SUBSTITUTIONAL EFFECT ON DIELECTRIC CONSTANTS OF $PB_XSR_{1-X}TIO_3$ IN PARA-ELECTRIC PHASE

Sunil Kumar[*]
M S Yadav[**]
Neetu Sharma[***]
S C Deorani[****]

ABSTRACT

Dielectric constants are calculated for $Pb_xSr_{1-x}TiO_3$ single crystals in para-electric phase. In our calculations, double time temperature-dependent Green's function technique along with a modified Hamiltonian for the crystals is used which includes anharmonicity up to the fourth order due to the interactions of the soft mode. Fourier transformation and Dyson's equation treatment is used in arriving the result. In this paper, temperature and composition dependent dielectric constants of $Pb_xSr_{1-x}TiO_3$ have been studied. Dielectric constant increases with increase in the composition x at constant temperature and decreases with increasing temperature at constant value of x.

Keywords: *Dielectric Constant, Perovskites, Anharmonicity, Retarted Green's Function, Hamiltonian, Fourier Transform, Dyson's Equation.*

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Introduction

It is easy to control the dielectric properties of the $Pb_xSr_{1-x}TiO_3$ (PST) by adjusting [Pb/Sr] ratio. Pure $SrTiO_3$ is intrinsic quantum para-electric. It is known that permittivity peaks can be induced in $SrTiO_3$ by introducing substitutional impurities into the lattice[1]. In the present paper, $Pb_xSr_{1-x}TiO_3$ with Pb contents x varies from 0 to 1.0. (Pb, Sr)TiO_3 solid solutions belong to ferro-electric materials of the form (A'A'')BO_3 type. Both constituents of this solution are ferro-electric.

Ferroelectric materials can be phenomenologically defined as the dielectric materials with Spontaneous polarization in a range of temperature. This is produced by lack of symmetry of The crystal structure with respect to higher symmetry of the para electric phase. Spontaneous polarization decreases with the increase in temperature up to where the phase transition occurs[2]. The study of dielectric properties helps in understanding intra and inter molecular interactions conformational changes in the macromolecules etc. The temperature and the frequency dependence of dielectric constant in external electric field have been the subject of much interest in the past years. The dielectric constants of $Pb_xSr_{1-x}TiO_3$ are large and show temperature dependence. This behaviour results from a near cancellation of the short range restoring forces and the long-range driving forces on the ions[3].

Solid solutions of lead strontium titanate (PST) are very interesting ferro-electric materials which gain considerable attention due to useful in broad range of electric devices such as pyro-electric detector, DRAM capacitors, tunable microwave devices etc. PST is the most promising material because it is good insulator with large value of relative dielectric constant and low value of dielectric loss near ambient temperature.

Due to their specific features, ferroelectrics are broadly employed to make small size capacitors of capacitance, non-linear capacitor with controllable capacitance, frequency multiplier, voltage stabilizers, modulators, dielectric amplifiers etc. They possess high dielectric co-efficients over a wide

[*]   Department of Physics, R. R. College, RRBM University, Alwar, Rajasthan, India.
[**]   Department of Physics, R. R. College, RRBM University, Alwar, Rajasthan, India.
[***]   Department of Physics, R. R. College, RRBM University, Alwar, Rajasthan, India.
[****] Department of Physics, R. R. College, RRBM University, Alwar, Rajasthan, India.

temperature and frequency range. They are used as dielectrics in integrated or surface mounted device capacitors. The remarkable piezo-electric effect is applied in a variety of electromechanical sensors, actuators and transducers. Infrared sensors need a high pyro-electric co-efficient which is available with this class of materials[4]. Tunablethermistor properties in semiconducting ferro-electrics are used in positive temperature co-efficient resistors (PTCR)[5]

Lead Strontium titanate $Pb_xSr_{1-x}TiO_3$ (PST) which is known to adopt ABO_3 type solution, is a continuous solid solution of $PbTiO_3$ (PTO) and $SrTiO_3$ (STO) over the whole concentration range. The properties of $Pb_xSr_{1-x}TiO_3$ are known to depend on the composition $x^{6,7,8}$. In our previous paper we found that$PbTiO_3$is usually added to$SrTiO_3$ as a shifter in order to move the Curie-point T_C to higher temperatures. It is well established that T_C of $SrTiO_3$ increases linearly with the amount of Pb in place of Sr^9. For bulk $Pb_xSr_{1-x}TiO_3$, the Curie-point varies from -236^0 C to 490^0 C with value of x.

G.A.Samara[10] studied pressure and temperature dependence of the dielectric properties and phase transitions of the ferro electric perovskite $-PbTiO_3$ and $BaTiO_3$. G Rupprecht and R. O. Bell[11] studied the dielectric constant at microwave frequencies over awide temperature range up to 1000 ^{0}K on a series of perovskites, (CaTiOs, SrTiOs, BaTiOs, PbTiOs, andKTaOs) and their mixtures.

The experimental Dielectric constant measured for PST thin films was in the range of 900-1500 at 1MHz, with maximum value obtained for PST30 thin film. The loss tangents at room temperature were in the range of 0.07-0.1 for PST thin films with different compositions[12].

Vijender et. al[13]study the synthesis, structural and electrical properties of pure $PbTiO_3$ ferroelectric ceramics and measured the dielectric constant with temperature at constant frequencies. M. Tarnaoui et al[15]have reported temperature dependence dielectric constant of $Pb_xSr_{1-x}TiO_3$ at various compositions(x=0.0,0.2,0.5,0.8 and 1.0) for cooling down simulation.

Dielectric and ferroelectric properties were investigated as afunction of temperature, frequency, and dc applied field. Pb0.6Sr0.4TiO3 films showed a dominant voltage dependence of dielectric constant with a high tunability in a temperature range of 25–230 °C. The compositionally graded PST thin films with x=0.3–0.6 also showed the high tunability. The graded thin films exhibited a diffused phase transition accompanied by a diffused peak in the temperature variations of dielectric constants[14].

The aim of present paper is to study theoretically the dielectric constant in $Pb_xSr_{1-x}TiO_3$ (PST) mixed crystals by forming a model hamiltonian for the PST mixed crystals. Double time thermal Green's function method is used to obtain observable quantities with the help of model Hamiltonian considering anharmonic effects up to fourth-order with substitutional defects. For simplicity the ions are assumed non polarizable. Such a formulation is particularly interesting as the real part of dielectric constant ε' will lead to an expression for the change in the Curie-temperature resulting from the presence of impurities. we have taken into account the contribution of the temperature and defects(impurities) towards dielectric constant. The effect of defects (impurities) on the real part of dielectric constant of a displacive ferroelectric materials in para-electric phase is discussed and thus theoretical results obtained are compared with previous results available elsewhere.

Theory

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 perovskite, in para-electric phase which includes defects (substitutional impurity) is used in present study and is exactly similar as used earlier[15] 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

$$H = \sum_k \frac{\hbar\omega_k^a}{4}(A_k^{a+}A_k^a + B_k^{a+}B_k^a) + \sum_k' \frac{\hbar\omega_k^0}{4}(A_k^{0+}A_k^0 + B_k^{0+}B_k^0) -$$

$$\frac{\hbar\omega_0^0}{4}(A_0^{0+}A_0^0 + B_0^0 B_0^0) + \sum_k \hbar F(k)A_0^0 A_k^{0+}A_k^a + \sum_k \hbar\beta^a(k) A_0^{02}A_k^{0+}A_k^0 +$$

$$\sum_k \hbar\beta^0(k) A_0^{02}A_k^{a+}A_k^a + \sum_{k_1k_2k_3} \hbar\varphi(k_1k_2k_3)A_0^0 A_{K_1}^0 A_{k_1}^a A_{k_3}^a +$$

$$\sum_{k_1k_2k_3} \hbar\psi(k_1k_2k_3)A_0^0 A_{K_1}^0 A_{k_1}^0 A_{k_3}^0 + \hbar V A_0^{04} +$$

$$\hbar E$$

$$\{-\alpha A_0^0 +$$

$$\sum_k A(k)A_k^a A_k^{0+} + \sum_{k,\lambda} B^\lambda(k)A_0^0 A_k^{\lambda+}A_k^\lambda + \sum_{k_1k_2k_3} C(k_1,k_2,k_3)A_{K_1}^0 A_{k_2}^a A_{k_3}^a +$$

$$\sum_{k_1k_2k_3} D_1(k_1,k_2,k_3)A_{K_1}^0 A_{k_2}^0 A_{k_3}^0 - D_1(0,0,0)A_0^{03}\}$$

$= H_1 + H_2 + H_0 + H_3 + …………H_{14},$ ………..(2)

Where

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

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

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

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

$$A(k) = \frac{A'(k)}{(\omega_k^0 \omega_k^a)^{\frac{1}{2}}}$$ ………………………………….(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, $Y(K_1, K_2, K_3)$ 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[16]

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

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

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

and

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

x exp .[i{k_1 .R(ℓ)+k_1 . R(ℓ')}], …………………………(3e)

where e(k) is the polarization vector, R(ℓ) the equilibrium position vector of the ℓ-th atom, C(k_1, K_2) vanishes when n is either zero or N.Δφ denotes the force constant change, ℓ and ℓ' refer to the impurity and its nearest neighbours and μ = [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[16]. 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') =<<A(t); A(t')>>ω + iε (5) or G_0^0 (ω+ iε) = G'(ω)−iG"(ω) (6)

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

ε'(ω) −1 =−8 π² Nμ² G'(ω)   (7)   and tan δ=G"(ω)/ G'(ω) (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(ω+ iε) = $\frac{\omega_0^0}{\pi[\omega^2 - v^2(\omega) - 2\omega i \Gamma_0^0(\omega)]}$ (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[17] and Naithani et al[16].

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

$$\frac{v2(\omega)}{\gamma_1} = --\frac{(\omega_0^0)2}{\gamma_1} + \frac{\gamma_1}{\gamma_1} T + \frac{\Delta(v_D^2(\omega))}{\gamma_1} + \frac{\gamma_2 T^2}{\gamma_1}$$

$$\frac{v2(\omega)}{\gamma_1} = (T - T_c' + \xi T^2)$$ … (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). …………………………..(11b)

Equation (11a) can be reduced now as

$v^2(\omega) = \gamma_1 (T - T_c' + \xi T^2)$ …………(12)

or $v^2(\omega) = \gamma_1(T - T_c')$ and $\xi = -1.49 \times 10^{-3}$ cm^{-1} k^{-2} is negligible for ST[17].   …………….. (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 Constant

The real part of the complex dielectric constant [with the help of Eq. (7) and (10)] is given by:

$\varepsilon'(\omega) - 1 = -8\pi N\mu^2(\omega^2 - v^2)\varepsilon_s/[(\omega^2 - v^2)^2 + 4\omega^2\Gamma_0^2]$,

or $\varepsilon'(\omega) = 8\pi N\mu^2(v^2 - \omega^2)\varepsilon_s/[(v^2 - \omega^2)^2 + 4\omega^2\Gamma_0^2]$, $\varepsilon' \gg 1$ …………………………………(14)

Neglecting second term (being small) in comparison to the first term in the denominator, as $\omega\tau \leq 1$, $v \gg \omega$,(the soft mode frequency is very large as compared to the microwave frequencies as $\omega/v = 10^{-3}$)

So the real part of the frequency dependent dielectric constant in the range $\omega\tau \leq 1$ can be written as $\varepsilon'(\omega) = 8\pi N\mu^2\varepsilon_s/v^2$…………………………………………(15)

or $\varepsilon'(\omega) = \lambda/v^2$…………………………………………(16)

here, v is the soft mode frequency which depend upon anharmonicity, temperature, defects and electric field. ε_s is the static dielectric constant of the material, μ is the dipole moment per unit cell, N the total number of unit cells in the specimen, and $\lambda = 8\pi N\mu^2\varepsilon_s$ is a constant independent of temperature and equal to 2.05×10^{27} Hz^2 from reference[17] and other symbols are usual.

- **Composition-dependence of Dielectric Constant**

Using relation $\varepsilon'(\omega) = \lambda/v^2$ as stated above (taking $\lambda = 2.05 \times 10^{27}$ from reference[17], Soft mode (v) and T_C from our previous paper), Dielectric constantε' for $Pb_xSr_{1-x}TiO_3$ crystals are calculated for various values of x at constant temperature 800K which are given in table 1. Dielectric constant as a function of defect(impurity) concentration is plotted in figure 1.

Table 1: Dielectric constant versus composition at T=800K.

X	T_C(K)	V x 10^{12}Hz	$\varepsilon'(T) = \lambda/v^2$(x$10^3$)
0	37	4.28	0.11
0.1	109.6	4.07	0.12
0.2	182.2	3.85	0.14
0.3	254.8	3.61	0.16
0.4	327	3.37	0.18
0.5	400	3.10	0.21
0.6	472.6	2.80	0.26
0.7	545.2	2.47	0.34
0.8	617.1	2.09	0.47
0.9	690.4	1.62	0.78
1.0	763	0.94	2.31
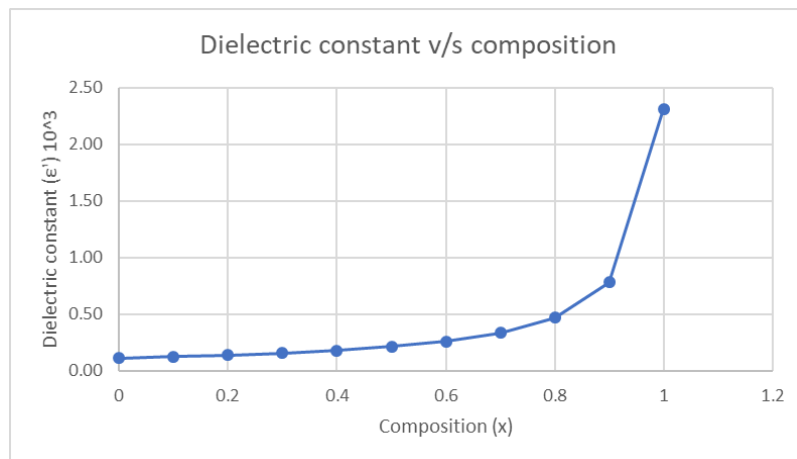
Fig. 1. Dielectric constant v/s composition for $Pb_xSr_{1-x}TiO_3$ at constant temperature T=800 K

- **Dielectric constant as a function of temperature**

For different value of x, Dielectric constant ε' is calculated as a function of temperature using the relation $\varepsilon' = \lambda/v^2$. The various calculated values are given in table 2(a) to 2(k) and are shown in figure 2.

Table 2(a): Dielectric constant v/s temperature of $SrTiO_3$, T_C = 37K, x=0

T(K)	50	60	70	80	90	100	300	400
Dielectric constant ε'	6581	3719	2592	1989	1614	1358	325	236

Table 2(b): Dielectric constant v/s temperature of $Pb_{0.1}Sr_{0.9}TiO_3$, T_C = 109.6K, x=0.1

T(K)	120	130	140	150	200	300	400
Dielectric constant ε'	8226	4194	2814	2118	946	449	295

Table 2(c): Dielectric constant v/s temperature of Pb0.2Sr0.8TiO3, TC = 182.2K, x=0.2

T(K)	190	200	210	220	230	300	400	500
Dielectric constant ε'	10968	4806	3077	2263	1790	726	393	269

Table 2(d): Dielectric constant v/s temperature of Pb0.3Sr0.7TiO3, TC = 254.8K, x=0.3

T(K)	260	270	280	290	300	400	500	600
Dielectric constant ε'	16452	5628	3395	2430	1893	589	349	248

Table 2(e): Dielectric constant v/s temperature of Pb0.4Sr0.6TiO3, TC = 327K, x=0.4

T(K)	330	340	350	360	370	400	500	600
Dielectric constant ε'	28516	6581	3719	2592	1989	1172	494	313

Table 2(f): Soft mode v/s temperature of Pb0.5Sr0.5TiO3, TC =400K, x=0.5

T(K)	410	420	430	440	450	500	600	700
Dielectric constant ε'	8555	4277	2852	2139	1711	855	428	285

Table 2(g): Dielectric constant v/s temperature of Pb0.6Sr0.4TiO3, TC =472.6K, x=0.6

T(K)	480	490	500	510	520	600	700	800
Dielectric constant ε'	11561	4917	3122	2287	1805	671	376	261

Table 2(h): Dielectric constant v/s temperature of Pb0.7Sr0.3TiO3, TC = 545.2K, x=0.7

T(K)	550	560	570	580	590	600	700	800
Dielectric constant ε'	17823	5780	3450	2458	1910	1561	553	336

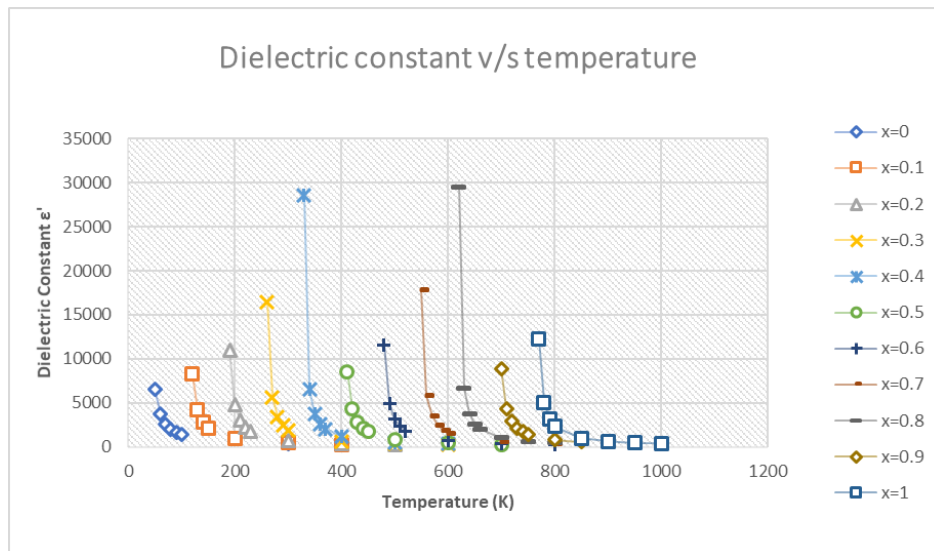**Table 2(i): Dielectric constant v/s temperature of Pb0.8Sr0.2TiO3, TC = 617.1K, x=0.8**

T(K)	620	630	640	650	660	700	750	800
Dielectric constant ε'	29499	6632	3736	2600	1994	1032	644	468

Table 2(j): Dielectric constant v/s temperature of Pb0.9Sr0.1TiO3, TC = 690.4K, x=0.9

T(K)	700	710	720	730	740	750	800	850
Dielectric constant ε'	8911	4365	2890	2160	1725	1435	781	536

Table 2(k): Dielectric constant v/s temperature of PbTiO$_3$, T$_C$ = 763K, x=1.0

T(K)	770	780	790	800	850	900	950	1000
Dielectric constant ε'	12221	5032	3168	2312	983	624	457	361

Fig. 2:Dielectric constant v/s Temperature for Pb$_x$Sr$_{1-x}$TiO$_3$at constant value of x

Discussion

The treatment adopted here leads one to see the comparison of dielectric constant with the variation of composition x, and temperature T ofPbxSr1-xTiO3. It is clear from equations (11) that the substitutional impurity dependence of dielectric constant is a clear consequence of defect (substitutional impurity) dependence of Soft mode frequency. So in order to discuss the defect dependence of dielectric constant of displacive ferroelectrics, we have first discussed the defect dependence of Soft mode frequency in our earlier paper[ ].It is clear from table1 that the Curie temperature(T$_c$) varies with variation

of lead concentration in the pure strontium titanate crystal and it continuously increase when impurity of lead increases. The dielectric constant of pure SrTiO3 has the lowest value as compared to dielectric constant for mixed crystals. The increment in dielectric constant with the increase of impurity concentration of Pb is due to the fact that the Curie temperature (T_c) increases with the increase of impurity of Pb on SrTiO3.

Figure 2 shows the variation of dielectric constant with temperature of PbxSr1-xTiO3 for different impurity concentration x in para-electric phase. The variation in dielectric constant for all values of x are of similar trend. It is also evident that in case of pure SrTiO3, dielectric constant increases sharply as soft mode (v) decreases and has highest value as T→Tc and dielectric constant decreases as temperature goes away from Tc in para-electric phase. The trend is same for all cases of x. Thus anomalous behaviour of dielectric constant near Tc can be explained by equation $ε'=λ/v^2$ as soft mode (v) tends to zero as T→Tc.

Equation (11b) shows that Curie-temperature increases with increase of defect concentration. The results of temperature and composition dependence of dielectric constants are good qualitative and close agreement with results of others[12,14,18].

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