Fabrication of Pure-Perovskite-Phase Sm-Doped $Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ Epitaxial Thin Film on Si by Magnetron Sputter using Powder Target

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The relaxor-based ferroelectric single crystal such as $Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ (PMN-PT) has attracted attention with a higher piezoelectricity beyond $Pb(Zr,Ti)O_3$ (PZT). Especially, Samarium-doped PMN-PT (Sm-PMN-PT) single crystal was recently reported to exhibit a world-leading high d_{33} value [1]. This monocrystalline thin film with the pure-perovskite phase and ideal orientation is also expected to provide a giant piezoelectricity, and has a potential to create high-performance piezoelectric MEMS devices. However, the epitaxial growth of such a film with a thickness of a few micrometers for MEMS is generally uneasy due to the thermodynamics instability [2].

In this study, we have tried to solve this problem by introducing a (001)-oriented PZT-based seed layer and separate deposition technique with a powder target. As a result, we successfully demonstrated that the proposing methods are effective to promote the (100)/(001) orientation and to suppress the pyrochlore phase generation of Sm-PMN-PT.

In this experiment, a Sm-PMN-PT epitaxial thin film was sputter-deposited by “three separate deposition” or “continuous deposition” on a (001)PZT-based seed layer covered on a $SrRuO_3$/Pt/ZrO_2/(100)Si substrate (purchased from KRYSTAL Inc.). The deposition temperature and pressure were set to 600°C and 0.4 Pa with a mixture of Ar and O_2 gas (flow rate ratio, Ar/O_2 = 20/1), respectively. The sputtering target with the composition ratio of $Pb_{96.25}Sm_{0.025}[(Mg_{1/3}Nb_{2/3})_{0.67}Ti_{0.33}]O_3$ was prepared. In addition, the excess 20 mol% of Pb was added for compensating loss of Pb component due to the vaporization during the sputtering process.

Figure 1 indicates that the thin film prepared via the “three separate deposition” had the pure perovskite phase and a preferential (001)/(100) orientation, while the sample prepared via the “continuous deposition” had the pyrochlore phase. Energy dispersive X-ray spectrometry (EDX) analysis of the transmission electron microscope (TEM) (Fig. 2) indicated that the sample via the separate deposition had a homogeneous distribution of Pb in the film-thickness direction. On the other hand, the Pb amount in the other sample was gradually decreased as the surface was approached. This proved that the separate deposition technique contributes to the improvement of homogeneousness of Pb distribution and thus to suppression of the pyrochlore phase generation.

We believe that this achievement is a great step to create a giant piezoelectric transducer thin film for MEMS actuator beyond PZT.

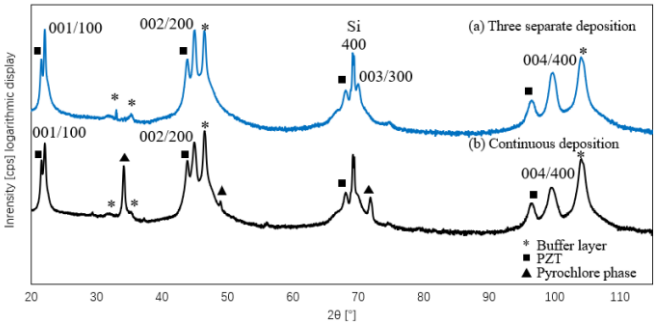

Fig. 1 XRD θ -2θ patterns of the thin films prepared via (a) three separate deposition and (b) continuous deposition.

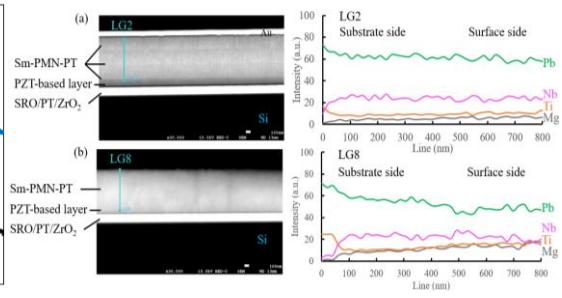

Fig. 2 TEM EDX analysis of the thin films from substrate side to surface side.

[1] F. Li, L. Q. Chen, T. R. Shrout, and S. Zhang, Nat. Mater., vol. 17, pp. 349-354, 2018.
[2] S. D. Bu, M. K. Lee, S. K. Streiffer, and J. J. KrajewskiAppl. Phys. Lett., vol. 79, pp. 3482-3484, 2001.

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