# ブリルアン散乱法によるチタン酸バリウム単結晶の 分極反転についての電場効果の研究

Effect of Electric Field on Domain Switching in BaTiO<sub>3</sub> Single Crystals Probed by Brillouin Scattering

H28海自66

派遣先	International Conference on Technologically Advanced Materials
	and Asian Meeting on Ferroelectricity (ICTAM-AMF10)
	(University of Delhi, New Delhi, India)
期 間	平成28年11月5日~平成28年11月12日(7日間)
申請者	筑波大学 数理物質科学研究科 物性·分子工学専攻
	博士後期課程2年 HELAL MD AL

# 海外における研究活動状況

#### 研究目的

Ferroelectric materials are widely used in sensing and transducing applications due to their superior electromechanical coupling along a certain crystallographic axes. The ability of ferroelectric materials to transform electromagnetic, thermal and mechanical energy into electrical energy has made it useful for the practical applications. In the last several decades, barium titanate (BaTiO<sub>3</sub>, abbreviated as BT) is one of the most extensively studied ferroelectric material due to its very simple and well known crystal structures. Recently, to enhance the piezoelectric properties of ferroelectric materials, considerable attention has been devoted owing to its possible applications in new generation devices. One way to enhance the piezoelectric response is to control the domain configuration by the application of electric field and mechanical stress. Domain switching can induce the mechanical

stress and electric fields which may lead to the failure behavior of ferroelectric materials. In this respect, it is necessary to study the field induced domain switching mechanisms for applications and development of ferroelectric materials.

# 海外における研究活動報告 Conference overview

The International Conference on Technologically Advanced Materials and Asian Meeting on Ferroelectricity (ICTAM-AMF10) was held during November 7-11, 2016 at University of Delhi, New Delhi, India. The total number of delegates joined there at around 694 from more than 30 countries including USA, UK, Japan, South Korea, China, Thailand, Germany, Bangladesh and many more. There were plenty of lectures and poster presentations. Among all of them, there were 12 plenary lectures, 127 invited talks, 82 oral lectures and around 473 poster presentations. The six parallel sessions were held in six different rooms and several interesting topics

including (i) Dielectric, Piezoelectric, Ferroelectric and Related Materials, (ii)Nanomaterials and Applications, (iii) Carbonmaterials: Graphene, Lead Free Materials, Oxides and Composites, (v) Electronic and Spintronic Materials, (vi) Piezoelectric Materials and Sensors, (vii) Multiferroic and magnetic Materials, (viii) Advancedand Smart Materials and Electronic Device Applications, (ix) Energy Harvesting and Thermoelectric Materials and Devices, (x) Bio-inspired Materials and Biomedical Applications, (xi) Functional Materials Thin Films and Functional Materials, (xii) Epitaxial Grown Ferroelectrics and Characterization, and (xiii) Advanced Materials Characterization were delivered. Therefore, I believe that this conference was a unique meeting place for the students, young scientists and very famous researchers from the different branches of materials science to interact with each other, share knowledge, and develop new materials for diverse applications.

#### The research presented at ICTAM-AMF10

I presented a poster and it was held on  $9^{th}$  November, 2016 entitled "Effect of electric field on domain switching in BaTiO<sub>3</sub> single crystals probed by Brillouin scattering". At present advanced materials, especially ferroelectric materials have played a major role in shaping the modern civilization. Ferroelectric materials and technologically very important due to their unique combination of physical properties. The ability of ferroelectric materials to transform electromagnetic, thermal and mechanical energy into electrical energy has made it useful for the practical applications such as capacitor, transducers,

ceramic filters, actuators, modulators, ultrasonicgenerators, high-voltage generators, optical displays and most recently non-volatile computer memories etc. In the last several decades, barium titanate (BaTiO<sub>3</sub>, abbreviated as BT) is one of the most extensively studied ferroelectric material due to its very simple and well known crystal structures.

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 Curie temperature,  $T_{\rm C}$  so called the Burns temperature  $(T_{\rm B})$ . In BT, some phenomena typically related to the relaxation dynamics of PNRs, namely, deviation from the linear temperature dependence of the index of refraction, significant softening of the longitudinal acoustic (LA) frequency, and deviations from the Curie-Weiss law for the permittivity were observed. In addition, inelastic light scattering technique revealed the existence of central peaks (CPs) above  $T_{C-T}$  indicating the existence of PNRs. 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 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. The domain switching mechanism and field induced phase transition are also studied.

### **Discussion and Conclusions**

Electric field induced domain switching and structural phase transition have been studied in (100)-orientated BaTiO<sub>3</sub> single crystals by

micro-Brillioun scattering. The deviation of the elastic constant (related to frequency shift of LA phonon) from its normal anharmonic behavior at around 533 K is correlated to the formation of dynamic PNRs and is assigned as Burns temperature,  $T_{\rm B}$ . On zero field cooling (ZFC), the sound attenuation of the LA phonon starts to increase rapidly towards the  $T_{C-T}$  at around 503 K where the permanent PNRs appears and is called the intermediate temperature,  $T^*$ . Upon further ZFC, both the frequency shift and FWHM of LA phonon show sudden jump at around 402 K suggesting the transition from cubic to tetragonal phase. During the field cooling (FC) process, the scattering of a LA phonon weakens and shift to higher frequency compared with that of the ZFC process, being attributed to the decrease of the number density or size of the static/dynamic PNRs. In the vicinity of the cubic to tetragonal phase transition, the order-disorder character of the ferroelectric phase transition is discussed by the analysis of the sound attenuation based on the perturbation theory. At 303 K, the field dependence of frequency shift and FWHM of LA phonon shows a sudden jump at around 3.9 kV/ cm which indicates the change of sound velocity from [100] to [001] direction. From these two different LA and transverse acoustic (TA) phonons, we have calculated the four different elastic constants which are consistent with the previous report. In a paraelectric cubic phase, the field induced cubic to tetragonal phase is observed from the abrupt changes in the frequency shift and FWHM of LA phonon. The critical electric field to induce the transition is found to shift to higher values with increasing temperature. In the previous study, the microscopic origin of the

strong CP observed in BT single crystals was ascribed to the off-centered motions of the Ti ions in the oxygen octahedral. The polarization fluctuation induced by the relaxational motion of the off-centered Ti ions was thought to induce the quasielastic CP from many BT-based perovskite ferroelectrics. Therefore, in the present study the appearance of the CP in BT single crystals can be the evidence of PNRs.

In summary, the significant softening of both the frequency shift and FWHM of LA phonon were observed in the vicinity of a cubic to tetragonal phase transition,  $T_{C-T} \sim 402$  K. Under the applied electric field at 533 K, the increase of frequency shiftand decrease of FWHM of LA phonon were found due to the reduced density of twin domain walls and static/dynamic PNRs. In the vicinity of  $T_{C-T}$ , 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. An E-T phase diagram was determined and the slope was calculated by the Clausius-Clapeyron law. Under the application of the electric field in a paraelectric cubic phase, two phenomena were observed: (i) the CP in [100]-orientated BT suppressed, and (ii) the CP in [111]-orientated BT strongly enhanced. These two phenomena provide a clear evidence that the local structure of PNRs in BT single crystals is perfectly rhombohedral. These findings will be helpful for further research on ferroelectrics with weak random fields.

## この派遣の研究成果等を発表した 著書、論文、報告書の書名・講演題目

Md Al Helal, Md Aftabuzzaman, and Seiji Kojima "Effect of electric field on domain switching in BaTiO<sub>3</sub> single crystals probed by Brillouin scattering", International Conference on Technologically Advanced Materials and Asian Meeting on Ferroelectricity (ICTAM-AMF10), 2016.11.07, University of Delhi, India. (Poster). Preparing manuscript for submission to the peer reviewed journal.