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Theoretical Study of 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 theoretically investigated for $Ca_xSr_{1-x}TiO_3$ and $Pb_xSr_{1-x}TiO_3$ mixed crystal perovskites in para-electric phase as a function of composition and temperature. In our calculation, we used the Silverman Joseph`s Hamiltonian augmented with fourth order phonon co-ordinates using double time temperature dependent Green`s Function technique. Dielectric loss is calculated for various value of x (x=0.0 to 1.0)). The substitutional impurity dependent dielectric losses $(\tan \delta)_D$ increases with increases in composition x and Dielectric loss tangent (tan δ) decreases with increasing temperature at constant values of x. In the para-electric phase, the dielectric loss decreases with increasing temperature showing the Curie-Weiss behavior of the tangent loss.

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

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I. Introduction

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

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. The applications of the perovskites lies in the field of ceramic industry, memory display, optical communication, holographic storage media, etc. 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 (Sr) divalent strontium in ST. Lattice parameter and Curie temperature vary almost linearly with composition.

The lattice defects in the ferroelectric materials greatly influence their static dielectric constant in para-electric phase. Various impurities change the Curie-temperature of the same material in different ways. In our previous papers we have discussed that The Curie-temperature of $SrTiO_3$ increases on the substitution of Ca or Pb ions for Sr [1-2]. The analogous effect is felt by dielectric loss.

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[3-5].

Using Green`s function method, microwave losses in displacive ferroelectrics ($BaTiO_3$, $SrTiO_3$ $KaTiO_3$) have been reported theoretically and experimentally (S.C.Deorani et.al[4]; Rupprecht and Bell 1961, 1962; Rupprecht et al 1961)[6-7]. A Linz and K. Herrington [8] measured microwave losses in pure $CaTiO_3$ with temperature. 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[9].

The microwave losses in both pure and doped $SrTiO_3$ have been investigated as a function of frequency and temperature by Hung SC et al [10], Zhang L et al [11], Kukreti et al [12]

Dielectric losses of $Ca_xSr_{1-x}TiO_3$ has been measured as a function of temperature and frequency for different composition values x by G.-F. Zhang et al [13]

Dielectric losses for $Pb_xSr_{1-x}TiO_3$ (PST) has been investigated as a function of Pb/Sr compositions at constant frequency by Kyoung-T. Kim and Chang-I Kim [14]. 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[15], Kyoung-Tae Kim et al [16], Yoshita Somiya et al [17].

In the para-electric phase, the dielectric losses deceases with increasing temperature obeys the Curie-Weiss law of dielectric loss (Deorani S .C. et al [4]). 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 [1-2,18-19] we have discussed variation of Curie temperature ,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.

In the present work, we have theoretically studied functional dependence of dielectric 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

Hamiltonian and Green's function:

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 and the value of H and H_D are similar as used in our previous paper[21].

In order to get the effect of defect on soft mode frequency, we transform the Hamiltonian H' as given by Naithani et. al [22]. The transformed Hamiltonian is obtained as

$H` = H + H_D + \hbar\omega_0^0 g A_0^0$ ..... (2)

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') =<<A(t); A(t')>>ω + iε ..... (3)

or G_0^0 (ω+ iε) = G'(ω)−iG"(ω) ..... (4)

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

$\varepsilon'(\omega) - 1 = -8\pi^2 N\mu^2 G'(\omega)$ ..... (5)

and

$\tan\delta = G''(\omega)/G'(\omega)$ ..... (6)

The equation of motion for Green's function is

$i\hbar\frac{d}{dt}G(t, t') = \hbar\frac{d}{dt}\theta(t - t') < [A(t),B(t')] > + << [A(t),H_T];B(t') >>$ .....(7)

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

$G_0^0(\omega+i\varepsilon) = \frac{\omega_0^0}{\pi[\omega^2-\nu^2(\omega)+i\Gamma_0^0(\omega)]}$ .....(8)

Here $\nu^2(\omega)=\nu_0^2(\omega)+\Delta(\nu_D^2(\omega))$ ..... (8a)

where $\nu_0^2(\omega)=-(\omega_0^0)^2 + 4\omega_0^0\bar{Q}+\Delta_0(\omega)$ ..... (8b)

and

$\Delta(\nu_D^2(\omega)) =4\omega_0^0 D(0,0) + 4\omega_0^0\sum_{k,\lambda} D^2(k_\lambda 0)[\tilde{\omega}_k^{\lambda}/\omega^2-(\tilde{\omega}_k^{\lambda})^2] - 4(\omega^2/\omega_0^0)\sum_{k,\lambda} C^2(k_\lambda 0)[\tilde{\omega}_k^{\lambda}/\omega^2-(\tilde{\omega}_k^{\lambda})^2]+ 4\omega\sum_{k,\lambda} C(k_\lambda 0)D^*(k_\lambda 0)\times[\tilde{\omega}_k^{\lambda}/\omega^2-(\tilde{\omega}_k^{\lambda})^2] - 4\omega\sum_{k,\lambda} C^*(k_\lambda 0) D(k_\lambda 0)[\tilde{\omega}_k^{\lambda}/\omega^2-(\tilde{\omega}_k^{\lambda})^2] +96V^2\times\omega_0^0([1+3(N^0_0)^2]*3\Omega/\omega^2-(3\Omega)^2-[1-((N^0_0)^2)]*\Omega/\omega^2-(3\Omega)^2)$ … (8c)

The notations used here are in the same sense as used in our previous paper[21] and Naithani et al[22].

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

$\nu^2(\omega)=-(\omega_0^0)^2+\gamma_1 T+\gamma_2 T^2+\Delta(\nu_D^2)$ …(9)

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

Thus from equation (11), we conclude

$\frac{\nu2(\omega)}{\gamma_1}=--\frac{(\omega_0^0)2}{\gamma_1}+\frac{\gamma_1}{\gamma_1}T+\frac{\Delta(\nu_D^2(\omega))}{\gamma_1}+\frac{\gamma_2 T^2}{\gamma_1}$

$\frac{\nu2(\omega)}{\gamma_1}=(T-T_c'+\xi T^2)$ …..... (9a)

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

Equation (11a) can be reduced now as

$\nu^2(\omega)=\gamma_1(T-T_c'+\xi T^2)$ …….(10)

or $\nu^2(\omega)\ \alpha\ (T-T_c'+\xi T^2)$ …….. (11)

Here $T_c'=T_c+\Delta(T_c)$ is the new Curie-temperature in presence of defect impurity.

Here, $\Delta(T_c)=-\frac{\Delta(\nu_D^2(\omega))}{\gamma_1}$ ……. (11a)

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 (11) shows that the change in Curie temperature depends on substitutional impurity. $\Delta(\nu^2_D(\omega))$ (temperature independent part due to defect) and γ_1 (anharmonic coupling constant) and hence $\Delta T_c$is a function of mass change due to defect and anharmonic constants.

Dielectric Loss

Using equation (5),(6),(7) and (8),the expression for Loss tangent is obtained as

$\tan\delta(\omega)=-\Gamma_o^o(\omega)/\nu^2(\omega)$ .....(12)

Naithani et al [23] has given

$\Gamma^o(\omega)=a+bT+cT^2$ .....(13)

Using equation (10),(12),(13) we get

$\gamma_1(T-T_c'+\xi T^2)\tan\delta=a+bT+cT^2$

or $(T-T_c'+\xi T^2)\tan\delta=\alpha+\beta T+\Upsilon T^2$ .....(14)

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 $SrTiO_3$ ((Panwar Thesis [24])

Equation (14) gives the dielectric loss in mixed crystal. The parameter α depends on the impurity contents (Ca and Pb) in the anharmonic crystal and is (α=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.

In the present paper, we are interested to calculated dielectric losses (i.e. α) due to impurities and temperature and 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 (14) can be approximated as

$(T-T_c')\tan\delta=\alpha+\beta T+\Upsilon T^2$ .....(15)

Or $\tan\delta=\frac{\alpha+\beta T+\gamma T^2}{T-T_c'}$ .....(16)

$(\tan\delta)_{DT}=\frac{\alpha}{T-T_c'}+\frac{\beta T+\gamma T^2}{T-T_c'}$ .....(17)

$(\tan\delta)_{DT}=(\tan\delta)_D+(\tan\delta)_T$ .....(17a)

Or $(\tan\delta)_D=(\tan\delta)_{DT}-(\tan\delta)_T$ .....(17b)

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

III. Calculations

3.1 Dielectric losses due to defect with composition x

Using the equations (17) and (17b) 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 755 K are calculated . Parameters β and γ for CT are obtained from Fig. (4) of A. Linz and K. Herrington, [8] and for PT from fig (3) of Talvinder Singh et.al.[9] by best fit of data and these parameters for ST are reported by G. Rupprecht and R. O. Bell [7]. A summary of these parameters along with T_c for these crystals are given in table 1.

Table 1. Parameters β, ϒ and T_c for $CaTiO_3$ (CT), $PbTiO_3$ (PT) and $SrTiO_3$ (ST)

Parameters	CT	PT	ST
β	-1.91×10^{-5}	8.794×10^{-6}	6.53×10^{-4}
ϒ	$5.54 \times 10^{-8} K^{-1}$	-1.043×10^{-9}	$2.54 \times 10^{-6} K^{-1}$
T_c	743 K	763 K	37 K

The parameter α for $Ca_xSr_{1-x}TiO_3$ (CST) and $Pb_xSr_{1-x}TiO_3$ (PST) with corresponding values of x have been obtained from Fig. (7) of G. F. Zhang et.al.[13] and Fig (6) of Kyoung-T. Kim et al [14] 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 [1,19].

With the help of α, β, γ, T_c and equations (17) and (17b), we have calculated dielectric losses due to defect are given in table 2 and table 3 and are shown in fig. 1 and Fig 2.

Table 2. Impurity dependent dielectric loss v/s composition for $Ca_xSr_{1-x}TiO_3$ at 755 K

X	T_c	$(Tan\delta)_D \times 10^{-2}$
0.0	37	0
0.2	178	3.79
0.4	319	4.16
0.6	461	3.55
0.8	602	1.73

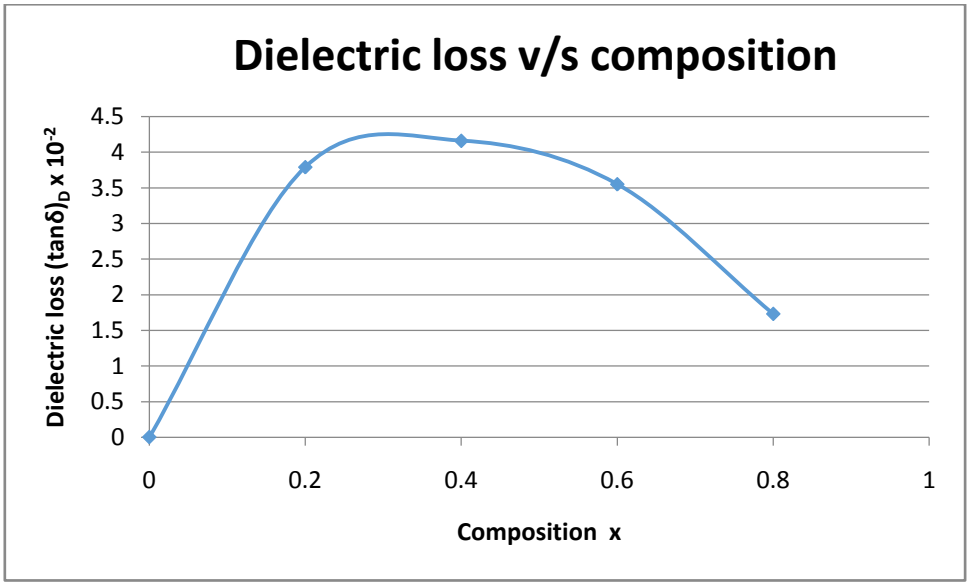

Fig.1. Impurity dependent dielectric loss as a function of impurity composition for $Ca_xSr_{1-x}TiO_3$ at 755K

Table 3. Impurity dependent dielectric loss v/s composition for $Pb_xSr_{1-x}TiO_3$ at 755K

x	T_c	$(Tan\delta)_D \times 10^{-2}$
0.0	37	0
0.2	182	0.33
0.4	327	0.40
0.6	472	2.41
0.8	617	3.54

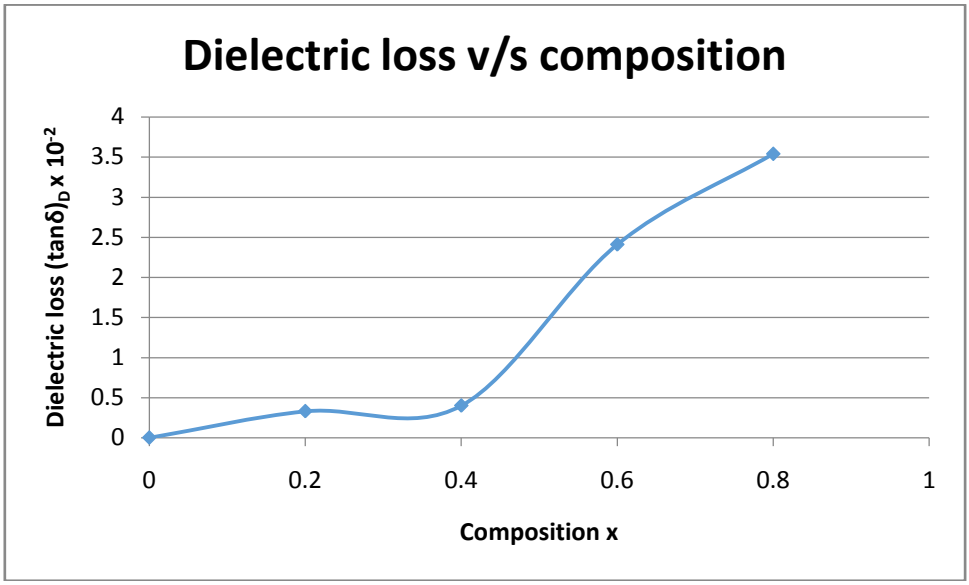

Fig.2. Impurity dependent dielectric loss as a function of impurity composition for $Pb_xSr_{1-x}TiO_3$ at 755 K

3.2 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 (4a) to (4e) for $Ca_xSr_{1-x}TiO_3$ and table (5a) to (5e) for $Pb_xSr_{1-x}TiO_3$ and are shown in Fig. 3 and Fig.4 respectively.

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

Temperature(T K)	755	760	765	770	775	780	785
Dielectric loss (tan δ)	0.002703	0.002716	0.002728	0.00274	0.002753	0.002765	0.002778

Table 4(b) Dielectric loss (tan δ) v/s temperature (T) of $Ca_{0.2}Sr_{0.8}TiO_3$, T_c=178K, x=0.2

Temperature(T K)	755	760	765	770	775	780	785
Dielectric loss (tan δ)	0.041263	0.040948	0.040637	0.040333	0.040033	0.039739	0.03945

Table 4(c) Dielectric loss (tan δ) v/s temperature (T) of $Ca_{0.4}Sr_{0.6}TiO_3$, T_c=319K, x=0.4

Temperature(T K)	755	760	765	770	775	780	785
Dielectric loss (tan δ)	0.046055	0.045584	0.045124	0.044674	0.044234	0.043805	0.043384

Table 4(d) Dielectric loss (tan δ) v/s temperature (T) of $Ca_{0.6}Sr_{0.4}TiO_3$, T_c=461K, x=0.6

Temperature(T K)	755	760	765	770	775	780	785
Dielectric loss (tan δ)	0.042096	0.041467	0.04086	0.040272	0.039704	0.039154	0.038621

Table 4(e) Dielectric loss (tan δ) v/s temperature (T) of $Ca_{0.8}Sr_{0.2}TiO_3$, T_c=602K, x=0.8

Temperature(T K)	755	760	765	770	775	780	785
Dielectric loss (tan δ)	0.030011	0.029204	0.028447	0.027736	0.027067	0.026436	0.02584

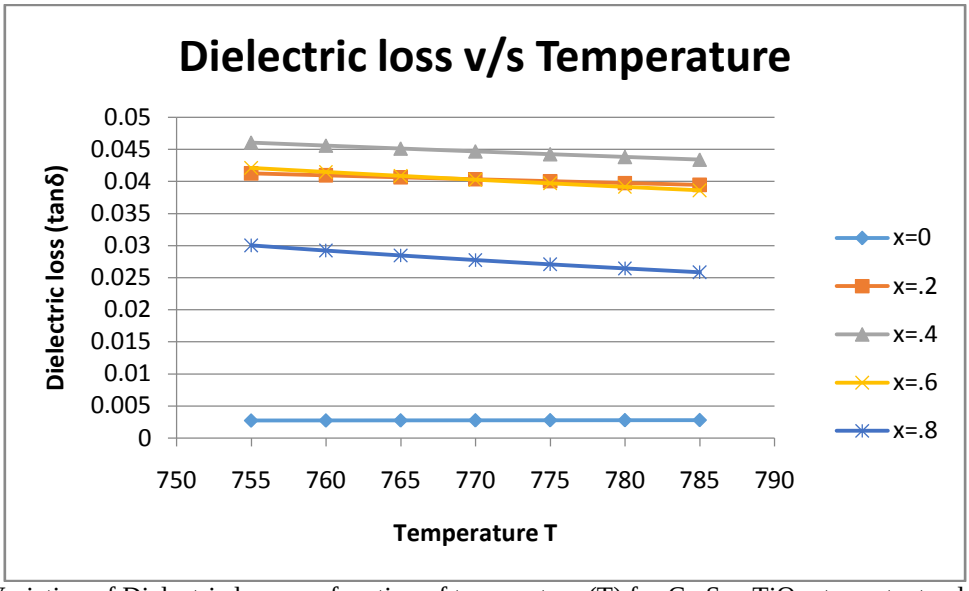

Fig.3.Variation of Dielectric loss as a function of temperature (T) for $Ca_xSr_{1-x}TiO_3$ at constant value of x.

Table 5(a) Dielectric loss (tan δ) v/s temperature (T) of $SrTiO_3$, T_c=37K, x=0.0

Temperature(T K)	755	760	765	770	775	780	785
Dielectric loss (tan δ)	0.002703	0.002716	0.002728	0.00274	0.002753	0.002765	0.002778

Table 5(b) Dielectric loss (tan δ) v/s temperature (T) of $Pb_{0.2}Sr_{0.8}TiO_3$, T_c=182K, x=0.2

Temperature(T K)	755	760	765	770	775	780	785
Dielectric loss (tan δ)	0.006641	0.006623	0.006605	0.006587	0.00657	0.006554	0.006538

Table 5(c) Dielectric loss (tan δ) v/s temperature (T) of $Pb_{0.4}Sr_{0.6}TiO_3$, T_c=327K, x=0.4

Temperature(T K)	755	760	765	770	775	780	785
Dielectric loss (tan δ)	0.008515	0.008468	0.008423	0.00838	0.008337	0.008296	0.008256

Table 5(d) Dielectric loss (tan δ) v/s temperature (T) of $Pb_{0.6}Sr_{0.4}TiO_3$, T_c=472K, x=0.6

Temperature(T K)	755	760	765	770	775	780	785
Dielectric loss (tan δ)	0.030947	0.030488	0.030045	0.029617	0.029204	0.028805	0.028418

Table 5(e) Dielectric loss (tan δ) v/s temperature (T) of $Pb_{0.8}Sr_{0.2}TiO_3$, T_c=617K, x=0.8

Temperature(T K)	755	760	765	770	775	780	785
Dielectric loss (tan δ)	0.049451	0.047879	0.046415	0.045047	0.043766	0.042565	0.041436

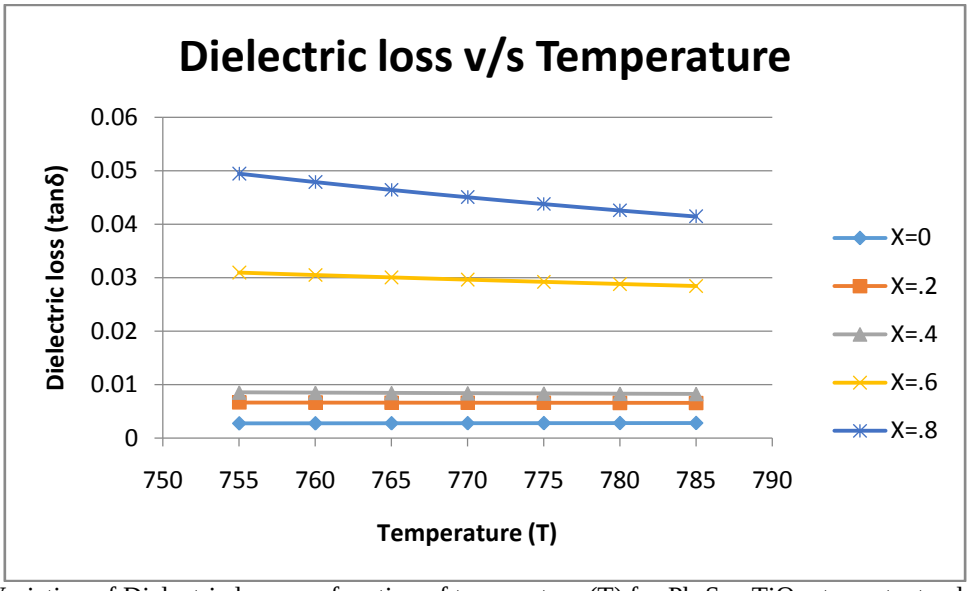

Fig.4.Variation of Dielectric loss as a function of temperature (T) for $Pb_xSr_{1-x}TiO_3$ at constant value of x.

IV. Discussion

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

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 T_c caused by impurity, depends upon the change in harmonic force constant between the impurity host lattice atoms mass change due to impurity and can be 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 Temperature 755K. It is clear from these figures that dielectric loss increases with the increases in the defects (impurity composition Ca or Pb).

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 decreases when temperature increases. 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.[4,8-9,14-17,25-26]

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