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Microwave dielectric tangent losses in KDP and DKDP crystals

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Abstract. By adding cubic and quartic phonon anharmonic interactions in the pseudospin lattice coupled mode (PLCM) model for KDP-type crystals and using double-time temperature dependent Green's function method, expressions for soft mode frequency, dielectric constant and dielectric tangent loss are obtained. Using model parameters given by Ganguli *et al* [9] the dielectric losses are calculated for KDP and DKDP crystals. In the microwave frequency range an increase in frequency (1–35 GHz) is followed by an increase in dielectric tangent loss (1–35) at 98 K and (1–15) $\times 10^{-2}$ at 333 K for KDP and DKDP crystals respectively. The dielectric tangent loss decreases from 0.052 to 0.042 for KDP crystals with increase in temperature from 130 to 170 K and for DKDP crystals it decreases from 0.0166 to 0.0074 with an increase in temperature from 230–343 K in their paraelectric phases at 10 GHz. This shows Curie–Weiss behavior of the dielectric tangent loss.

Keywords. Ferroelectrics; Green's function; soft mode; anharmonicity; dielectric loss.

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1. Introduction

In recent years there has been considerable interest in investigating the dielectric properties of KDP-type (KH_2PO_4-type) ferroelectric crystals due to their promising applications in electrooptic and memory devices, optical communication, temperature control devices and ceramic industry [1]. In the ferroelectric crystals, it is the soft mode frequency which largely determines the dielectric, thermal and scattering properties [2]. At transition temperature the frequency of polar soft mode tends to zero and lattice displacement associated with this mode becomes unstable which explains the anomalous behavior of many of the physical properties of ferroelectric crystals [3] such as dielectric constant and loss. In KH_2PO_4 crystal ($T_c = 123$ K) the soft mode is connected with the pseudospin-type motion of the proton between two equilibrium sites in the double minimum type O–H..O bond potential. De Gennes [4], Tokunaga and Matsubara [5], Blinc and Zeks [6], Kobayashi [7], Ramakrishnan and Tanaka [8], and Ganguli *et al* [9] have investigated theoretically the dielectric and other properties of KDP-type crystals using pseudospin model and its extension, i.e., pseudospin lattice coupled mode model. Wang *et al* [10] and Jhang *et al* [11] have applied undetermined constant method to pseudospin model with four spin coupling term. They have not considered phonon part in their calculations which however has a very important contribution in crystals. Ganguli *et al* [8] have used PLCM model with fourth-order phonon anharmonic interaction term. They however decoupled the correlations at an early stage. In doing so some important interactions were disappeared. In this way they could not obtain better and convincing results to explain dielectric and phase transition properties of KDP-type crystals.

In the present work, we shall consider the third- and fourth-order phonon anharmonic interaction terms [12,13] into pseudospin lattice coupled mode (PLCM) model of KH_2PO_4 crystal. The phonon anharmonic interactions are found to be very important in explaining dielectric, thermal and scattering properties of solids by many authors [14–17] in the past. We use the double time thermal Green's function method [18] of quantum dynamics, for the development. We shall evaluate expressions for frequency, shift, width, soft mode frequency, dielectric constant and loss. Using the model parameters given by Ganguli *et al* in the theoretical expression for $\langle S^x\rangle$, $\langle S^z\rangle$, soft mode frequency (Ω'') etc. their thermal variations will be calculated.

The frequency and temperature dependences of dielectric loss in the range (1–35 GHz) and in the temperature ranges (130–170 K) and (230–343 K) at 10 GHz for KDP and DKDP respectively will be calculated and will be compared with experimental results of Kaminow and Harding [19], Kaminow [20] and Gervais and Simon [21].

2. Hamiltonian and Green's function

We modify [12,13] the PLCM Hamiltonian by including third- and fourth-order phonon anharmonic interaction terms which is expressed as

$$H = -2\Omega\sum_i S_i^x - \frac{1}{2}\sum_{ij} J_{ij} S_i^z S_j^z + \frac{1}{4}\sum_k \omega_k (A_k^+ A_k + B_k^+ B_k) + \sum_{ik} V_{ik} S_i^z A_k + \sum_{k_1k_2k_3} V^3(k_1k_2k_3) A_{k_1}A_{k_2}A_{k_3} + \sum_{k_1k_2k_3k_4} V^4(k_1k_2k_3k_4) A_{k_1}A_{k_2}A_{k_3}A_{k_4} \quad (1)$$

where $S_q^z = \sum_i S_i^z \exp(-iq\cdot R_i), R_i$ denotes the Bravais lattice site of the ith bond, Ω is the proton tunneling frequency, J_{ij} represents exchange interaction between pseudospins, S_i^x and S_i^z are the components of spin-half operator, V_{ik} is the linear pseudospin–lattice coupling constant, A_k is the displacement operator, B_k is the momentum operator, ω_k is the harmonic phonon frequency and V^3 and V^4 are the third- and fourth-order atomic potentials respectively defined by Born and Huang [22].

We consider the Green's function [18] as

$$G(t-t') = \langle\langle S_t^z; S_j^z\rangle\rangle = -i\theta(t-t')\langle[S_t^z; S_j^z]\rangle \quad (2)$$

where θ is the unit step function, $\theta = 1$ for $t > t'$ and zero otherwise.

Writing equation of motion for Green's function (2) using Hamiltonian (1) Fourier transforming and writing it in Dyson's equation form one gets

$$G(\omega) = \pi^{-1}\Omega\langle S_q^x\rangle[(\omega^2 - \Omega''^2 - i\Gamma\Omega)]^{-1}, \quad (3)$$

where

$$\Omega''^2 = \Omega'^2 + \Delta. \tag{4}$$

Solving eq. (4) self-consistently one gets

$$\Omega''^2_{\pm} = \frac{1}{2}(\Omega'^2 + \omega'^2_q) \pm \frac{1}{2}[(\omega'^2_q - \Omega'^2)^2 + 8V_q^2\Omega\langle S_q\rangle]^{1/2}, \tag{5}$$

where

$$\Omega'^2 = \Omega^{*2} + (2a^4 + V_q a^2 N_q)^{1/2} \tag{6}$$

with $\Omega^{*2} = a^2 + b^2 - bc, a = J_q\langle S^z\rangle, b = 2\Omega$, and

$$c = J_q\langle S^x\rangle. \tag{7}$$

Shift $\Delta(\omega)$ is given as

$$\Delta(\omega) = \Delta_1 + \Delta_2 + \Delta_3 \tag{8a}$$

with

$$\Delta_1 = 2a^4(\omega^2 - \Omega^{*2})^{-1}, \quad \Delta_2 = V_q^2 a^2 N_q(\omega^2 - \Omega^{*2})^{-1} \quad \text{and}$$
$$\Delta_3 = 4V_q^2\langle S_q^x\rangle\omega_q(\omega^2 - \omega''^2_q)[(\omega^2 - \omega''^2_q)^2 + \Gamma_q^2(\omega)]^{-1}. \tag{8b}$$

Width $\Gamma(\omega)$ is given as

$$\Gamma(\omega) = \Gamma_1 + \Gamma_2 + \Gamma_3 \tag{9a}$$

where

$$\Gamma_1 = \pi a^4\Omega^{*-1}[\delta(\omega - \Omega^*)],$$
$$\Gamma_2 = \pi V_q^2 a^2 N_q(\Omega^*)^{-1}[\delta(\omega - \Omega^*) - (\omega + \Omega^*)], \quad \text{and}$$
$$\Gamma_3 = 4V_q^2\langle S_q^x\rangle\Gamma_q(\omega)[(\omega^2 - \omega''^2_q)^2 + \Gamma_q^2(\omega)]^{-1}. \tag{9b}$$

In the expressions (8b) and (9b), frequency ω''_q, phonon shift $\Delta_q(\omega)$ and phonon width $\Gamma_q(\omega)$ are the result of anharmonic phonon interactions, which are obtained by solving phonon Green's function with the help of Hamiltonian (third, fifth and sixth terms of eq. (1))

$$\langle\langle A_q; A_q^+\rangle\rangle = \omega_q\pi^{-1}[(\omega^2 - \omega''^2_q) - i\Gamma_q(\omega)]^{-1} \tag{9c}$$

with $\omega''^2_q = \omega'_q + \Delta_q(\omega)$, $\omega'^2_q = \omega_q^2 + \omega_q\langle[F, B_q^+]\rangle(2\pi)^{-1}$ and $F_q = 2\pi\sum 3\sum V_s(q_1 \dots q_{s-1}, -q)A_{q1}\dots A_{qs-1}$.

Phonon shift is given as

$$\begin{aligned}\Delta_q(\omega) = {} & 36\omega_q \sum_{q_1 q_2} |V^3(q_1 q_2, -q)|^2 \frac{\omega_{q_1}\omega_{q_2}}{\omega'_{q_1}\omega'_{q_2}}[(N_{q_1}+N_{q_2}) \\ & \times\{(\omega'_{q_1}+\omega'_{q_2})/(\omega^2-(\omega'_{q_1}+\omega'_{q_2})^2) \\ & +(N_{q_2}-N_{q_1})\{(\omega'_{q_1}+\omega'_{q_2})/(\omega'^2-(\omega'_{q_1}+\omega'_{q_2})^2)\}] \\ & +96\omega_q \sum_{q_1 q_2 q_3} |V^4(q_1 q_2 q_3, -q)|^2 \frac{\omega_{q_1}\omega_{q_2}\omega_{q_3}}{\omega'_{q_1}\omega'_{q_2}\omega'_{q_3}} \\ & \times[(1+N_{q_1}N_{q_2}+N_{q_2}N_{q_3}+N_{q_3}N_{q_1}) \\ & \times\{(\omega'_{q_1}+\omega'_{q_2}+\omega'_{q_3})/(\omega^2-(\omega'_{q_1}+\omega'_{q_2}+\omega'_{q_3})^2)\} \\ & +3|[(1-N_{q_1}N_{q_2}+N_{q_2}N_{q_3}-N_{q_3}N_{q_1}) \\ & \times\{(\omega'_{q_1}-\omega'_{q_2}-\omega'_{q_3})/(\omega^2-(\omega'_{q_1}-\omega'_{q_2}-\omega'_{q_3})^2)\}]. \end{aligned} \tag{9d}$$

Phonon width is given as

$$\begin{aligned}\Gamma_q(\omega) = {} & 18\pi\omega_q \sum_{q_1 q_2} |V^3(q_1 q_2, -q)|^2 \frac{\omega_{q_1}\omega_{q_2}}{\omega'_{q_1}\omega'_{q_2}}[(N_{q_1}+N_{q_2}) \\ & \times\{\delta(\omega+\omega'_{q_1}+\omega'_{q_2})-\delta(\omega-\omega'_{q_1}-\omega'_{q_2})\}(N_{q_2}-N_{q_1}) \\ & \times\{\delta(\omega+\omega'_{q_1}-\omega'_{q_2})-\{\delta(\omega-\omega'_{q_1}+\omega'_{q_2})\}] \\ & +48\pi\omega_q \sum_{q_1 q_2 q_3} |V^4(q_1 q_2 q_3, -q)|^2 \frac{\omega_{q_1}\omega_{q_2}\omega_{q_3}}{\omega'_{q_1}\omega'_{q_2}\omega'_{q_3}} \\ & \times[(1+N_{q_1}N_{q_2}+N_{q_2}N_{q_3}+N_{q_3}N_{q_1}) \\ & \times\{\delta(\omega+\omega'_{q_1}+\omega'_{q_2}+\omega'_{q_3})-\delta(\omega-\omega'_{q_1}-\omega'_{q_2}-\omega'_{q_{34}})\} \\ & +3|[(1-N_{q_1}N_{q_2}+N_{q_2}N_{q_3}-N_{q_3}N_{q_1})\{\delta(\omega+\omega'_{q_1}-\omega'_{q_2}-\omega'_{q_3}) \\ & -\delta(\omega-\omega'_{q_1}-\omega'_{q_2}-\omega'_{q_{34}})\}]. \end{aligned} \tag{9e}$$

In the vicinity of transition temperature in the paraelectric phase one may expand $\Omega_{-}''^2$ in powers of $(T-T_c)$ around its value at T_c getting immediately

$$\Omega''^2 = \left(\frac{\partial\Omega_{-}''^2}{\partial T}\right)_{T=T_c}(T-T_c) \tag{10a}$$

$$\Omega''^2 = K(T-T_c) \tag{10b}$$

with

$$K = \Omega^2 J_q [kT_c^2 \cosh^2 \beta c\Omega]^{-1}. \tag{11}$$

When $T \to T_c, \Omega'' \to 0$, eq. (5) gives

$$T_c = \Omega\left[k \tanh^{-1}\left(\frac{4\Omega}{J'_q}\right)\right]^{-1} \tag{12}$$

where

$$J_q' = J_q + \omega_q^2 V_q^2 [\omega_q''^4 + \Gamma_q^2]^{-1}. \tag{13}$$

Now following Kubo [23] and Zubarev [18] the real part of dielectric constant ε of KDP crystal can be expressed with the help of Green's function (4) as

$$\varepsilon - 1 = -8\pi^2 N\mu^2 \text{ Re } alG(\omega) = -8\pi N\mu^2 \Omega \langle S_q^x \rangle (\omega^2 - \Omega''^2) \times [(\omega^2 - \Omega''^2)^2 + 4\Omega^2\Gamma^2(\omega)]^{-1}. \tag{14}$$

3. Dielectric tangent loss

The dielectric tangent loss for the dissipation of power is defined as the ratio of imaginary and real parts of dielectric constant. The dielectric tangent loss can be calculated with the help of eq. (14) as

$$\tan\delta = \frac{\varepsilon''}{\varepsilon'} = -2\Omega\Gamma(\omega)[(\omega^2 - \Omega''^2)]^{-1}. \tag{15}$$

At the microwave frequencies, $\omega, (\omega/\Omega''_- = 10^{-3})$, one may approximate $\Omega'' \gg \omega$ and $\Omega'' \gg \Gamma(\omega)$ so that eq. (15) reduces to

$$\tan\delta = -2\Omega\Gamma(\omega)\Omega''^{-2}. \tag{16}$$

Now writing $\Gamma(\omega)$ in the form $\Gamma(\omega) = \alpha + \beta T + \gamma T^2$ and by making use of eq. (10) for Ω'' we obtain eq. (16) in the from

$$(T - T_c)\tan\delta = -2\Omega(A + BT + CT^2). \tag{17}$$

where $A = \alpha K^{-1}, B = \beta K^{-1}, C = \gamma K^{-1}, K$ being given by eq. (10) and α, β and γ are obtained with the help of eqs (9a)–(9e) which come out as

$$\alpha = \frac{2\pi V_q^4 \omega_q^2 \Omega^2}{\Omega'(\omega^2 - \omega_q''^2)^2}[\delta(\omega - \Omega') - \delta(\omega + \Omega')] \tag{18}$$

$$\beta = \frac{36\pi V^2 \omega_q^2 \Omega k}{(\omega^2 - \omega_q''^2)^2} \sum |V^3(q_1, q_2, -q)|^2 \frac{\omega_{q_1}\omega_{9q_2}}{(\omega_{q_1}\omega_{q_2})^2} \times [(\omega_q'' + \omega_{q_2}'')\{\delta(\omega + \omega_{q_1}'' + \omega_{q_2}'') - \delta(\omega + \omega_{q_1}'' - \omega_{q_2}'')\}] + (\omega_{q_2}'' - \omega_{q_1}'')\{\delta(\omega_{q_1}'' + \omega_{q_1}'' - \omega_{q_2}'') - \delta(\omega - \omega_{q_1}'' + \omega_{q_2}'')\} \tag{19}$$

$$\gamma = \frac{192\pi V^2 \omega_q^2 \Omega K}{(\omega^2 - \omega_9''^2)^2} \sum_{q_1 q_2 q_3} |V^4(q_1 q_2 q_3 - q)|^2 \times \left[\frac{\omega_{q_1}\omega_{q_2}\omega_3}{(\omega_{q_1}''\omega_{q_2}''\omega_{q_3}'')^2}\right][\omega_{q_1}'' + \omega_{q_2}'' + \omega_{q_3}''] \{\delta(\omega + \omega_{q_1}'' + \omega_q'' + \omega_{q_3}'')\} - \delta(\omega + \omega_{q_1}'' + \omega_{q_2}'' + \omega_{q_3}'') + 3(\omega_{q_1}'' - \omega_{q_2}'' - \omega_{q_3}'') \times \{\delta(\omega + \omega_{q_1}'' - \omega_{q_2}'' - \omega_{q_3}'') - \delta(\omega - \omega_{q_1}'' + \omega_{q_2}'' + \omega_{q_3}'')\}]. \tag{20}$$

3.1 *Numerical calculation of width, shift and soft mode frequency*

Using Blinc–de Gennes model parameter values for KH_2PO_4 and KD_2PO_4 crystals as given by Ganguli *et al* [8] (see table 1), we have calculated $\langle S_q^x \rangle = (2\Omega/\Omega')$ tanh $\beta\Omega', \langle S_q^z \rangle = (J_q \langle S_q^z \rangle/\Omega')$ tanh $\beta\Omega'$. Using eqs (5)–(9) we have calculated Ω', Ω^*, soft mode frequency Ω'', shift $\Delta(\omega)$ and width $\Gamma(\omega)$ for KDP and DKDP crystals (see tables 2 and 3).

3.2 *Frequency dependence of dielectric losses*

Putting our calculated values of $\Gamma(\omega)$ and Ω'' for different temperatures into eq. (16) or (17) we obtain dielectric loss for KDP and DKDP crystals in 1–35 GHz range at 98 K and 333 K respectively. The variations are shown in graphs (figures 1 and 2). The increase in frequency (1–35 GHz) is followed by an increase in loss (1–35) and (2–15) $\times 10^{-2}$ in KDP and DKDP crystals respectively. Our theoretical results fairly agree with experimentally reported results [19–21] within experimental errors.

Table 1. Blinc–de Gennes model parameters for KDP and DKDP as given by Ganguli *et al* [8].

Crystal	Ω (cm^{-1})	J (cm^{-1})	J' (cm^{-1})	T_c (K)	V/kT_c
KDP	82	334	440	123	0.299
DKDP	0.5	472	626	229	0.144

Table 2. Calculated values for KDP crystal.

T (K)	$\langle S_q^x \rangle$	$\langle S_q^z \rangle$	Ω' (cm^{-1})	Ω'' (cm^{-1})	$\Gamma = \Gamma_1 + \Gamma_2 + \Gamma_3$ (10^3 cm^{-1})
122	0.475	0.156	62.95	87.50	2.78+2.77+0.96
123	0.500	0.000	1.15	0.00	0.00+000+0.12
130	0.483	0.000	30.24	30.20	0.00+0.28+12.94
140	0.461	0.000	45.80	45.76	0.00+0.35+12.01
150	0.441	0.000	56.34	56.30	0.00+0.00+11.21
160	0.421	0.000	65.19	65.15	0.00+0.00+10.51
170	0.404	0.000	71.86	71.81	0.00+0.00+9.86

Table 3. Calculated values for DKDP crystal.

T (K)	$\langle S_q^x \rangle$	$\langle S_q^z \rangle$	Ω' (cm^{-1})	Ω'' (cm^{-1})	$\Gamma = \Gamma_1 + \Gamma_2 + \Gamma_3$ (10^3 cm^{-1})
230	0.0016	0.00	0.0	0.0	0+0+0
240	0.0015	0.00	5.0	5.0	0+0+1.41
260	0.0014	0.00	6.0	6.0	0+0+1.45
279	0.0013	0.00	7.0	7.0	0+0+1.48
297	0.0013	0.00	8.0	8.0	0+0+1.52
316	0.0011	0.00	9.0	9.0	0+0+1.57
343	0.0010	0.00	10.0	10.0	0+0+2.12

3.3 *Temperature dependence of dielectric losses*

Using eq. (16) or (17) and our calculated values from table 2, we have calculated dielectric loss at 10 GHz in temperature ranges (130–170 K) and (230–343 K) for KDP and DKDP crystals respectively. We have calculated losses at 10 GHz frequency because we have experimental data available only at this frequency range. The dielectric loss vs. $(T - T_c)^{-1}$ for KDP and DKDP crystals are shown in figures 3 and 4 respectively. Our theoretical results are in good agreement with experimental results of Kaminow [20] and Gervais and Simon [21].

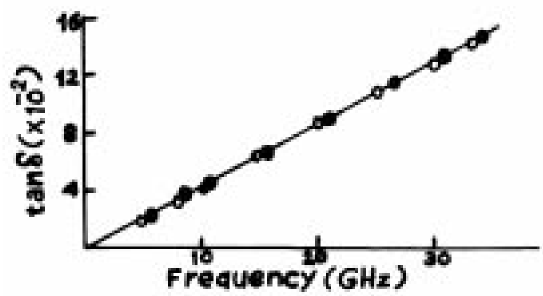

Figure 1. Dielectric loss vs. frequency in KDP at 98 K. ○ Kaminow [20], • our calculation.

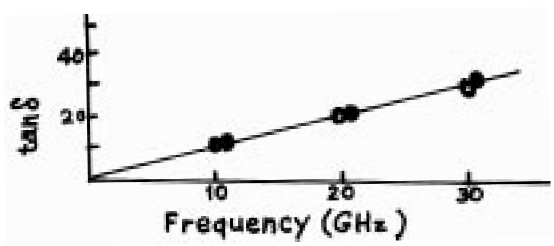

Figure 2. Dielectric loss vs. frequency in DKDP at 333 K. ○ Kaminow and Harding [20], • our calculation.

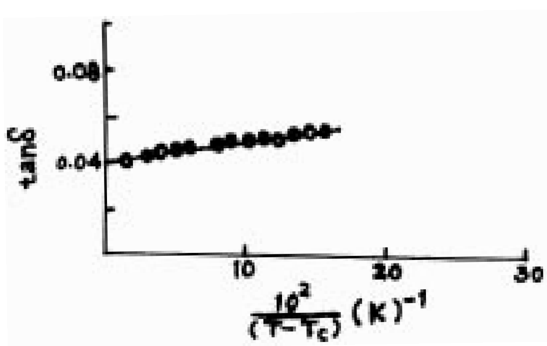

Figure 3. Microwave loss vs. $(T - T_c)^{-1}$ at 10 GHz in a single KDP crystal. ○ Gervais and Simon [21], • our calculation.

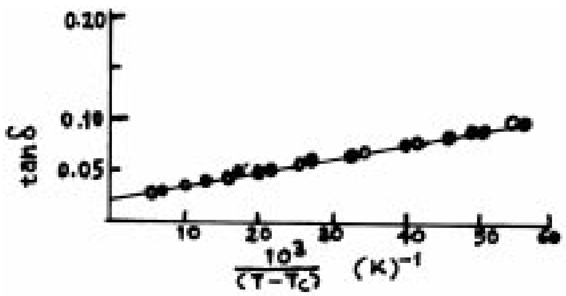

Figure 4. Microwave loss vs. $(T - T_c)^{-1}$ at 10 GHz in a single DKDP crystal. ◦ Kaminow and Harding [19], • our calculation.

4. Conclusion

In this paper, by modifying the pseudospin lattice coupled mode (PLCM) model for KDP-type ferroelectric crystals by adding the third- and fourth-order phonon anharmonic interaction terms, we have evaluated theoretically the expressions for the soft mode frequency, dielectric constant and dielectric tangent loss. Using Blinc–de Gennes model parameter values given by Ganguli *et al* [8] we have calculated temperature variations of these quantities for KDP and DKDP crystals. Our work differs from the works of some other works [5,7,9,11] in the sense that they have not considered phonon anharmonic terms. Only Ganguli *et al* [8] have considered fourth-order anharmonic term but they also could not do it in a convincing way as they truncated the spin correlations at an early stage, and so could not obtain width and shift. In this way they could not explain dielectric loss and also could not obtain better results as reported by us. Our eqs (16) and (17) along with figures 1 and 2 show that dielectric losses vary linearly with frequency which is in agreement with experiments [19–21]. The loss can be explained as follows: A transverse radiation field derives the low-lying transverse mode of the material in a forced vibration. Energy is transferred from the electromagnetic field to this lattice mode and is then degraded into other vibrational modes of the material. Due to anharmonic phonon interactions, decay processes take place. For example, third-order interaction leads to the decay of a virtual phonon into two real phonons or the virtual phonon may be destroyed by scattering a thermally excited phonon. Similar processes occur for fourth and higher order interactions. In figures 3 and 4 the losses show Curie–Weiss behavior, i.e., losses are proportional to $(T - T_c)^{-1}$ in the vicinity of T_c which is in agreement with experiments [19–21]. In the ferroelectric phase the losses are high because both pseudospin and the phonon parts contribute whereas dielectric losses are low in paraelectric phase because only phonon part contributes in this phase $(\langle S^z \rangle = 0)$ which is well explained by eqs (5) and (16).

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