonic interactions, they could not obtain quantitatively good results and could not describe some interesting properties, like dielectric, ultrasonic attenuation, etc. Our theoretical calculations fairly agree with experimentally [45-48] reported results within experimental errors.

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