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Electric Field Dependent Inelastic Scattering Cross Section of Neutrons in $Ba_x Sr_{1-x}TiO_3$ Displacive Ferroelectric Perovskites

Abstract

Using the method of double time thermal Green's functions, a general expression has been derived for the electric field dependence of the one phonon differential scattering cross section of neutrons in $Ba_xSr_{1-x}TiO_3$ displacive ferroelectric perovskites in the presence of external electric field by considering higher order anharmonic terms in the Hamiltonian. At any temperature well above the Curie temperature T_C, the differential scattering cross section increases with the increasing defect, temperature and electric field in these perovskites. In the vicinity of T_C it is the long wavelength Cochran soft mode which gives the anomalous behaviour of the scattering cross section.

Keywords: Displacive Ferroelectrics; Anharmonic Effect; Soft Mode; Neutron Scattering Cross Section

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Introduction

A ferroelectric crystal shows spontaneous polarization whose direction can be reversed by an external electric field. Broadly ferroelectrics may be classified into two main groups : Order-disorder and displacive, according to whether the transition is associated with the individual ordering of ions or is associated with the displacement of a whole sub lattice of ions of one type relative to another sub lattice. Ferroelectrics are the most typical non-linear dielectrics. Due to their specific features they are broadly employed in many devices and have found a wide range of practical applications such as in memory display, optical communication, coherent optical processing, modulator beam reflectors and holographic storage media. Besides these, ferroelectrics are broadly used in ceramic industry. Semiconducting ferroelectric ceramics having positive temperature coefficient of resistivity (PTCR) are used in temperature control and many other devices. The most intensively studied and widely used PTCR materials are those based on barium titanate ($BaTiO_3$) and strontium titanate ($SrTiO_3$). Barium strontium titanate ($Ba_xSr_{1-x}TiO_3$) is a solid solution family composed of barium titanate and strontium titanate with its Curie temperature covering a wide range. When strontium atoms were introduced to A site in perovskite barium titanate matrix to replace barium atoms, the phase transition temperature of paraelectric to ferroelectric decreases and the phase transition behaviour changes from sharp to diffuse.[1]

It is now well known that several interesting temperature dependent properties of ferroelectrics result from the temperature dependence of the low lying transverse optic mode of vibration. One of the very interesting properties of these crystals is the electric field dependence of the low frequency transverse optic mode. With the further evolution of nuclear physics, particularly following the construction of nuclear reactors, neutrons, deuterons and α-rays have been diffracted from crystals. Of these different nuclear particles, neutrons have been shown to be particularly useful in crystal structure analysis. Unlike x-rays and electrons, neutrons interact with atomic nuclei and are sensitive to their magnetic properties. Consequently, neutron diffraction can be used in the elucidation of many structures whose detailed atomic arrangements do not affect their x-ray diffraction spectra in the same way.

The neutron scattering experiments[2-4] on displacive ferroelectrics have shown that several properties of ferroelectric crystals can be understood in terms of lattice dynamics[5] of these crystals. The coherent scattering of neutrons by an harmonic crystals has been the subject of theoretical investigations.[6-9] Gairola and Semwal[10,11] have obtained the expressions for differential cross section for the coherent inelastic scattering of neutrons by the ferroelectric crystals, by one phonon processes using modified Silverman-Joseph Hamiltonian in presence of an external electric field. The Silverman-Joseph Hamiltonian[12] is modified using the transformation according to the general scheme[12,13], taking into account anharmonic effects up to fourth order in presence of electric moment terms.

The present study differs with Gairola and Semwal[10,11] in view that they have obtained general expressions only, while we have taken the specific case of $Ba_xSr_{1-x}TiO_3$ perovskites. The effect of temperature and electric field upon inelastic scattering cross section of neutron in these perovskites is discussed using modified Silverman-Joseph Hamiltonian containing anharmonicity, defect and electric field and is exactly same as used in our previous studies[15,16] and thermal Green's function technique.[17]

Formulation

The temperature and field dependence of differential scattering cross section[11], is proportional to $\Gamma_k^{\lambda}(\omega)$ (half width) and is given as

$$\left(\frac{d^2\sigma_{coh}^{\lambda}}{d\Omega\, d\varepsilon}\right) = \int_{-\infty}^{+\infty}\sum_k A^{\lambda}(k)\,\frac{\Gamma_k^{\lambda}(\omega)}{\left(\omega^2-\sigma_k^{\lambda^2}(\omega)\right)^2}\,d\omega \quad , \qquad (1)$$

Where,

$$A^{\lambda}(k) = \left(\frac{Na^2}{\Pi h}\right)\frac{|q|}{|q_0|}\Delta(Q-K)\,|\,f(Q,k)\,|^2\,\omega_k^{\lambda}\,\frac{\exp(\beta hw)}{\exp(\beta hw)-1} \qquad (2)$$

Here,

$$\varepsilon_k^{\lambda^2}(\omega) = \overline{\overline{\omega}}_k^{\lambda^2} + 2\omega_k^{\lambda}\Delta_k^{\lambda}(\omega) \qquad (3)$$

With $\lambda = 0$, a and $\overline{\overline{\omega}}_k^{\lambda}$ field and defect dependent frequency. $\Delta_k^{\lambda}(\omega)$ is temperature, field and defect dependent shift.

Also Γ_k^{λ} is the half width of the response function. The notations used are the same and in same sense as used in reference.[11]

The values of the acoustical shift and width are the same and used in the same sense as given in reference 18, while the optical shift and width are given in reference 19.

Result, Discussion and Conclusion

In the present study using the approach of reference 10 and 11, we have tried to show the temperature and field variation with the inelastic differential scattering cross section of neutrons for $Ba_xSr_{1-x}TiO_3$ ferroelectric perovskites. The differential scattering cross section in these crystals can be expressed as

$$\left(\frac{d^2\sigma_{coh}^{\lambda}}{d\Omega\, d\varepsilon}\right) = A_0 + A_1T + A_2T^2 + A_3E^2 + A_4E^2T \qquad (4)$$

Here A_0 is purely defect dependent and A_1-A_4, are temperature and field independent constants in equation (4). A_3 and A_4 are also modified in presence of impurity. The values of A_0-A_4 for these perovskites are summarized in the table 1. Table 2 represent Curie temperature in $Ba_xSr_{1-x}TiO_3$ for different values of x and electric field (E).

Table 1

Perovskite	A_0	A_1	A_2	A_3	A_4
$BaTiO_3$	-	698.04555	0.2346774	4.74×10^5	1200
$Ba_{0.5}Sr_{0.5}TiO_3$	107.85	700.14320	0.7045213	12.8200×10^5	1272
$Ba_{0.2}Sr_{0.8}TiO_3$	211.96	2.90012	11.3637×10^{-3}	2.3500×10^5	3194
$SrTiO_3$	-	2.54014	9.9546×10^{-3}	1.15884×10^5	3132

Table 2 : Calculated values of Curie temperature in $Ba_xSr_{1-x}TiO_3$ for different values of x and electric field (*E*)

	E (k V/cm)	0	100	200	300	400
T_C(K)	$SrTiO_3$	37	37.19	37.38	37.57	37.76
	$Ba_{0.2}Sr_{0.8}TiO_3$	105	105.19	105.38	105.57	105.76
	$Ba_{0.5}Sr_{0.5}TiO_3$	218	218.19	218.38	218.57	218.76
	$BaTiO_3$	395	395.19	395.38	395.57	395.76

Using the above constants we have shown the variation of differential scattering cross section (logarithmic) with temperature taking electric field as a parameter in the cases of $Ba_xSr_{1-x}TiO_3$.

It is evident that the cross section increases with increasing temperature and electric field in all the three cases. In the cases of $SrTiO_3$, the electric field enhances the scattering cross section remarkably while in the case of $BaTiO_3$; such remarkable effect is not observed. This is due to the high Curie temperature of $BaTiO_3$.

The soft mode frequency $\Omega\left(\approx \varepsilon_k^{\lambda}(\omega)\right)$ field in these perovskites have been well discussed by Kukreti et al[20] and it has been shown that the square of effective soft mode frequency varies as the square of the electric field in agreement with the experimental results.[21] The influence of electric field on this mode also affects the interaction of soft modes $\left(\Omega \approx (T-T_C)^{1/2}\right)$ with other modes in presence of higher order anharmonic terms, thus giving electric field dependence of various properties. Soft mode frequency is held responsible for the anomalous behaviour of these perovskites near the phase transition temperature. The scattering cross-section increases with increasing electric field and defect.

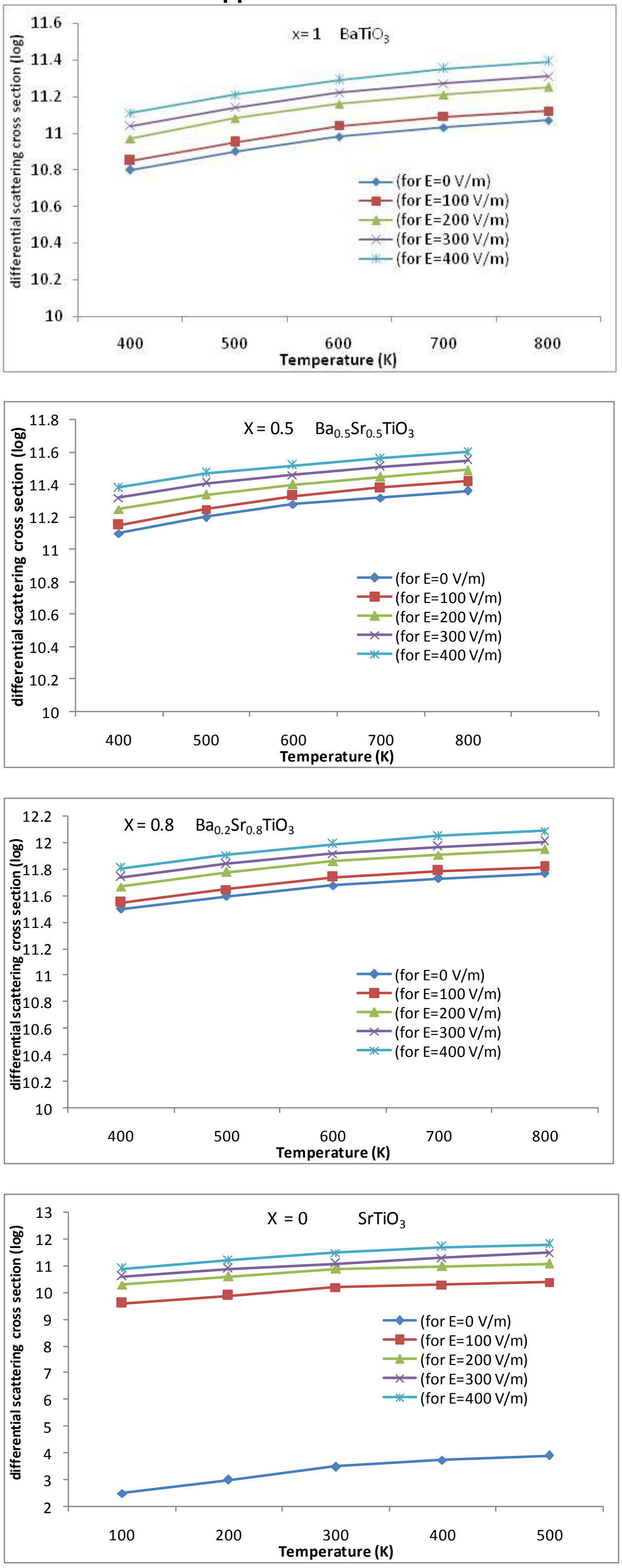
Recently we have used Green's function technique[17], in obtaining the expression for dielectric constant and loss tangent[20,22,23] in pure and mixed ferroelectric perovskites. The structural, dielectric and electrical properties of Leas Zirconate Titanate and $CaCu_3Ti_4O_{12}$ ceramic composite[24] and (1-x) $BaTiO_3$-x PZT ceramics have been experimentally measured by us.

Neutron scattering techniques have been used to study ceramics almost from the birth of neutron scattering in the 1940's, and provides microscopic information on the atomic structure and dynamics of materials. As well as advancing our fundamental understanding of condensed matter, neutron scattering has made important contributions to a wide range of technologically important materials, ranging from bio-polymers to exotic superconductors. A review on neutron scattering in condensed matter can be found in the literature.[25]

Conclusion

There has been considerable interest in the physical properties of mixed crystals, as they find interesting applications and their study helps in understanding basic mechanism of mixed crystal formation. Dependence of physical properties of mixed crystals varies from system to system. The property may change in a linear or non-linear manner. For example solid solutions of $BaTiO_3$ with other ferroelectrics of the same class and also with certain compounds, which are not themselves ferroelectric, possess ferroelectric properties and change in the composition of the solid solution makes it possible to regulate the Curie point within broad range of temperature. Among all these perovskites of mixed systems $Ba_xSr_{1-x}TiO_3$ has been identified as the leading material for under cooled detector fabrication[1], photo refractive mirrors [26,27] and as a gate insulator of the oxide super conductors FET in the thin film.[28] A recent review of Neutron scattering cross-section in mixed crystals is available in references 28 and 29 and references therein.

Differential scattering cross section of neutrons in $Ba_xSr_{1-x}TiO_3$ (logarithmic) vs. temperature for different values of applied external electric field.



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