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# Successive Phase Transition of Potassium Tantalate Niobate Crystals Studied by Broadband Brillouin Scattering

広帯域ブリルアン散乱による KTN 結晶の逐次相転移

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## 1. Introduction

Dynamical properties of ferroelectrics are very important topics, and they are closely related to a structural phase transitions. Brillouin scattering is one of the powerful tools to obtain the elastic property of materials at a GHz frequency range. The frequency shifts and full widths at half maximum (FWHMs) of acoustic modes are very sensitive to a phase transition. In addition a relaxation mode can be observed as a central peak (CP). Therefore, we can study not only elastic anomaly but also slowing down of relaxation time in the vicinity of a phase transition point.

Ferroelectric potassium tantalite niobate ($KTa_{1-x}Nb_xO_3$, KTN) with perovskite structure have attracted attention by the huge quadratic electro-optic coefficient [1]. Therefore, KTN is attractive for use in fabricating optical components [2]. KTN undergoes a successive phase transition of cubic–tetragonal–orthorhombic-rhombohedral sequence. In addition, phase transition temperatures linearly increase with increasing the niobate concentration [3]. And it shows the relaxor behavior due to the off-centering of Nb ions and the resulting formation of polar nanoregions (PNRs) [4].

In this study, we report the observation of a successive phase transition of KTN crystals by Brillouin scattering to make clear the elastic and relaxation properties of KTN crystal.

## 2. Experimental

Brillouin scattering spectra were measured by using a high contract 3+3 pass Sandercock tandem Fabry-Perot interferometer (JRS Scientific Instruments) combined with a reflection optical microscope (Olympus BH-2), a custom-built micro optical system and a conventional photon-counting system [5]. The free spectral range (FSR) of the spectrometer is fixed to be 75 GHz. Brillouin scattering was excited by a single frequency green YAG laser at wavelength of 532 nm with a power of 50 mW.

The synthetic KTN crystal was grown by the top seed flux growth, and cut into 5× 5× 1 $mm^3$ along the *a*, *b* and *c* axes in cubic coordinate system, respectively. The incident beam is focused on to a *c*-plane along the *c*-axis. The scattering geometry is the backward scattering geometry with the scattering angle =180° ($q||c$). Sample was put into a cooling/heating stage (Linkam THMS600) to change temperature of the sample from -190 °C to 600 °C.

## 2. Result and Discussion

**Fig. 1** shows the spectrum at four temperatures. Brillouin peaks scattered by longitudinal acoustic (LA) mode and transverse acoustic (TA) mode were clearly observed. At near zero frequency a broad central peak (CP) was also observed. Spectra show remarkable changes with temperature. In order to analyze the spectra, Voigt functions were used for fitting.

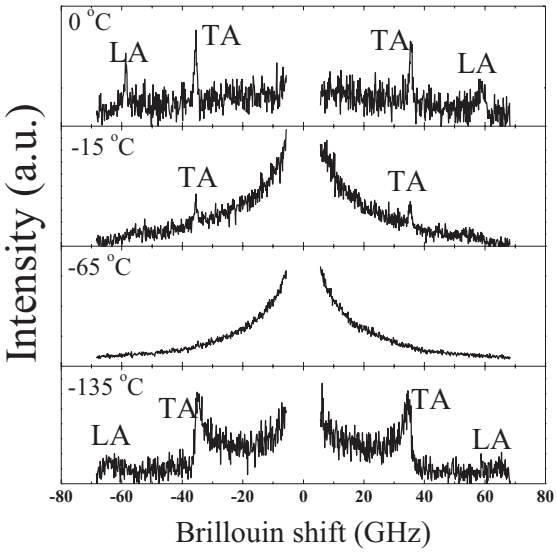


Fig. 1 Brillouin spectra of a KTN crystal at four temperatures. LA and TA denote the Brillouin components scattered by LA and TA phonons, respectively.

FWHM of a broad CP is originated from the dielectric relaxation of polarization fluctuations thorough the generalized LST relation [5, 6]. The relaxation time determined by assuming a Debye relaxation ($\tau = 1/\pi\Delta\Gamma$), where $\tau$ is the relaxation time and $\Delta\Gamma$ is FWHM of a CP. **Fig. 2** shows the

temperature dependence of relaxation time. The three anomalies of the relaxation time were clearly observed around each phase transition temperature of a KTN crystal at -15 $^{o}$C, -65$^{o}$C and -107 $^{o}$C. It is attributed to a cubic–tetragonal–orthorhombic-rhombohedral sequence of a successive phase transition. The increase of the relaxation time towards a cubic-tetragonal transition temperature reflects a growth in the volume fraction of PNRs and a decrease in the inter-site relaxation rate at the B-site [7, 8].

The temperature dependence of sound velocity of LA and TA modes is shown in **Fig. 3**. The softening of the sound velocity is clearly observed in the vicinity of the cubic-tetragonal transition temperature. On cooling from a high temperature, the LA velocity gradually decreases towards a phase transition temperature. In contrast, the TA velocity of TA mode is nearly constant except the very vicinity of a phase transition temperature. In the studies by the ultrasonic pulse echo technique, similar anomalies were observed [9]. The softening of these sound velocities was caused by the interaction of acoustic phonons with dynamically reorienting PNRs.

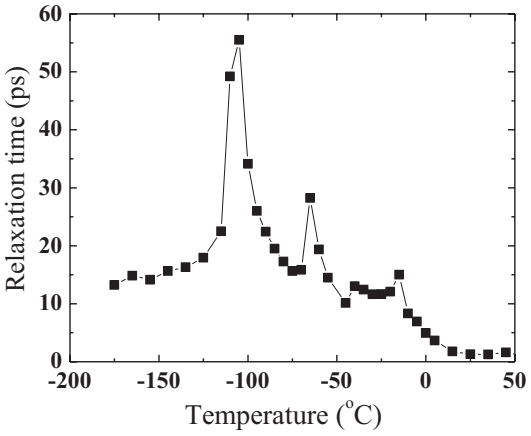


Fig. 2 Temperature dependence of relaxation time of polarization fluctuations determined by a broad CP.

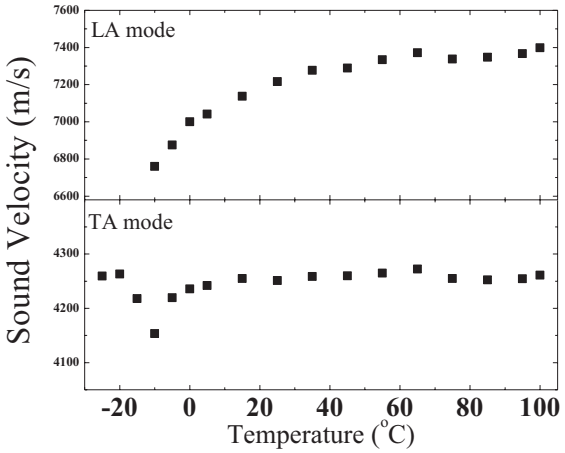


Fig. 3 Temperature dependence of sound velocity of LA and TA modes along the *c*-axis.

## 4. Conclusion

We study a successive phase transition of a ferroelectric KTN crystal with perovskite structure by broadband Brillouin scattering measurement. The relaxation time of polarization fluctuations is determined by the width of CP. The LA and TA sound velocities are determined by the LA and TA Brillouin shifts, respectively. In the vicinity of a phase transition temperature about $T_1$ = -15 $^{o}$C, the slowing of relaxation time and the softening of sound velocity of LA and TA modes towards $T_1$ are clearly observed. These anomalies are probably originated from polarization fluctuations of dynamic PNRs.

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