

Domain Structure of LSAT Single Crystal

Shunsuke OKADA¹, Katsuhiko SASAKI¹, Tomoharu TOKUNAGA¹, Shunsuke KOBAYASHI²,
Takahisa YAMAMOTO^{1,2,3}

¹Department of Quantum Engineering, NAGOYA University, Nagoya, 464-8603

²Department of Advanced Materials Science, The University of Tokyo, Chiba, 277-8561

³Nanostructures Research Laboratory, Japan Fine Ceramic Center, Nagoya, 456-8587, Japan

The detail occupation manner of La, Sr, Al, Ta at A/B-sites was directly investigated for LSAT single crystals mainly by high angle annular dark-field STEM (HAADF-STEM) from a viewpoint of order/disorder domain structure. The clear regularity of the contrast variation in HAADF images was obtained only at B-sites in the ordered domains, suggesting that Al and Ta cations at B-sites exhibit ordering. The atomic ratio of Al/Ta at B-sites consists of two types of Al-rich and Ta-rich. On the other hand, the contrast variation at A-sites in ordered domains and at A/B-sites in disordered domains was almost constant. However, the contrast at B-sites sometimes exhibit very low intensity in the disordered domains. It means that the atomic ratio of Al/Ta even at B-sites in the disordered domains is not fully constant.

The crystal thin films have been studied energetically for harvesting new physical properties. Crystallinity and physical property of thin films often depend on lattice mismatch between films and substrates. To obtain the high-quality epitaxial film growth, the lattice constants and thermal expansion coefficients of the substrates need to match those of the growing thin films. $(\text{La}_{0.3}\text{Sr}_{0.7})(\text{Al}_{0.65}\text{Ta}_{0.35})\text{O}_3$ (LSAT) is one of the most preferable substrates for GaN and high temperature superconductor ceramics thin film growth in terms of a lattice constant matching, high melting temperature, and no phase transition below the melting temperature. To grow high quality thin films, the substrate surface is necessary to have atomically flat, *i.e.*, step-terrace structure. The step-terrace structure is closely related to the lattice structure of the substrates. LSAT is reported to have order/disordered domain structure[1], suggesting that the domain structure possibly affects the step-terrace structure. In this study, the domain structure of LSAT single crystal was investigated directly at single atomic column scale mainly by using HAADF-STEM from a viewpoint of order/disordered domains.

Figure 1 shows a dark-field image obtained from 111 reflection of LSAT single crystal, which is peculiar to an order domain near 110 zone. In the image, bright areas correspond to the ordered domains. A size of the ordered domains ranges from 5 to 40 nm, and the interfaces between order/disordered domains are very wavy. Fig. 2 is HRTEM image taken from an area including order/disordered domains. Two types of image structures can be seen in the image, however, it is very difficult to distinguish a domain type. To directly observe B-site ordering, we carried out Z-contrast (HAADF-STEM) observation. Fig. 3(a) is HAADF-STEM image of an area including both ordered/disordered domains, which was taken from [110] of LSAT single crystal. A/B site columns of LSAT single crystal is separately observed from the direction as seen in (b) schematically showing a lattice structure. As seen the insets (d), contrast intensity is almost constant in the ordered domains, indicating a total Z number including respective B-site columns does not change largely. On the other hand, contrast intensity periodically changes at B-sites in the ordered area as in the inset (c), indicating that Z number at B-site atomic columns, *i.e.*, a ratio of Al/Ta, changes regularly. Considering the intensity variation at respective B-site columns in the ordered domain, B-site ordering consists of Al-rich and Ta-rich columns although the two types of B-site columns are reported to be Al full columns and Al-rich ones[1]. On the other hand, some unique contrast can be seen at B-site even in a disordered domain. The contrast intensity sometimes decreases largely

as indicated by the arrows in Fig. 3(a). As for A-site, strong intensity can be sometimes observed in both types of domains, suggesting that atomic ratio of La/Sr is La-rich.

As shown in this study, the occupation manner of cations in LSAT is very complicated even in a single crystal. The complexity would be closely related to the step-terrace structure of LSAT surfaces.

Acknowledgement

A part of this study was financially supported by a Grant-in-Aid for Scientific Research on Innovative Areas "Nano Informatics" (Grant No. 25106003) from Japan Society for the Promotion of Science (JSPS).

Reference

- 1) Hao Li et al. J. Mater. Res. 17, 1698-1704 (2003).

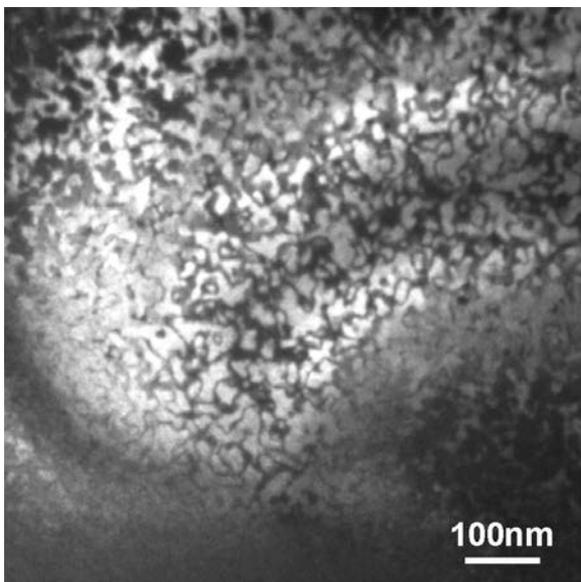


Fig.1 TEM bright field image of LSAT

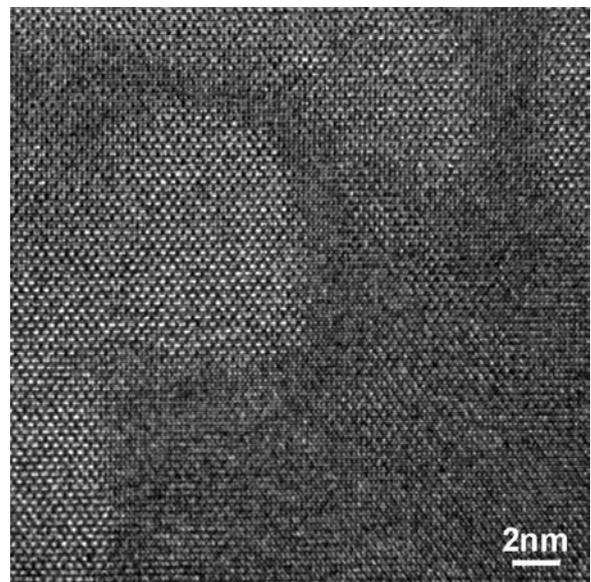


Fig. 2 HRTEM image of LSAT

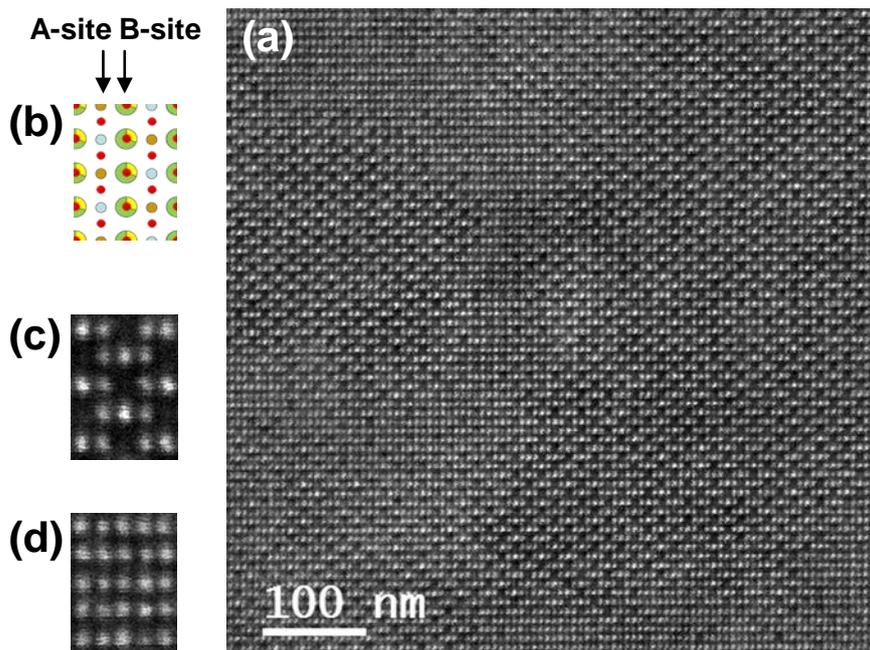


Fig. 3
 (a) HAADF-STEM image of LSAT
 (b) Schematic drawing of LSAT structure viewed from [110]
 (c) HAADF-STEM image taken from ordered domains
 (d) HAADF-STEM image taken from disordered domains