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Effect of electric field on elastic properties of $BaTiO_3$ single crystals: A micro-Brillouin scattering study

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1. Introduction

Ferroelectric materials are widely used in sensing and transducing applications due to their superior electromechanical coupling along certain crystallographic axes. In the last several decades, barium titanate ($BaTiO_3$, abbreviated as BT) is one of the most extensively studied ferroelectric material due to its very simple and well known crystal structures. In spite of extensive studies on BT crystals, the nature of the ferroelectric phase transitions still a controversial matter of discussion. Displacive-type phase transition has been considered to occur in BT [1,2]. The existence of the order-disorder character of phase transitions has been studied by means of several experiments and theories [3,4]. Burns and Dacol observed the deviation of the optical index of refraction from the high-temperature linear behavior and ascribed it as the existence of precursor local polarization in the paraelectric phase above the Curie temperature, T_C [5,6]. These precursor polar clusters are local, non-cubic regions having a broken inversion symmetry and a local polarization.

Nowadays, it is believed that the relaxor nature of a ferroelectric material is mainly caused by the existence of polar nanoregions (PNRs) which appears few hundreds degree above T_C so called the Burns temperature (T_B). In BT, some phenomena typically related to the relaxation dynamics of PNRs, were observed. In addition, inelastic light scattering technique revealed the existence of central peaks (CPs) above T_C indicating the existence of PNRs [7,8]. In spite of all previous studies on this important class of ferroelectric materials, a detailed microscopic view on the exact role of PNRs during the phase transition still remain unclear. In addition, there has been no discussion on the electric field induced acoustic anomalies in a paraelectric cubic phase and/or in a ferroelectric tetragonal phase. In this respect, in the present study, the Brillioun scattering technique has been applied to examine the effect of electric field on the acoustic properties and PNRs in BT single crystals.

2. Experimental

The (100) oriented BT single crystals with the dimensions of 5 × 5 mm^2 and thickness of 1 mm was used after polishing its surfaces to optical quality. A micro-Brillouin scattering system with a 3+3 pass Sandercock-type tandem Fabry-Pérot interferometer was used to measure the Brillouin spectra at a backward scattering geometry. The exciting light source was a single frequency diode pump solid state (DPSS) laser with a wavelength of 532 nm and a power of about 100 mW. In order to study the effect of an electric field, the two surfaces, perpendicular to [100] direction, were coated with silver paste along [100] direction. In addition, gold contact wires were attached to the electrodes.

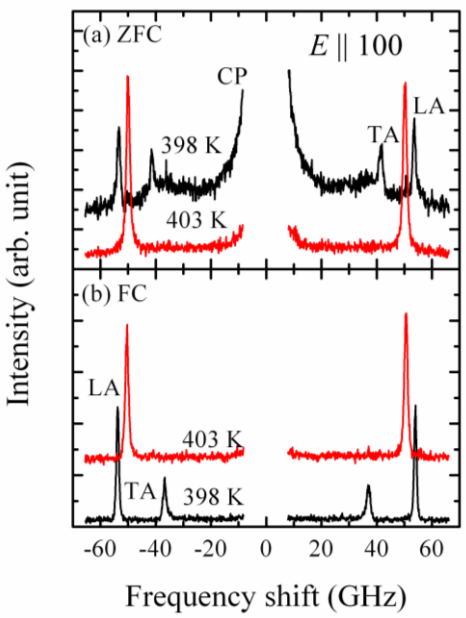
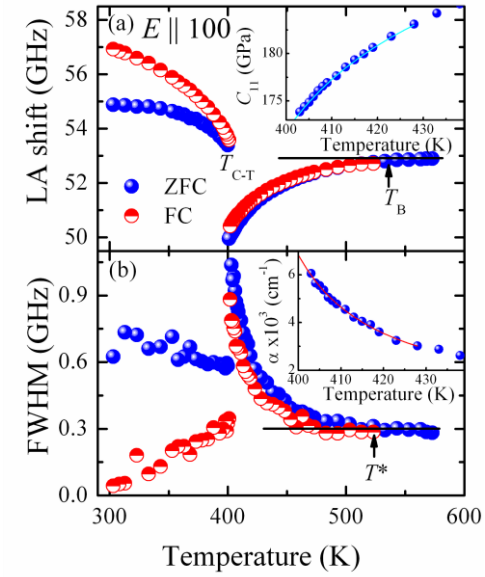


Fig. 1. Brillouin spectra of 100-BT single crystal in (a) ZFC and (b) FC process, respectively, at selected temperatures.

3. Results

Figures 1(a) and (b) show a set of measured Brillouin scattering spectra of a BT crystal under zero field cooling (ZFC) and field cooling (FC) process, respectively, at selected temperatures. At 403 K, as shown in Fig. 1 (a),

Brillouin spectrum consists of one longitudinal acoustic (LA) mode and a weak central peak (CP). The appearance of a transverse acoustic (TA) mode below T_{C-T} (~ 402 K) clearly reflects a ferroelectric tetragonal phase. In addition, the intensity of CP becomes stronger in the tetragonal phase. In order to observe the effect of an electric field on the Brillouin spectrum, the 1 kV/cm field was applied at 533 K and then the spectra were measured on FC process. The temperature dependences of the frequency shift (υ_{LA}) and FWHM (Γ_{LA}) of LA phonon under the ZFC and FC process are shown in Figs. 2 (a) and (b), respectively. The deviation of the elastic constant (related to υ_{LA}) from its normal anharmonic behavior at around 533 K is correlated to the formation of dynamic PNRs and is assigned as T_B. In Fig. 2 (b), on ZFC, the sound attenuation (related to Γ_{LA}) of the LA phonon starts to increase rapidly towards the T_C at around 503 K where the permanent PNRs appear and is called the intermediate temperature, T^*.



ig. 2. The temperature dependences of (a) frequency shift (υ_{LA}) and (b) FWHM (Γ_{LA}) of LA phonons of 100-BT single crystals measured in ZFC (blue solid symbols) and FC (red solid half-filled symbols) processes.

In BT, the weak random fields (RFs), which creates the PNRs, exist. It is seen from Fig. 2, upon FC, the υ_{LA} and Γ_{LA} show noticeable differences in both the cubic and tetragonal phases. It is already established that in the cubic phase the local dipole moment in PNRs fluctuates along [111] directions. Therefore, under the electric field along a [100] direction, the density of static/dynamic PNRs decreases which is expected to weaken the scattering of LA phonons by PNRs, resulting in higher frequency and lower damping of LA phonons. Since the electric field was applied in a paraelectric cubic phase, therefore, below the T_{C-T}, the present ferroelectric axis along [100] changes to [001] direction. As a result, the υ_{LA} and Γ_{LA} shows significant changes. Therefore, the large difference of υ_{LA} and Γ_{LA} between ZFC and FC process in the tetragonal phase indicates the transformation from the ferroelectric [100] to [001] direction.

The frequency shifts of the LA and TA phonons as a function of electric field were also investigated at 303 K. At around 2.94 kV/cm, an additional TA mode begins to appear and it persists up to 3.9 kV/cm which indicates the coexistence of *a*- and *c*-domains, respectively. On the other hand, from 2.94 kV/cm to 3.9 kV/cm, the intensity of the additional TA mode increases rapidly. This fact shows that *a*-domains begins to transform towards the *c*- domains.

4. Conclusions

In summary, the significant softening of υ_{LA} and the increase of Γ_{LA} were observed in the vicinity of a cubic to tetragonal phase transition, T_C ~ 402 K. Under the applied electric field at 533 K, the increase of υ_{LA} and decrease of Γ_{LA} were found due to the reduced density of twin domain walls and static/dynamic PNRs. In the vicinity of T_C, the temperature dependence of sound attenuation was analyzed by the theoretical model indicating the order-disorder nature of the ferroelectric phase transition in BT. A complete 90 degree domain switching process at 303 K was observed which could be very helpful for deep understanding of domain switching mechanism in the field of ferroelectrics.

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