Heterostructure oxide LaAlO$_3$(LAO)/SrTiO$_3$(STO) with perovskite structure exhibits excellent high dielectric constant, good magnetic, ferroelectric and insulating properties, high wear resistance, and outstanding resistance against oxidation and are candidates for memory devices and spintronics application.[1] One of the most prominent properties of the material is formation of electrically conducting layer, that is, two-dimensional electron gas (2DEG) at the interface between two insulating oxides LAO and STO. Interestingly, the conductivity was shown in amorphous LAO grown on STO as well as the crystalline LAO [2]. However, the issue of instability of the oxide under electron beam was also raised reporting that amorphous STO, formed by irradiation with 1.0 MeV Au at 400 K, is accelerated to recrystallize epitaxially at the a/c interfaces by electron beam in transmission electron microscopy [1]. For the applications of these oxide materials, it is necessary to study the thermodynamic stability and phase transformation behavior of the heterostructure oxides under electron beam.

In this study, amorphous LAO films were grown on TiO$_2$-terminated STO substrates by pulsed laser deposition (PLD) at room temperature in an oxygen atmosphere. The sample was annealed at 500°C in vacuum subsequently.[3] The atomic structure change during nucleation and growth at the interface of LAO/STO was studied with HAADF STEM images and EEL spectra collected using a Cs-corrected microscope Titan S 80-300 operated at 300 kV equipped with Gatan Quantum 966 spectrometers. Cross-sectional samples for STEM analysis were prepared by mechanical thinning, precision polishing, and ion milling (PIPS 691).

In HAADF image of annealed a-LAO/STO it was found that epitaxial one atomic layer of La(Al,Ti)O formed on STO. Exposure to electron beam makes this La(Al,Ti)O layer grows from the interface. Figure 1 shows that electron beam positioned even slightly apart from the surface (marked by an arrow in Fig. (b)) for 20 sec can cause epitaxial growth from the interface. Electron irradiation induced nucleation and growth in this material could be explained by a radiation enhanced diffusion, which is resulted from both ionization processes and a strong thermodynamic driving force for crystallization.[4] In this study, the growth behavior of crystalline phase is investigated under systematic microscopic conditions and controlling of size and shape of the crystalline region will be suggested, which can be applied for patterning of crystalline phase in a-LAO.

References
[2] S.Y. Moon et al. (under submission)

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Fig. 1: STEM HAADF image of (a) before and (b) after electron beam positioning for 20 sec at a-LAO annealed at 500℃ in air. La(Al,Ti)O layer grows from top surface (yellow line) in a pyramidal shape.