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Electric Field Dependent Inelastic Scattering Cross Section of Neutrons in $BaTiO_3$, $SrTiO_3$ and $KTaO_3$ Displacive Ferroelectric Perovskites

Gaurav Kumar Aggarwal*, Ashok Kumar and U.C. Naithani

Department of Physics, H.N.B. Garhwal Central University,
Pauri Campus, Pauri Garhwal, Uttarakhand, India
**Corresponding Author Email: gkagg007@yahoo.co.in*

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 $BaTiO_3$, $SrTiO_3$ and $KTaO_3$ displacive ferroelectric perovskites in the presence of external electric field by considering higher order harmonic terms in the Hamiltonian. At any temperature well above the Curie temperature T_C, the differential scattering cross section increases with the increasing temperature and electric field in these perovskites. In the vicinity of T_C it is the long wavelength Cochran soft modes which gives the anomalous behaviour of the scattering cross section.

Keywords: Displacive ferroelectrics; anharmonic effect; soft mode; neutron scattering cross section.

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 ($BaTiO_3$), strontium titanate ($SrTiO_3$) and potassium tantalate ($KTaO_3$).

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 [1-3] on displacive ferroelectrics have shown that several properties of ferroelectric crystals can be understood in terms of lattice dynamics [4] of these crystals. The coherent scattering of neutrons by anharmonic crystals has been the subject of theoretical investigations [5-8]. Gairola and Semwal [9, 10] 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 [10] in presence of an external electric field. The Silverman-Joseph Hamiltonian [11] 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 [9, 10] in view that they have obtained general expressions only, while we have taken the specific cases of $BaTiO_3$, $SrTiO_3$ and $KTaO_3$ 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 equation (1) of reference 10 and thermal Green's function technique [14]. The notations used here are the same and in the same sense as in references 9 and 10.

Formulation

The temperature and field dependence of differential scattering cross section [10], is proportional to $\Gamma_k^\lambda(\omega)$ (half width) and is given as

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

where,

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

here,

$$\varepsilon_k^{\lambda^2}(\omega) = \varpi_k^{\lambda^2} + 2\omega_k^\lambda \Delta_k^\lambda(\omega) \tag{3}$$

With λ = 0, a and ϖ_k^λ is the field dependent frequency.

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 10.

The values of the acoustical shift and width are given by equations 10(a) and 10(b) of reference 10 while the optical shift and width are given by equations 13(a) and 13(b) of reference 10. Obtaining the general expression Gairola and Semwal have discussed the field and temperature dependence of scattering cross section.

Result, Discussion and Conclusion

In the present study using the approach of reference 9 and 10, we have tried to show the temperature and field variation with the inelastic differential scattering cross section of neutrons for $BaTiO_3$, $SrTiO_3$ and $KTaO_3$ ferroelectric perovskites. The differential scattering cross section in these crystals can be expressed as

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

where A_1-A_4, are temperature and field independent constants in equation (4).

The values of A_1-A_4 for these perovskites are summarized in the following table [15].

Perovskite	T_C	A_1	A_2	A_3	A_4
$BaTiO_3$	395 K	698.04555	0.2346774	4.74×10^5	1200
$SrTiO_3$	37 K	2.54014	9.9546×10^{-3}	1.15884×10^5	3132
$KTaO_3$	13 K	7.885873	6.30378×10^{-3}	7.8×10^4	6000

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 $BaTiO_3$, $SrTiO_3$ and $KTaO_3$ in the figures 1, 2 and 3 respectively.

It is evident that the cross section increases with increasing temperature and electric field in all the three cases. In the cases of $SrTiO_3$ and $KTaO_3$, 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 $\Omega(\approx \epsilon_k^\lambda(\omega))$ variations with temperature and electric field in these perovskites have been well discussed by Kukreti et al [16] 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 [17]. The influence of electric field on this mode also affects the interaction of soft modes $(\Omega \approx (\mathrm{T} - \mathrm{T_c})^{\frac{1}{2}})$ 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.

Recently we have used Green's function technique [14], in obtaining the expression for dielectric constant [16, 18 and 19] and loss tangent [20, 21 and 22] in pure and mixed ferroelectric perovskites. The structural, dielectric and electrical properties of Leas Zirconate Titanate and $CaCu_3Ti_4O_{12}$ ceramic composite [23] 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 [24].

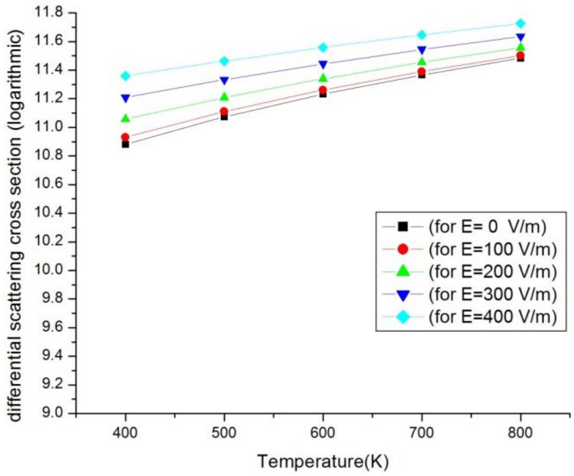

Figure 1: Differential scattering cross section of neutrons in $BaTiO_3$ (logarithmic) vs. temperature for different values of applied external electric field.

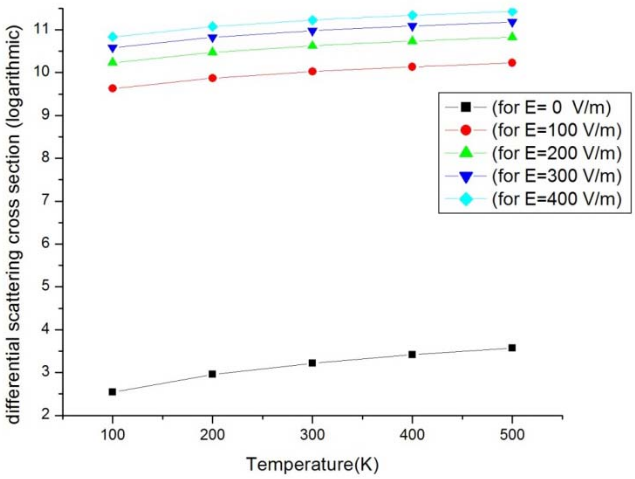

Figure 2: Differential scattering cross section of neutrons in $SrTiO_3$ (logarithmic) vs. temperature for different values of applied external electric field.

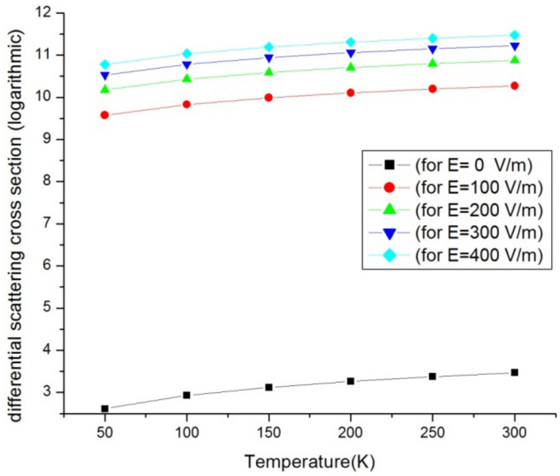

Figure 3: Differential scattering cross section of neutron in $KTaO_3$ (logarithmic) vs. temperature for different values of applied external electric field.

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