Composite Materials
2021; 5(2): 30-36
http://www.sciencepublishinggroup.com/j/cm
doi: 10.11648/j.cm.20210502.11

Dependence of Electrical and Thermal Properties on Applied Electric Field in La Doped Barium Titanate Displacive Ferroelectric Perovskites

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To cite this article:
Dushyant Pradeep, Jagdev Singh. Dependence of Electrical and Thermal Properties on Applied Electric Field in La Doped Barium Titanate Displacive Ferroelectric Perovskites. *Composite Materials*. Vol. 5, No. 2, 2021, pp. 30-36. doi: 10.11648/j.cm.20210502.11

Received: April 6, 2021; **Accepted**: May 12, 2021; **Published**: August 31, 2021

Abstract: The high dielectric constant and low dielectric loss are the most desirable characteristics of the dielectric materials. Considering the hazardous nature of lead-based ceramics, many research groups have paid great attention to the research on non-lead-based perovskites. The effect of the electric field on the electrical and thermal properties of the Lanthanum doped barium Titanate, one of the main important members of the ferroelectric Perovskites family is studied using the method of double time thermal Green's function and Kubo formalism. With the help of the Silverman-Joseph Hamiltonian a general expression is derived. The electric field and temperature dependent dielectric constant and specific heat capacity of $Ba_{1-x}La_xTiO_3$ crystal has been investigated. The dielectric constant decreases with increase in temperature and applied electric field component. The variation shows a consequence of the gradual decrease in the Curie temperature of the doped material with the increase in defect concentrations. The results also compared with some previous studies done by the author and other workers on similar other materials.

Keywords: Ferroelectrics, Perovskites, Curie Temperature, Green's Function, Kubo Formalism

1. Introduction

Barium Titanate is one of the most important and significant member of the ferroelectric Perovskites family. At room temperature, $BaTiO_3$ adopts a tetragonal Perovskites type structure and is a ferroelectric with high permittivity. It transforms to the cubic, paraelectric state at the Curie temperature, T_C of approximates to 125°C. Ferroelectricity in tetragonal $BaTiO_3$ is due to an average relative displacement along the *c*-axis of titanium from its Centro symmetric position in the unit cell and consequently the creation of a permanent electric dipole. The study of the electrical and thermal properties of the BT ceramic is a very important research topic because of its great technical importance as it is used in multi- layer ceramic capacitor (MLCC), Ferroelectric Random-Access Memories (FRAM) and so forth because of its excellent dielectric, piezoelectric and ferroelectric properties and difficulty of explaining the behaviour thoroughly. Because of the great demand of these non-conducting ceramic materials peoples are going to reduce the size of all the communication devices as small as possible. Because of which materials with high dielectric constant like Barium titanate are very important in ceramic materials. The changes in the physical properties are also remarkable when one system is mixed with the other to form a composite system and their study helps in understanding basic mechanism of mixed crystal formation. For example, solid solution of the BT ceramics with the other ferroelectric Perovskites of same class and also with certain compounds which are not themselves ferroelectric materials possess ferroelectric properties and change in the composition of their solid solutions leads to change in the Curie point within the broad range of the temperature.

One of the very interesting properties of the BT ceramics is the electric field dependence of the low frequency transverse optic mode. Also, various temperature dependent properties of ferroelectrics result from the temperature dependence of the low lying transverse optic mode of

vibration [1, 2]. So, these temperature dependent properties indirectly depend upon the applied electric field on these ceramics. The dielectric properties and ferroelectric transition temperature of the BT ceramic can be controlled by the various kinds of the substitutions at Ba^{2+} on 'A' sites or Ti^{4+} on 'B' sites. Since its discovery, BT has been used as a high permittivity capacitor material because of its high dielectric constant. Variation in Chemical composition led to quite drastic changes in physical properties while retaining their piezoelectric properties. The effect of change in chemical composition by adding isovalent substitution such as Ca^{2+}, Sr^{2+}, Pb^{2+} for Ba^{2+} and Zr^{4+}, Hf^{4+} for Ti^{4+} on dielectric properties and phase transition temperature of BT ceramics has been done [1]. The effect of the heterovalent substitutions such as Ni^{3+}, Co^{3+}, Er^{3+}, La^{3+}, Nb^{5+}, W^{6+} etc. for Ti^{4+} or Ba^{2+} on dielectric properties and phase transition temperature is still a great field of study. These heterovalent impurities causes charge imbalance and creates vacancies at 'A' or 'B' or Oxygen sites in Perovskite structure [2-8]. Ferroelectrics have many applications like in holographic storage media, optical communication, memory displays, modulator beam reflectors etc. and also most of the semiconducting ferroelectrics ceramics due to their positive temperature coefficient of resistivity (PTCR) these are also used in the temperature control and many other devices. The barium titanate, strontium titanate, and potassium titanate are the broadly used PTCR material and most intensively studied [9].

The aim of the present paper is the theoretical study of the field dependent dielectric properties of La doped Barium titanate ($BaTiO_3$) displacive ferroelectric Perovskites by forming a model Hamiltonian for the polycrystalline mixture of ferroelectric Perovskites and then applying it to $Ba_{1-x}La_xTiO_3$. Double time temperature dependent Green's function technique is used to obtain thermally averaged correlation function and hence the observable quantities with the help of modified model Hamiltonian, taking into account the anharmonic effect up to the fourth order with substitutional defect and electric moment terms. The softmode frequency contribution towards dielectric constant has been taken in to consideration too. The effect of defects and electric field on the real part of dielectric constant of La displaced ferroelectric material in paraelectric phase is discussed and results obtained are compared with the previous theoretical and experimental studies.

2. General Formulation/Model Hamiltonian

Retarded double time dependent Green's function is recommended to evaluate the expressions for the various dynamic properties for optical phonon, when both defect and external electric field is considered as [10]

$$G_0^0(t-t') = << A_0^0(t); A_0^0(t') >> \omega + i \in = -i\theta(t-t') < [A_0^0(t) : A_0^0(t')] > \quad (1)$$

$$\text{or } G_0^0(\omega + i\varepsilon') = G'(\omega) - G''(\omega) \quad (2)$$

where $G'(\omega)$ and $G''(\omega)$ are real and imaginary parts of the Green's function $G(\omega)$. Differentiating $G_0^0(t-t')$ as given in Eq. (1) with respect to *t*, its equation of motion is given by

$$i\hbar \frac{d}{dt} G_0^0(t-t') = \hbar\delta(t-t') < [A_0^0(t); A_0^0(t')] > + << [A_0^0(t), \bar{H}_T]; A_0^0(t') >>, \quad (3)$$

Where the equation of motion is to be solved via the modified Hamiltonian $\bar{H}_T$.

Differentiating Eq. (3) with respect to *t,* and Fourier transforming it, we get

$$(\omega^2 + \omega_3\omega_4)\, G^0(\omega) = (\omega_3/\pi) + (\omega_3/\pi) \times << M_1(t); A_0^0(t') >>, \quad (4)$$

$$\text{Where } \omega_3 = \omega_0^0 + 4C(0,0), \quad (5)$$

$$\omega_4 = \omega_0^0 - 4D(0,0) + 24gD_1'E^2 - 96g^2E^2V, \quad (6)$$

$$\text{And } M_1(t) = F(t) + 2\pi Y + \left(\frac{2\omega}{\omega_3}\right)\pi \times (t), \quad (7)$$

The expressions for the defect, field and temperature dependent renormalized frequencies of soft mode, acoustic mode and optic mode can be easily obtained by starting with

The soft phonon Green's function $(= << A_0^0(t)\,;\, A_0^0(t') >>)$,

Acoustic phonon Green's function $(= << A_k^a(t)\,;\, A_{k'}^a(t') >>)$,

and optic phonon Green's function $(= << A_k^0(t)\,;\, A_{k'}^0(t') >>)$ respectively and solving their equations of motion with the help of $\bar{H}_T$.

Solving these Green's functions, one gets the values of $\bar{\Delta}_0^0(\omega)$ as follows:

$$\bar{\Delta}_0^0(\omega) = \tilde{\Delta}_0^0(\omega) + \Delta_D(\omega) + \Delta_{D,E}(\omega) \tag{8}$$

Where $\tilde{\Delta}_0^0(\omega)$ describes the contribution towards $\bar{\Delta}_0^0(\omega)$ in presence of anharmonicity and electric field terms and is given by

$$\tilde{\Delta}_0^0(\omega) = \text{Re}\ \ \omega_0^0[\Delta_1(\omega) + \Delta_2(\omega) + \Delta_3(\omega) + \Delta_4(\omega) + \Delta_5(\omega) + \Delta_6(\omega) + \Delta_7(\omega)], \text{ (say)} \tag{9}$$

The value of Δ_1 - Δ_7 (ω) are same as studied by the previous workers [11]. The third term $\Delta_{D,E}(\omega)$ gives the combined effect of defect and electric field. In a similar manner we write

$$\bar{\Gamma}_0^0(\omega) \quad = \quad \tilde{\Gamma}_0^0(\omega) \ + \ \Gamma_D(\omega) \ \ + \Gamma_{D,\ E}(\omega) \tag{10}$$

Where $\tilde{\Gamma}_0^0(\omega)$ describes the contribution towards $\bar{\Gamma}_0^0(\omega)$ in presence of anharmonicity and electric field and is given by

$$\tilde{\Gamma}_0^0(\omega) = \ \ \omega_0^0\pi[\Gamma_1(\omega) + \Gamma_2(\omega) + \Gamma_3(\omega) + \Gamma_4(\omega) + \Gamma_5(\omega) + \Gamma_6(\omega) + \Gamma_7(\omega)], \tag{11}$$

The Γ_1 - Γ_7 have the same values as derived by the previous workers [11].

3. Soft Mode Frequency

The real part of the pole of $G_0^0(\omega + i\in', \ E)_D$ would give the defect, electric field and temperature dependent soft mode frequency $\bar{\Omega}$ as the self-consistent solution of the equation

$$\bar{\Omega}^2 \ = -(\omega_0^0)^2 + 4\,\omega_0^0 D(0,0) - 24 g\omega_0^0 D_1'\ E^2 + 96\omega_0^0 g^2 E^2 V + 4\omega_0^0 \sum_{k,\lambda} \beta^\lambda(k) < A_k^{\lambda+}\ A_k^\lambda > + \bar{\Delta}_0^0(\bar{\Omega}) \tag{12}$$

A knowledge of $\bar{\Delta}_0^0(\bar{\Omega})$ gives the expression for soft mode frequency as

$$\bar{\Omega}^2 \quad = \quad \tilde{\Omega}^2 \quad + \quad \Omega_D^2 \quad + \quad \Omega_{D,E}^2 \tag{13}$$

Here $\tilde{\Omega}$ is the field and temperature dependent element of the square of the efficient soft mode frequency $\bar{\Omega}$.

It is evident from Eq. (13) that the square of the defect, temperature and field dependent soft mode frequency varies with the defect and applied external field parameters in presence of anharmonicity. The presence of these effects stabilizes the soft mode frequency. The temperature independent part of the effective soft mode frequency ($\bar{\Omega}$) is due to defect. The influence of defect and external applied field on this mode also affects the interaction [12] of soft mode with other modes, thus giving rise to defect and field dependences of various dynamic properties. In the present work we have obtained the expression of effective soft mode frequency ($\bar{\Omega}$) and it has been shown that the combined effect of defect and field dependence in presence of anharmonicity on the frequency is observed. Now it is clear that the defects contribution towards effective soft mode frequency is temperature independent. However, the indirect temperature dependence due to defect terms is obtained from $\Delta_{D,E}(\omega)$, in the classical limit of high temperature. It is interesting to note here that this term $\Delta_{D,E}(\omega)$ is also missing in the absence of electric field. It is contended that this cross dependence or better to say the indirect temperature dependence due to defect is caused by applied field. $\Delta_{D,E}(\omega)$ Will vanish either defect is zero or field is not applied. Thus one can easily express the dependence of the effective soft mode frequency on the temperature, given in Eq. 3.2 as

$$\bar{\Omega}^2 = -(\omega_0^0)^2 + \omega_0^0(96g^2V - 24\ g\ D_1'\ )\ E^2 + \Omega_D^2 + \gamma' T + \gamma_2 T^2, \tag{14}$$

With $\gamma' \ = \ \gamma_1 \ + \ \gamma_3$; which have their predefined values.

Thus our expression (14) gives the soft mode frequency dependence for an anharmonic ferroelectric Perovskite crystal on temperature and external applied electric field, subjected to an external applied field. The square of the effective soft mode frequency shows the gradual increase when the strength of applied electric field varies [13], in presence of defect, which is in agreement with the previous results [14].

4. Field Dependent Dielectric Constant of $Ba_{1-x}La_xTiO_3$

The effects of applied electric field on the complex dielectric constant in anharmonic ferroelectric material are calculated in its paraelectric phase with the help of Silverman –Joseph Hamiltonian augmented with fourth order phonon co-ordinates using double time Green's function technique [15]. The Hamiltonian used here is given by:

The soft phonon Green's function is linked to real part of dielectric constant as

$$G_0^0(\omega + i\in') = << A_0^0(t)^0; A_0^0(t') >> \quad (15)$$

$$\text{As } \in'(\omega) - 1 = -8\pi^2 N\mu^2 G'(\omega) \quad (16)$$

The real dielectric constant [with the help of Eq. (16)] element is given by

$$\in' -1 = \frac{-8\pi^2 N\mu^2(\omega^2 - \Omega^2)^2 \in_s}{(\omega^2 - \Omega^2)^2 + 4\omega^2\Gamma^{02}};\ v_0^0 \approx \Omega$$

$$\text{Or } \in' = \frac{8\pi N\mu^2(\Omega^2 - \omega^2)\in_s}{[(\Omega^2 - \omega^2)^2 + 4\omega^2\Gamma^{02}]};\ \in' >> 1 \quad (17)$$

Where $\in_s$, is the invariable dielectric constant of the material, μ is the dipole moment/unit cell and N is the total number of unit cell in the specimen and other symbols are as usual.

The dependence of dielectric constant on frequency [in the range $\omega\tau \le 1$] may be calculated using the relation [16].

$$\in' = \frac{\in_s \omega_0^2(\omega_0^2 - \omega^2)}{\left(\omega_0^2 - \omega^2\right)^2 + 4\omega^2\Gamma^{02}};\ (\omega_0 \approx \Omega \approx \upsilon_0) \text{ or}$$

$$\in' = \frac{\lambda\,(\omega_0^2 - \omega^2)}{\left(\omega_0^2 - \omega^2\right)^2 + 4\omega^2\Gamma^{02}} \quad (18)$$

Where $\lambda = \in_s (T)\omega_0^2$ is independent of thermal variation at particular point.

The soft mode frequency is high as compared to the micro wave frequency (as $\omega/\Omega \approx 10^{-3}$) and no relaxation effects are realized and half width [$\Gamma^0(\omega)/2\omega_0$] is such that $\Gamma^0(\omega) << 2\omega_0$.

For optic mode the soft mode frequency can be given as [17]

$$\Omega_{ADE} = -(\omega_k^0)^2 - 2\omega_k^0 V + Y_1 + Y_2 T + Y_3 T^2, \ldots \quad (19)$$

Where the symbols Y_1, $Y2$ and Y_3 are the temperature independent, coefficients of T and T^2 respectively in the expression of square of soft mode frequency.

From Eqns. (17), (18) and (19), (v_0^0 is same as ω_0^0) we achieved that

$$\in'(T) = \frac{\lambda}{K(T + \xi\ T^2 - T'_c)}, \ldots \quad (20)$$

Where K is temperature independent constant, $T'_c (= \alpha/\beta)$ is paraelectric field dependent phase transition temperature, ξ $(= \gamma/\beta)$ is non-linearity constant, This constant characterizes the deviated variation of the temperature dependence of the $\in'$ from the Curie-Weiss law and is corelated to the third-and fourth-order coupling coefficients [15].

$$\alpha = -(\omega_k^0)^2 - 2\omega_k^0 V + Y_1,\ \beta = Y_2,\ \gamma = Y_3$$

If ξ is very less and T is not very large (i.e., though in paraelectric phase but closer to vicinity of Curie point (T_c) then the term $\xi\ T^2$ can be neglected from the denominator term.

Where

$$T'_c = T_c + \Delta T \ \ldots \quad (21)$$

With

$$\Delta T = 0.0019 \times E \ \ldots \quad (22)$$

Where E is external applied field in Volt/cm [18].

5. General Formulation

The relation of soft mode frequency with temperature of ferroelectric materials is given by [19]

$$\Omega_{(T)} = [K\ (T - T_c)]^{1/2} \ldots \quad (23)$$

Where K is a constant depend on temperature and electric field parameter. Also the electric field dependant soft mode frequency can be given as

$$\Omega_{(E)} \cong K\ (E^2 + 1)^{1/2}\ (T - T'c)^{1/2} \ \ldots \quad (24)$$

Where $T'_c = T_c + \Delta T$ with $\Delta T = 1.9 \times 10^{-3} \times E$, where E is applied electric field and measured in Volt/cm [18].

From Eqs. (20) and (21)

$$\frac{\Omega_{(E)}}{\Omega_{(T)}} = (E^2 + 1)^{1/2}\ \frac{(T - T'_C)^{1/2}}{(T - T_C)^{1/2}} \text{ Or}$$

$$\Omega_{(E)} = \Omega_{(T)}\ (E^2 + 1)^{1/2}\ \frac{(T - T'_C)^{1/2}}{(T - T_C)^{1/2}}$$

$$\text{Or } \Omega_{(ADE)} = \Omega_{(AD)}\ (E^2 + 1)^{1/2}\ \frac{(T - T'_C)^{1/2}}{(T - T_C)^{1/2}} \ldots \quad (25)$$

$$(\Omega\text{ADE} \sim \Omega_E\ \&\ \Omega_{AD} \sim \Omega_T)$$

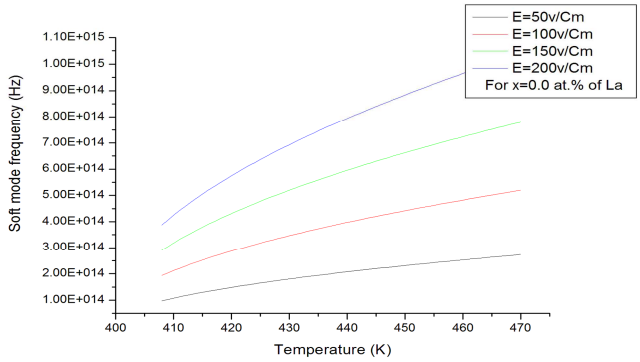
Where Ω_{ADE} is field and defect dependent soft mode frequency and Ω_{AD} is defect dependent soft mode frequency.

6. Varialtion of Softmode Frequency with Temperature

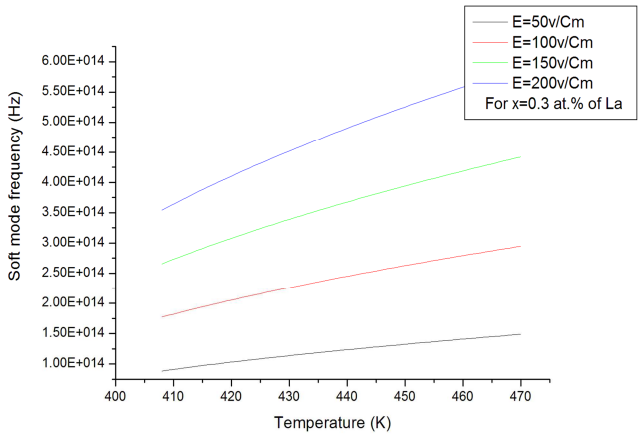
Using Eq. (25) frequency at soft mode of $Ba_{1-x}La_xTiO_3$ for distinct values of x (i.e. x=0.0, 0.3, 0.5, & 1.0) at. % of La in different electric field strengths is determined. The Curie temperature of $Ba_{1-x}La_xTiO_3$ (BLT) for distinct values of x is obtained from Wei Cai et al. [1] by using best fit of data. The values of Ω_{AD} (defect dependent soft mode frequency) is obtained from the earlier study of Cai et al. (In which they have studied the case of zero field) [3].

We computed values of frequency at soft mode of $Ba_{1-x}La_xTiO_3$ with various values of x at. % of La (i.e. x=0, 0.3, 0.5, 1.0) and Soft mode frequency verses temperature curves

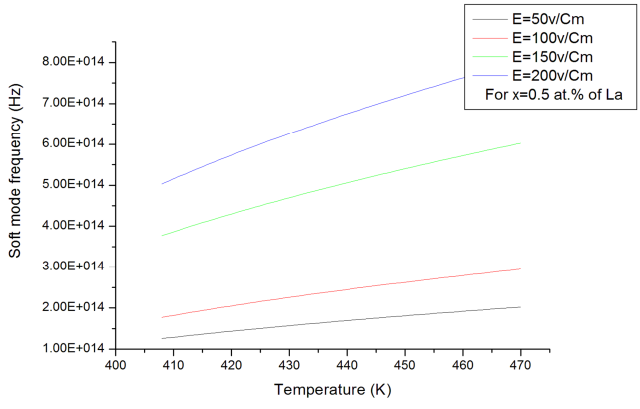
of $Ba_{1-x}La_xTiO_3$ (BLT) for different values of x at. % of La (i.e. x=0.0, 0.3, 0.5, 1.0) are drawn in figures 1(a) to 1(d) respectively. The trend of the curves obtained are showing the variations in agreement with the earlier experimental and theoretical results of others [16, 17, 18 and 19].



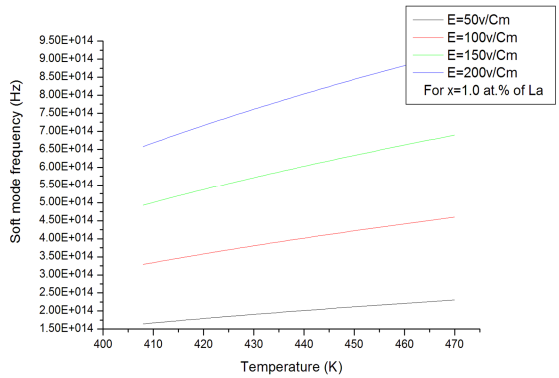
a) $Ba_{1-x}La_xTiO_3$=(x=0 at. % of La)



b) $Ba_{1-x}La_xTiO_3$=(x=0.3 at. % of La)



c) $Ba_{1-x}La_xTiO_3$=(x=0.5 at. % of La)

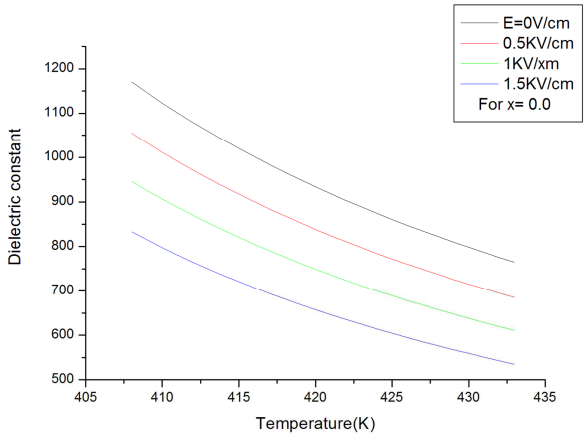


d) $Ba_{1-x}La_xTiO_3$=(x=1.0 at. % of La)

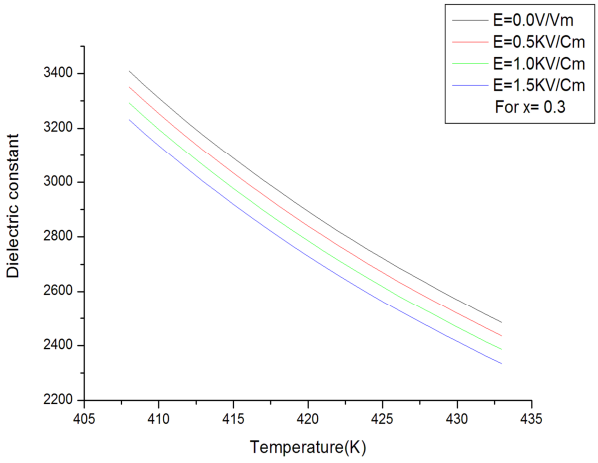
Figure 1. *Variation of soft mode frequency (Ω_{ADE}) of $Ba_{1-x}La_xTiO_3$ with temperature (K) at different applied electric field strengths are as.*

7. Varialtion of Dielectric Constant With Temperature

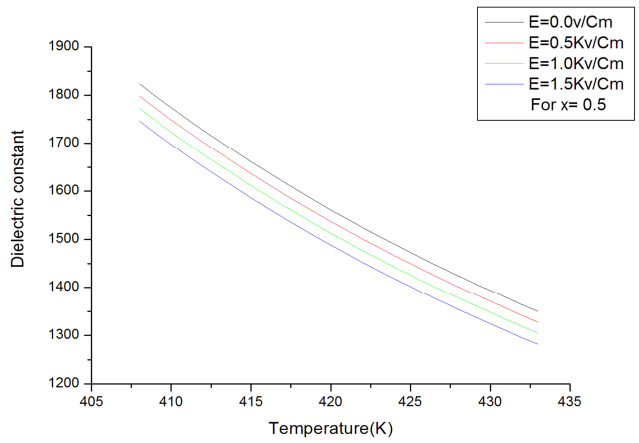
We have calculated the temperature dependent dielectric constant in para electric phase of $Ba_{1-x}La_xTiO_3$. The value of Curie constant C ($=8.54\times10^4$ K) are taken from Naithani [13] by best fit of data.



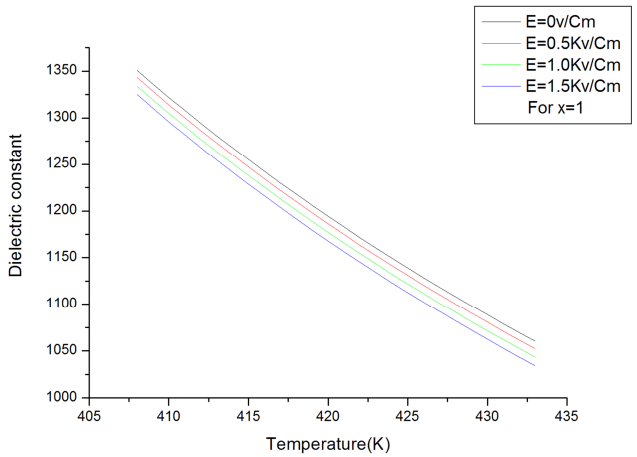
a) $Ba_{1-x}La_xTiO_3$ (x=0.0 at. % of La)



b) $Ba_{1-x}La_xTiO_3$ (x=0.3 at. % of La)



c) $Ba_{1-x}La_xTiO_3$ (x=0.5 at. % of La)



d) $Ba_{1-x}La_xTiO_3$ (x=1.0 at. % of La)

Figure 2. *Variation of dielectric constant (ε') of $Ba_{1-x}La_xTiO_3$ with temperature (K) at different applied electric field strengths are as.*

The values of dielectric constant ($\in'$) of $Ba_{1-x}La_xTiO_3$ for distinct values of x at. % of La (i.e. x=0.0, 0.3, 0.5, & 1.0) in presence of external applied electric field are calculated and corresponding variations of dielectric constant with temperature at different electric field are shown in Figures 2(a) to 2(d) respectively. Taking a reference temperature the value of Dielectric Constant increases with rise in applied electric field for all cases. It is clearly visible that taking electric field as a parameter, dielectric constant decreases with rise of temperature in these cases.

8. Results

From the above graphs as the Curie temperature changed (Increases) in the presence of electric field for these Perovskites. Using eqn. (25) we have determined frequency at soft mode of $Ba_{1-X}La_XTiO_3$ for distinct concentrations x at. %of La (i.e. x=0.0, 0.3, 0.5 & 1.0) in different electric field strengths. Hence we have studied the variation of soft mode for different electric fields from set of figures {1(a) to 1(d)}. The trend of the curves obtained are showing the variations in agreement with the earlier experimental and theoretical results of others [14, 19-23].

We have also discussed the variation of dielectric constant in the paraelectric phase of BT and have shown these variations with the temperature in the set of figures {2(a) to 2(d)} (5.2) for the different electric field strengths. Taking a particular temperature it is observed that the dielectric constant decreases as the strength of the externally applied electric field increases in all the cases (i.e. of x=0.0, 0.3, 0.5 & 1.0 at. % of La) because the Curie temperature decreases as the concentration of the Lanthanum increases in BLT [24]. At a particular single value of applied electric field, the dielectric constant decreases as the temperature increases for all the cases that we have discussed.

9. Discussion and Conclusion

The expression for the dielectric constant for anharmonic displacive ferroelectric crystal $Ba_{1-X}La_XTiO_3$ when subjected to the external electric field is obtained. The converted Hamiltonian is used for calculations. This conversion is important because the external field dependence of the frequency at soft mode and hence the dielectric constant is governed by $g\,(=\alpha/\omega_0^0)$. If the term $(-\alpha\,E\,A_0^0)$ in the Hamiltonian is treated without conversion, it will give zero Green's function. It may be concluded that higher order anharmonic terms in the Hamiltonian stabilize the soft modes because of high occupation number should be responsible for an appreciable scattering of the rest of the modes.

The treatment pursued here shows the change in the dielectric constant with the changing values of frequency and temperature of $Ba_{1-X}La_XTiO_3$ for distinct values of x (x=0.0, 0.3, 0.5 & 1.0) at. % of La in the presence of electric field. It is now clear from Equations that the dependence of dielectric constant on applied field is clear consequence of the field dependence of soft mode frequency. Therefor if we want to study the field dependence of dielectric constant of ABO_3 type ferroelectrics, so firstly we have to study the dependence of applied field on soft mode frequency. It is clear from Eq. (9) that soft mode frequency is directly proportional to the square of applied electric field strength.

Figures 1(a) to 1(d) show the variation of frequency at soft mode with temperature for different electric field strengths of $Ba_{1-X}La_XTiO_3$ for different values of x (x=0.0, 0.3, 0.5, & 1.0) at. % of La. It is clear from the figures that as and when temperature rises soft mode frequency first rises for all values of x and at higher temperature soft mode frequency becomes constant for all values of x in $Ba_{1-X}La_XTiO_3$. So the soft mode frequency rises with rise of applied electric field and is in good agreement with the experimental result of other worker's [13] and in the absence of the electric field the results are in good agreement with the result of Kumar et al. [14], and in the absence of defect in pure crystals the results are in good agreement with the result of Lingwal et al. [13]

Figures 2(a) to 2(d) show the variation of dielectric constant with temperature in distinct values of applied external electric field strengths of $Ba_{1-X}La_XTiO_3$ for distinct

values of x (x=0, 0.3, 0.5 & 1.0). According to our outcomes as temperature rises and approaches to the Curie temperature (i.e. $T \rightarrow T_c$) dielectric constant increases sharply but a rapid fall of dielectric constant is observed as the difference (*T-Tc*) increases. This variation obtained is in acordance with experimental results observed by Rupprecht and Bell [25], Kumar et. al. [13] and Jona and Shirane [26].

The field effect is effective and noticeable at low temperature side and less effective along higher temperature side as it is clear from figures. As we move away from the Curie temperature towards the high temperature side the effect due to higher order anharmonic effect dominates upon the applied external effect.

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