1J2-2

Large electromechanical coupling and temperature characteristic of free-standing sputter-epitaxial $PbTiO_3$ plates.

自立構造エピタキシャル $PbTiO_3$ 薄片の
高い電気機械結合係数と温度特性

Yuka Matsuda[1,2†] and Takahiko Yanagitani[1,2,3]
([1] Waseda Univ. [2] ZAIKEN [3] JST PRESTO)
松田友佳 [1,2†]，柳谷隆彦 [1,2,3] ([1] 早稲田大院　先進理工, [2] 材研, [3]JST さきがけ)

1. Introduction

Abnormal characteristics sputter-growth of epitaxial films of $Pb(Zr_xTi_{1-x})O_3$ (PZT) family such as higher Curie temperature than that of bulk PZT [1] and monotonic increase of electromechanical coupling coefficient k_t^2 with increasing concentration of Ti [2] were reported. These characteristics are peculiar to sputter-epitaxial PZT films but not to PZT in bulk ceramic form. These differences may be caused by epitaxial substrate constraints [3]. However, it is reasonable to assume that stress relaxation occurs in very thick epitaxial films more than a few μm. In this study, we fabricated free-standing epitaxial $PbTiO_3$ (PTO) plates by RF sputtering. Resonance characteristics were measured to examine the effect of substrate removing.

2. Epitaxial growth of $PbTiO_3$ plates

PTO plates were grown on a single crystalline La-STO substrate by RF magnetron sputtering. The thickness of the PTO epitaxial plates was 46 μm. **Fig. 1** shows the XRD patterns of PTO plates. (002) peak rocking curve FWHM of the PTO was measured to be 1.1°. As shown in **Fig. 2** X-ray, pole figure analysis shows four folds symmetry indicating an epitaxial growth of the PTO plates on the STO substrate.

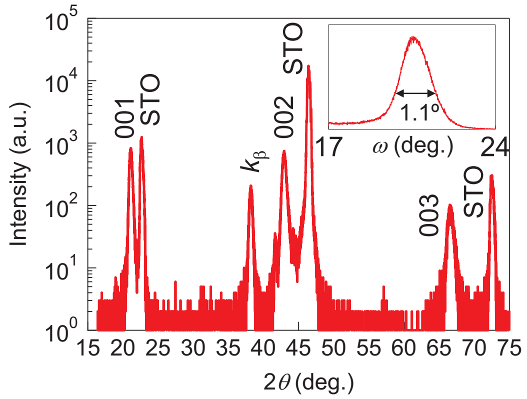

Fig. 1　2θ-ω scan XRD pattern and rocking curve of the PTO plates.

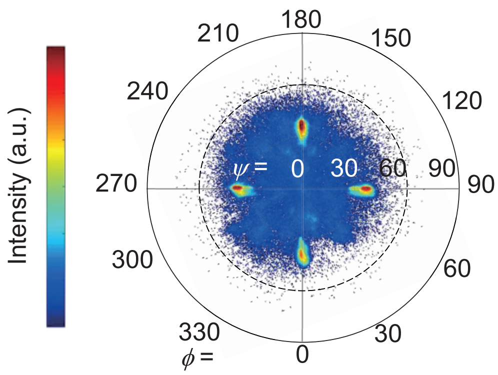

Fig. 2　Pole figure of the PTO plates of plane orientation (101).

3. Electromechanical coupling coefficient k_t^2

3.1 PTO/STO structure without removing substrate

The high-overtone bulk acoustic resonator (HBAR) structure (Au/PTO/La-STO) were fabricated. Longitudinal wave conversion loss (CL) was calculated using S_{11} parameter measured by a network analyzer (E5071C, Agilent Technology). **Fig. 3** shows experimental CL and theoretical one simulated by Mason's model. The minimum CL of 3.7 dB was found at 42 MHz where is thickness extensional mode frequency. k_t^2 estimated by comparison of experimental and theoretical CLs was 16.9%.

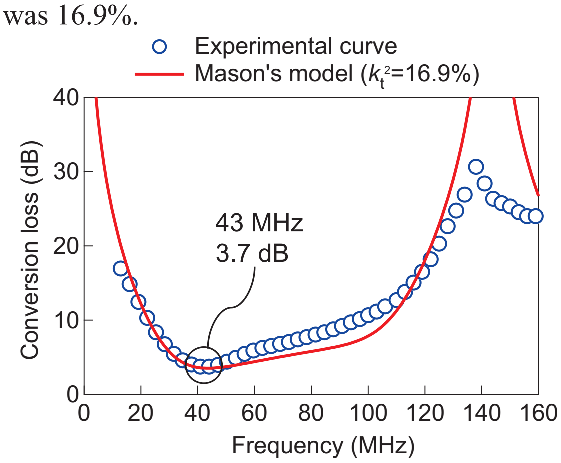

Fig. 3 Comparison of experimental and theoretical conversion losses simulated by Mason's model.

3.2 Free-standing plate structure.

Next, we obtained the free-standing PTO plates by peeling off the plates from the substrates to estimate k_t^2 by resonance-antiresonance method. The real part of admittance (Y_{real}) and the real part of impedance (Z_{real}) were measured using a network analyzer. **Fig. 4** shows the experimental admittance and the Y_{real} and the Z_{real} of the PTO free-standing plates. k_t^2 of the PTO were estimated to be 35% by substituting the resonance frequency (f_s:91.03 MHz) and the anti-resonance frequency (f_p:108.9 MHz) into Eq. (1).

$$k_t^2 = \frac{\pi}{2}\frac{f_s}{f_p}\tan\left(\frac{\pi}{2}\cdot\frac{f_p - f_s}{f_p}\right) \cdots (1)$$

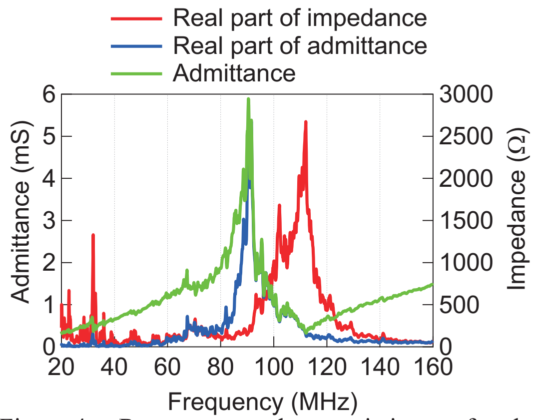

Fig. 4 Resonance characteristics of the free-standing PTO plates.

These results suggest that removing substrate may increase k_t^2 of the PTO plates, but further investigation are needed in order to determine the certain mechanism. k_t^2 of 35% of the free-standing PTO plates is higher than that of polycrystal bulk PTO (k_t^2=21% [4]).

4. Temperature characteristic

The temperature coefficient of frequency (TCF) for resonance and anti-resonance frequencies in the range of 140-35 ºC were measured to be -25 ppm/ºC and -21 ppm/ºC, respectively. These values compare well with those of AlN (-28 ppm/ºC [5]) and ZnO (-61.5 ppm/ºC [6]).

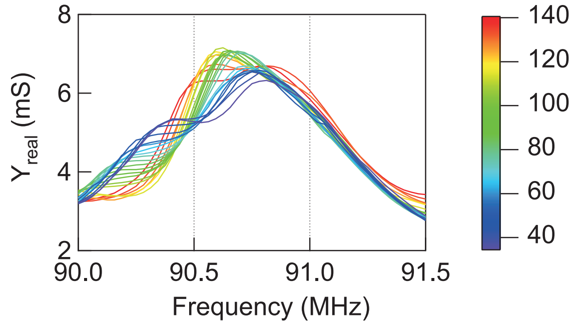

Fig. 5 The shift of the fundamental resonance frequency with temperature.

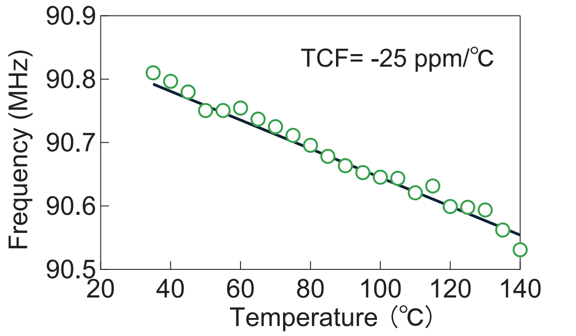

Fig. 6 TCF of the fundamental resonance frequency.

4. Conclusion

we achieved PTO free-standing sputter-epitaxial growth by RF magnetron sputtering. The electromechanical coupling k_t^2 of the free-standing PTO plates of 35% is high compared with that of HBAR structure. Moreover, this PTO plates exhibits lower TCF of -25ppm/ºC and -21ppm/ºC than those of AlN, ZnO, and Pb ($Zr_{0.4}Ti_{0.6}$) O_3 in bulk ceramic form (TCF=-60 ppm/ºC [7], k_p mode). Further experiments are needed to examine these unexpected results.

The low-frequency resonance of 90 MHz were also achieved in this PTO plate. The frequency range of 20-100 MHz ultrasonics are promising for photoacoustic imaging, which is useful to observe blood *in vivo* at high resolution. However, k_t^2 of PVDF commonly used for transducers in the 20-100 MHz is too low for practical application. Therefore, PTO free-standing sputter-epitaxial films have a bright promise for photoacoustic imaging and medical ultrasonic applications.

Acknowledgment

This work was supported by the JST PRESTO (No. JPMJPR16R8) and KAKENHI (Grant-in-Aid for Scientific Research No.16H04356, No.19H02202, and No.18K19037).

References

[1] T. Yanagitani, et al., IFCS2014, 6859872-1 (2014).
[2] R. Noda, et al., Proc. of 2018 IUS, 8580154 (2018).
[3] K. Wasa, et al., JAP., **117**, 124106-1 (2015).
[4] G. S. Kino, “Acoustic waves : Devices, Imaging, and Analog Signal Processing,” (1987).
[5] K. M. Lakin, et al., Proc. of 2000 IUS, **1**, 855 (2000).
[6] E. F Tokarev, et al., Sov. Phy. Sol. State, **17**, 629, (1975).
[7] H. Thomann, et al., Ferroelectrics, **40**, 189 (1981).