

## By using PLCM model variation of ferroelectric properties of ammonium iron alum along with temperature

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Applying double-time thermal Green's function technique<sup>24</sup> and modifying pseudospin-lattice coupled mode (PLCM) model by adding third-and fourth-order phonon anharmonic interactions and extra spin-lattice interaction term dielectric properties of AFeSD alum have been studied. Expressions for shift, width, normal mode frequency, dielectric constant and loss tangent have been derived for AFeSD alum. Numerical calculations have been done. Theoretical results have been compared with experimental results of Pepinsky *et al.*<sup>25</sup>. Agreement has been found.

**Keywords:** Dielectric constant, PLCM model, Phase transition, Tangent loss.

### 1 Introduction

Ferroelectric materials have got potential applications in memory devices, transducers and infrared detectors. Some alums show ferroelectric properties. Liu *et al.*<sup>1</sup> have studied applications of ferroelectrics in photovoltaic devices. Ammonium iron alum  $(\text{NH}_4)\text{Fe}(\text{SO}_4)_2 \cdot 12\text{H}_2\text{O}$  is ferroelectric below 88K. Below 88K, it is cubic while above 88K it is paraelectric. In AFeSD alum  $\text{NH}_4^+$  group gives rise to order-disorder mechanism in proton subsystem linked with these groups. This is responsible for ferroelectric phase transition in alums. Due to order-disorder nature of the ammonium group in AFeSD, H-bonds associated with these groups undergo ordering. It is possible to apply pseudospin model similar to case of KDP, after suitable modification. The proton motion is associated with the "active" ammonium ion. In this crystal there are small isotope effect on  $T_c$  and C, pseudospin motion should be highly damped with strong anharmonic phonon interactions. Derby<sup>2</sup> has carried out crystal growth of AFeSD alums. Weber<sup>3</sup> has carried out experimental study of dielectric properties of AFeSD alum. Boujelben and Mihiri<sup>4</sup> have done Raman spectroscopic studies in AFeSD alum. Sachdeva *et al.*<sup>5</sup> have done experimental crystal growth studies on AFeSD alum.

Petrusevki<sup>6</sup> has carried out vibrational spectra of AFeSD, Selenate and other alums. Gu and Li<sup>7</sup> have

carried out special properties of AFeSD and potassium alum. Gu and Hao<sup>8</sup> have done spectral properties on AFeSD alum ammonium alum. Shaxin *et al.*<sup>9</sup> have studied application of AFeSD alum in inorganic synthesis. Bow *et al.*<sup>10</sup> have studied experimentally the adjuvant action of AFeSD alums. Earlier theoretical studies on AFeSD alums has been done by O'Reilly and Tsang<sup>11</sup>. Basara *et al.*<sup>12</sup> have studied electrical conductivity of some alums experimentally. Singh *et al.*<sup>13</sup> have studied physical properties of polymer mixed alums. Ghanem *et al.*<sup>14</sup> have studied experimentally the effect of doping different polymers on alums. Compbell and Debenedetti<sup>15</sup> have done Mossbauer study of AFeSD alum. They observed the phase transition of this alum about 88K. Abdeen *et al.*<sup>16</sup> have done x-ray and neutron diffraction on studies of alums. Chaudhury *et al.*<sup>17</sup> gave phenomenological explanation of the anomalous dielectric behaviour of alums with pseudo-spin-lattice coupled-mode model. Robert and Sambles<sup>18</sup> have carried out spin relaxation phenomena in ammonium ferric alum using Mossbauer spectroscopy. Mokoto and Kazuyuki<sup>19</sup> has carried out Luminescence from alums. Torgashev and Yuzyuk<sup>20</sup> have carried out Raman spectroscopy of alums. Venkatesh and Narayanan<sup>21</sup> have carried out spectroscopic studies of ferroelectric alums. Frost and Klopogge<sup>22</sup> have carried out Raman Microscopy study of AFeSD alum and related alums.

Thereafter Chaudhury *et al.*<sup>17</sup> have done theoretical study of AFeSD alum. They have applied pseudospin-lattice coupled-mode model. These authors have not

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considered third order phonon anharmonic interaction term. They have decoupled the correlations at an early stage. As a result some important interactions were disappeared from their calculations. In the present work, we shall modify pseudospin-lattice coupled mode model by adding the third-and fourth-order phonon anharmonic interactions and extra terms. By using double-time thermal Green's function method<sup>24</sup>, expressions for ferroelectric mode frequency, dielectric constant and loss tangent will be obtained. Theoretical results will be compared with experimental results of Pepinsky *et al.*<sup>25</sup>.

## 2 Modified Model Hamiltonian

For ferroelectric AFeSD alum, the Chaudhury *et al.*<sup>17</sup> have applied pseudospin-lattice coupled mode model. We shall add third-and fourth-order phonon anharmonic interaction terms:-

$$H_1 = -2\Omega \sum_i S_i^x - \frac{1}{2} \sum_{ij} J_{ij} S_i^z S_j^z + \frac{1}{4} \sum_{ik} \omega_k (A_k^+ A_k + B_k^+ B_k) - \sum_{ik} V_{ik} S_i^z A_k, \quad \dots (1)$$

we shall add:

$$H_2 = \sum_{ijk} V^{(3)}(k_1, k_2, k_3) A_{k_1} A_{k_2} A_{k_3} + \sum_{k_1 k_2 k_3 k_4} V^{(4)}(k_1 k_2 k_3 k_4) A_{k_1} A_{k_2} A_{k_3} A_{k_4} - \sum_{ik} V_{ik} S_i^x A_k, \quad \dots (2)$$

where,  $\Omega$  is proton tunnelling frequency between double well potential O-H...O bonds,  $S_i^x$  is x-component of the spin variables,  $J_{ij}$  is exchange interaction constant,  $S_i^z$  is z-component of pseudospin variable S,  $V_{ik}$  is spin- lattice interaction constant,  $A_k$  and  $B_k$  are position and momenta coordinates.  $V^3(k_1, k_2, k_3)$  and  $V^4(k_1, k_2, k_3, k_4)$  are the third and fourth order phonon anharmonic interactions terms. The third term in Eq. (2) describes an indirect coupling between the tunnelling motion of one proton and other proton. This term describes the modulation of distance between the two equilibrium sites in the O-H...O bonds. This modulates  $\Omega$  by non polar optic phonons. We shall consider the total Hamiltonian:

$$H = H_1 + H_2, \quad \dots (3)$$

for our study of MASD crystal.

## 3 Green's Function, Shifts and Widths

We consider the following Green's function given by Zubarev<sup>24</sup>:

$$G_{ij}(t-t') = \langle\langle S_i^z(t); S_j^z(t') \rangle\rangle = -i\theta(t-t') \langle [S_i^z(t), S_j^z(t')] \rangle \quad \dots (4)$$

Where  $\theta(t-t')$  is step function.  $\theta(t-t')=1$ , for  $t=t'$  and  $\theta(t-t')=0$ , for  $t=t'$ . The angular bracket  $\langle \dots \rangle$  denotes ensemble average over a grand canonical ensemble.

Differentiation of Green's function (4) with respect to time t using model Hamiltonian (3) and multiplying both sides by i we obtain:

$$i \frac{dG(t-t')}{dt} = \delta(t-t') \langle [S_i^z, S_j^z] \rangle + \langle\langle [S_i^z, H]; S_j^z(t') \rangle\rangle \quad \dots (5)$$

Hamiltonian (3) yields:

$$[S_i^z, H] = -2\Omega i \sum_i S_i^y \quad \dots (6)$$

We again differentiate Eq. (5) with respect to time t and obtain:

$$i^2 \frac{d^2 G(t-t')}{dt^2} = \delta(t-t') \langle [-2\Omega i S_i^y, S_j^z] \rangle + \langle\langle [-2\Omega i S_i^y, H]; S_j^z \rangle\rangle \quad \dots (7)$$

We obtain ,

$$[-2\Omega i S_i^y, H] = 4\Omega^2 S_i^z \quad \dots (8)$$

We now Fourier transforms Eq.(4) using formulae:

$$G(t-t') = \int_{-\infty}^{\infty} G(\omega) e^{-i\omega(t-t')} d\omega \quad \dots (9)$$

$$\frac{dG(t-t')}{dt} = (-i\omega) \int_{-\infty}^{\infty} G(\omega) e^{-i\omega(t-t')} d\omega \quad \dots (10)$$

$$\frac{d^2 G(t-t')}{dt^2} = -\omega^2 \int_{-\infty}^{\infty} G(\omega) e^{-i\omega(t-t')} d\omega \quad \dots (11)$$

We obtain,

$$\omega^2 G(\omega) = \frac{\Omega \langle S_{li}^x \rangle \delta_{ij}}{\pi} + \langle\langle F_i(t); S_j^z(t') \rangle\rangle + 4\Omega^2 G(\omega) \quad \dots (12)$$

Now we consider Green's function

$$\Gamma(t-t') = \langle\langle F_i(t); S_j^z(t') \rangle\rangle, \quad \dots (13)$$

Differentiating Eq.(13) with respect to time  $t'$  we obtain:

$$i \frac{d\Gamma(t-t')}{dt'} = -\delta(t-t') \langle [F_i(t), S_j^z(t')] \rangle + \langle\langle F_i(t); [H, S_j^z(t')] \rangle\rangle \quad \dots (14)$$

and,

$$[H, S_j^z(t')] = -2\Omega i S_j^y \quad \dots (15)$$

We differentiate Eq.(14) with respect to time  $t'$ :

$$i \frac{d^2 \Gamma(t-t')}{dt'^2} = \delta(t-t') \langle -2\Omega i S_j^y \rangle + \langle\langle F_i(t); [-2\Omega i S_j^y, H] \rangle\rangle \quad \dots (16)$$

we obtain:

$$[-2\Omega i S_j^y, H] = -2\Omega i S_i^y \quad \dots (17)$$

We fourier transform Eq(16) and obtain:

$$\omega^2 \Gamma(\omega) = 4\Omega^2 \Gamma(\omega) + \langle\langle F_i(t); F_j(t') \rangle\rangle \quad \dots (18)$$

Putting value of  $\Gamma(\omega)$  from Eq.(16) into Eq.(12) and writing the resulting equation in the form of Dyson's equation:

$$G(\omega) = G^0(\omega) + G^0(\omega)P(\omega)G(\omega) \quad \dots (19)$$

where unperturbed Green's function  $G^0(\omega)$  is given as:

$$G^0(\omega) = \frac{\Omega \langle S_i^x \rangle \delta_{ij}}{(\omega^2 - 4\Omega^2)} \quad \dots (20)$$

and ,

$$P(\omega) = \frac{\pi}{\Omega \langle S_i^x \rangle \delta_{ij}} \langle\langle F_i(t); F_j(t') \rangle\rangle \quad \dots (21)$$

We have from Eq(19) value of Green's function as:

$$G_{ij}(\omega) = \frac{\Omega \langle S_i^x \rangle \delta_{ij}}{\pi[\omega^2 - \hat{\Omega}^2 - \hat{P}(\omega)]} \quad \dots (22)$$

where,

$$G_{ij}^0(\omega) = \frac{\Omega \langle S_i^x \rangle}{\pi(\omega^2 - 4\Omega^2)} \quad \dots (23)$$

$$\hat{\Omega}^2 = (J_{ij} \langle S_j^z \rangle)^2 + 4\Omega^2 - 2\Omega J_{ij} \langle S^x \rangle \quad \dots (24)$$

and,

$$\hat{P}(\omega) = \frac{\pi \langle [F_i, B'] \rangle}{2\Omega^2 \langle S_i^x \rangle^2} + \frac{\pi^2 \langle\langle F_i, F_j \rangle\rangle}{\Omega^2 \langle S_i^x \rangle^2} \quad \dots (25)$$

Where,

$$F_i = -\Omega \sum_i \sum_{ij} J_{ij} S_i^x S_j^z - \Omega \sum_i \sum_{ij} J_{ij} S_i^z S_i^x - 2\Omega \sum_i \sum_{ik} V_{ik} A_k S_i^x + 4\Omega \sum_i \sum_{ik} V_{ik} A_k S_i^z - \frac{1}{2} \sum_{ik} \sum_{ij} V_{ik} A_k J_{ij} S_i^x S_j^z - \frac{1}{2} \sum_{ik} \sum_{ij} V_{ik} A_k J_{ij} S_i^z S_i^x - \sum_{ik} V_{ik}^2 A_k^2 S_i^x + \sum_{ik} V_{ik}^2 A_k^2 S_i^z \quad \dots (26)$$

The Green function ( GF ) can be now be written as:

$$G(\omega) = \frac{\Omega \langle S_i^x \rangle}{\pi[\omega^2 - \tilde{\Omega}^2 - \tilde{P}(\omega)]}, \quad \dots (27)$$

where,  $\tilde{\Omega}^2 = 4\Omega^2 + \frac{i}{\langle S_i^x \rangle} \langle [F_i(t), S_i^y(t')] \rangle, \dots (28)$

and,  $\tilde{P}(\omega) = \frac{\pi}{\Omega \langle S_i^x \rangle} \langle\langle F_i(t); F_j(t') \rangle\rangle. \dots (29)$

Solving Eq. (28) we obtain:

$$\tilde{\Omega}^2 = a^2 + b^2 - bc, \text{ where:} \quad \dots (30)$$

$$a = J \langle S_i^z \rangle$$

$$b = 2\Omega$$

$$c = J \langle S_i^x \rangle$$

In the denominator of Eq.(27),  $P(\omega)$  consists of higher order Green's functions. These are like  $\langle\langle AB;CD \rangle\rangle$ ,  $\langle\langle ABC;DEF \rangle\rangle$  etc. These can be evaluated using decoupling schemes. These are broken into simpler Green's functions, which are solved and substituted. In this way  $P(\omega)$  is evaluated. Now  $P(\omega)$  is separated into real and imaginary parts. Real part is called  $\Delta(\omega)$  while imaginary part is known as width  $\Gamma(\omega)$ . We obtain them as:

$$\begin{aligned} \Delta(\omega) = & \frac{a^4}{(\omega^2 - \tilde{\Omega}^2)} + \frac{b^2 c^2}{(\omega^2 - \tilde{\Omega}^2)} + \frac{V_{ik}^2 N_k a^2}{(\omega^2 - \tilde{\Omega}^2)} + \\ & \frac{2bV_{ik} \langle S_i^x \rangle \omega_k \delta_{kk'}}{(\omega^2 - \tilde{\omega}_k^2)} + \frac{4b^2 V_{ik}^2 N_k}{(\omega^2 - \tilde{\Omega}^2)} + \\ & \frac{8aV_{ik}^2 \langle S_i^z \rangle \omega_k \delta_{kk'}}{(\omega^2 - \tilde{\omega}_k^2)} + \frac{c^2 V_{ik}^2 N_k}{(\omega^2 - \tilde{\Omega}^2)} + \frac{V_{ik}^2 N_k a^4}{b^2 (\omega^2 - \tilde{\Omega}^2)} + \\ & \frac{2V_{ik}^4 N_k \langle S_i^x \rangle \omega_k \delta_{kk'}}{\Omega (\omega^2 - \tilde{\omega}_k^2)} + \frac{6V_{ik}^4 N_k \langle S_i^z \rangle \omega_k \delta_{kk'} a}{b\Omega (\omega^2 - \tilde{\omega}_k^2)} \end{aligned} \quad \dots (31)$$

and,

$$\begin{aligned} \Gamma(\omega) = & \frac{a^4}{2\Omega} \{ \delta(\omega - \tilde{\Omega}) - \delta(\omega + \tilde{\Omega}) \} + \frac{b^2 c^2}{2\tilde{\Omega}} \{ \delta(\omega - \tilde{\Omega}) - \delta(\omega + \tilde{\Omega}) \} \\ & + \frac{V_{ik}^2 N_k a^2}{2\tilde{\Omega}} \{ \delta(\omega - \tilde{\Omega}) - \delta(\omega + \tilde{\Omega}) \} + \frac{2bV_{ik} \langle S_i^x \rangle \omega_k \delta_{kk'}}{2\tilde{\omega}_k} \{ \delta(\omega - \tilde{\Omega}) - \delta(\omega + \tilde{\Omega}) \} \\ & + \frac{4b^2 V_{ik}^2 N_k}{2\tilde{\Omega}} \{ \delta(\omega - \tilde{\Omega}) - \delta(\omega + \tilde{\Omega}) \} + \frac{8aV_{ik}^2 \langle S_i^z \rangle \omega_k \delta_{kk'}}{2\tilde{\omega}_k} \{ \delta(\omega - \tilde{\Omega}) - \delta(\omega + \tilde{\Omega}) \} \\ & + \frac{c^2 V_{ik}^2 N_k}{2\tilde{\Omega}} \{ \delta(\omega - \tilde{\Omega}) - \delta(\omega + \tilde{\Omega}) \} + \frac{V_{ik}^2 N_k a^4}{2\tilde{\Omega} b^2} \{ \delta(\omega - \tilde{\Omega}) - \delta(\omega + \tilde{\Omega}) \} \\ & + \frac{2V_{ik}^4 N_k \langle S_i^x \rangle \omega_k \delta_{kk'}}{2\Omega \tilde{\omega}_k} \{ \delta(\omega - \tilde{\omega}_k) - \delta(\omega + \tilde{\omega}_k) \} \\ & + \frac{6V_{ik}^4 N_k \langle S_i^z \rangle \omega_k \delta_{kk'} a}{2b\Omega \tilde{\omega}_k} \{ \delta(\omega - \tilde{\omega}_k) - \delta(\omega + \tilde{\omega}_k) \} \end{aligned} \quad \dots (32)$$

In Eqs(31) and (32)  $\tilde{\omega}_k$  is modified phonon frequency while  $\Gamma_k(\omega)$  and  $\Delta_k(\omega)$  are phonon width and shift functions. These are obtained in the evaluation of phonon Green's function  $\langle\langle A_k; A_k^+ \rangle\rangle$  which appear in spin-lattice interaction term. We obtain:

$$\langle\langle A_k(t); A_k^+(t') \rangle\rangle = \frac{\omega_k \delta_{kk'}}{\pi[\omega^2 - \tilde{\omega}_k^2 - 2i\omega_k \Gamma_k(\omega)]} \quad \dots (33)$$

with,

$$\tilde{\omega}_k^2 = \tilde{\omega}_k^2 + 2\omega_k \Delta_k(\omega) \quad \dots (34)$$

The phonon shift  $\Delta_k(\omega)$  is obtained as:

$$\begin{aligned} \Delta_k(\omega) = & Re P_k(\omega) = 18P \sum_{k_1 k_2} |V^{(3)}(k_1, k_2, -k)|^2 \times \\ & \frac{\omega_{k_1} \omega_{k_2}}{\tilde{\omega}_{k_1} \tilde{\omega}_{k_2}} \left\{ \begin{aligned} & \left( n_{k_1} + n_{k_2} \right) \frac{\tilde{\omega}_{k_1} + \tilde{\omega}_{k_2}}{\omega^2 - (\tilde{\omega}_{k_1} + \tilde{\omega}_{k_2})^2} \\ & \left( n_{k_2} - n_{k_1} \right) \frac{\tilde{\omega}_{k_1} - \tilde{\omega}_{k_2}}{\omega^2 - (\tilde{\omega}_{k_1} - \tilde{\omega}_{k_2})^2} \end{aligned} \right\} \\ & + 48P \sum_{k_1 k_2 k_3} |V^{(4)}(k_1, k_2, k_3, -k)|^2 \frac{\omega_{k_1} \omega_{k_2} \omega_{k_3}}{\tilde{\omega}_{k_1} \tilde{\omega}_{k_2} \tilde{\omega}_{k_3}} \times \\ & \left\{ \begin{aligned} & \left( 1 + n_{k_1} n_{k_2} + n_{k_2} n_{k_3} + n_{k_3} n_{k_1} \right) \frac{\omega_{k_1} + \omega_{k_2} + \omega_{k_3}}{\omega^2 - (\tilde{\omega}_{k_1} + \tilde{\omega}_{k_2} + \tilde{\omega}_{k_3})^2} \\ & + 3 \left( 1 - n_{k_1} n_{k_2} + n_{k_2} n_{k_3} - n_{k_3} n_{k_1} \right) \frac{\omega_{k_1} - \omega_{k_2} - \omega_{k_3}}{\omega^2 - (\tilde{\omega}_{k_1} - \tilde{\omega}_{k_2} - \tilde{\omega}_{k_3})^2} \end{aligned} \right\} \\ & + \text{higher terms} \quad \dots (35) \end{aligned}$$

And the phonon width  $\Gamma_k(\omega)$  is:

$$\begin{aligned} \Gamma_k(\omega) = & Im P_k(\omega) = 9\pi \sum_{k_1 k_2} |V^{(3)}(k_1, k_2, -k)|^2 \times \\ & |V^{(4)}(k_1, k_2, k_3, k_4)|^2 \times \left[ \begin{aligned} & \delta(\omega + \tilde{\omega}_{k_1} + \tilde{\omega}_{k_2} + \tilde{\omega}_{k_3}) \\ & - \delta(\omega - \tilde{\omega}_{k_1} - \tilde{\omega}_{k_2} - \tilde{\omega}_{k_3}) \end{aligned} \right] \\ & + 48\pi \sum_{k_1 k_2 k_3} \left[ \begin{aligned} & \left( 1 + n_{k_1} n_{k_2} + n_{k_2} n_{k_3} + n_{k_3} n_{k_1} \right) \frac{\omega_{k_1} \omega_{k_2} \omega_{k_3}}{\tilde{\omega}_{k_1} \tilde{\omega}_{k_2} \tilde{\omega}_{k_3}} \\ & + \text{higher terms} \end{aligned} \right] \quad \dots (36) \end{aligned}$$

The Green's function (4) finally takes value:

$$G_{ij}^{(\omega)} = \frac{\Omega \langle S_i^x \rangle \delta_{ij}}{\pi[\omega^2 - \hat{\Omega}^2 - 2\Omega i\Gamma(\omega)]} \quad \dots (37)$$

where,  $\hat{\Omega}^2 = \tilde{\Omega}^2 + \Delta(\omega)$  ... (38)

**4 Ferroelectric Mode Frequency**

Cochran<sup>23</sup> has suggested that at transition temperature, some of normal vibrational mode becomes very small so that phase transition occurs. If we solve Eq.(38), we obtain:

$$\hat{\Omega}^2 = \frac{1}{2} \left( \tilde{\omega}_k^2 + \tilde{\Omega}^2 \right) \pm \frac{1}{2} \left[ \left( \tilde{\omega}_k^2 - \tilde{\Omega}^2 \right)^2 + 4 \left( \frac{2bV_{ik} \langle S_i^x \rangle \omega_k \delta_{kk'} + 8aV_{ik}^2 \langle S_i^z \rangle \omega_k \delta_{kk'}}{\Omega} + \frac{6V_{ik}^4 N_k \langle S_i^z \rangle \omega_k \delta_{kk'} a}{b\Omega} \right) \right]^{1/2} \quad \dots (39)$$

The frequency  $\hat{\Omega}_-$  corresponding to negative sign is the ferroelectric mode frequency.

**5 Dielectric Constant**

We may obtain formula for dielectric constant  $\epsilon$  as:

$$\epsilon = 1 + 4\pi\chi \quad \dots (40)$$

**6 Curie Temperature**

At transition or Curie temperature ( $T_c$ )  $\tilde{\Omega} \rightarrow 0$ , the value of  $T_c$  becomes:

$$T_c = \frac{\Omega}{2k_B \tanh^{-1} \left( \frac{4\Omega}{J} \right)} \quad \dots (41)$$

where,

$$J = J + \frac{2V_{ik}^2 \omega_k^2}{\tilde{\omega}_k^2} \quad \dots (42)$$

**7 Dielectric Constant**

Following Zubarev<sup>24</sup> we have:

$$\chi = -\lim_{\epsilon \rightarrow 0} 2\pi N \mu^2 G_{ij}(\omega + i\epsilon), \dots (43)$$

$\chi$  is related to dielectric constant  $\epsilon$  as:

$$\epsilon = 1 + 4\pi\chi$$

$$= (1 + 4\pi) \lim_{x \rightarrow 0} -2\pi N \mu^2 G_{ij}(\omega + ix) \quad \dots (44)$$

since in ferroelectrics  $\epsilon \gg 1$ . So Eq.(44) becomes:

$$\epsilon = \frac{-8\pi^2 N \mu^2 \langle S_i^x \rangle \delta_{ij}}{\pi[\omega^2 - \hat{\Omega}^2 - 2\Omega i\Gamma(\omega)]} \quad \dots (45)$$

**8 Loss Tangent**

In ferroelectrics or dielectric substances some power is lost in the form of heat. This loss of power is called loss tangent ( $\tan\delta$ ), given by:

$$\tan \delta = \frac{\text{imaginary}\epsilon}{\text{real}\epsilon} \quad \dots (46)$$

From Eq.(45), it becomes:

$$= \frac{2\Omega\Gamma(\omega)}{(\omega^2 - \hat{\Omega}^2)} \quad \dots (47)$$

**9 Numerical Calculations and Results**

By fitting model values of various physical quantities, given in Table -1 for AFeSD alum we have calculated temperature dependence of ferroelectric mode frequency, dielectric constant and loss tangent for AFeSD alum (Fig. 1-3). The results have been compared with experimental data of Pepinsky *et al.*<sup>25</sup> for dielectric measurements for AFeSD alum and correlated data for ferroelectric mode frequency. It can be observed that theoretical results agree with experimental data.

Table 1 — Model values of physical quantities for AFeSD alum:

$\Omega(\text{cm}^{-1})$	$J(\text{cm}^{-1})$	$J^*(\text{cm}^{-1})$	$V_{ik}(\text{cm}^{-1})$	$T_c(\text{K})$	$\omega_k(\text{cm}^{-1})$	$C(\text{K})$	$\mu * 10^{18}(\text{cgs})$	$A_k * 10^{17}(\text{erg/K})$
0.17	177.61	128	5.12	88	5	425	1.405	0.05

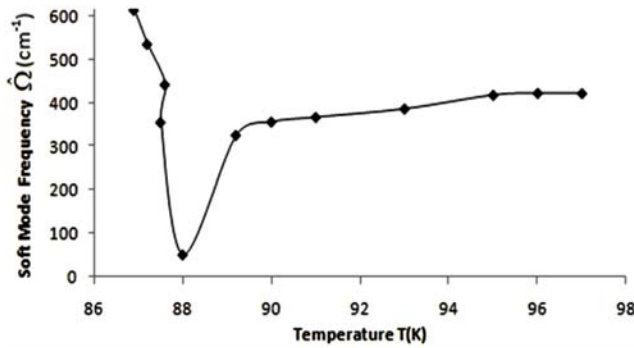


Fig. 1 — Temperature dependence of soft mode frequency  $\hat{\Omega}$  ( $\text{cm}^{-1}$ ) of AFeSD alum (— Our calculation,  $\blacklozenge$  correlated with experimental values of Pepinsky *et al.*<sup>25</sup>).

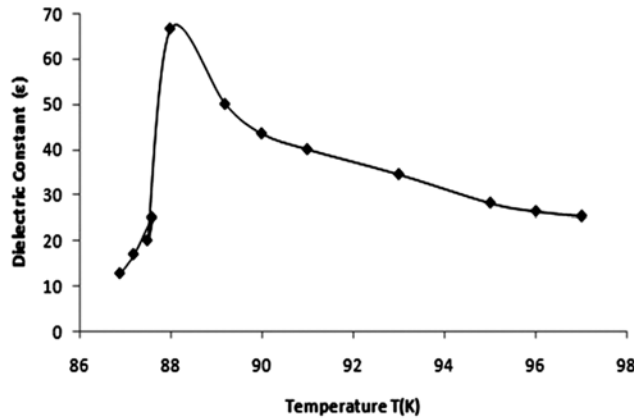


Fig. 2 — Temperature dependence of dielectric constant ( $\epsilon$ ) of AFeSD alum (— Our calculation,  $\blacklozenge$  correlated with experimental values of Pepinsky *et al.*<sup>16</sup>).

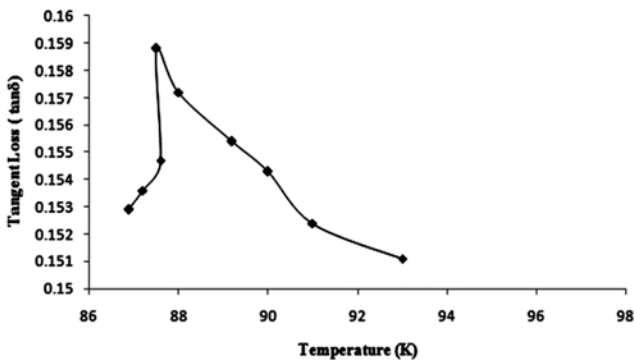


Fig. 3 — Temperature dependence of loss tangent ( $\tan\delta$ ) of AFeSD alum (— Our calculation,  $\blacklozenge$  correlated with experimental values of Pepinsky *et al.*<sup>16</sup>).

## 10 Discussion

In the present work we have considered pseudospin-lattice coupled mode model along with third-and fourth-order phonon anharmonic interactions terms as well as extra spin lattice terms. Unlike previous authors

we have not decoupled correlations at an early stage. So that all possible interactions could be included in the results. Our Eq.(39) shows that ferroelectric mode frequency decreases from below as temperature is increased. It becomes small at  $T_c$  and then increases. This is quite in agreement with experimental observations. Our Eq.(45) shows that dielectric constant  $\epsilon$  first increases from below if we increase temperature. It becomes largest at  $T_c$  and then decreases. This agrees with experimental observations. Our Eq.(41) shows that if temperature is increased from below, loss tangent increases becoming largest at  $T_c$  then it decreases. This behaviour is in accordance with experimental results.

## 11 Conclusions

It is for the conclusion that pseudospin-lattice coupled mode model along with third-and fourth-order phonon anharmonic interactions explains ferroelectric transition and dielectric properties of AFeSD alum. Present formulae may also be used to explain nature of other alums like AFeSD, ACrSD, ANdSD alums.

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## References

- 1 Liu Y, Wang S, Chen Z & Xiao L, *Sci China Mat*, 59 (2016) 851.
- 2 Derby J J, *14<sup>th</sup> Int Summar School on Crystal Growth (ISSIG-141, Dalian, China, Aug (2010) 1.*
- 3 Weber H J, *Zeits fur kvistall*, 149 (2010) 269.
- 4 Boujelben M & Mihiri T, *Int J Spectrosc*, 2011 (2011) 12840.
- 5 Sachdeva, *Mater Technol*, 4 (2013) 467.
- 6 Petrusovski V M, *Maced J Chem Chem Eng*, 34 (2015) 73.
- 7 Gu H & Li Y, *Spectrochim Acta Part A*, 139 (2015) 342.
- 8 Gu H & Hao X, *Spectrochim Acta Part A*, 146 (2015) 273.
- 9 Shaxin Y, *Org Synth*, 17 (2005) 27.
- 10 Bow M & Ennio G L M, *Ulmer Jeffrey B, Nature Medicine*, 17 (2011) 415.
- 11 O' Reilly D E & Tsang T, *Phys Rev*, 157 (1967) 417.
- 12 Basara S M A, Iqbal Z, Rahman K, Rahman H & Ejaz M F, *J Appl Res Tech*, 12 (2014) 560.
- 13 Singh P K, Bhattacharya B, Sachdeva A & Singh R, *Mater Technol*, 47 (2013) 467.

- 14 Ghanem H M E, Saquam S, Saadi M & Abdul Zawad S, *J Mod Phys*, 2 (2011) 1533.
- 15 Compbell L & Debenedetti S, *Phys Lett*, 20 (1966) 102.
- 16 Abdeen A M, Will G, Schater W, Kirtel A, Bargouth M O & Recker K, *Zeits Fur Krist*, 157 (1981)147.
- 17 Chaudhury K R, Nath D, Banerjee S & Chaudhuri B K, *Phys Rev B*, 26 (1982) 6276.
- 18 Roberts J M & Sambles, *J Phys Chem Solids*, 45 (1984) 937.
- 19 Mokoto M & Kazuyuki M, *J Phys Soc Jpn*, 38 (1975) 1048.
- 20 Torgashev V I, Yuzuk Y U I, Kirpichnikova L F & Shuvalov L A, *Ferroelectrics*, 1101 (1989)13.
- 21 Venkatesh G M & Narayanan P S, *J Ind Inst Sci*, 52 (1970) 2133.
- 22 Frost R L & Kloprogge J T N, *Jb Miner Mh Jg*, 1 (2001) 27.
- 23 Cochran W, *Phil Mag Suppl*, 10 (1961) 401.
- 24 Zubarev D N, *Sov Phys Usp*, 3 (1960) 320.
- 25 Pepinsky R, Jona F & Shirane G, *Phys Rev*, 102 (1956) 1181.