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Impedance Study of Eptaixial $Li_{3x}La_{2/3-x}TiO_3$ Thin Films on Perovskite Substrates Deposited by Pulsed Laser Deposition

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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 due to the A-site vacancy, which is promising for potential applications in lithium-ion batteries [1]. Recently, epitaxial thin film growth of LLT was reported on $NdGaO_3$ (NGO) (110) [2] but its ionic conductivity was significantly lower than that of bulk LLT. Considering the high oxygen partial pressure during thin film growth, the low ionic conductivity could be attributed to Li-loss in the film. In this study, we used a Li-rich target to compensate the Li-loss, and controlled Li composition by laser fluence of PLD method. We evaluated the in-plane and out-of-plane impedances to investigate the effect of crystalline axis and lattice strain on the ionic transportation.

LLT epitaxial thin films were grown on atomically flat NGO (100) substrates by PLD method. A $Li_{0.85}La_{0.56}TiO_{3+x}$ target was used for ablation to fabricate stoichiometric $Li_{0.33}La_{0.56}TiO_3$ thin film. The energy fluence of pulsed KrF excimer laser was varied in a range of 0.5 - 2 J/cm^2. Substrate temperature and oxygen pressure were varied respectively in the ranges of 900 - 1000 ℃ and 1 - 100 mTorr. For in-plane impedance measurements, Ti/Au comb electrodes were fabricated by photolithography and EB evaporation. For out-of-plane impedance measurements, 5 nm thick epitaxial $LaNiO_3$ buffer layer was fabricated for bottom electrode. The capacitance and ionic conductivity were measured by an AC impedance analyzer with frequency from 1 Hz to 10 MHz.

Figure 1 shows the 2θ-θ XRD pattern of LLT (001) epitaxial thin film on NGO (110) substrate. The rocking curve FWHM for LLT (004) was 0.04° indicating high crystalline quality. Reciprocal space mapping indicated that epitaxial LLT had the same in-plane lattice constants as NGO. Figure 2 shows an Arrhenius plot of Li ionic conductivity (σ) for LLT thin film on NGO substrate at [1-10] direction. The σ value at room temperature was 0.6 mS cm^{-1} with an activation energy of 0.32 eV, the former of which is one order higher than that in the previous report [2] and is closed to 1.3 mS cm^{-1} in bulk research. In this report, we shall also discuss the effect of strain on σ and the results of out-of-plane impedance measurements.

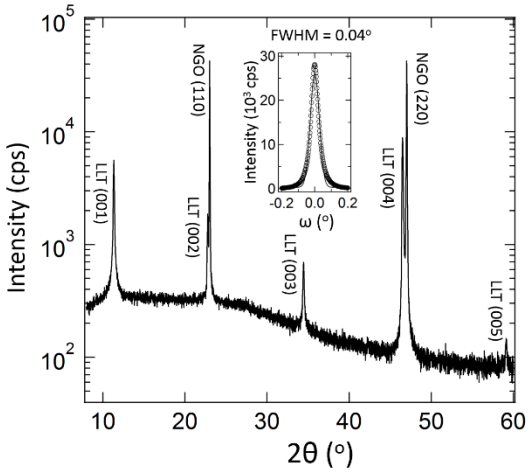

Figure 1 XRD pattern of LLT thin film on NGO (110) substrate. The inset shows rocking curve of LLT (004) with a FWHM of 0.04°.

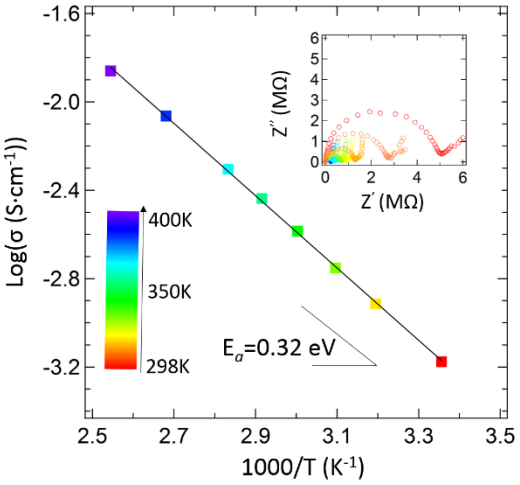

Figure 2 Arrhenius plot of 60 nm thick LLT film on NGO at [1-10] direction. The inset shows complex impedance plots at each temperature.

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

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