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Atomically sculptured heart in oxide film using convergent electron beam

Gwangyeob Lee[1], Seung-Hyub Baek[2] and Hye Jung Chang[1,3*]

Abstract

We demonstrate a fabrication of an atomically controlled single-crystal heart-shaped nanostructure using a convergent electron beam in a scanning transmission electron microscope. The delicately controlled e-beam enable epitaxial crystallization of perovskite oxide $LaAlO_3$ grown out of the relative conductive interface (i.e. 2 dimensional electron gas) between amorphous $LaAlO_3$/crystalline $SrTiO_3$.

Keywords: Epitaxial crystallization, Electron beam irradiation, Spherical aberration corrected scanning transmission electron microscope, Nanoarchitectonics

Description

Lithography techniques utilizing various sources including light, X-rays, electron beams (e-beams), and ion beams have been investigated to obtain better performance (Levenson et al. 1982; Ehrfeld and Lehr 1995; Watt et al. 2005). Among these techniques, e-beam lithography is one of the most promising methods of fabricating nanostructures because of its excellent spatial resolution (Tseng et al. 2003; Altissimo 2010). Atomically controlled nanostructure sculpting can be conducted using recent advances in aberration-corrected scanning transmission electron microscopy (STEM) (Song et al. 2011; Jesse et al. 2015). In this study, we investigated the e-beam controlled epitaxial crystallization of an amorphous $LaAlO_3$ (a-LAO) thin film that had a conductive interface with a $SrTiO_3$ (STO) substrate (Moon et al. 2016).

The a-LAO thin film was grown on TiO_2-terminated STO substrates at room temperature by pulsed laser deposition in an oxygen atmosphere. Cross-sectional TEM specimens were prepared by standard mechanical polishing (Struers; Labopol-5) and subsequent argon-ion milling (PIPS 691; Gatan). Observation of the crystallization of the a-LAO under e-beam irradiation was performed using aberration-corrected STEM (Titan S80–300; FEI), and the convergent e-beam was controlled by STEM software. The acceleration voltage and dose rate of the incident e-beam were 300 keV and 0.169×10^9 e^- A^{-2} s^{-1}, respectively.

Under delicately controlled e-beam irradiation, the amorphous structure changed into a crystalline structure with epitaxy with the STO substrate. Using this technique, we sculptured heart-shaped crystallized LAO in a-LAO layer (Fig. 1). The atomically controlled nanostructure sculpting was conducted using several control parameters such as the interfacial conductivity, dose rate, and e-beam's distance from the heterointerface (Lee et al. 2017). The heart-shaped crystallized region with a brighter contrast in the high-angle annular dark-field (HAADF) STEM image was perovskite-type pseudocubic LAO, which was confirmed by the chemical composition and diffraction pattern analyses (Lee et al. 2017). We hope that e-beam lithography using sub-nano scale e-beams in STEM can be applied to manipulate the structures and properties of materials and devices.

* Correspondence: almacore@kist.re.kr
[1]Advanced Analysis Center, Korea Institute of Science and Technology, Seoul 02792, Republic of Korea
[3]Division of Nano and Information Technology, KIST School, University of Science and Technology, Seoul 02792, Republic of Korea
Full list of author information is available at the end of the article

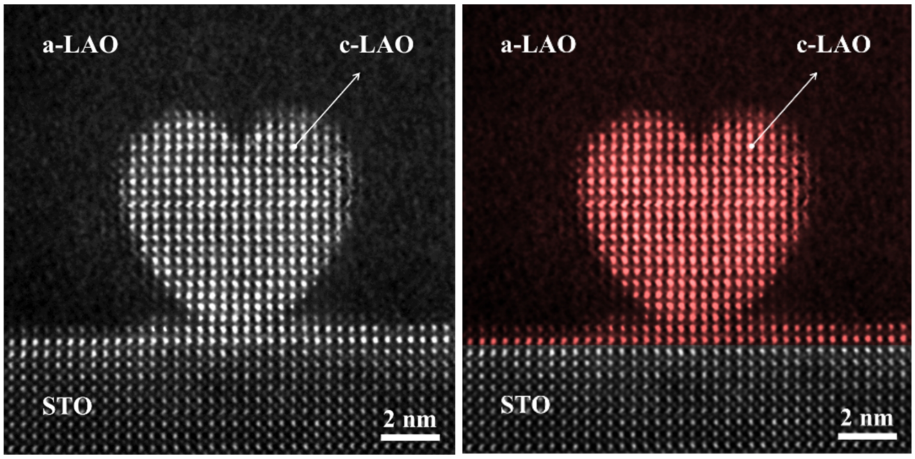

Fig. 1 A high-angle annular dark-field scanning transmission electron microscopy (HAADF STEM) (left) and colored image (right) of the sculptured heart-shaped crystallized LAO in the a-LAO region through e-beam irradiation in STEM

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Not applicable.

Authors' contributions
Hye Jung Chang supervised the project and wrote the manuscript. Gwangyeob Lee conducted the STEM experiment to generate the data and prepared the manuscript. Seung-Hyub Baek prepared the samples. All the authors discussed the results and approved the final manuscript.

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Availability of data and materials
Not applicable.

Competing interests
The authors declare that they have no competing interests.

Author details
[1]Advanced Analysis Center, Korea Institute of Science and Technology, Seoul 02792, Republic of Korea. [2]Electronic Materials Research Center, Korea Institute of Science and Technology, Seoul 02792, Republic of Korea. [3]Division of Nano and Information Technology, KIST School, University of Science and Technology, Seoul 02792, Republic of Korea.

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