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Epitaxial Growth of $Li_{3x}La_{2/3-x}TiO_3$ Thin Films on Perovskite Substrates by Pulsed Laser Deposition

Univ. of Tokyo[1], JST-CREST[2], KAST[3]

○Jie Wei[1], Tomoteru Fukumura[1,2], Yasushi Hirose[1,2,3], Tetsuya Hasegawa[1,2,3]

E-mail: weijie@chem.s.u-tokyo.ac.jp

A double perovskite, lithium lanthanum titanate $Li_{3x}La_{2/3-x}TiO_3$ (LLT, $c \approx 2a_p$), has attracted great interest because of its high ionic conductivity induced by A-site vacancy, which is promising for potential applications in lithium-ion batteries [1]. Recently, epitaxial thin film growth was reported on $SrTiO_3$ (STO) (100) [2]. However, the ionic conductivity of epitaxial LLT films on STO has not been successfully measured due to the presence of conductive interface. Considering the high oxygen partial pressure during thin film growth, it is difficult to attribute the high interface conductivity only to the formation of oxygen vacancy. Here, we study the intermixing between LLT and STO substrate, which could be the main reason for the reported interface conductivity.

LLT epitaxial thin films were grown on atomically flat STO (100) substrates by pulsed laser deposition. An $x = 0.11$ polycrystalline target was used for ablation. The energy fluency of pulsed KrF excimer laser was varied in a range of 0.5 − 2 J/cm^2. Substrate temperature at 900 − 1000 ℃ and oxygen pressure from 1 − 100 mTorr were investigated. Amorphous LLT thin films were also grown in the same laser condition at room temperature with oxygen pressure of 1×10^{-6} Torr. The obtained amorphous films were post-annealed in air at 1100 ℃ for 2 h. Secondary ion mass spectrometry (SIMS) was used for measuring depth profiles of the elements in the LLT films.

Figure 1 shows the depth profiles of Li, Ti, La and Sr in each LLT film. Obviously, Sr from STO substrate diffused into the as-deposited LLT epitaxial thin film. In the LLT layer treated at high temperature in air, Sr-intermixing was surprisingly serious. This suggests that Sr-diffusion leads to the formation of an perovskite (La, Sr)TiO_3 nanostructure, which could show high electric conduction. Thus, the above-mentioned scenario can give a reasonable explanation for the fact that STO/LLT interface became conductive even in an oxidative atmosphere. In the presentation, the effect of laser condition and oxygen pressure on lithium composition and crystal structure will also be discussed.

References

[1] Y. Inaguma et al., Solid State Commun. **86** (1993) 689.

[2] T. Ohnishi et al., Solid State Ionics **228** (2012) 80.

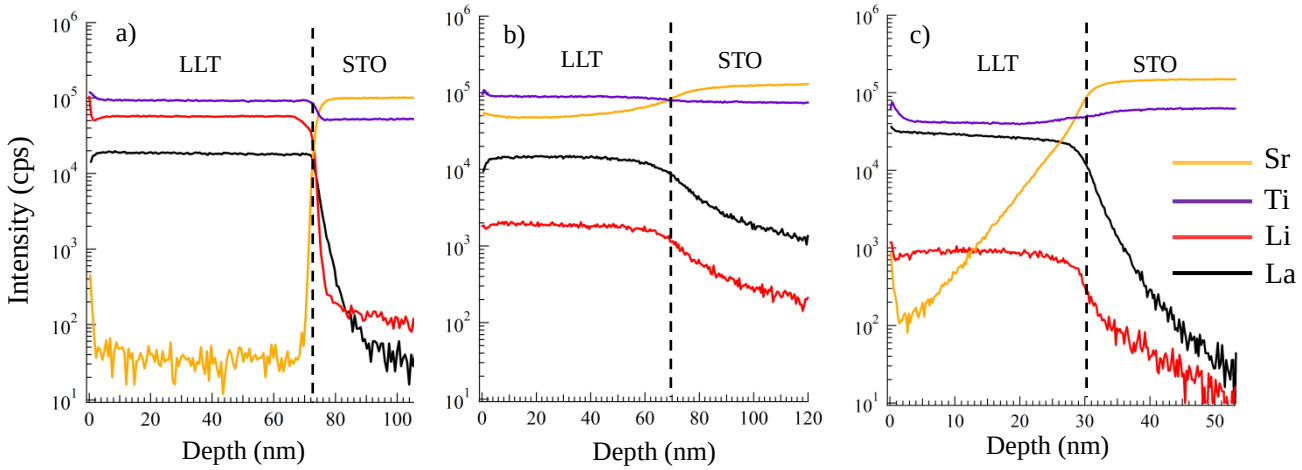

Figure 1 SIMS depth profile of a) amorphous LLT grown at room temperature; b) LLT post-annealed in air at 1100°C; c) LLT as-deposited at 950 ℃ in oxygen pressure of 75 mTorr.