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DOI: 10.1017/S1431927606063161

Microsc Microanal 12(Supp 2), 2006
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Quantitative Analysis of Atomic Resolution HAADF-STEM (Z-contrast) Imaging for $PbTiO_3$ / $SrTiO_3$ Substrate Dielectric Thin Films

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It is an important issue in non-volatile memory applications to understand finite size effects in dielectric thin films [1]. Moreover, the dielectric property will be strongly influenced by interfaces with electrodes or other materials [2]. For an understanding of the behavior of dielectric thin films it is important to survey accurate thickness of dielectric thin films and interface structure with an electrode with an atomic scale.
High-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) is now an accepted high resolution imaging approach in materials research. However, image contrast of experimental HAADF-STEM is not clear due to background noise because of intensity of high angle scattered electrons are too weak. Furthermore, in the case of general analytical TEM (C_s is about 1.0 mm), atomic resolution HAADF-STEM images contain artificial bright spots on no atomic columns caused by the effect of spherical aberration of probe-forming lens [3]. Therefore, we can not apply it for precise quantitative analysis of Z-contrast images.

In this study, we performed direct observation of atomic species by high resolution HAADF-STEM imaging for $PbTiO_3$ epitaxially grown on $SrTiO_3(100)$ substrate by MOCVD. From the result of HAADF-STEM images, we confirmed accurate thickness of $PbTiO_3$ thin films and evaluated the interface between $PbTiO_3$ and $SrTiO_3$ related to deposition conditions. The HAADF-STEM images were obtained by a JEOL JEM-2010F with the C_s = 1.1 mm and thermal-Schottky field-emission gun in probe-forming mode at 200 kV. The specimen thickness was measured by electron-energy-loss spectroscopy (EELS) for accurate quantitative analysis of Z-contrast image [4]. The thickness of all specimens matched to 30 nm. Noise reduction for HAADF-STEM images was made by maximum entropy method (MEM) [5]. The artificial images contained in the experimental images removed by deconvolution technique [6]. The experimental images were analyzed and compared with the calculated atomic intensities obtained from HAAD-STEM image calculation [7].

Fig. 1 shows HAADF-STEM images of $PbTiO_3$ thin films prepared with thicknesses of 2 mono-layer (ML) and 8 ML. The image intensity corresponds to the atomic numbers of Pb =82, Sr = 38 and Ti = 22, respectively. It is found that $PbTiO_3$ films grow epitaxially on $SrTiO_3$ substrate and the thickness is controlled by the deposition time. The process of MEM and deconvolution for HAADF-STEM image of $PbTiO_3$ of 2 ML is shown in Fig. 2. As a result of image processing, HAADF-STEM image becomes clear and completely removes the effect of probe intensity distribution. Fig. 3 shows the line profiles of atomic column height intensity from Sr substrate to Pb thin films. The image brightness and contrast are calibrated in using the intensity ratio of Sr to Ti. The intensity of Pb first layer is low in comparison with maximum the Pb intensity. Consequently, it is observed that Pb and Sr are mixed by the interface. It is estimated that there were about 60% Pb replacement for Sr site by HAADF-STEM image calculation.

https://doi.org/10.1017/S1431927606063161 Published online by Cambridge University Press

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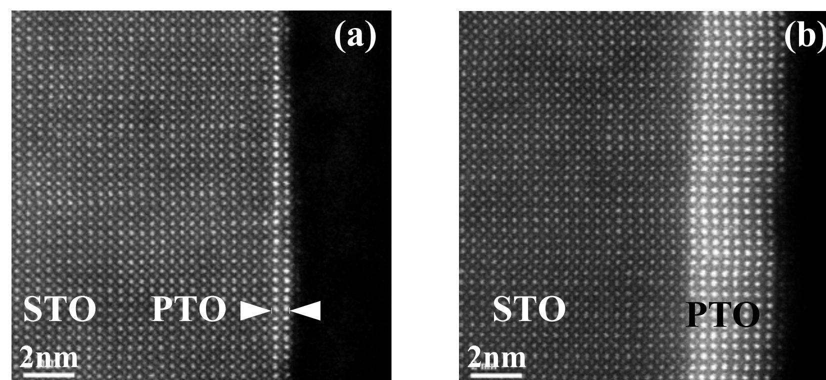

Fig.1 HAADF-STEM images of $PbTiO_3$ thin films on $SrTiO_3$ substrate for (a) 2 ML (b) 8 ML.

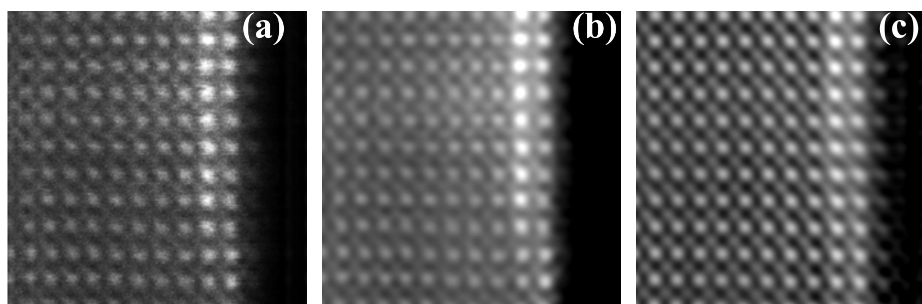

Fig.2 MEM and deconvolution process for $PbTiO_3$ of 2 layers (a) Experimental HAADF-STEM image (b) MEM processed image (c) Deconvolution processed image.

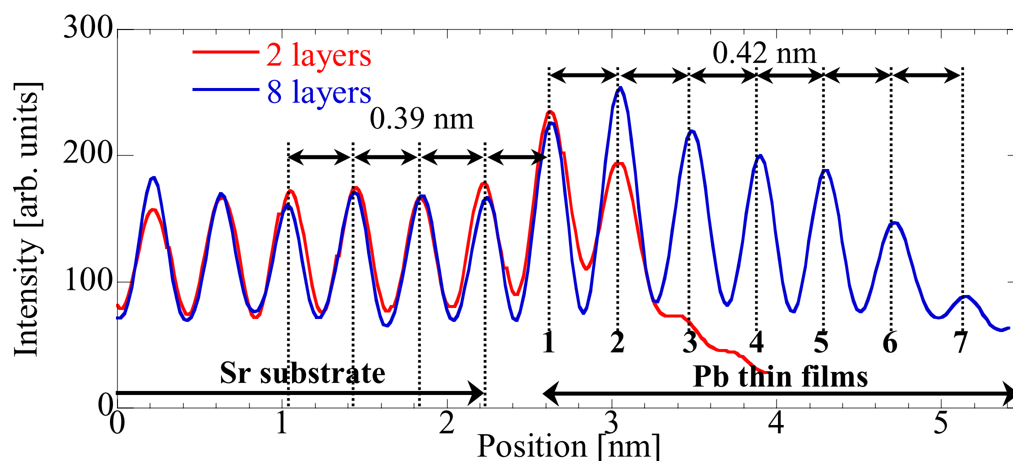

Fig.3 Line profiles of atomic column height intensity from Sr substrate to Pb thin films.

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