INTERNATIONAL JOURNAL FOR RESEARCH

IN APPLIED SCIENCE & ENGINEERING TECHNOLOGY

Substitutional Effect on Dielectric Losses in Polycrystalline $Ca_xSr_{1-x}TiO_3$ and $Pb_xSr_{1-x}TiO_3$ Ferro-Electric Mixed Crystals

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Abstract: The Dielectric losses are theoretical investigated for $Ca_xSr_{1-x}TiO_3$ and $Pb_xSr_{1-x}TiO_3$ single crystal perovskites in para-electric phase as a function of composition and temperature. In our calculation, the Silverman Joseph`s Hamiltonian augmented with fourth order phonon co-ordinates using double time temperature dependent Green`s Function technique is used. Dielectric loss is estimated for various value of x. The substitutional impurity depend dielectric losses increases with increases in composition x at room temperature and Dielectric loss tangent (tan δ) increases with increasing temperature at constant values of x.

Keywords: Dielectric losses, Single crystal, Perovskites, anharmonicity, Retarted Green's function, Hamiltonian.

I. INTRODUCTION

Strontium titanate $(SrTiO_3)$ST is one of the most interesting and more studied member of ABO_3 perovskites family due to its strong ferro-electricity, semi-conductivity, low dielectric losses ,used in super conductivity and potential applications in many devices. It is well known that the ferro-electricity in ABO_3 results from the delicate balance between the short-range repulsions favouring cubic structure, and the long-range coulomb interactions favouring tetragonal structure in the case of ST. It is of interest to find out how foreign ions will influence the losses in ST. As an example, divalent (Pb or Ca) may be substituted for divalent strontium in ST. Lattice parameter and curie temperature vary almost linearly with composition.

The investigation of the dielectric properties provides an important approach in understanding inter and intra molecular interactions models of the motion and conformational change in the macromolecules. The temperature defect and frequency dependence of dielectric loss in ferroelectric perovskites has been the subject of considerable interest due to their extensive use in various applications.

It is very interested to study physical properties of mixed crystals, as it helps in understanding basic mechanism of mixed crystal formation which finds interesting applications. The dependence of physical properties of mixed crystals varies from system to system. The variations in the properties may be linear or non- linear. Also it is very well known that several interesting temperature dependent properties of ferroelectrics results from the temperature dependence of low lying transverse optic mode of vibration[1-3]. The applications of the perovskites lies in the field of ceramic industry, memory display, optical communication, holographic storage media, etc.

Microwave losses in displacive ferroelectrics (BT,ST etc.) have been reported experimentally (Rupprecht and Bell 1961, 1962; Rupprecht et al 1961)[4].The microwave losses in both pure and doped $SrTiO_3$ have been investigated as a function of frequency and temperature. It is found that the loss tangent is proportional to frequency in the range 3-36 kMc/sec. (Rupprecht and Bell 1961, 1962; Rupprecht et al 1962) [5]. In the low-frequency range at room temperature Linz[6] has reported a loss tangent of $SrTiO_3$ which is independent of frequency in the range between 10^2 and 10^7 cps. The microwave losses in mixed and pure $SrTiO_3$ crystals have been experimentally measured as a function of frequency and temperature by Hung SC et al [7],Zhang L et al [8], Kukreti et al [9].

A Linz and K. Herrington [10] measured microwave losses in pure $CaTiO_3$ with temperature. Dielectric losses of $Ca_xSr_{1-x}TiO_3$ has been measured as a function of temperature and frequency for compositions by G.-F. Zhang et al [11]

Kyoung-T. Kim and Chang-I Kim has been measured Dielectric losses for $Pb_xSr_{1-x}TiO_3$ (PST) as a function of Pb/Sr compositions at constant frequency [12]. Dielectric losses of $Pb_xSr_{1-x}TiO_3$ (PST) has been measured experimentally and theoretically as a function of temperature and frequency by Y. P. Jiang et al[13], Kyoung-Tae Kim et al [14], Yoshita Somiya et al[15].

Electric field, Frequency and Temperature dependent Microwave losses of pure $PbTiO_3$, $KNbO_3$ ferroelectric perovskites have been theoretically calculated in para-electric phase from the Silverman –Joseph Hamiltonian augmented with fourth order phonon coordinates using Double time temperature Green`s function by Talvinder Singh et al[16]

Microwave loss obeys the Curie-Weiss law (Deorani S .C. et al [2]). This may be taken as a direct evidence for the temperature dependence of the soft mode frequency. At transition temperature, soft mode frequency tends to zero and lattice displacement associated with this mode becomes unstable. This explains the anomalous behavior of the dielectric loss near the phase transition. So theoretically it is now clear that absorption of microwave is not due to the absorption or creation of single phonon.

Energy and momentum cannot be simultaneously conserved in the process, since the microwave photon is negligible ($\omega/\Omega=10^{-3}$) compared to that of an excited phonon. It is possible, however, to excite a virtual phonon which subsequently decay into a real phonon due to interactions with lattice imperfections. Imperfection simply play the role of absorbing the excess momentum of the phonons and decay into other vibrational mode of the crystal is also possible.

In our previous paper [17-19] we have discussed variation of dielectric constant and soft mode of $Ca_xSr_{1-x}TiO_3$ and $Pb_xSr_{1-x}TiO_3$ mixed crystals with impurity concentration and temperature.$Ca_xSr_{1-x}TiO_3$ (CST) and $Pb_xSr_{1-x}TiO_3$ (PST) solid solutions are basic materials for microwave devices.

The aim of the present work is to discuss the variation of microwave loss tangent (tanδ) with concentration of impurity and temperature in $Ca_xSr_{1-x}TiO_3$ and $Pb_xSr_{1-x}TiO_3$ mixed crystals. Using Green`s function method, dielectric losses are theoretically calculated in para-electric phase and the results are compared with the results available elsewhere.

II.   THEORY

A. Hamiltonian and Green's function

The Hamiltonian which includes the anharmonicity upto fourth -order in the potential energy due to interaction of soft mode coordinates, resonant interaction and scattering terms are considered. The impurities introduced are characterized by different mass than the host atoms and with modified nearest neighbour harmonic force constants around their sites. The influence on the anharmonic coupling coefficients in the Hamiltonian, being small is neglected. The modified Hamiltonian of a mixed displasive perovskite, in para-electric phase which includes defects (substitutional impurity),third and fourth order anharmonicity and higher order electric moment term are used in present study and is exactly similar as used earlier[20] is given

H' = H + H$_D$ …….   (1)

Where H is Hamiltonian for pure crystal and H$_D$ is the contribution by the defect in Hamiltonian which involves the effect of mass change and harmonic force constant change between the impurity and host lattice atoms due to substitutional defects.

Where

$H = \sum_k \frac{\hbar\omega_k^a}{4}(A_k^{a+}A_k^a + B_k^{a+}B_k^a) + \sum_k' \frac{\hbar\omega_k^0}{4}(A_k^{0+}A_k^0 + B_k^{0+}B_k^0) - \frac{\hbar\omega_0^0}{4}(A_0^{0+}A_0^0 + B_0^0 B_0^0) + \sum_k \hbar F(k)A_0^0 A_k^{0+}A_k^a + \sum_k \hbar\beta^a(k)A_0^{02}A_k^{0+}A_k^0 +$

$\sum_k \hbar\beta^0(k)A_0^{02}A_k^{a+}A_k^a + \sum_{k_1 k_2 k_3}\hbar\varphi(k_1 k_2 k_3)A_0^0 A_{K_1}^0 A_{k_1}^a A_{k_3}^a + \sum_{k_1 k_2 k_3}\hbar\psi(k_1 k_2 k_3)A_0^0 A_{K_1}^0 A_{k_1}^0 A_{k_3}^0 + \hbar V A_0^{04} + \hbar E\{-\alpha A_0^0 +$

$\sum_k A(k)A_k^a A_k^{0+} + \sum_{k,\lambda}B^\lambda(k)A_0^0 A_k^\lambda A_k^+ + \sum_{k_1 k_2 k_3}C(k_1,k_2,k_3)A_{K_1}^0 A_{k_2}^a A_{k_3}^a + \sum_{k_1 k_2 k_3}D_1(k_1,k_2,k_3)A_{K_1}^0 A_{k_2}^0 A_{k_3}^0 - D_1(0,0,0)A_0^{03}\}$ =H$_1$ + H$_2$ + H$_0$ + H$_3$ +…………H$_{14}$,   ……..(2)

Where

$F(k) = \frac{P(k)}{\sqrt{N}}.\frac{1}{(\omega_k^0 \omega_k^a)^{\frac{1}{2}}}$ ……………………..(2a)

$\beta^a(k) = \frac{G^0(k)}{N}.\frac{1}{\omega_k^0}$ …………………………(2b)

$\beta^0(k) = \frac{G^a(k)}{N}.\frac{1}{\omega_k^a}$ ………………………………………(2c)

$\varphi(k_{1,}k_2,k_3) = \frac{1}{\sqrt{N}}\Upsilon(k_1,k_2,k_3)\frac{\hbar^{\frac{1}{2}}}{(\omega_{k_1}^0 \omega_{k_2}^a \omega_{k_3}^a)^{\frac{1}{2}}}$ ………………………….(2d)

$\psi(k_{1,}k_2,k_3) = \frac{1}{\sqrt{N}}\mu(k_1,k_2,k_3)\frac{\hbar^{\frac{1}{2}}}{(\omega_{k_1}^0 \omega_{k_2}^0 \omega_{k_3}^0)^{\frac{1}{2}}}$ ………………………..(2e)

International Journal for Research in Applied Science & Engineering Technology (IJRASET)
ISSN: 2321-9653; IC Value: 45.98; SJ Impact Factor: 7.538
Volume 10 Issue XII Dec 2022- Available at www.ijraset.com

$A(k) = \frac{A'(k)}{(\omega_k^0 \omega_k^a)^{\frac{1}{2}}}$ ………………………………(2f)

$B^\lambda(k) = \frac{B'^\lambda(K)}{\sqrt{N}} \cdot \frac{1}{\omega_k^\lambda}$ ……………………………………..(2g)

$C(k_1, k_2, k_3) = C'(k_1, k_2, k_3) \frac{\hbar^{\frac{1}{2}}}{(\omega_{k_1}^0 \omega_{k_2}^a \omega_{k_3}^a)^{\frac{1}{2}}}$ …………………………………..(2h)

$D(k_1, k_2, k_3) = D'(k_1, k_2, k_3) \frac{\hbar^{\frac{1}{2}}}{(\omega_{k_1}^0 \omega_{k_2}^0 \omega_{k_3}^0)^{\frac{1}{2}}}$ ……………..(2i)

$V = \Psi(0,0,0)$ …………………………………………………… (2j)
$D_1' = D_1(0,0,0)$ ……………………………………… (2K)

In the above equations $G^0(k)$ and $G^a(K)$ are the Fourier transforms of 3rd order anharmonic force constants, $\Upsilon(K_1, K_2, K_3)$ and $\mu(K_1, K_2, K_3)$ are the Fourier transformed 4th order anharmonic force constants. (α); $A(K)$, $B^\lambda(K)$ and $C'(K_1, K_2, K_3)$, $D'(K_1, K_2, K_3)$ represent the linear, second and third-order electric moment co-efficients respectively. The primed sum in H_2 is taken for $k \neq 0$.
H_D is the contribution by the defect in Hamiltonian which involves the effect of mass change and harmonic force constant change between the impurity and host lattice atoms due to substitutional defects and is given by[21]

$H_D = -\hbar C(0,0) B_0^0 B_0^0 + \hbar D(0,0) A_0^0 A_0^0 - \hbar B_0^0 X + \hbar A_0^0 Y + \hbar Z,$ …………………………….(3)

With

$X = \sum_{k,\lambda} C(k^\lambda, 0) B_k^\lambda$ ……………………………………………(3a)

$Y = \sum_{k,\lambda} D(k^\lambda, 0) A_k^\lambda$ …………………………………(3b)

And

$Z = \sum_{k_1,k_2,\lambda} [D(k_1^\lambda, k_2^\lambda) A_{k_1}^\lambda A_{k_2}^\lambda - C(k_1^\lambda, k_2^\lambda) B_{k_1}^\lambda B_{k_2}^\lambda]$
$+ \sum_{k_1,k_2} [D(k_1^a, k_2^0) A_{k_1}^a A_{k_2}^0 - C(k_1^a, k_2^0) B_{k_1}^a B_{k_2}^0]$ ………………………(3c)

Here $\lambda = a, 0$ are for acoustic and optic modes respectively. The defect parameters $C(k_1, k_2)$ and $D(k_1, k_2)$ depend upon the changes in the mass and force constants due to the substitutional defects respectively and are given by

$C(k_1, k_2) = \frac{1}{2\mu}(M_0/2N)(\omega_{k_1} \omega_{k_2})^{\frac{1}{2}} e(k_1) e(k_2)$

$\times \{\sum_\ell^n f \exp.[i(k_1 + k_2).R(\ell)] - \sum_\ell^n \exp.[i(k_1 + k_2).R(\ell)]\}$ …………………….(3d)

and

$D(k_1, k_2) = \frac{1}{4N}(\omega_{k_1} \omega_{k_2})^{-\frac{1}{2}} \times \sum_{\ell\alpha\ell\beta} [\Delta\phi_{\alpha\beta}(\ell\ell')/M_0] \times e(k_1) e(k_2)$

$\times \exp.[i\{k_1.R(\ell) + k_1.R(\ell')\}],$ ……………………(3e)

Where $e(k)$ is the polarization vector, $R(\ell)$ the equilibrium position vector of the ℓ-th atom,
$C(k_1, K_2)$ vanishes when n is either zero or N. $\Delta\varphi$ denotes the force constant change, ℓ and ℓ' refer to the impurity and its nearest neighbours and $\mu = [M M'/(M'-M)]$. M_0 is the weighted harmonic mean of the masses of all atoms and is defined by the relation
$\frac{1}{M_0} = \frac{f}{M'} + \frac{1-f}{M}$ ………(3f)

With $f = \frac{n}{N}$. Here N is the total number of atoms in the crystal whose (N-n) lattice sites are occupied by atoms of mass M while n sites are occupied by randomly distributed substitutional impurities each of mass M'.
In order to get the effect of defect on soft mode frequency, we transform the Hamiltonian H' as given by Naithani et. Al[21]. The transformed Hamiltonian is obtained as
$H_T = H + H_D + \hbar\omega_0^0 g A_0^0$ ……………………………………………………..(4)
Where $g = \frac{\alpha}{\omega_0^0}$.
The retarded Green's function $G_0^0(t-t')$ for optic phonon is defined as
$G_0^0(t-t') = \langle\langle A(t); A(t')\rangle\rangle_\omega + i\varepsilon$ .... (5)
or $G_0^0(\omega + i\varepsilon) = G'(\omega) - iG''(\omega)$ .... (6)

International Journal for Research in Applied Science & Engineering Technology (IJRASET)

ISSN: 2321-9653; IC Value: 45.98; SJ Impact Factor: 7.538
Volume 10 Issue XII Dec 2022- Available at www.ijraset.com

The real (ε ') and imaginary parts (tan δ) of dielectric constant are related to Green's function as

$\varepsilon'(\omega) - 1 = -8 \pi^2 N\mu^2 G'(\omega)$ .... (7)

and

$\tan \delta = G''(\omega) / G'(\omega)$ .... (8)

The equation of motion for Green's function is

$i\hbar \frac{d}{dt}G(t, t') = \hbar \frac{d}{dt}\theta(t - t') < [A(t), B(t')] > + << [A(t), H_T]; B(t') >>$ ....(9)

Using this equation of motion, modified Hamiltonian, Fourier transforming and shaping in Dyson's equation form, we get

$G_0^0(\omega + i\varepsilon) = \frac{\omega_0^0}{\pi[\omega^2 - v^2(\omega) + i\Gamma_0^0(\omega)]}$ ....(10)

Here $v^2(\omega) = v_0^2(\omega) + \Delta(v_D^2(\omega))$ .... (10a) where $v_0^2(\omega) = -(\omega_0^0)^2 + 4\omega_0^0 \bar{Q} + \Delta_0(\omega)$ .... (10b)

and

$\Delta(v_D^2(\omega)) = 4\omega_0^0 D(0,0) + 4\omega_0^0 \sum_{k,\lambda} D^2(k_\lambda, 0)[\tilde{\omega}_k^\lambda / \omega^2 - (\tilde{\omega}_k^\lambda)^2] - 4(\omega^2/\omega_0^0) \sum_{k,\lambda} C^2(k_\lambda, 0)[\tilde{\omega}_k^\lambda / \omega^2 - (\tilde{\omega}_k^\lambda)^2] + 4\omega \sum_{k,\lambda} C(k_\lambda, 0)D^*(k_\lambda, 0) \times [\tilde{\omega}_k^\lambda / \omega^2 - (\tilde{\omega}_k^\lambda)^2] - 4\omega \sum_{k,\lambda} C^*(k_\lambda, 0) D(k_\lambda, 0) [\tilde{\omega}_k^\lambda / \omega^2 - (\tilde{\omega}_k^\lambda)^2] + 96V^2 \times \omega_0^0 ([1+3(N_0^0)^2]*3\Omega / \omega^2 - (3\Omega)^2 - [1-((N_0^0)^2)]*\Omega / \omega^2 - (3\Omega)^2)$ ... (10c)

The notations used here are in the same sense as used by Yadav et al[20] and Naithani et al[21].

Temperature dependence of $v^2(\omega)$ can be written as

$v^2(\omega) = -(\omega_0^0)^2 + \gamma_1 T + \gamma_2 T^2 + \Delta(v_D^2)$ ...(11)

Where $\Delta_0(\omega)$ (shift in phonon frequency corresponds to pure crystal, $\Delta(v_D^2(\omega))$ is temperature independent part due to defect and γ_1 and γ_2 are temperature dependent parts in $v^2(\omega)$ and depend on anharmonic force- constant and electric dipole moment terms. Thus from equation (11), we conclude

$\frac{v^2(\omega)}{\gamma_1} = - \frac{(\omega_0^0)^2}{\gamma_1} + \frac{\gamma_1}{\gamma_1}T + \frac{\Delta(v_D^2(\omega))}{\gamma_1} + \frac{\gamma_2 T^2}{\gamma_1}$

$\frac{v^2(\omega)}{\gamma_1} = (T - T_c' + \xi T^2)$ …... (11a)

Where $T_c' = -\frac{(\omega_0^0)^2}{\gamma_1} + \frac{\Delta(v_D^2(\omega))}{\gamma_1}$ and $\xi = \frac{\gamma_2}{\gamma_1}$ (non linearity constant).

Equation (11a) can be reduced now as

$v^2(\omega) = \gamma_1(T - T_c' + \xi T^2)$ ………….(12)
or $v^2(\omega) \alpha (T - T_c' + \xi T^2)$ …………….. (13)
Here $T_c' = T_c + \Delta(T_c)$ is the new Curie-temperature in presence of defect impurity.

Here, $\Delta(T_c) = -\frac{\Delta(v_D^2(\omega))}{\gamma_1}$ …………. (13a)

Thus T_c is one of the parameters which is very sensitive to impurity (x).The above results show that T_c varies linearly with x.
Equation 13 shows that the change in Curie temperature depends on substitutional impurity. $\Delta(v_D^2(\omega))$ (temperature independent part due to defect) and γ_1 (anharmonic coupling constant) and hence ΔT_c is a function of mass change due to defect and anharmonic constants.

Dielectric Loss
Using equation (7),(8),(9) and (10),the expression for Loss tangent is obtained as
$\tan \delta(\omega) = -\Gamma_o^o(\omega) / v^2(\omega)$ ....(14)
Naithani et al [22] has given
$\Gamma^o(\omega) = a + bT + cT^2$ ....(15)
Using equation (12), we get
$\gamma_1(T - T_c' + \xi T^2) \tan \delta = a + bT + cT^2$
or $(T - T_c' + \xi T^2) \tan \delta = \alpha + \beta T + \Upsilon T^2$ ....(16)

Where $\alpha = \frac{a}{\gamma_1}$, $\beta = \frac{b}{\gamma_1}$, $\Upsilon = \frac{c}{\gamma_1}$, and ξ =is very small $\approx 10^{-5}$ for SrTiO₃ (Panwar Thesis [23])

Equation (16) gives the dielectric loss in mixed polycrystal. The parameter α depends on the impurity contents (Ca and Pb) in the anharmonic crystal and is ($\alpha = 0$) zero for a pure single crystal. The parameters β and Υ, which are related to third and fourth order anharmonic terms in the interionic potential, are shown to be intrinsic properties of the perfect lattice and unaffected by imperfections.

International Journal for Research in Applied Science & Engineering Technology (IJRASET)

ISSN: 2321-9653; IC Value: 45.98; SJ Impact Factor: 7.538
Volume 10 Issue XII Dec 2022- Available at www.ijraset.com

In the present paper, we are interested in losses (i.e. α) due to impurities and temperature. How α changes with the composition x in $Ca_xSr_{1-x}TiO_3$ and $Pb_xSr_{1-x}TiO_3$.

The value of ξ is very small order thus the equation (16) can be approximated as

$(T-T_c')$ tan δ= α+βT+ϒT^2 ....(17)

Or $\quad$ tanδ=$\frac{α+βT+γT^2}{T-T_c'}$ ....(18)

$\quad\quad$ (tanδ)$_{DT}$ = $\frac{α}{T-T_c'}$ + $\frac{βT+γT^2}{T-T_c'}$ ....(19)

$\quad\quad$ (tanδ)$_{DT}$ = (tanδ)$_D$+(tanδ)$_T$ ....(19a)

Or $\quad\quad$ (tanδ)$_D$ = (tanδ)$_{DT}$ -(tanδ)$_T$ ....(19b)

Here , (tanδ)$_{DT}$ is loss tangent due to defect and temperature variation, (tanδ)$_D$ is loss due to defect only, (tanδ)$_T$ is loss due to temperature variation.

III. CALCULATIONS

A. Dielectric Losses Due To Defect With Composition X

Using the equations (19) and (19b) the dielectric loss for $Ca_xSr_{1-x}TiO_3$ (CST) and $Pb_xSr_{1-x}TiO_3$ (PST) mixed crystals with defect compositions (x=0.0,0.2,0.4,0.6,0.8) in the para-electric phase at the Room Temperature are calculated . The parameters α, β, γ and T_c for $Ca_xSr_{1-x}TiO_3$ (CST) and $Pb_xSr_{1-x}TiO_3$ (PST) with corresponding values of x (= 0.0,0.2,0.4,0.6,0.8) have been calculated from Fig.(2) of Arther Linz et al [6] , Fig. (6) of G. F. Zhang et.al.[11] and Fig (6) of Kyoung-T. Kim et al [12] by best fit of data. The curie temperature of $Ca_xSr_{1-x}TiO_3$ (CST) and $Pb_xSr_{1-x}TiO_3$ (PST) have been taken from our previous paper [19,24].

With the help of α, β, γ, T_c and equations (19) and (19b), we have calculated dielectric losses at room temperature due to defect are given in table 1 and table 2 and are shown in fig. 1 and Fig 2.

Table1. Impurity dependent dielectric loss v/s composition for $Ca_xSr_{1-x}TiO_3$ at Room Temperature

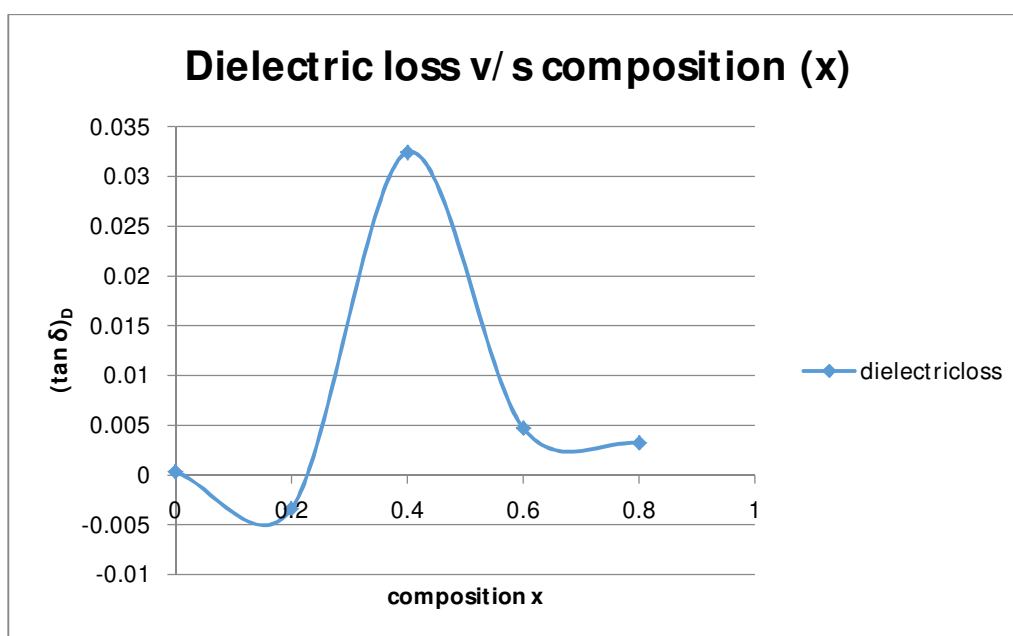| X	T_c	(Tanδ)$_D$
0.0	37	0.000327155
0.2	178.2	-0.003370763
0.4	316.5	0.032426557
0.6	460.7	0.004724344
0.8	602	0.003204701

Fig.1. Impurity dependent dielectric loss as a function of impurity composition for $Ca_xSr_{1-x}TiO_3$ at Room Temperature

Table2. Impurity dependent dielectric loss v/s composition for $Pb_xSr_{1-x}TiO_3$ at Room Temperature

x	T_c	$(Tan\delta)_D$
0.0	37	0.000327155
0.2	182.2	0.011917197
0.4	327	0.037366503
0.6	472.6	0.037367907
0.8	617.1	0.038746382
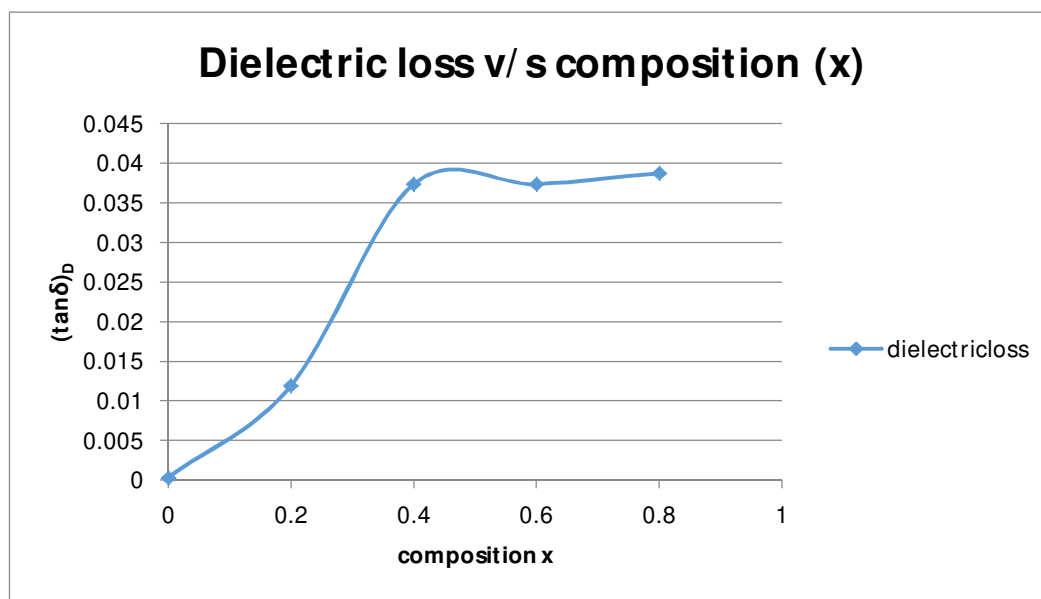
Fig.2. Impurity dependent dielectric loss as a function of impurity composition for $Pb_xSr_{1-x}TiO_3$ at Room Temperature

B. Variation of Dielectric Loss With Temperature

With the help of α, β, γ, T_c and equation (19), we have calculated dielectric losses as a function of temperature for different value of composition (x) are given in table (3a) to (3e) for $Ca_xSr_{1-x}TiO_3$ and table (4a) to (4e) for $Pb_xSr_{1-x}TiO_3$ and are shown in Fig. 3 and Fig.4 respectively.

Table 3(a) Dielectric loss (tan δ) v/s temperature (T) of $SrTiO_3$, T_c=37K, x=0.0

T	760	770	780	790	800	810	820	830	840	850	860
tan δ	0.00533	0.0054	0.0055	0.0055	0.0056	0.0056	0.0057	0.0058	0.0058	0.0059	0.006

Table 3(b) Dielectric loss (tan δ) v/s temperature (T) of $Ca_{0.2}Sr_{0.8}TiO_3$, T_c=178.2K, x=0.2

T	760	770	780	790	800	810	820	830	840	850	860
tan δ	0.0058	0.0059	0.0059	0.006	0.0061	0.0061	0.0062	0.0063	0.0063	0.0064	0.0065

Table 3(c) Dielectric loss (tan δ) v/s temperature (T) of $Ca_{0.4}Sr_{0.6}TiO_3$, T_c=316.5K, x=0.4

T	760	770	780	790	800	810	820	830	840	850	860
tan δ	0.0071	0.0072	0.0073	0.0073	0.0074	0.0075	0.0075	0.0076	0.0076	0.0077	0.0078

Table 3(d) Dielectric loss (tan δ) v/s temperature (T) of $Ca_{0.6}Sr_{0.4}TiO_3$, T_c=460.7K, x=0.6

T	760	770	780	790	800	810	820	830	840	850	860
tan δ	0.0068	0.0068	0.0069	0.007	0.007	0.0071	0.0072	0.0073	0.0073	0.0074	0.0075

International Journal for Research in Applied Science & Engineering Technology (IJRASET)
ISSN: 2321-9653; IC Value: 45.98; SJ Impact Factor: 7.538
Volume 10 Issue XII Dec 2022- Available at www.ijraset.com

Table 3(e) Dielectric loss (tan δ) v/s temperature (T) of $Ca_{0.8}Sr_{0.2}TiO_3$, T_c=602K, x=0.8

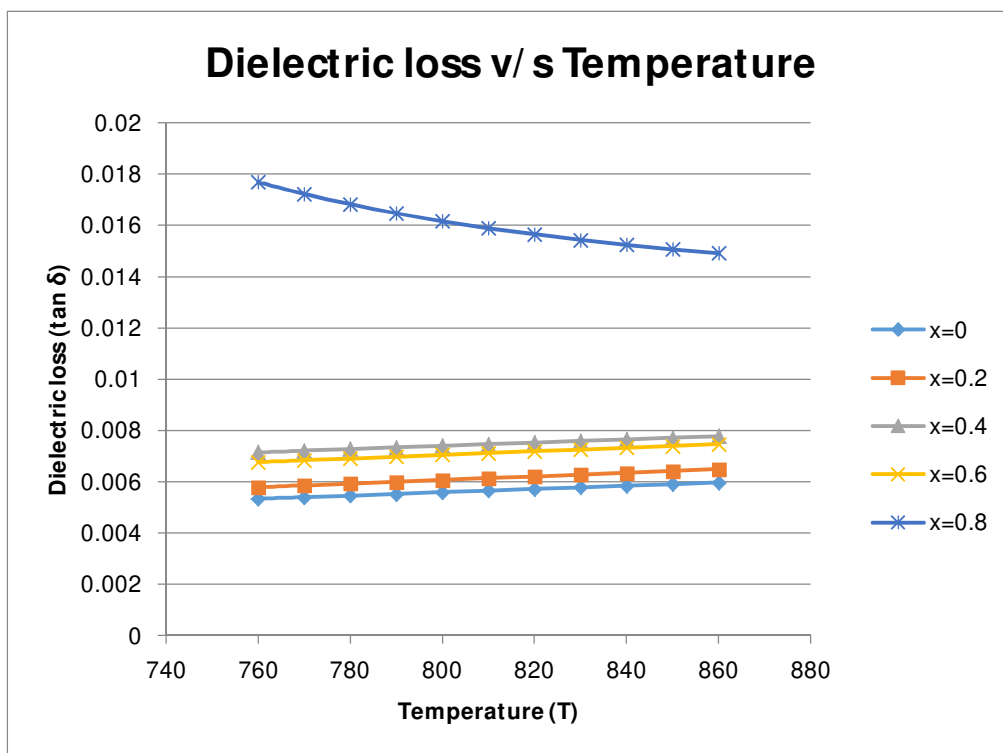| T	760	770	780	790	800	810	820	830	840	850	860
tan δ	0.0177	0.0172	0.0168	0.0165	0.0162	0.0159	0.0157	0.0154	0.0152	0.0151	0.0149

Fig.3.Variation of Dielectric loss as a function of temperature (T) for $Ca_xSr_{1-x}TiO_3$ at constant value of x

Table 4(a) Dielectric loss (tan δ) v/s temperature (T) of $SrTiO_3$, T_c=37K, x=0.0

T	760	770	780	790	800	810	820	830	840	850	860
tanδ	0.0053	0.0054	0.0055	0.0055	0.0056	0.0056	0.0057	0.0058	0.0058	0.006	0.006

Table 4(b) Dielectric loss (tan δ) v/s temperature (T) of $Pb_{0.2}Sr_{0.8}TiO_3$, T_c=182.2K, x=0.2

T	760	770	780	790	800	810	820	830	840	850	860
tanδ	0.0088	0.0089	0.0089	0.0089	0.0089	0.009	0.009	0.009	0.009	0.009	0.0091

Table 4(c) Dielectric loss (tan δ) v/s temperature (T) of $Pb_{0.4}Sr_{0.6}TiO_3$, T_c=327K, x=0.4

T	760	770	780	790	800	810	820	830	840	850	860
tanδ	0.0061	0.0061	0.0062	0.0063	0.0064	0.0065	0.0066	0.0066	0.0067	0.007	0.0069

Table 4(d) Dielectric loss (tan δ) v/s temperature (T) of $Pb_{0.6}Sr_{0.4}TiO_3$, T_c=472.6K, x=0.6

T	760	770	780	790	800	810	820	830	840	850	860
tanδ	-0.0062	-0.0059	-0.0055	-0.005	-0.005	-0.005	-0.004	-0.004	-0.004	-0.003	-0.003

Table 4(e) Dielectric loss (tan δ) v/s temperature (T) of $Pb_{0.8}Sr_{0.2}TiO_3$, T_c=617.1K, x=0.8

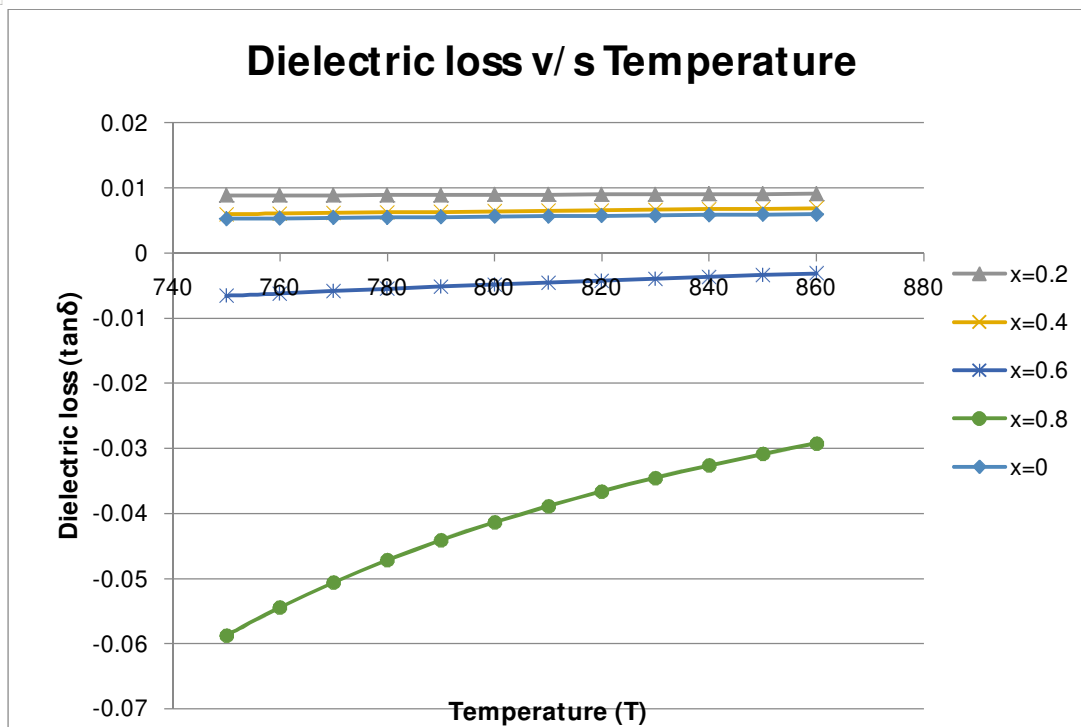| T	760	770	780	790	800	810	820	830	840	850	860
tanδ	-0.0544	-0.0506	-0.0472	-0.044	-0.041	-0.039	-0.037	-0.0345	-0.033	-0.031	-0.029

International Journal for Research in Applied Science & Engineering Technology (IJRASET)
ISSN: 2321-9653; IC Value: 45.98; SJ Impact Factor: 7.538
Volume 10 Issue XII Dec 2022- Available at www.ijraset.com

Fig.4.Variation of Dielectric loss as a function of temperature (T) for $Pb_xSr_{1-x}TiO_3$ at constant value of x

IV.   DISCUSSION

The calculated values show the comparative variation of loss tangent with defect, frequency

and temperature in $Ca_xSr_{1-x}TiO_3$ and $Pb_xSr_{1-x}TiO_3$ mixed crystals.The dielectric loss tangent depends upon anharmonic, resonsnt interaction and scattering terms due to defects.In the present study, the Hamiltonian proposed by Pytte (1970) has been designed in terms of creation and annihilation operators. To evaluate the higher order correlation functions, the renormalized Hamiltonian has been evaluated using the Green's function technique and Dyson's equation. At microwave frequencies the results are in good agreement with the experimental results.

Curie - Weiss law behaviour of microwave losses may be taken as a direct evidence for the temperature dependence of the polarisation mode frequency. At transition temperature, soft mode frequency tends to zero and lattice displacement associated with this mode becomes unstable. This explains the anomalous behaviour of the dielectric loss near the phase transition. At very high temperatures, loss tangent deviates from Curie-Weiss law due to non linear parameter ξ. The change in Tc caused by impurity, depends upon the change in harmonic force constant between the impurity host lattice atoms mass change due to impurity and canbe negative or positive. In mixed crystals, the major contribution to the loss orginates from the impurity part α in comparision to second and third term i.e. β and Υ dependent terms (or third & fourth order anharmonic terms).

Figure 1 and Figure 2 shows the variations of Impurity dependent dielectric loss as a function of impurity composition for $Ca_xSr_{1-x}TiO_3$ and $Pb_xSr_{1-x}TiO_3$ mixed crystals at Room Temperature. It is very clear from these figures that dielectric loss increases with the increases in the defects (impurity composition Ca or Pb) but in $Ca_xSr_{1-x}TiO_3$ loss tangent decreses after x=0.4.

Figure 3 and Figure 4 shows the variations of the dielectric loss as a function of temperature in $Ca_xSr_{1-x}TiO_3$ and $Pb_xSr_{1-x}TiO_3$ for different concentration   x (x=0.0-0.8) in para electric phase at (1-100) KHz. The dielectric losses increases when temperature increase. The variation in dielectric loss for all values of x are similar trend above the curie temperature. The results of temperature and composition dependence of the dielectric losses are good qualitative and close agreements with experimental and theoretical results of others.[2,6,10-16]

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