doi:10.1017/S1431927616008746
Microsc. Microanal. 22 (Suppl 3), 2016

Interfacial electrical conductivity controlled crystallization of amorphous $LaAlO_3$ under electron-beam irradiation

G. Y. Lee[1,2], J. Y. Kim[1,3], S. Y. Moon[4], S. H. Baek[4], D. H. Kim[2], H. J. Chang[1,#]

[1] Advanced Analysis Center, Korea Institute of Science and Technology, Seoul, South Korea
[2] Center for Noncrystalline Materials, Department of Materials Science and Engineering, Yonsei University, Seoul, South Korea
[3] Research Institute of Advanced Materials, Seoul National University, Seoul, South Korea
[4] Electronic Materials Research Center, Korea Institute of Science and Technology, Seoul, South Korea

Perovskite structured oxide $LaAlO_3/SrTiO_3$ shows various unique physical properties such as superconductivity, magnetic effect and high electron mobility which are generated from the 2-dimensional electron gas (2DEG) at the interface [1-4]. Particularly, electric conductivity at the interface of two insulating oxide has attracted many researchers' attention since the first report by H. Y. Hwang et al. in 2004 [4]. The formation of 2DEG occurs regardless of the crystalline orientation of $SrTiO_3$ and more interestingly, it has been demonstrated that 2DEG also can be generated even at the heterointerface of amorphous(a)-$LaAlO_3$ overlayer on $SrTiO_3$ substrate [5]. The conductivity disappear when it is annealed under O_2 pressure, which reveals that 2DEG might be induced by oxygen vacancy rather than the charge catastrophe.

In this study, we investigated crystallization behaviors of a-$LaAlO_3$ at the interface with $SrTiO_3$ under electron-beam irradiation and the effect of interfacial conductivity on the crystallization. The a-$LaAlO_3$ thin films were grown on TiO_2-terminated $SrTiO_3$ substrates by pulsed laser deposition (PLD) in an oxygen atmosphere. The grown all a-$LaAlO_3$ thin film thickness were about 26 nm and it was confirmed by transmission electron microscope (TEM). The 2DEG was formed when the a-$LaAlO_3$ thin film was grown at room temperature while it disappeared after ex-situ annealing at 500℃ for 1 h under the oxygen pressure of 300 mTorr. The observation of nucleation and crystallization behavior of a-$LaAlO_3$ under electron-beam irradiation was performed in aberration-corrected STEM (Titan S80-300; FEI) operated at 300 kV while irradiation was conducted with different electron-beam current (0.16 ~ 0.67 nA) as well as irradiation time (0 ~ 300s). When a convergent electron beam positioned at a-$LaAlO_3$, in most cases, local crystallization occurs with a random crystal orientation due to knock-on energy of electron accelerated as 300 keV. In the meantime, epitaxial crystallization of $LaAlO_3$ from the $SrTiO_3$ interface was observed when the electron beam positioned within a distance of 3 nm from the interface. It demonstrate that epitaxial crystallization pattern can be obtained by controlling the e-beam position with a consideration of the effective radius for the atomic displacements. In addition, it was found that the crystal growth shape and its kinetics depend on the a-$LaAlO_3/SrTiO_3$ interfacial conductivity, which could be controlled with oxygen pressure during thermal annealing. This indicates that the accumulated electron charge near the insulating interface can activate the crystallization while the conductive interface annihilates the charge causing slower crystallization kinetics. The compositional intermixing and bonding state after the crystallization was studied by electron energy loss spectroscopy (EELS; Gatan Quantum 966 spectrometers) and interlayer distance measurement.

References:

[1] Rijnders, G. and D. H. A. Blank., Nature 433 (2005) p. 369.
[2] Reyren, N., S. Thiel, A. D. Caviglia, L. F. Kourkoutis, G. Hammerl, C. Richter, C. W. Schneider, T. Kopp, A. S. Ruetschi, D. Jaccard, M. Gabay, D. A. Muller, M. J. Triscone, J. Mannhart, Science 317 (2007) p. 1196.
[3] Wang, X., W. M. Lu, A. Annadi, Q. Z. Liu, K. Gopinadhan, S. Dhar, T. Venkatesan, Ariando, Physical Review B 84 (2011) p. 075312.
[4] Ohtomo, A. and H. Y. Hwang., Nature 427 (2004) p. 423.
[5] A. Sambri et al., Phys. Lett. 100 (2012) p. 231605

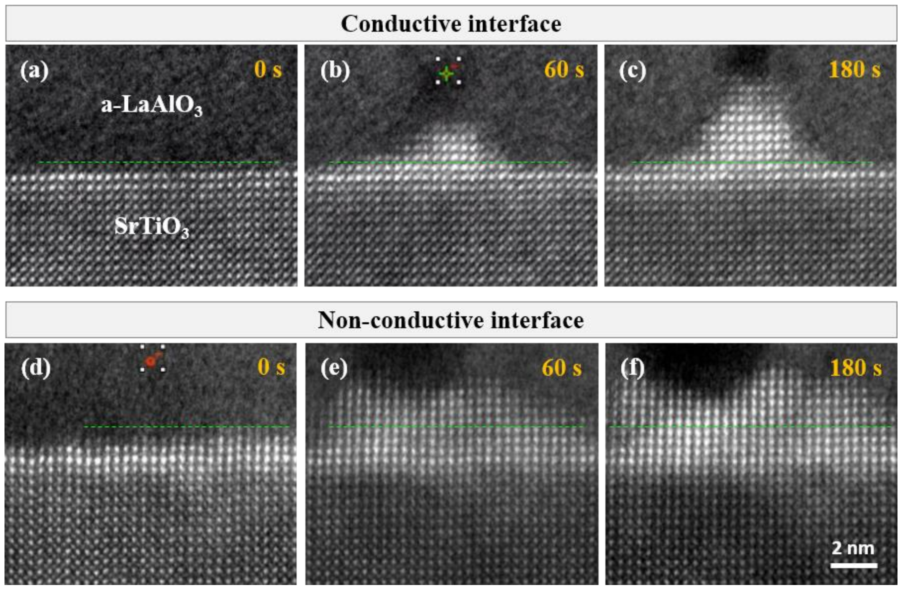

Figure 1. HAADF STEM images of a-$LaAlO_3$/$SrTiO_3$ under focused electron-beam irradiation which positioned about (a)-(c) 3.5 nm and (d)-(f) 3 nm away from the interface.

https://doi.org/10.1017/S1431927616008746 Published online by Cambridge University Press