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Composition variation of elastic anomalies in uniaxial relaxor $Ca_xBa_{1-x}Nb_2O_6$ crystals

一軸性リラクサー結晶 $Ca_xBa_{1-x}Nb_2O_6$ の弾性異常の組成依存性

Kohei Suzuki[1†], Kazuya Matsumoto[1], J. Dec[2], T. Łukasiewicz[3], W. Kleemann[4], and Seiji Kojima[1]
([1]University of Tsukuba, [2]University of Silesia, Katowice, [3]Institute of Electronic Materials Technology, Warsaw, [4]Duisburg-Essen University)
鈴木康平 [1†]，松本和也 [1]，小島誠治 [1]，J. Dec[2], T. Łukasiewicz[3], W. Kleemann[4]([1] 筑波大院, [2] シレジア大学, [3] 電子材料研究所, [4] デュースブルク·エッセン大学)

1. Introduction

Tungsten-bronze ferroelectrics such as strontium barium niobate $Sr_xBa_{1-x}Nb_2O_6$ (SBN) are technologically important because of excellent dielectric, pyroelectric, and photorefractive properties. SBN shows a crossover from normal to relaxor ferroelectric behavior with increasing Sr content because of the enhancement of random fields mostly originating from the missing charges at A (*i.e.* A1 and A2) site vacancies in the tetragonal tungsten bronze (TTB) structure as shown in Fig. 1 [1]. In the TTB structure, the A1+A2 site occupancy is 5/6. It has, hence, a fraction of 1/6 vacancies [2]. On heating from room temperature, it undergoes a ferroelectric phase transition from *4mm* to *4/mmm*.

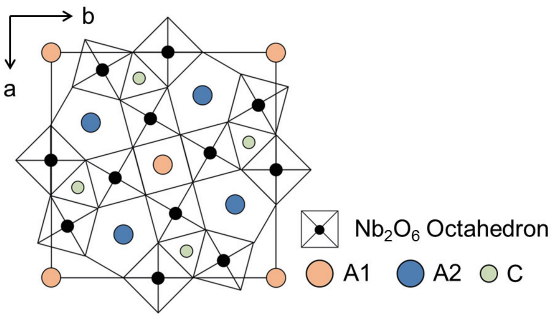

Fig. 1 Atomic arrangement in a unit cell of the tetragonal tungsten-bronze structure.

Calcium barium niobate $Ca_xBa_{1-x}Nb_2O_6$ (CBN) has physical properties similar to SBN, while its higher T_c is better for applications. The temperature dependences of the elastic properties of CBN ($0.18 \leqq x \leqq 0.35$) were studied very recently by the resonant ultrasonic spectroscopy (RUS) [3]. However, in RUS, it is difficult to measure elastic properties around T_c because of the large acoustic absorption. In contrast, in Brillouin scattering it is possible to measure even, if a noticeable increase of absorption occurs.

In the present study, elastic anomalies and relaxation dynamics of CBN ($0.26 \leqq x \leqq 0.32$) were investigated by Brillouin scattering.

2. Experimental

$Ca_xBa_{1-x}Nb_2O_6$ (x=0.26, 0.28, 0.30, 0.32) single crystals (CBN26, CBN28, CBN30, CBN32) were grown by the Czochralski method [4]. Single crystal plates were cut along [100] (*a*-plate) and [001] (*c*-plate) with optically polished 5mm×5mm surfaces with 1mm thickness. Brillouin scattering was measured using a tandem Fabry-Perot interferometer and a single frequency green YAG laser with wavelength 532 nm [5]. The elastic anomalies were studied in the large temperature range from room temperature up to 750 °C with the free spectral range (FSR) of 75GHz for the study of elastic anomalies. A broad central peak (CP) was also studied with a FSR of 300 GHz to clarify relaxation processes.

3. Result and discussion

The temperature dependence of the Brillouin scattering spectra observed in an *a*-plate of CBN26 (T_c = 276°C) is shown in Fig. 2. The doublets of longitudinal acoustic (LA) and transverse acoustic (TA) peaks are clearly observed at all temperatures. The broadening of the CP is also observed near T_c.

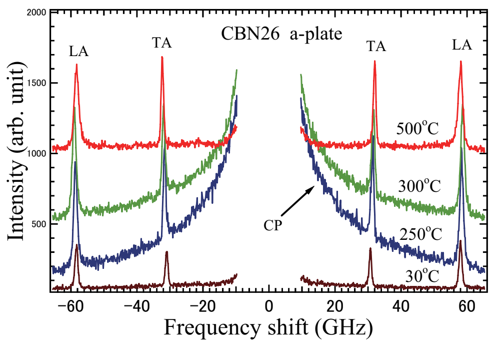

Fig. 2 Temperature dependence of Brillouin scattering spectra of an *a*-plate of CBN26.

The LA mode frequency observed in an *a*-plate shows the monotonic increase on heating from room temperature, while that observed in a *c*-plate shows a sharp minimum in the vicinity of T_C. Sound velocity and acoustic absorption coefficient were calculated by the equations, $V=\pi\nu_B/2n_o$ and $\alpha=\pi\Gamma_B/V$, respectively, where λ and n_o denote the laser wavelength (532 nm) and the refractive index of ordinary ray. Figures 3 and 4 show the temperature dependences of sound velocity and acoustic absorption coefficient, respectively, of the LA mode observed in *c*-plates. It is found that the elastic anomaly becomes diffusive as the Ca content increases. The observed Curie temperatures are 276 °C (CBN26), 244 °C (CBN28), 218 °C (CBN30) and 190 °C (CBN32). In CBN, as the Ca content increases, T_c increases and the ferroelectric phase transition becomes diffusive because of the increase of the disorder. For further heating, it is found that the acoustic absorption coefficient shows a remarkable decrease at around 500°C, which can be the intermediate temperature T^* at which the rapid growth of PNRs starts.

The temperature dependence of a broad CP was also studied using an *a*-plate to determine the relaxation time of polarization fluctuations along the *c*-axis. The intensity of a broad CP becomes maximum in the vicinity of T_c as shown in Fig. 2. The width of a broad CP is inversely proportional to the relaxation time. The temperature dependences of relaxation time are shown in Fig. 5. In CBN26, the critical slowing down was clearly observed in the vicinity of T_c, reflecting the order-disorder nature of a ferroelectric phase transition. However, in CBN32, the critical slowing down is suppressed and a broad diffusive peak was observed. The phase transition temperatures well coincide with that stemming from dielectric measurements.

4. Conclusion

Elastic anomalies and slowing down of polarization fluctuations along the ferroelectric *c*-axis were studied in CBN ($0.26 \leqq x \leqq 0.32$) by Brillouin scattering. The LA mode propagating along an *c*-axis shows remarkable softening of sound velocity and marked increase of absorption coefficient in the vicinity of T_c. With the increase of Ca content, the anomaly becomes diffusive. The relaxation time of polarization fluctuations along a *c*-axis shows critical slowing down in CBN26, while a broad diffusive peak was observed in CBN32. These dynamical behaviors show that the anomaly near T_c becomes diffusive with the increase of Ca content.

References

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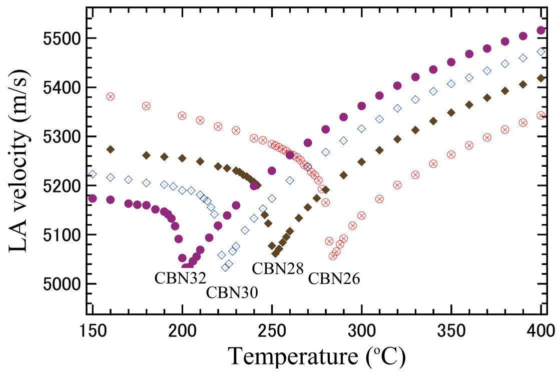

Fig. 3 Temperature dependences of LA sound velocity of CBN26, CBN28, CBN30, and CBN32 observed in *c*-plates.

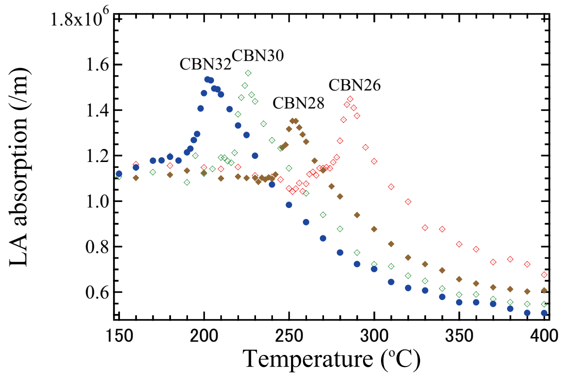

Fig. 4 Temperature dependences of LA absorption coefficient of CBN26, CBN28, CBN30, and CBN32 observed in *c*-plates.

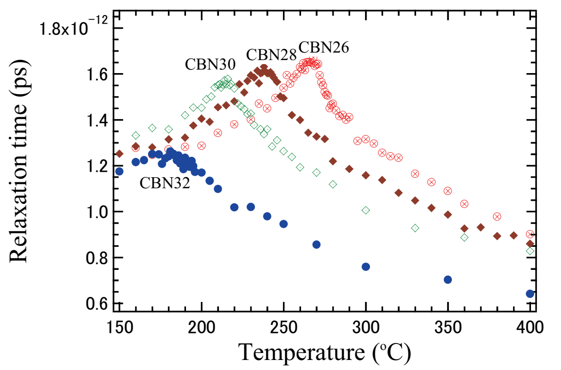

Fig. 5 Temperature dependences of relaxation time of CBN26, CBN28, CBN30, and CBN32 determined by a broad CP width observed in *a*-plates.