

Controlling atomic scale epitaxial crystallization by an interfacial conductivity of the insulating oxides under an electron beam

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Device structures have become miniaturized with a recent rapid progress in nanotechnology, and the demand for fabricating and manipulating nanostructures has dramatically increased. An e-beam lithography (EBL) is one of the most promising for nanoscale patterning owing to its excellent resolution and maskless patterning among the several other techniques utilizing various sources including light, X-ray and ion-beam sources.

Meanwhile, researches on the perovskite-structured oxide materials are rapidly increasing recently and interesting physical properties of perovskite material have observed [1]. For instance, interfacial conductivity between two insulating oxides, that is two-dimensional electron gas (2DEG), shows a number of intriguing properties such as on/off switching with external electric fields, nanoscale electronic devices and tunable conductivity. A formation of the conductive interface has been reported in crystalline (c)- as well as amorphous (a)-LaAlO₃/SrTiO₃ heterostructures, and the interfacial conductivity can be degraded by annealing in high oxygen pressure [2].

Here, we demonstrate an e-beam-controlled crystallization of an a-LaAlO₃ thin film grown epitaxially at the a-LaAlO₃/SrTiO₃-substrate interface. Also, we report the effect of the interfacial conductivity on the kinetic behavior of epitaxial crystallization. The observation on the crystallization behavior of a-LaAlO₃ under the e-beam irradiation was performed in aberration-corrected STEM (Titan S80-300; FEI) operated at 300 kV. The irradiation current was conducted from 0.16 nA to 0.67 nA.

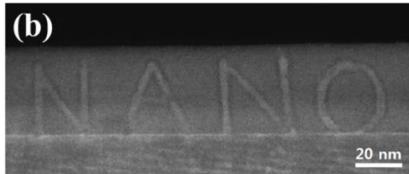
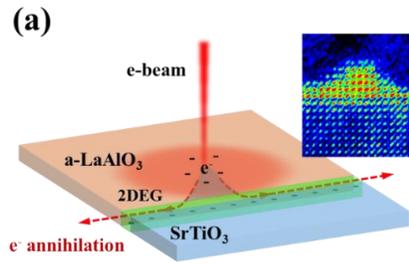
When a convergent e-beam positioned at a-LaAlO₃ overlayer, in most cases, local crystallization occurs with a random crystal orientation due to knock-on energy of electron accelerated as 300 keV. However, epitaxial crystallization from the interface occurs when the e-beam is located within a critical distance from the interface. The 2DEG acting as a current path delays the crystallization kinetic, thus delicate control of the crystallized pattern shape and size was available. Schematics showing the distribution of electrons around the incident e-beam in the a-LAO overlayer is shown in the figure below.

As a result, successful pattern writing with a width of about 5 nm was performed as shown in the figure [3]. This work provides useful guidelines for coherent atomic scale e-beam patterning considering critical distance of the e-beam from the interface for the epitaxial growth, e-beam dose rate effect on the growth rate and the heterostructure interfacial conductivity.

[1] Rijnders, G. and D. H. A. Blank., *Nature*, 433, 2005, 369-370.

[2] S. Y. Moon, C. W. Moon, H. J. Chang, T. Kim, C.-Y. Kang, H.-J. Choi, J.-S. Kim, S.-H. Baek and H. W. Jang, *Electron. Mater. Lett.*, 12, 2016, 243 - 250.

[3] Lee, G., Moon, S. Y., Kim, J., Baek, S. H., Kim, D. H., Jang, H. W., & Chang, H. J., *RSC Advances*, 7(64), 2017, 40279-40285.



This research was supported by Basic Science Research Program through the National Research Foundation of Korea (NRF) funded by the Ministry of Science, ICT and future Planning (2017R1A2B2012514) and the KIST Institutional Program (2V05210).