Temperature Dependence of Polarizability in Sodium Potassium Tantalate Mixed Ceramic System

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ABSTRACT

Using a quadratic anharmonic model Hamiltonian and using double time temperature dependent Green's function method and Dyson's equation treatment, expression for polarizability in the frequency response for mixed perovskite type ferroelectrics have been obtained. Using the experimentally observed temperature dependent dielectic constant, loss tangent, soft mode frequency, width for $Na_{1-x}K_xTaO_3$ (x=0,0.2,0.4,0.5 and 0.6) the polarizability has been calculated for these samples at 10kHz

Keywords : Polarizability, ferroelectrics, Model Hamiltonian

I. INTRODUCTION

The study of polarizability factor for $Na_{1-x}K_xTaO_3$ ceramics is important because it was realized that the large nonlinear polarizability of ferroelectrics makes them one of the most promising classes of materials for electro-optical and optical parametric devices.

A self-consistent of the soft mode frequencies was first given by Boccara and Sarma[1] by employing at the onset a renormalized phonon basis. Their formal treatment represented (the lowest order of) what is now called the self-consistent phonon approximation (SPA)[2]. This approximation has been very successful in describing the anharmonic rare-gas solids, including the quantum crystals of solid helium.

Numerical calculations have shown that the SPA gives a first – order transition for a model ferroelectric containing only fourth order anharmonic interactions [3]. This result is surprising because the phenomenological Landau (Devonshire) theory predicts the transition to be second order when only terms up to fourth order in the polarization are included [4-6].

II. METHODS AND MATERIAL

To illustrate the essential features of the self-consistent phonon approximation (SPA)[2] and to understand why a first-order transition is obtained, it is instructive to consider a simple model with a single degree of freedom,

$$H = 1/2\sum P_l^2(l) + \tfrac{1}{2}\Omega_o^2\sum_l Q_l^2(l) - 1/2\sum_{ll}V(ll`)Q_l(l)Q_{l`}(l`) + 1/2\Gamma_1\sum Q_l^4(l) \quad \ldots \ldots (1)$$

Here Q_l is a localized normal mode coordinate describing the ion displacements in cell l, and P(l)is the canonical conjugate momentum

$$[Q_l, P_l^{'}] = i\delta_{ll'} \qquad \ldots\ldots\ldots(2)$$

We set $Q_l = Q_0 + U_l$, where the thermal average $Q_0 = \langle Q_l\rangle$ measures the distortion from the high temperature structure, while U_l describes the fluctuations about the average value. In the SPA the free energy $F = \langle H\rangle - TS$ is obtained by using a harmonic trial density matrix[2]. The distortion Q_0 and the effective harmonic force constants are determined

by minimizing the free energy. For the Hamiltonian given by Eq. (7) the extremum condition, $\partial F/\partial Q_0 = 0$ takes the form

$$Q_0[\Omega_0^2 - v(0) + r_1 Q_0^2 + 3 r_1 \Delta] = 0 , \quad \ldots\ldots(3)$$

Where $v(0) = \sum_{ll`} V(ll`)$, and Δ is defined below. The effective force constants determine the self-consistent normal mode frequency. For this model it is given by

$$\omega_q^2 = \Omega^2 + v(0) - v(q) \sim \Omega^2 + \alpha^2 q^2 , \quad \ldots\ldots(4)$$

where

$$\Omega^2 = \Omega_0^2 - v(0) + 3 r_1 Q_0^2 + 3 r_1 \Delta , \quad \ldots\ldots\ldots(5)$$

Or, in the distorted phase, using equation (9)

$$\Omega^2 = 2 r_1 Q_0^2 \quad \ldots\ldots\ldots\ldots(6)$$

The correlation function $\Delta = \langle U_i U_j \rangle$ is determined with the help of the fluctuation dissipation theorem[6]

$$\Delta = N^{-1} \sum_q \tfrac{1}{2} \omega_q^{-1} \coth \tfrac{1}{2}\beta \omega_q , \quad \ldots\ldots\ldots(7)$$

for simplicity we consider the limit $\omega_q/kT \ll 1$, Δ may be approximated by the Ornstein- Zernike form

$$\Delta = kTN^{-1}\sum_q (\Omega^2 + \alpha^2 q^2)^{-1} \quad \ldots\ldots\ldots\ldots(8)$$

Evaluating the summation in the Debye-approximation, we obtain

$$\Delta = \Delta_0 - \Delta_t \quad \ldots\ldots\ldots\ldots(9)$$

where $\Delta_0 = 3kT/\omega_d^2$ and

$$\Delta_T = \Delta_0(\Omega/\omega_d)\tan^{-1}(\omega_d/\Omega),$$

On substituting in equation (9), we get

$$\Delta = 3kT/\omega_d^2 - \Delta_0(\Omega/\omega_d)\tan^{-1}(\omega_d/\Omega)$$
$$= 3kT/\omega_d^2 - 3kT/\omega_d^2 (\Omega/\omega_d)\tan^{-1}(\omega_d/\Omega)$$
$$= 3kT/\omega_d^2 [1 - (\Omega/\omega_d)\tan^{-1}(\omega_d/\Omega)] , \ldots(10)$$

where $\omega_d = \alpha q_D$, q_D being the Debye wave vector. Δ_T denotes the contribution to Δ due to long wavelength fluctuations. For $Q_0 \neq 0$, Eq. (9) may be written

$$a(T-T_c) + r_1 Q_0^2 - 3 r_1 \Delta_T = 0 , \ldots\ldots (11)$$

where

$$a = 9 r_1 K_B/\omega_d^2 , \quad T_c = a^{-1}[v(0) - \Omega_0^2] \ldots (12)$$
$$\text{and } v(0) = T_c a + \Omega_0^2 \ldots(13)$$

Because Δ_T is linear in Ω as $\Omega \to 0$, it follows from Eqs. (6) and (11) that the transition is first order. However it is important that the linear term giving rise to the first order transition is entirely due to the long wavelength fluctuations.

On substituting Eq.6, 7 and 13 in Eq.5, we get the expression as

$$Q_0^2 = 9 K_B/\omega_d^2 [T_c - \{1 - \Omega/\omega_d \tan^{-1}\omega_d/\Omega\}T] \quad \ldots(14)$$

On substituting the experimental results of soft mode frequency (Ω), natural frequency of the system (ω_d) for $Na_{1-x}K_xTaO_3$ (x=0,0.2,0.4,0.5 and 0.6) system , we get the variation of polarizability factor (Q_0^2) with temperature is given in table 1 and fig.1 at 10Khz. Experimental results for $Na_{1-x}K_xTaO_3$ system has been obtained by S.Glinsek et.al.[7]. The result has been obtained by using our previously experimental results[8-13] and the theoretical data by earlier workers[4,6,14-16].

Table-1: Variation of Polarizability factor with temperature for $Na_{1-x}K_xTaO_3$ system at 10 kHz

Temp(k)	$NaTaO_3$ $(\times 10^{-47})$	$Na_{0.8}K_{0.2}TaO_3$ $(\times 10^{-47})$	$Na_{0.6}K_{0.4}TaO_3$ $(\times 10^{-47})$	$Na_{0.5}K_{0.5}TaO_3$ $(\times 10^{-47})$	$Na_{0.4}K_{0.6}TaO_3$ $(\times 10^{-47})$
423	8.293	8.69	8.866	9.668	10.558
443	8.245	8.645	8.766	9.588	10.508
463	8.191	8.601	8.697	9.561	10.466
483	8.142	8.522	8.655	9.443	10.333
503	8.093	8.501	8.635	9.357	10.245
523	8.038	8.498	8.581	9.297	10.198
543	7.99	8.47	8.542	9.262	10.101

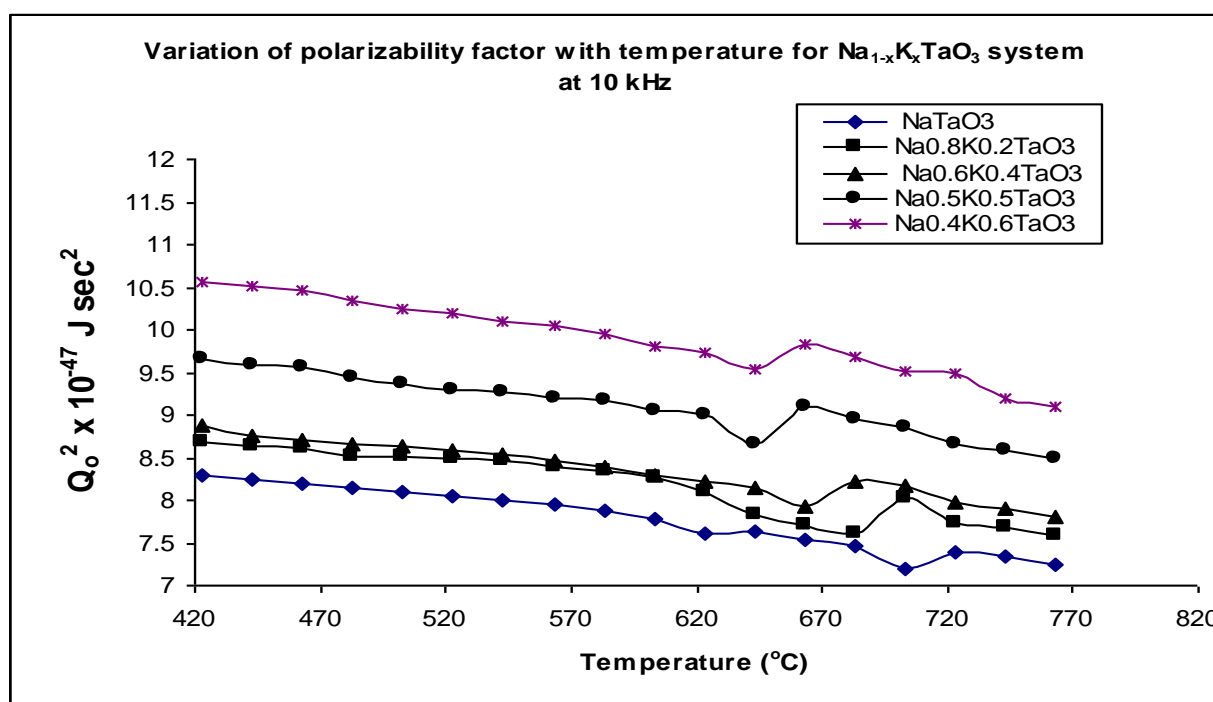| 563 | 7.941 | 8.402 | 8.467 | 9.196 | 10.057 |
583	7.885	8.35	8.388	9.16	9.945
603	7.781	8.258	8.301	9.048	9.807
623	7.606	8.105	8.214	8.997	9.721
643	7.626	7.821	8.143	8.662	9.535
663	7.541	7.705	7.939	9.09	9.82
683	7.457	7.601	8.226	8.95	9.675
703	7.197	8.018	8.17	8.86	9.502
723	7.402	7.721	7.964	8.667	9.485
743	7.331	7.688	7.907	8.589	9.207
763	7.244	7.587	7.815	8.488	9.106

Figure 1

III. RESULT AND DISCUSSION

Polarizability factor (Q_0^2) is calculated by using the formula obtained by Pytte [14]. The temperature dependence of polarizability factor at 10 kHz frequency for $Na_{1-x}K_xTaO_3$ has been shown in figure 1. For $Na_{1-x}K_xTaO_3$ [x=0, 0.2, 0.4, 0.5 and 0.6] transition temperature are found at 703°C, 683°C, 663°C, 643°C and 640°C respectively. From these figures it is observed that polarizability factor tends to minimum up to the transition temperature thereafter increases with the increasing temperature and hence showing anomaly at transition temperature. The nature of polarizability factor is found to be in good agreement with the results obtained theoretically by Pytte [15].

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