Electric field dependent attenuation constant in Ba$_x$Sr$_{1-x}$TiO$_3$ perovskites

Ashish Kukreti*, Ashok Kumar & U C Naithani
Department of Physics, Garhwal University, Pauri Campus, Pauri, Garhwal, Uttarakhand 246 001

*E-mail: ashishkukreti07@gmail.com

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A theoretical expression is derived for attenuation constant in para-electric phase in anharmonic Ba$_x$Sr$_{1-x}$TiO$_3$ ferroelectric crystal, using double time temperature dependent Green’s function technique. Taking electric field as a parameter, the variation of attenuation constant with frequency and temperature has been studied by considering third and fourth order anharmonic interactions. The mass and force constant change due to the presence of impurity atoms in Silverman-Joseph Hamiltonian augmented with higher order anharmonic terms. The effect of electric field, defect and anharmonicity on stabilization of the soft mode frequency has also been studied. The attenuation constant increases with increasing temperature and frequency in the presence of an electric field. In the vicinity of the Curie temperature, the attenuation constant increases anomalously.

Keywords: Attenuation constant, Curie temperature, Anharmonicity, Electric dipole moment, Displacive ferroelectrics

1 Introduction

Ultrasonic technique is used to study the phase transition in solids. The low frequency ultrasonic velocity yields information about the static aspects, while the absorption data give information about the dynamic aspects. Anomalous acoustic behaviour in the vicinity of structural phase transitions in solids is a widespread and well known phenomenon. There have been several experiments on the propagation of ultrasonic waves near the ferroelectric phase transition temperature in displacive type ferroelectrics. In recent years, ultrasonic studies have played an increasing important role in characterizing the behaviour of the systems near the co-operative phase transitions (co-operative ordering of the dipoles which gives rise to the spontaneous polarization which is destroyed by thermal agitation above the Curie – temperature) and critical points. One advantage of ultrasonic measurements is that the static and dynamic properties of the system can be simultaneously measured.

Ultrasonic attenuation data provide information about the dynamic behaviour and from the frequency, temperature, defect and as well as electric field dependence, the mechanism involved can be understood. Theoretically new ways of describing critical phenomena in terms of fluctuation correlation have been of great importance. The large variations in the strength of attenuation near the transition are usually referred ‘anomalous’. Such special variations are now described as critical. As revealed experimentally as well as theoretically,$^{1,5}$ the soft or ferroelectric mode plays an essential role in displacive ferroelectrics. As the temperature approaches the Curie temperature($T_c$), the soft mode frequency becomes vanishingly small ($\Omega \sim (T-T_c)$) resulting in an increase in its amplitude, which should influence the acoustic mode via the phonon-phonon interactions and is expected to give rise to an anomalous behaviour of sound near $T_c$. A considerable review on pure and mixed ferroelectric crystals is available in the literature$^{6-10}$ and references therein.

The phenomenological theory of sound attenuation near the transition point has been developed by Landu and Kalatnikov.$^{11}$ The degeneracy of the soft mode with the longitudinal acoustic mode for large wave numbers gives the characteristic behaviour of the soft mode in displasive ferroelectrics like SrTiO$_3$. The idea of coupling of elastic strain and the soft phonon mode was suggested by many researchers,$^{12-14}$ where the coupling is described in terms of the electro-strictive coupling, which is quadratic in polarization and linear in strain. The velocity of the transverse wave does not show any appreciable critical behaviour. In general, it is agreed that the main contribution to low frequency attenuation is from an interaction of soft phonons with each other and with acoustic phonons. In the vicinity of the Curie temperature, when soft mode frequency tends to zero only interactions with longitudinal sound remains. Ba$_x$Sr$_{1-x}$TiO$_3$(BST) exhibits a number of advantages over the formerly known ferroelectrics such as high mechanical strength to heat and...
moisture, the presence of ferroelectric properties within a broad range of temperature and ease of manufacturing.

Barium strontium titanate (BST) has attracted universal attention of researchers and has found a wide practical application. Several researchers have derived expression for attenuation constant in displacive ferroelectrics. Kumar et al., have also discussed the attenuation constant in Ba

The aim of the present work is to study theoretically attenuation constant in polycrystalline mixtures of Ba

The Hamiltonian of the defect ferroelectric crystal is constructed by a method similar to that used in our previous study from the crystal model proposed by Silverman and Joseph by augmenting it with fourth order anharmonic interaction terms involving ferroelectric optic modes of lowest wave vector. It is assumed that the introduction of defects in the lattice causes changes in the harmonic force constants besides mass change, their influence upon anharmonic coefficients is neglected. This is true for isovalent and non-polarizable defects because they cause change in the short-range forces only. For small concentration of defects, the impurity–impurity interaction can be neglected. It is known that this Hamiltonian provides a satisfactory interpretation of anomalous behaviour of sound near the Curie points, electric field dependence of Curie-temperature and temperature dependence of thermal conductivity of ferroelectrics and contains essential characteristic of a displacive type ferroelectric with perovskite structure.

To define the response function it is now introduced by retarded double time thermal Green's function for acoustical phonon as:

\[
G^a_{kk'}(t-t') = (\omega - \omega^a_k)G^a_{kk'}(\omega) = \delta_{kk'}/2\pi + \langle\alpha F^a_1(t); A^{a^\dagger}_k(t')\rangle 
\]

(\omega - \omega^a_k)G^a_{kk'}(\omega) = \delta_{kk'}/2\pi + \langle\alpha F^a_1(t); A^{a^\dagger}_k(t')\rangle 

(3)

and

\[
\omega^a_k = \omega^a_k + 2D(-k, k^a) + 2C(-k, k^a) 
\]

(5)

The equation of motion of \(\langle\alpha F^a_1(t); A^{a^\dagger}_k(t')\rangle\) with respect to time argument \(t'\) gives:

\[
(\omega - \omega^a_k) \langle\alpha F^a_1(t); A^{a^\dagger}_k(t')\rangle = (1/2\pi)\langle\alpha F^a_1(t); A^{a^\dagger}_k(t')\rangle + \langle\alpha F^a_1(t); F^{a_2}_2(t')\rangle 
\]

(6)

where

\[
F^a_2(t') = D(-k', 0)A^0_0 + C(-k', 0)B^0_0 + [D(-k', k^0)]A^0_{kl} + [C(-k', k^0)]B^0_{kl} + \alpha(-k')A^0_k 
\]

(7)
Substituting Eq. (6) in Eq. (3), the equation of motion for \( G_{k,k'}^a(\omega) \) in the lowest approximation, in the form of Dyson equation can be written as:

\[
G_{k,k'}^a(\omega) = \{\delta_{k,k'}\} / 2\pi \left[ \omega - \bar{\omega}_k^a - \langle F_i^a(t) ; A_k^a(\omega) \rangle \right] + \bar{P}(\omega) , \quad \cdots (8)
\]

With

\[
\bar{P}(\omega) = 2\pi << F_i^a(t) ; F_2^a(t') >>_\omega . \quad \cdots (9)
\]

Writing the equation of motion for the Green's function [Eq.(2)] with the help of the modified Hamiltonian, Fourier transforming and writing it in the Dyson's equation form, one obtains:

\[
G(\omega + i\varepsilon) = \delta_{\omega,\omega'} / 2\pi [\omega + \bar{\omega}_k^a + i\bar{\Gamma}_k(\omega)] \quad \cdots (10)
\]

where \( \bar{\Gamma}_k(\omega) \) is the damping constant and \( \bar{\omega}_k^a \) is the field-dependent stabilized acoustical frequency and is given as:

\[
\bar{\omega}_k^a = \omega_k^a + 2\omega_k^a \bar{A}_k(\omega) \quad \cdots (11)
\]

where

\[
\omega_k^a = \omega_k^a + 8\omega_k^a gE^2[2g\beta^a(k) - B^a(k)] \quad \cdots (11a)
\]

\[
\bar{A}_k(\omega) = A_k(\omega) + AE^2 \quad \cdots (11b)
\]

\( \Delta_k(\omega) \) and \( \Gamma_k(\omega) \) are identified as the expression for the shift and width of acoustical phonons in the absence of the field, in the lowest approximation. \( \Delta_k(\omega) \) depends upon third and fourth order anharmonic coefficients in the presence of electric moment terms and the constant \( A \) depends upon third order anharmonic terms only.

In the presence of electric field, for displacive ferroelectric damping constant \( \bar{\Gamma}_k(\omega) \) is expressed as:

\[
\bar{\Gamma}_k(\omega) = \Gamma_A(\omega) + \Gamma_D(\omega) + \Gamma_E(\omega) \quad \cdots (12)
\]

where \( \Gamma_A(\omega) \), \( \Gamma_D(\omega) \) and \( \Gamma_E(\omega) \) are the damping constants which are dependent upon anharmonic contributions, defect and electric field terms, respectively and their values are obtained as:

\[
\Gamma_A(\omega) = -\pi |F(\kappa)|^2 \left( \omega_k^0 / \bar{\omega}_k^0 \right) \sum (\bar{N}_k^0 \pm N_0) \times [\delta(\omega - \Omega - \bar{\omega}_k^0) - \delta(\omega - \Omega + \bar{\omega}_k^0)]
\]

\[
+4\pi |\beta^a(k)|^2 (\omega_k^a / \bar{\omega}_k^a) \times [\delta(\omega + 2\Omega - \bar{\omega}_k^0) - \delta(\omega + 2\Omega + \bar{\omega}_k^0)]
\]

\[
+ (1 + N_0^2 - 2N_0\bar{N}_k^a) [\delta(\omega - 2\Omega - \bar{\omega}_k^0) - \delta(\omega + 2\Omega - \bar{\omega}_k^0)]
\]

\[
+ \pi \delta^3 \sum_{k_1,k_2} \phi(k_1,k_2,k_1,k_2,k_1,k_2)
\]

\[
x (\omega_k^0\omega_{k_1}^a \omega_{k_2}^a / \bar{\omega}_k^0 \bar{\omega}_{k_1}^a \bar{\omega}_{k_2}^a) < (1 + N_0\bar{N}_k^a + \bar{N}_k^0 \bar{N}_k^a)
\]

\[
+ N_0\bar{N}_k^a) [\delta(\omega - \Omega - \bar{\omega}_k^0 - \bar{\omega}_{k_1}^a - \bar{\omega}_{k_2}^a)
\]

\[
- \delta(\omega + \Omega + \bar{\omega}_k^0 + \bar{\omega}_{k_1}^a + \bar{\omega}_{k_2}^a)]
\]

\[
- \delta(\omega - \Omega - \bar{\omega}_k^0 - \bar{\omega}_{k_1}^a - \bar{\omega}_{k_2}^a)] > \quad \cdots (13)
\]

and

\[
\Gamma_D(\omega) = -\text{Im} 2\pi << F_i^a(t) ; F_2^a(t') >>_{\omega,\text{inc}} . \quad \cdots (14)
\]

where \( F_i^a(t) & F_2^a(t') \) are given by Eqs (4 and 7), respectively.

Also

\[
\Gamma_E(\omega) = 2\pi E^2[|2g\beta^a(K)|^2 (\omega_k^a / \bar{\omega}_k^a) \Sigma(\bar{N}_k^0 \pm N_0)
\]

\[
\times [\delta(\omega - \Omega - \omega_k^0) - \delta(\omega + \Omega + \omega_k^0)]
\]

\[
+ 4\pi E^2 [A(k) - 2gF(k)] (\omega_k^0 / \bar{\omega}_k^0)
\]

\[
\times [\delta(\omega + \bar{\omega}_k^0) - \delta(\omega - \bar{\omega}_k^0)]
\]

\[
+ 2\pi E^2 \sum_{k_1,k_2} |C(k_1,k_2,k_1,k_2)|^2 g \phi(k_1,k_2,k_1,k_2)
\]

\[
\times (\omega_k^0\omega_{k_1}^a \omega_{k_2}^a / \bar{\omega}_k^0 \bar{\omega}_{k_1}^a \bar{\omega}_{k_2}^a) \sum (\bar{N}_k^0 \pm N_0)
\]

\[
(\omega - \bar{\omega}_k^0 + \bar{\omega}_k^a) - (\omega + \bar{\omega}_k^0 + \bar{\omega}_k^a)] . \quad \cdots (15)
\]

Here

\[
\delta^3 = \delta_{123} + \delta_{213} + \delta_{321} \quad \cdots (16)
\]

\[
\delta_{123} = \delta_{k_1,k_2} \delta_{k_3,k_4} + \delta_{k_2,k_3} \delta_{k_4,k_1} \quad \cdots (17)
\]

\[
\bar{N}_k^0 = < A_k^\lambda > \quad \quad \quad \text{cosh} (1/2\beta\hbar\omega_k^\lambda k) \quad \cdots (18)
\]

\[
N_0 = < A_0^\lambda A_0^\lambda > \quad \text{cosh} (1/2\beta\hbar\Omega) \quad \cdots (19)
\]
dependence of 

discussed quantitavely and qualitatively by the 

In the absence of an external electric field (zero field 

\[ \alpha = \alpha_0 + \alpha_{E}(\omega) \] 

where \( \Omega \) is effective soft mode frequency and 
temperature and field dependence of \( \Omega \) is given as \( \Omega_{E}^{2} \sim k(T - T_{c}) \). In Eqs (13) and (15) \( F(k), \) 

\[ \beta^{4}(k); \varphi(k_{1}, k_{2}, k_{3}) \) and \( C(k_{1}, k_{2}, k_{3}) \) are third-order, 

fourth-order and electric moment coefficients, respectively.

3.1 Temperature and field dependence of attenuation constant 
in \( Ba_{x}Sr_{1-x}TiO_{3} \) perovskites 

The expression for the attenuation constant is given as:

\[ \alpha = \frac{\tilde{\Gamma}_{k}}{\omega} \]  

where \( c \) is the sound velocity and \( \tilde{\Gamma}_{k} \) is the damping 
constant and is also defined as the width of frequency 
response of acoustic phonon mode \( k \), the value of 
\( \tilde{\Gamma}_{k}(\omega) \) is given by Eq. (12). Here, the attenuation 
constant can be expressed as:

\[ \alpha = \alpha_{\lambda}(\omega) + \alpha_{\varphi}(\omega) + \alpha_{E}(\omega). \]  

In the absence of an external electric field (zero field 

\[ \alpha = \alpha_{\lambda}(\omega) + \alpha_{\varphi}(\omega) \], which have already been 
discussed quantitavely and qualitatively by the 
authors\(^ {19} \). There, the temperature defect and field 
dependence of \( \alpha(\omega) \) is expressed as:

\[ \alpha(\omega) = A_{1} + (A_{2} + A_{3}T + A_{4}E^{3})T + (A_{5} + A_{6}T + A_{7} E^{5}) \times[T/(T - T_{c})^{1/2}] + A_{8}[T^{3}/(T - T_{c})^{1/2}] + A_{9}[T^{5}/(T - T_{c})^{1/2}] \]  

... (23)

where \( A_{i}'s( i = 1-9) \) denote the temperature and 
electric field independent term in \( \tilde{\Gamma}_{k}(\omega) \) [Eq. (12)]. \( A_{1} \) 
is only defect-dependent while the rest depend upon 
impurity concentration, anharmonic force constants and 
electric moment term. The expression given in 
Eq. (23) does not give the explicit temperature 
dependence of \( \alpha \) because of the renormalization effects which are of two-types. The renormalized 
frequencies of each phonon mode appearing in real 
and imaginary parts of polarization operator may 

produce some change in the temperature dependence. 
It is clear from the relation given in Eq. (23) that as 
\( T \rightarrow T_{c} \) attenuation constant increases anomalously in 
agreement with the results of Tani\(^ {25} \) and Pytte\(^ {26} \). In the 
presence of electric field and low temperature range 
for the reduced temperature, Eq. (23) becomes as:

\[ \alpha(\omega) \approx A_{1}(\omega) + A_{2}(\omega)/T - T_{c}^{3/2} + A_{3}E^{2}T/(T - T_{c})^{1/2}, \]  

... (23a)

which is same as obtained by Tani\(^ {25} \). Here in the 
absence of electric field the attenuation constant can 
be written as:

\[ \alpha_{AD}(\omega) = \alpha_{\varphi}(\omega) + \alpha_{E}(\omega) \]  

Comparing Eqs (23) and (24), we get:

\[ \alpha_{\lambda}(\omega) \approx A_{1}(\omega)/(T - T_{c})^{3/2} \]  

and \( \alpha_{\varphi}(\omega) \approx A_{1} \)  

\[ \alpha_{E}(\omega) \) for \( SrTiO_{3} \) is obtained by best fit of data from 
authors' previous study\(^ {19} \) and assumed that it remains 
unchanged by addition of impurity of barium atom.

\[ A_{1} = [D/T_{c}^{2}(\omega_{0}^{0})] \alpha_{0} \]  

where \( D \) is defect parameter determined by lattice 
imperfection and equal to zero for pure single crystal. 
It was also assumed that \( \omega_{0} \) remains the same for pure 
and mixed crystal for simplicity. The defect 
contribution and Curie temperature for pure and 
mixed \( SrTiO_{3} \) are obtained from Rupprecht and 
Bell\(^ {27} \). \( \alpha_{0} \) for pure \( SrTiO_{3} \) is equal\(^ {18} \) to \( 0.25 \times 10^{5} \) 
dBm\(^ {1} \).

Considering the effect of electric field on the 
attenuation constant, the electric field dependent 
contribution to \( \alpha(\omega) \) is given by \( \alpha_{E}(\omega) (= \Gamma_{E}(\omega)/c) \). At 
any temperature, well above \( T_{c} \) the temperature 
dependence of \( \alpha_{E}(\omega)[III^{rd} \) term in Eq. (23a) \)] is given by:

\[ \alpha_{E}(\omega) = [T/(T - T_{c})^{1/2}] E^{2}A_{7} \]  

where \( T_{c}' = T_{c} + \Delta T \) and \( \Delta T = 1.9 \times 10^{-3} \times E \). \( E \) is 
external electric field and measured in V/cm, \( A_{7} \) 
(2.20\times10^{-4} \text{S/m}^{1/2}) is the coefficient of temperature 
and electric field in Eqs (15) and (26) and has been 
calculated by best fit of data from Lingwal \textit{et al.}\(^ {18} \) for 
zero defect cases.
In the present study, only the net critical relaxation attenuation \( (\alpha_c) \) associated with the critical fluctuations above phase transition temperature \( T_c \) in the presence of field is concerned. By plotting attenuation constant \( (\alpha) \) versus temperature \( (T) \) for \( \text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3 \) with different value of defect concentration \( (x) \) in the presence of electric field, for each value of \( x \) this variation is linear which is in agreement with the authors’ previous results\(^{18,19}\). The attenuation constant increases with increase of impurity concentration and electric field.

With the help of Eq. (23a) we have calculated the field dependent attenuation constant for \( \text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3 \) mixed crystal with different values of \( x \) \((x = 0, 0.2, 0.5, 0.7, 0.8, 1.0) \) and plotted attenuation constant versus temperature curves for \( \text{SrTiO}_3, \text{Ba}_0.2\text{Sr}_{0.8}\text{TiO}_3, \text{Ba}_0.5\text{Sr}_{0.5}\text{TiO}_3, \text{Ba}_0.7\text{Sr}_{0.3}\text{TiO}_3, \text{Ba}_0.8\text{Sr}_{0.2}\text{TiO}_3 \) and \( \text{BaTiO}_3 \) in Fig. 1(a-d), respectively for different biasing fields. When the temperature approaches to the Curie temperature \( (T \rightarrow T_c) \) the attenuation constant increases \([\text{Fig. 1(a-d)}]\). The variations are in good agreement with previous experimental and theoretical results of other researchers\(^{16-19}\).

3.2 Frequency variation of field dependent attenuation constant in \( \text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3 \) ferroelectric perovskites

The frequency dependent attenuation constant can be represented as:

\[
\alpha(\omega) = a_1 + a_2 \omega^2 + a_3 \omega + a_4 \omega^4, \quad \ldots (27)
\]

where first three terms are due to the anharmonic and fourth term is due to the defect contributions to the attenuation constant, respectively. The \( \omega^2 \) dependence arises due to fourth order anharmonic interactions (quadratic anharmonic interaction), \( \omega \) dependence arises due to third order anharmonic interactions terms and \( \omega^4 \) dependence arises due to impurity concentrations, if we consider change in harmonic force constant only.

If we consider third order anharmonic interaction only then Eq. (27) gives:

\[
\alpha(\omega) \approx a_1 + a_2 \omega^2 + a_3 \omega + a_4 \omega^4 \quad \ldots (28)
\]

Garland \textit{et al.}\(^{28}\) expression together with defect term for attenuation constant can be rewritten as:

\[
\alpha = a_0 + \alpha_E + \alpha_D, \quad \ldots (29)
\]

Here \( a_0 \) is the critical attenuation constant (for a pure crystal). \( \alpha_E \) and \( \alpha_D \) are electric field and defect dependent attenuation constant, respectively.

Comparing Eqs (28) and (29), we get:

\[
\alpha_D \approx a_4 \omega^4 \quad \ldots (30)
\]

\[
\alpha_E \approx a_2 \omega^2 \quad \ldots (30a)
\]

This Eq. (30) shows that defect dependence attenuation varies linearly with \( \omega^4 \).
\[ a_d = (D/T_c)^2 a_2 \text{ MHz}^{-4} \text{s}^{-2} \text{m}^{-1} \quad \ldots (31) \]

where \( D \) is defect dependence parameter and \( T_c \) is Curie temperature. \( D \) and \( T_c \) in \( \text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3 \) for different values of \( x \) are obtained from Ruprecht and Bell\(^2\) by best fit of data and the parameter \( a_2 \) for pure \( \text{SrTiO}_3 \) is calculated from Kumar et al\(^1\)\(^9\). The calculated values of \( D, T_c, a_2 \) and \( a_4 \) of \( \text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3 \) for different values of \( x \) are presented in Table 1.

Eq. (30a) shows the field dependent attenuation constant, where \( a_2 \) is calculated by best fit of data and has value \( a_2 = 4.1 \times 10^{-14} \text{ S}^2\text{m}^{-1} \) at temperature 398K and frequency 10 MHz and gives the value of field dependent attenuation as:

\[ a_E = 1.62 \times 10^{-3} \Omega \times \omega \text{ dBm}^{-1} \quad \ldots (32) \]

where \( \omega \) is in MHz. Since soft mode frequency \( \Omega \) is very large as compared to microwave frequency \( \omega(\omega/\Omega \approx 10^{-3}) \), therefore, in the presence of electric field Eq. (32) can be rewritten as:

\[ a_E = 1.62 \times 10^{-3} \Omega \times \omega \text{ dBm}^{-1} \quad \ldots (33) \]

where \( \Omega \) is the stabilized soft mode frequency and it depends upon third and fourth-order anharmonic terms in presence of defect and electric moment terms. In a pure harmonic approximation, this frequency is purely imaginary and only the presence of anharmonicity and electric moment terms stabilizes this frequency. The variation of soft mode frequency with temperature is approximated as:

\[ \Omega^2 \sim K (T-T_c) \quad \text{or} \quad \Omega \sim K (T-T_c)^{1/2} \]

where \( K \) is temperature independent constant.

Using Eqs (29) and (33), we have calculated the attenuation constant for \( \text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3 \) mixed crystal with different values of \( x(x = 0, 0.2, 0.5, 0.7, 0.8, 1.0) \) and plotted attenuation constant versus frequency curves for \( \text{SrTiO}_3, \text{Ba}_0.2\text{Sr}_0.8\text{TiO}_3, \text{Ba}_0.5\text{Sr}_0.5\text{TiO}_3, \text{Ba}_0.7\text{Sr}_0.3\text{TiO}_3, \text{Ba}_0.8\text{Sr}_0.2\text{TiO}_3 \) and \( \text{BaTiO}_3 \) in Fig. 2(a-d), respectively, for different biasing fields. Taking a particular electric field as a reference, it is observed that attenuation constant increases with increase of frequency in all the cases, which is in

<table>
<thead>
<tr>
<th>Crystal</th>
<th>( D ) (GHz(^{-10})K)</th>
<th>( T_c ) (K)</th>
<th>( a_4 ) (MHz(^{-2}\text{S}^2\text{m}^{-1})</th>
<th>( a_0 ) (dB)m(^{-1})</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \text{Ba}<em>x\text{Sr}</em>{1-x}\text{TiO}_3 )</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>( x = 0 )</td>
<td>0.0027 \times 10^{-3}</td>
<td>105</td>
<td>0.522 \times 10^{-28}</td>
<td>0.81 \times 10^{-1}</td>
</tr>
<tr>
<td>( x = 0.2 )</td>
<td>0.0910 \times 10^{-3}</td>
<td>218</td>
<td>1.376 \times 10^{-28}</td>
<td>2.14 \times 10^{-1}</td>
</tr>
<tr>
<td>( x = 0.5 )</td>
<td>0.1000 \times 10^{-3}</td>
<td>280</td>
<td>0.752 \times 10^{-28}</td>
<td>1.17 \times 10^{-1}</td>
</tr>
<tr>
<td>( x = 0.7 )</td>
<td>0.0730 \times 10^{-3}</td>
<td>324</td>
<td>0.271 \times 10^{-28}</td>
<td>0.42 \times 10^{-1}</td>
</tr>
</tbody>
</table>

\[ \text{Fig. 2 — (a-d)-Variation of attenuation constant with frequency at different electric field in } \text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3 \text{ ferroelectric perovskites} \]
good agreement with previous theoretical results.\textsuperscript{18,19,25}

4 Results and Discussion

In the present study, we have derived an expression for attenuation constant of sound, and discussed its defect, frequency, temperature and electric field dependence in Ba\(_{x}\)Sr\(_{1-x}\)TiO\(_3\) ferroelectric perovskites, using double time thermal Green’s function technique. Both the mass and force constant changes due to doping, are taken into account, in the crystal Hamiltonian augmented with higher order anharmonic and higher order electric moment terms which give their contribution to various scattering processes. There are two scattering mechanisms present in the crystal: One is the various three and four-phonon scattering amongst phonons due to higher order anharmonicity and the other is the electric field induced scattering due to the electric moment terms in the presence of field. These two distinct scattering processes operate simultaneously, having the different values of relaxation times for a particular acoustic phonon mode \(a\) and the scattering rates are additive. It is shown that the acoustic width in the phonon frequencies is temperature dependent indicating the temperature dependence of the attenuation constant. The temperature dependence is the direct consequence of anharmonicity.

Figure 1(a-d) show the variation of attenuation constant with temperature at different fields for Ba\(_{x}\)Sr\(_{1-x}\)TiO\(_3\) with different values of defect concentration \(x\) (\(x = 0, 0.2, 0.5, 0.7, 0.8, 1.0\)), respectively. According to the present results as temperature approaches towards Curie temperature, attenuation constant increases. This variation is in good agreement with experimental results observed by Rupprecht and Bell\textsuperscript{27} and our previous studies.\textsuperscript{18,19} The trend is the same for all the values of \(x\) with different field strengths.

Figure 2(a-d) show the variation of attenuation constant with frequency for different electric fields in the case of Ba\(_{x}\)Sr\(_{1-x}\)TiO\(_3\). The attenuation constant increases linearly with the frequency, which is in good agreement with the results obtained theoretically and experimentally by many researchers\textsuperscript{16,18,19,29} [Fig. 2(a-d)]. The trend is the same for all the values of \(x\) with different field strengths.

Figure 3(a-d) shows the variation of attenuation constant with electric field, taking temperature as a parameter. It is clear from Fig. 3(a-d) that attenuation constant increases with the increasing field, which is in agreement with experimental results.\textsuperscript{8} The temperature and field dependence of attenuation constant is a clear consequence of the temperature and field dependence of soft mode frequency.

Fig. 3 — (a-d)—Variation of attenuation constant with electric field at different temperature in Ba\(_{x}\)Sr\(_{1-x}\)TiO\(_3\) ferroelectric perovskites
Figure 4(a-d) shows the variation of attenuation constant with electric field, taking frequency as a parameter. The attenuation constant increases with the increasing field [Fig. 4(a-d)]. It is also evident from Fig. 4(a-d) that at low values of electric fields, there is no sharp difference between the attenuation constant for different values of $x$ but at high fields the attenuation constant has a remarkable high value in case of SrTiO$_3$ as compared to BaTiO$_3$. Thus, at high fields, increasing value of $x$ decreases the attenuation constant. The attenuation constant at $\omega = 40$ MHz [Fig. 4(d)] has high values for different values of $x$ as compared to attenuation constant at $\omega = 10$ MHz [Fig. 4(a)]. Thus, increasing frequency increases the attenuation constant.

It is clear from Eq.(21) that the field dependence of attenuation constant is a clear consequence of the field –dependence of $\Gamma_E(\omega)$ [Eq. (15)], and hence of $\tilde{\Gamma}_k(\omega)$ [Eq. (12)]. $\Gamma_E(\omega)$ varies directly as the square of electric field strength and thus, the attenuation constant increases with the increase of applied biasing field. It is obvious that $\alpha_D$ (defect dependent attenuation constant) also increases linearly with $\omega^4$, for all impurity concentrations which show that the inverse relaxation time of scattering of phonons by mass defect is proportional to the fourth power of phonon frequency ($\omega^4$) which is in agreement with the authors’ previous results$^{30}$ and references therein. The results obtained are also in good agreement with the results of Tani and Tsuda$^7$ and Heuter and Neuhaus$^8$. In the vicinity of the Curie temperature $T_c$, the soft mode frequency $\Omega$, which is imaginary in harmonic approximations and becomes temperature dependent due to anharmonic terms, becomes vanishingly small and hence, the attenuation constant increases anomalously. Recently, we have applied the Green’s function technique in observing the field dependent dielectric constant$^{21}$ and microwave loss$^{31}$ in Ba$_x$Sr$_{1-x}$TiO$_3$ displacive ferroelectrics. A current review on Ba$_x$Sr$_{1-x}$TiO$_3$ is available in the literature.$^{32}$

5 Conclusions

In the present study, we have derived an expression for the attenuation constant of sound and discussed its impurity, electric field and temperature dependence, in a doped displacive anharmonic crystal (Ba$_x$Sr$_{1-x}$TiO$_3$). Both the mass and force constant changes due to doping, which are taken into account, in the crystal Hamiltonian augmented with higher order anharmonic and electric dipole moment terms. In deriving the expression for attenuation constant, different researchers have used different methods, approximations and symbols. In the treatment adopted here we have used Green’s function technique and Dyson’s equation treatment. The Dyson’s equation treatment has been found convenient to derive shift and width of the frequency response function and hence, to describe the properties of mixed crystals.
It is obvious that attenuation constant in case of Ba$_x$Sr$_{1-x}$TiO$_3$ perovskite varies as $T/(T-T_c)^{3/2}$. The attenuation constant in case of Ba$_x$Sr$_{1-x}$TiO$_3$ for any particular value of $x$, increases with increase of $T/(T-T_c)^{3/2}$. For phonons of small half width in the harmonic approximation, the inverse relaxation time and hence, the attenuation constant is proportional to fourth power of phonon frequency, neglecting force constant changes. But due to the modification of force constant changes, this dependence is no more of the form $\omega^4$ but contains a frequency independent term and a term proportional to $\omega^2$ arising from force constant changes. The experiments on the field-dependent sound attenuation in displacive ferroelectrics in the vicinity of the Curie temperature $T_c$ seems not to have been done except in Ba$_x$Sr$_{1-x}$TiO$_3$ ceramics by Heuter and Neuhaus, many years ago. They have measured the attenuation of 10 mega cycle sound as a function of temperature and applied electric field, and found an anomalous increase of the attenuation near $T_c$ (393 K). Thus our results are in good agreement with those of Heuter and Neuhaus. The expressions predict the variation of attenuation constant of sound with the electric field, showing that, at a constant temperature, the sound attenuation constant increases in the presence of electric field from the value obtained in the absence of electric field.

In the vicinity of the Curie temperature $T_c$, the soft modes frequency, which is imaginary in harmonic approximations and becomes temperature dependent due to anharmonic terms, becomes vanishingly small and $N_0$ (Eq. (19)) becomes anomalously large and hence, the attenuation constant increases anomalously. These results agree with the results of Tani and Tsuda. The soft mode frequency is stabilized in the presence of defects, anharmonicity and electric field. In the absence of all the above effects, this frequency is imaginary which is due to the cancellation of the competing forces. The impurity, anharmonicity and the increasing electric field, increase the soft mode frequency. The treatment adopted here leads one to see the comparative variation of the attenuation constant of sound with the variation of the defect and electric field parameters.

References