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# Temperature dependence of microwave loss in antiferroelectric squaric acid crystal

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A model of pseudospin for layers 1 and 2 of squaric acid (SQA) crystal proposed by Li and Qin, *Ferroelectrics,* 82 (1988), 69 has been modified to include pseudospin-phonon interaction and cubic and quartic phonon anharmonic interactions. With the help of double-time temperature dependent Green's function method and modified Hamiltonian, expressions for antiferroelectric soft mode frequency, dielectric constant and loss have been evaluated. By fitting model values of physical quantities in the theoretical expressions, temperature dependences of soft mode frequency and loss have been calculated. In the microwave frequency range, an increase in frequency is followed by increase in loss. The loss decreases with increase in temperature for SQA crystal in its paraelectric phase. This shows Curie-Weiss behaviour of the dielectric tangent loss. Theoretical results are compared with experimental results of Muser [*Proc. 6th Int'l Meeting on Ferroelectricity, Kobe* (1985)], Maier *et al*. [*Phys Status Solidi* (b) 89, (1978)], and Samara and Semmingsen [*J Chem Phys* 71 (3) (1979) 1401] which show a good agreement.

**Keywords:** Antiferroelectrics, Anharmonic interactions, Dielectric, Loss tangent

## 1 Introduction

Antiferroelectric squaric acid crystal ($C_4O_4H_2$) or SQA has layered structure. Each molecule is a square with oxygen ions at its four vertices linked by hydrogen bonds on equal footing. At room temperature, the layers are ferroelectrically ordered and antiferroelectrically stacked. At about 371K the crystal undergoes a first order phase transition to paraelectric phase[1-6]. Because of the planar H-bonds, the interaction between molecules in the same layers is stronger than in the interlayers. Below transition temperature, protons are arranged in ordered pattern. The directions of lattice spontaneous polarization are the same for one molecular plane and opposite for neighbouring ones.

Experimental studies on SQA crystal have been carried out by many workers. Semmingsen and Feder[2] have made the birefringence study and measured temperature dependence of partial birefringence $\Delta n$ near transition. Crystal structure of SQA crystal at room temperature has been determined by x-ray diffraction by Semmingsen[7] and neutron diffraction by Stucky *et al*.[8]. Samara and Semmingsen[9] have studied pressure dependences of dielectric properties and transition temperature. Maier[5] *et al*. have carried out dielectric measurements and proton NMR in SQA crystal. Morimoto and Tokura[10] have performed high pressure Raman scattering in SQA crystal.

Deininghaus and Mehring[11] discussed phase transition in SQA crystal on the basis of three dimensionally coupled Ising chains. The Ising model is solved in an approximation treating the chains exactly and the interactions between them in mean field approximation. It is shown that the coupled chain approximation is better than the pure mean field approximation. With the weakly coupled chains of Ising spins, Muser[12] has used the monte carlo method to evaluate temperature dependence of order parameter of spin correlation function and of homogeneous susceptibility in SQA and compared with experiments. Zinenko[13] has used the cluster approximation with tunnelling of the protons to explain phase transition in SQA crystal. Li and Qin[14] have considered pseudospin model to 1 and 2 kinds of layers stacked alternatively along the *b*-axis. By using Green's function method, they evaluated soft mode frequency and the conditions of para- and antiferroelectric phases, order parameter, dispersion relation, internal energy and transition entropy.

In the present study, the pseudospin model for layers 1 and 2 of Lin and Qin[14] has been modified by adding pseudospin-lattice interaction term[15] and third-

and fourth-order phonon anharmonic interaction terms.[16,19]

By using the modified model and the method of double-time thermal Green's function[20], expressions for width, shift, renormalized soft mode frequency, transition temperature, dielectric constant and loss have been obtained. By fitting model values of physical quantities in the expressions, their temperature dependences have been calculated. The frequency and temperature dependences of dielectric loss in the frequency range (1-35 GHz) and temperature range (296-482K) at 10 GHz for SQA crystal have been calculated and compared with experimental results of Maier *et al*.[5], Muser[12] and Samera and Semmingsen[9].

## 2 Theory

### 2.1 Model Hamiltonian

For SQA crsytal, one of the molecular planes of squaric acid in which hydrogen bond is shared between two $C_4O_4$ radicals, one can assign two of the four neghbouring hydrogen bonds as belonging to the central $C_4O_4$ radical. It is sufficient to consider one of these two neighbouring hydrogen bonds only. As for layer-layer interactions, only the interaction of neighbouring layers is taken into account. The $H_{sp}$ and $H_{anh}$ terms are addodl in the present study to Li and Qin model $H_s$, so that the total model for SQA is expressed as :

$$H = H_s + H_{sp} + H_{anh} \qquad \ldots (1a)$$

with

$$H_s = -2\Omega \sum_i \left( S_{i1}^x + S_{j2}^x \right) - \sum_{ik} \left[ J_{ij} \left( S_{i1}^z S_{i1}^z + S_{i2}^z S_{i2}^z \right) + K_{ij} S_{i1}^z S_{i2}^z \right], \qquad \ldots (1b)$$

$$H_{sp} = -\sum_{ik} V_{ik} S_{1i}^z A_k - \sum_{ik} V_{ik} S_{2i}^z A_k^+ + \frac{1}{4} \omega_k \sum_k \left( A_k^+ A_k + B_k^+ B_k \right), \qquad \ldots (1c)$$

$$H_{anh} = \sum_{k_1 k_2 k_3} V^{(3)} \left( k_1, k_2, k_3 \right) A_{k_1} A_{k_2} A_{k_3} + \sum_{k_1 k_2 k_3 k_4} V^{(4)} \left( k_1, k_2, k_3 k_4 \right) A_{k_1} A_{k_2} A_{k_3} A_{k_4}, \qquad \ldots (1d)$$

where $\Omega$ is tunnelling frequency of proton, $S_i^x$ and $S_i^z$ are the $x$ and $z$ components of the pseudospin variable respectively. The indices 1 and 2 refer to the two layers; $J_{ij}$ is spin-spin exchange interaction constant within a given chain and $K_{ij}$ is the effective interaction constant between neighbouring chains, $V_{ik}$ is pseudospin-lattice interaction constant, $\omega_k$ is bare phonon frequency, $A_k$ and $B_k$ are position and momentum operators, $V^3$ and $V^4$ are Fourier transforms of the third- and fourth-order atomic force constants defined by Born and Huang[16].

### 2.2 Green's function, shift, width, soft mode frequency and dielectric constant

Following Zubarev[18], the Green's function (GF) is as follows:

$$G_{ij}(t - t') = \left\langle\left\langle S_{1i}^z(t); S_{1j}^z(t') \right\rangle\right\rangle = -i\theta(t-t') \left\langle \left[ S_{1i}^z(t); S_{1j}^z(t') \right] \right\rangle, \qquad \ldots (2)$$

Differentiating Green's function (Eq. 2) with respect to times $t$ and $t'$ respectively twice with the help of modified Hamiltonian (1a), Fourier transforming and writing in Dyson's equation form one obtains :

$$G_{ij1}^z(\omega) = \frac{\Omega \left\langle S_i^x \right\rangle}{\pi \left[ \omega^2 - \tilde{\omega}^2 - \mathrm{P}(\omega) \right]}, \qquad \ldots (3)$$

where

$$\tilde{\Omega}^2 = 4\Omega^2 + \frac{i \left\langle \left[ F, S_{j1}^y \right] \right\rangle}{\left\langle S_{1j}^x \right\rangle}, \qquad \ldots (4)$$

and

$$P(\omega) = \frac{\pi}{\Omega \left\langle S_{1i}^x \right\rangle} \left\langle\left\langle F(t); F(t') \right\rangle\right\rangle, \qquad \ldots (5)$$

with

$$\left\langle\left\langle F(t); F(t') \right\rangle\right\rangle = \left[ \left\langle\left\langle -2\Omega J_{ij} \left( S_{1i}^x S_{1i}^z + S_{1i}^z S_{1i}^x \right) - 2\Omega K_{ij} S_{1j}^x S_{1j}^z \right\}; \left\{ -2\Omega J_{ij} \left( S_{1j}^x S_{1j}^z + S_{1j}^z S_{1j}^x \right) - 2\Omega K_{ij} \left( S_{1j}^x S_{2j}^z \right) \right\} \right\rangle\right\rangle + 4\Omega^2 V_{ik}^2 \left\langle A_k A_k^+ \right\rangle \left\langle\left\langle S_{1j}^x; S_{1j}^x \right\rangle\right\rangle$$

$$+4\Omega^2 V_{ik}^2 \left\langle S_{1i}^x S_{1i}^x \right\rangle \left\langle\left\langle A_k ; A_{k'}^+ \right\rangle\right\rangle \Big] \quad \ldots (6)$$

In $\left\langle\left\langle F(t); F(t') \right\rangle\right\rangle$ there are higher order Green's functions, which are evaluated by decoupling them using scheme <abcd> = <ab> <cd> + <ac> <bd> + <ad> <bc>. The simpler Green's functions are then evaluated in zeroth-order approximation.

Substituting values of various Green's functions in Eq. (6) and then resolving $P(\omega)$ into its real and imaginary parts, one obtains shift $\Delta(\omega)$ and width ($\Gamma$) respectively. Green's function (Eq. (3)) finally becomes

$$G_{ij1}(\omega + i\in) \frac{\Omega \left\langle S_{1i}^x \right\rangle}{\pi \left[ \omega^2 - \hat{\Omega}^2 - 2i\Omega\Gamma(\omega) \right]}, \quad \ldots (7)$$

with

$$\hat{\Omega}^2 = \tilde{\tilde{\Omega}}^2 + \Delta_{s-p}(\omega), \quad \ldots (8)$$

$$\tilde{\tilde{\Omega}}^2 = \tilde{\Omega}^2 + \Delta_s(\omega), \quad \ldots (9)$$

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

$$a = (2J - K)\left\langle S_1^z \right\rangle, \quad \ldots (11)$$

$$b = 2\Omega, \quad \ldots (12)$$

$$c = (2\mathrm{J} - \mathrm{K})\left\langle S_{1i}^x \right\rangle, \quad \ldots (13)$$

$$\left\langle S_1^z \right\rangle = -\left\langle S_2^z \right\rangle \text{ for } T < T_c, \quad \ldots (14)$$

$$\left\langle S_1^z \right\rangle = \left\langle S_2^z \right\rangle \text{ for } T > T_c, \quad \ldots (15)$$

Shift and width are obtained as

$$\Delta_s(\omega) = \frac{a^4}{2\Omega\left(\omega^2 - \tilde{\Omega}^2\right)} + \frac{V_{ik}^2 N_k a^2}{2\Omega\left(\omega^2 - \tilde{\Omega}^2\right)}, \quad \ldots (16)$$

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

$$+ \frac{\pi V_{ik}^2 N_k a^2}{4\Omega\tilde{\Omega}} \left[ \delta\left(\omega - \tilde{\Omega}\right) - \delta\left(\omega + \tilde{\Omega}\right) \right], \quad \ldots (17)$$

$$\Delta_{sp}(\omega) = \frac{2V_{ik}^2 \left\langle S_{1i}^x \right\rangle \omega_k \delta_{kk'} \left(\omega^2 - \tilde{\tilde{\omega}}_k^2\right)}{\left[ \left(\omega^2 - \tilde{\tilde{\omega}}_k^2\right)^2 + 4\omega_k^2 \Gamma_k^2(\omega) \right]}, \quad \ldots (18)$$

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

In Eqs (18) and (19), $\tilde{\tilde{\omega}}_k$ and $\Gamma_k(\omega)$ are renormalized phonon frequency and phonon width respectively obtained in the evaluation of phonon Green's function $\left\langle\left\langle A_k ; A_{k'}^+ \right\rangle\right\rangle$. The phonon Green's function is obtained as[17-19] :

$$\left\langle\left\langle A_k ; A_{k'}^+ \right\rangle\right\rangle = \frac{\omega_k \delta_{kk'}}{\left[ \omega^2 - \tilde{\tilde{\omega}}_k^2 - 2i\omega_k \Gamma_k(\omega) \right]} \quad \ldots (20)$$

Phonon frequency is obtained as :

$$\tilde{\tilde{\omega}}^2 = \tilde{\omega}_k^2 + 2\omega_k \Gamma_k(\omega) \quad \ldots (21)$$

Phonon width in obtained as :

$$\Gamma_k(\omega) = 9\pi \sum_{k_1 k_2} \left| V^{(3)}(k_1, k_2, -k) \right|^2 \frac{\omega_{k_1} \omega_{k_2}}{\tilde{\omega}_{k_1} \tilde{\omega}_{k_2}}$$

$$\Big\{ \left(n_{k_1} + n_{k_2}\right) \Big[ \delta\left(\omega + \tilde{\omega}_{k_1} + \tilde{\omega}_{k_1}\right) - \left(\omega - \tilde{\omega}_{k_1} - \tilde{\omega}_{k_1}\right)$$

$$+ \left(n_{k_2} - n_{k_1}\right) \Big[ \delta\left(\omega + \tilde{\omega}_{k_1} + \tilde{\omega}_{k_2}\right) - \delta\left(\omega + \tilde{\omega}_{k_1} + \tilde{\omega}_{k_2}\right) \Big] \Big\}$$

$$+ 48\pi \sum_{k_1 k_2 k_3} \left| V^{(4)}(k_1, k_2, k_3, k_4) \right.$$

$$\times \Big\{ \left(1 + n_{k_1} n_{k_2} + n_{k_2} n_{k_3} + n_{k_3} n_{k_4}\right)$$

$$\times \Big[ \delta\left(\omega + \tilde{\omega}_{k_1} + \tilde{\omega}_{k_2} + \tilde{\omega}_{k_3}\right) - (\omega - \tilde{\omega}_{k_1} - \tilde{\omega}_{k_2} - \tilde{\omega}_{k_3}) \Big]$$

$$+ 3\left(n_{k_1} n_{k_2} + n_{k_2} n_{k_3} - n_{k_3} n_{k_4}\right)$$

$$\times \Big[ \delta\left(\omega + \tilde{\omega}_{k_1} - \tilde{\omega}_{k_2} - \tilde{\omega}_{k_3}\right) - \delta(\omega - \tilde{\omega}_{k_1} + \tilde{\omega}_{k_2} + \tilde{\omega}_{k_3}) \Big] \Big\}$$

+ higher terms. $\ldots (22)$

Phonon shift is obtained as:

$$\Delta_k(\omega) = 18P \sum_{k_1 k 2} \left| V^{(3)}(k_1 k_{2-k}) \right|^2$$

$$\frac{\omega_{k1}\omega_{k2}}{\tilde{\omega}_{k1}\tilde{\omega}_{k2}}\left\{\left(n_{k1}+n_{k2}\right)\frac{\tilde{\omega}_{k1}+\tilde{\omega}_{k2}}{\omega^2-\left(\tilde{\omega}_{k1}+\tilde{\omega}_{k2}\right)^2}\right.$$

$$\left.+\left(n_{k_2}+n_{k_1}\right)\frac{\tilde{\omega}_{k1}+\tilde{\omega}_{k2}}{\omega^2-\left(\tilde{\omega}_{k1}+\tilde{\omega}_{k2}\right)^2}\right\}$$

$$+48P\sum_{k1_1k_{2k_3}}\left|V^{(4)}\left(k_1k_2,k_{3,-k}\right)\right|^2\frac{\omega_{k_1}\omega_{k2}\omega_{k3}}{\tilde{\omega}_{k_1}\tilde{\omega}_{k_2}\tilde{\omega}_{k3}}$$

$$\left\{\left(1+n_{k_1}n_{k_2}+n_{k_2}n_{k_3}+n_{k_3}n_{k_1}\right)\frac{\tilde{\omega}_{k_1}+\tilde{\omega}_{k2}+\tilde{\omega}_{k3}}{\omega^2-(\tilde{\omega}_{k_1}+\tilde{\omega}_{k_2}+\tilde{\omega}_{k3})^2}\right.$$

$$+3\left(1-n_{k_2}n_{k_1}+n_{k2}n_{k_3}-n_{k_3}n_{k_1}\right)$$

$$\left.\frac{\tilde{\omega}_{k_1}+\tilde{\omega}_{k2}+\tilde{\omega}_{k3}}{\omega^2-(\tilde{\omega}_{k_1}+\tilde{\omega}_{k_2}+\tilde{\omega}_{k3})^2}+\text{higher terms}\right. \quad \ldots (23)$$

Solving Eq. (8) self consistently one obtains antiferroelectric soft mode frequency as :

$$\hat{\Omega}^2=\frac{1}{2}\left[\left(\tilde{\tilde{\Omega}}^2+\tilde{\tilde{\omega}}_k^2\right)\pm\left\{\left(\tilde{\tilde{\omega}}_k^2-\tilde{\tilde{\Omega}}^2\right)^2+8V_{ik}^2\left\langle S_{1i}^x\right\rangle\Omega\omega_k\right\}^{1/2}\right], \quad \ldots (24)$$

Soft mode frequency $\hat{\Omega}_-$ explicitly depends upon modified pseudospin frequency, $\tilde{\tilde{\Omega}}$ as well as renormalized phonon frequency $\tilde{\tilde{\omega}}_k$ .

By applying condition, $\hat{\Omega}_- \to 0$ as $T \to T_c$ gives

$$T_c=\frac{\Omega}{2k_B\tanh^{-1}\left(\frac{4\Omega}{\tilde{J}}\right)}, \quad \ldots (25)$$

where

$$\tilde{J}=\left(2J-K\right)+\frac{2V_{ik}^2\omega_k^2}{\tilde{\tilde{\omega}}_k^2}, \quad \ldots (26)$$

Eq. (25) shows that antiferroelectric transition temperature $T_c$ is explicit function of tunnelling frequency, $\Omega$, inter layer and intra-layer interactions, spin-lattice interaction constant as well as phonon anharmonic interactions.

### 2.3 Dielectric tangent loss

Following Kubo[21] and Zubarev[20] the dielectric susceptibility is expressed as :

$$\chi=-\lim_{x\to 0}2\pi\mathrm{N}\mu^2\mathrm{G}_{ij}\left(\omega+ix\right), \quad \ldots (27)$$

where $N$ is no of dipoles in sample each with dipole moment $\mu$

and the dielectric constant is related to $\chi$ as

$$\in=1+4\pi\chi \quad \ldots (28)$$

With the help of Eqs (27), (28) and (7) one obtains expression for dielectric constant as ($\in>>1$) :

$$\in=-\frac{8\pi\mathrm{N}\mu^2\Omega\left\langle\mathrm{S}_{ij}^x\right\rangle}{\left[\left(\omega^2-\hat{\Omega}_{\pm}^2-2\Omega i\Gamma(\omega)\right)\right]} \quad \ldots (29)$$

In ferroelectric and antiferroelectrics crystals usually $\in\rangle\rangle 1$ and so Eq. (28) may be easily approximated. The loss tangent is expressed as :

$$\tan\delta=\frac{\in''}{\in'} \quad \ldots (30)$$

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

With the help of Eqs (29) and (30) one obtains expression for loss tangent as

$$\tan\delta=-\frac{2\Omega\Gamma(\omega)}{\left(\omega^2-\hat{\Omega}^2\right)} \quad \ldots (31)$$

At microwave frequency $\omega<<\hat{\Omega}$, Eq. (31) reduces to

$$\tan\delta=\frac{2\Omega\Gamma(\omega)}{\hat{\Omega}^2} \quad \ldots (32)$$

### 2.4 Numerical calculation of shift, width and antiferroelectric mode frequency

By using model values[13,14] of physical quantities appearing in expressions, derived for SQA crystal, i.e. $\Omega$ =107cm$^{-1}$, $T_c$ =371K, $\tilde{J}$ =1086cm$^{-1}$, $V_{ik}$ = 64 cm$^{-1}$ $N$ = 35.59cm$^{-1}$ $\mu\times10^{18}$(cgs) = 4.86, $\omega_k$ = 100cm$^{-1}$, $C$ =18000K, (2J-K) = 416cm$^{-1}$, $A_k\times10^{17}$(ergK$^{-1}$) = 35. The temperature variations for $\left\langle S_1^z\right\rangle,\left\langle S_1^x\right\rangle$, shift, width and soft mode frequency are calculated using

Table 1 — Calculated values of $\langle S_1^z \rangle, \langle S_1^x \rangle$, and frequencies, $\tilde{\Omega}, \tilde{\tilde{\Omega}}$ and $\hat{\Omega}$ for squaric acid crystal

| $T$(K) | 296 | 315 | 334 | 353 | 371 | 390 | 408 | 427 | 445 | 464 | 482 | 501 |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| $\langle S_1^z \rangle$ | 0.250 | 0.247 | 0.241 | 0.206 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 |
| $\langle S_1^x \rangle$ | 0.433 | 0.438 | 0.446 | 0.475 | 0.500 | 0.483 | 0.461 | 0.441 | 0.421 | 0.404 | 0.380 | 0.360 |
| $\tilde{\Omega}$ ($cm^{-1}$) | 129 | 126 | 121 | 98 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 |
| $\tilde{\tilde{\Omega}}$ ($cm^{-1}$) | 152 | 140 | 131 | 112 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 |
| $\hat{\Omega}$ ($cm^{-1}$) | 182 | 174 | 168 | 158 | 0 | 38 | 46 | 51 | 55 | 59 | 62 | 66 |

Table 2 — Calculated values of shifts ($\Delta_1, \Delta_2, \Delta_3$) and widths ($\Gamma_1, \Gamma_2, \Gamma_3$) for squaric acid crystal

| $T$ (K) | 296 | 315 | 334 | 353 | 371 | 390 | 408 | 427 | 445 | 464 | 482 | 501 |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| $\Delta_1$($cm^{-1}$) | 34.32 | 33.75 | 33.16 | 27.26 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 |
| $\Delta_2$($cm^{-1}$) | 36 | 34 | 38 | 29 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 |
| $\Delta_3$($cm^{-1}$) | 14.08 | 11.12 | 8.14 | 5.06 | 1.32 | 2.29 | 2.30 | 2.33 | 2.35 | 2.73 | 3.0 | 3.6 |

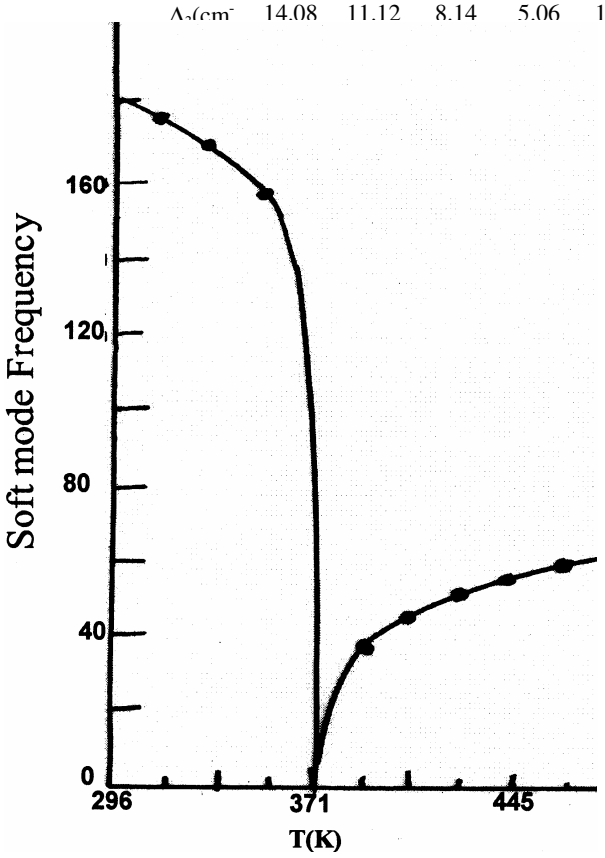


Fig. 1 — Temperature dependence of pseudospin lattice coupled soft mode frequency $\hat{\Omega}$ in SQA crystal (— Present calculation, ● Exp.[12])

respective equations and are presented in Tables 1 and 2. Variations of $\langle S_1^z \rangle$ and $\langle S_1^x \rangle$ are shown in Fig. 1.

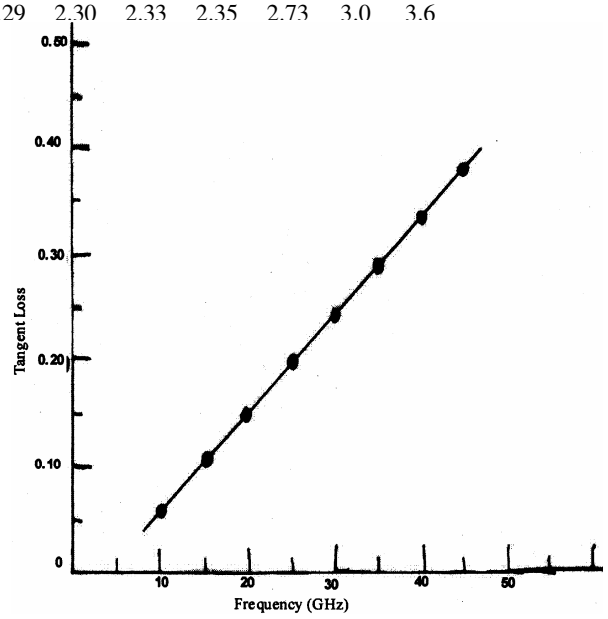


Fig. 2 — Frequency dependence of dielectric loss in SQA crystal (Present calculation —, Exp.[5,9] ●).

### 2.5 Frequency dependence of dielectric losses

Putting calculated values of $\Gamma(\omega)$ and $\hat{\Omega}$ for different temperatures into Eq. (31), dielectric loss is obtained for SQA crystals in 1-35 GHz range at 296K. The variation is shown in Fig. 2. The increase in loss is followed by an increase in frequency in SQA crystal.

### 2.6 Temperature dependence of dielectric loss

Using Eq. (31) and the calculated values from Tables 1 and 2, dielectric loss has been calculated in

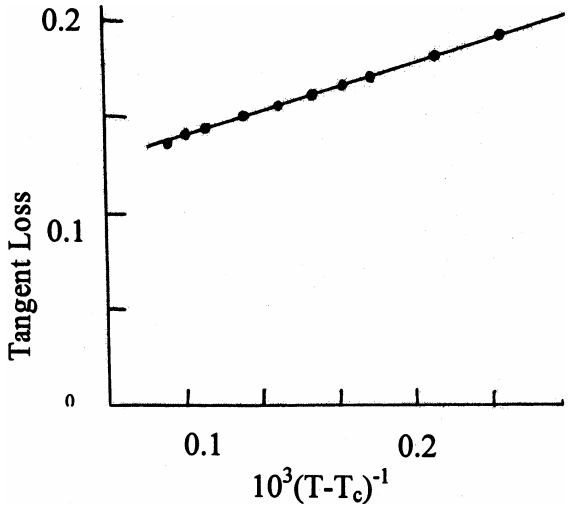


Fig. 3 — Temperature dependence of loss in SQA crystal (Present calculation —, Exp.[5,9] ●).

the temperature range (296-482K) at 10 GHz for SQA crystal. The loss versus $(T-T_c)^{-1}$ plot for SQA crystal is shown in Fig 3. The theoretical results are in fair agreement with experimental results of Muser[12], Maier *et al*.[5] and Samara and Semmingsen[9].

## 3 Conclusions

In the present study the modified pseudospin model for layers 1 and 2 coupled with phonon along with third-and fourth order phonon anharmonic interaction terms has been used to obtain expressions for shift, width, antiferroelectric soft mode frequency dielectric earlier workers constant and loss tangent. The method of statistical double-time thermal Green's function has been used for the evaluation have decoupled the correlations in the early stage and have not considered spin-phonon interaction and phonon anharmonic interaction terms. Therefore, they could not obtain better and convincing results to explain dielectric properties loss. in SQA crystal. In the present work, the correlations have been solved by decoupling them at proper stage. Hence all the important interactions could be contained in the expressions. Therefore, present results are much better and convincing, which agree with experimental results of Muser[12], Maier *et al*.[5] and Samara and Semmingsen[9]. If spin-lattice interaction and phonon anharmonic interaction terms and sifts and widths are neglected from our calculations, our results reduce to the expressions of earlier workers.[14] The earlier workers[14] could not explain tangent loss. The loss can be explained as follow. 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 Fig. 3 the loss shows Curie-Weiss behaviour, i.e. loss is proportional to $(T-T_c)^{-1}$ near antiferroelectric transition temperature.

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