c-Axis tilted epitaxial $PbTiO_3$ thin film/ 25° off -angle La-$SrTiO_3$ substrate resonators with high k'_{35}

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1. Background

c-Axis tilted tetragonal piezoelectric films excite thickness quasi-shear mode. These films are attractive for immunosensors operating in liquids and SH-SAW sensor devices.[1,2] Since tetragonal epitaxial $PbTiO_3$ (PTO) has high electromechanical coefficient e_{33}, quasi-shear mode electromechanical coupling coefficient k'_{35} is expected to be high when c-axis is tilted to the electric field E_3. In previous studies, we reported (001) PZT thin films epitaxially grown on single crystal substrates by RF magnetron sputtering. It is found that all PZT crystals are tetragonal regardless of the Zr/Ti ratio. $PbTiO_3$ (PTO) exhibit the highest electromechanical coupling coefficient (k_t^2=33.5%) rather than PZT.[3, 4] High quasi-shear mode $k'_{35}{}^2$ is expected due to high e_{33} of PTO epitaxial film. However, GHz quasi-shear mode excitation have not been reported because c-axis normal (001) PTO is usually grown on (001) $SrTiO_3$ or (001) MgO.

In this study, c-axis tilted epitaxial growth of PTO and shear wave excitation were first achieved by using (001) La-$SrTiO_3$ substrate with 25° off-angle to the a-plane direction. Thickness extensional mode k'_{33} and quasi-shear mode k'_{35} of the PTO films was estimated from HBAR conversion loss.[5]

2. Relationship between c-axis tilt angle and piezoelectricity of $PbTiO_3$

In tetragonal and rhombohedral PZT near MPB, it has been reported that the extensional mode k'_{33} decreases monotonically with increasing cut angle.[6] **Fig. 1** shows the calculated relationship between c-axis tilt angles (from the substrate normal direction (γ=0°)) and $k'_{33}{}^2$ and $k'_{35}{}^2$ for the PTO crystal. Physical constants in Ref 7 were used. In the tetragonal PTO, thickness extensional mode k'_{33} decreases monotonically with increasing c-axis tilt angle. On the other hand, quasi-shear mode k'_{35} reaches a temporal extreme value (41%) nearby a 30° tilt angle and indicates a maximum value (44%) at a plane-grown PTO with a 90° tilt angle.

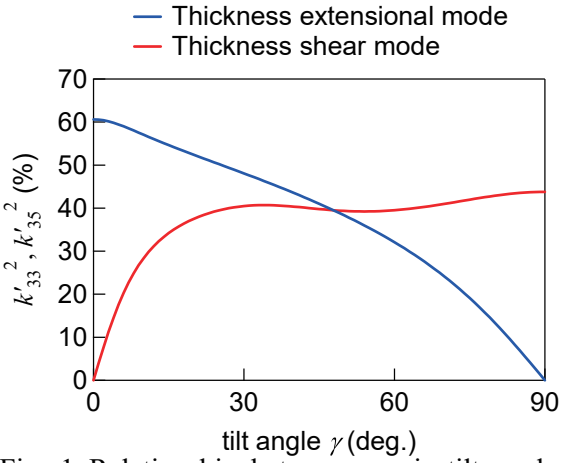

Fig. 1 Relationship between c-axis tilt angle and electromechanical coupling coefficient $k'_{33}{}^2$ and $k'_{35}{}^2$ of $PbTiO_3$

3. In-plane and out-plane orientation of $PbTiO_3$ films

XRD pole figures of PTO thin films grown on La-$SrTiO_3$ 25° off-angle substrates were measured (PANalytical X'Pert Pro). **Fig. 2** and **3** show (002) and (101) XRD pole figures of PTO films, respectively. XRD pole were shifted to χ=25° direction (left direction) as shown in **Fig. 2**. In addition, four-fold symmetry shifted 25° from origin were observed in **Fig. 3**. These indicate that the PTO is epitaxially grown following to the 25° off-angle to the substrate.

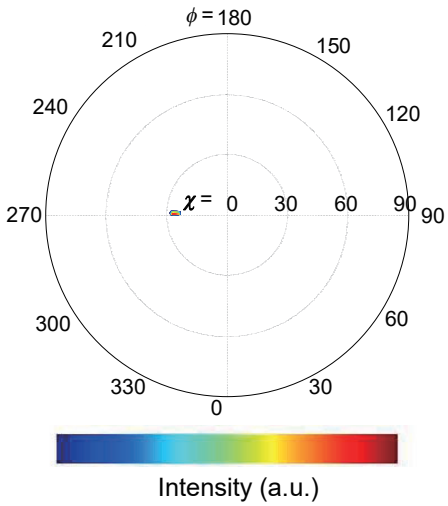

Fig. 2 (002) XRD pole figure of c-axis tilted epitaxial $PbTiO_3$ films of $SrTiO_3$ 25° off-angle substrates

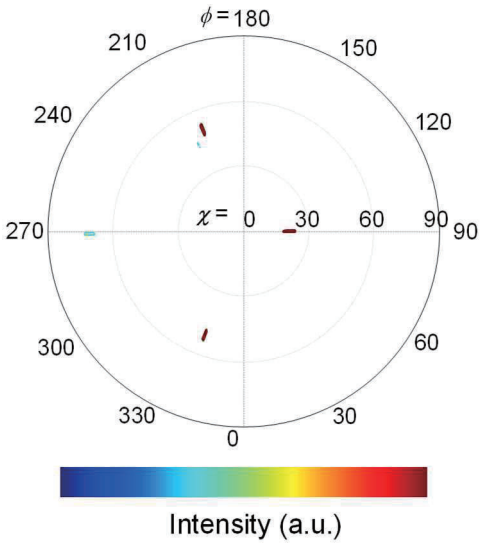

Fig. 3 (101) XRD pole figure of c-axis tilted epitaxial $PbTiO_3$ films of $SrTiO_3$ 25° off-angle substrates

4. Electromechanical coupling coefficients of $PbTiO_3$ films

Electromechanical coupling coefficients of PTO were estimated from HBAR conversion loss,[5] measured by a network analyzer (Keysight, E5071C). Electromechanical coupling coefficients k'_{33} and k'_{35} was estimated by comparing experimental conversion loss and theoretical one calculated by a Mason's equivalent circuit model. **Fig. 4** shows conversion loss curves of the PTO-HBAR with substate. Bule plots show experimental conversion loss of thickness extensional mode of PTO resonator, and red plots show quasi-shear mode one. Low conversion loss shows high electromechanical coupling. Comparing experimental curves and theoretical ones, thickness extensional mode $k'_{33}{}^2$ and quasi-shear mode $k'_{35}{}^2$ was estimated to be 13.5%, and 17.9%, respectively. The trend agrees well with the tetragonal (001) PZT properties[7] and the expected properties in **Fig. 1**. A large $k'_{35}{}^2$ was observed due to the c-axis tilt of PTO film.

5. Conclusion

c-Axis tilted PTO-HBAR were fabricated by epitaxially grown on 25° off-angle (001) La-$SrTiO_3$ substrate. Thickness extensional mode $k'_{33}{}^2$ and quasi-shear mode $k'_{35}{}^2$ of PTO was estimated to be 13.5%, and 17.9%, respectively from HBAR conversion loss. The $k'_{33}{}^2$ decrease and $k'_{35}{}^2$ improvement with increasing c-axis tilt angle were observed.

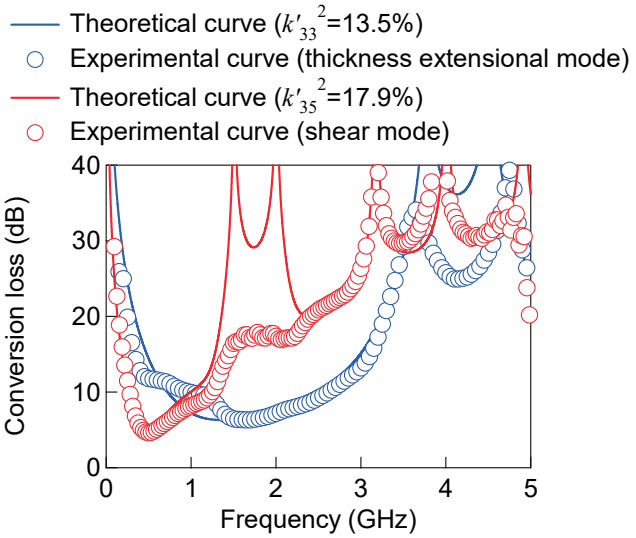

Fig. 4 Conversion loss curves of $PbTiO_3$-HBAR

Acknowledgment

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