

Structure Analysis for Clarification of Spatial Distribution of Crystals and Boundary Region between Crystals in CAAC-IGZO

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Abstract

To understand correlation between the c-axis aligned crystalline indium-gallium-zinc oxide (CAAC-IGZO) structure and device electric characteristics, dark-field TEM imaging technique was performed to analyze the aggregation state of the CAAC-IGZO film. According to a dark-field cross-sectional TEM image obtained from the (009) spot, texture of the CAAC-IGZO film starts to grow at a point several angstroms apart from the interface with the base layer. Textures are mainly aligned in a direction perpendicular to the substrate surface while some textures are tilting at several degrees. Furthermore, in a plan-view dark-field TEM image obtained from the 100 diffraction spots having the equivalent d-spacing, a region lying just perpendicular to the substrate surface has a width of approx. 100 nm, and is connected to the region consisting of tilted textures.

1. Introduction

A bulk crystal of indium-gallium-zinc oxide (IGZO) was first synthesized in 1985 by Kimizuka et al., who proposed the use of IGZO as a material for semiconductor devices in 1987 [1,2]. We have constantly analyzed physical properties of IGZO thin films we made and found c-axis aligned crystalline IGZO (CAAC-IGZO) [3] and nano-crystalline IGZO (nc-IGZO) [4], which have crystal structures different from those of single crystal IGZO and amorphous IGZO [5,6]. Recently, Waseda et al. have reported that CAAC and nc-IGZO both include fine crystalline clusters; the size of the crystalline clusters in CAAC-IGZO (approx. 2.2 nm) is slightly larger than that in nc-IGZO (approx. 1.8 nm) [7].

The CAAC-IGZO, which has a new aggregation state, can be deposited by sputtering at relatively high substrate temperatures [8]. A high-resolution cross-sectional TEM image of the CAAC-IGZO film reveals that the film has a region in which c-axis are aligned in a direction perpendicular to a substrate surface [8]. A fine structure in this region can be confirmed by nanobeam electron diffraction analysis [8]. Moreover, fast Fourier transform (FFT) filtering is performed on a high-resolution plan-view TEM image and lattice points are extracted, so that a Voronoi polyhedron can be obtained. This FFT image shows continuous crystal connection between grains with no clear grain boundaries [9].

There is a lot of uncertainty about the influence of fluctuation in CAAC-IGZO crystal structure and the boundary region between crystals on the device electric characteristics. To understand more details, it is important to evaluate the aggregation state at a mesoscopic level, which is between a nanometer level (which is larger than an atomic level) and a macroscopic level. For this reason, we analyzed the CAAC-IGZO film by dark-field TEM observation, which is an imaging technique using particular diffraction wave. Practically, the dark-field TEM image is obtained by locating the objective aperture to the target diffraction spot and switching to the imaging mode. For comparison, bright-field image observation was also performed.

2. Experiment

The dark-field TEM observation is a method for capturing defects with regularity and distinct texture in crystal, and needs to be performed with relatively low magnification, which is different from the high-resolution TEM analysis. Therefore, to obtain an electron beam spot with high intensity, CAAC-IGZO was deposited to be as thick as 3 μm by sputtering onto a 100-nm-thick thermal oxide film on a silicon substrate. In this study, cross-sectional and plan-view TEM observations were performed, where thinned samples were made by a focused ion beam (FIB) system.

3. Structural Analysis of CAAC-IGZO by TEM

Dark-field TEM observation of cross-sectional sample

First, selected area electron diffraction (SAED) was performed to observe the CAAC structure of the cross-sectional sample. Fig. 1 shows the results.

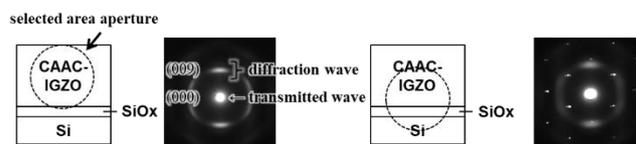


Fig. 1 SAED image of CAAC-IGZO cross-sectional sample (the diameter of the selected area denoted by the dotted line circle is approx. 3 μm).

Next, a bright-field image was formed by inserting an objective aperture with respect to transmitted wave (000) of the cross-sectional sample. Fig. 2 shows the bright-field image.

In the bright-field image, a distinct contrast unique to the CAAC-IGZO film was observed. This contrast probably comes from different tilting angle for each crystal domain. That is, regions having different orientation orders in scale of several tens of nanometers exist in the CAAC-IGZO film.



Fig. 2 Bright-field image of CAAC-IGZO cross-sectional sample.

To capture partial orientation state in the CAAC-IGZO film on the basis of the contrast in the bright-field image, a dark-field image of an arched (009) spot was partly observed by using an objective aperture. Fig. 3 shows the results. These images were formed near a surface which is the uppermost part of the CAAC-IGZO film, and correspond to the same view.

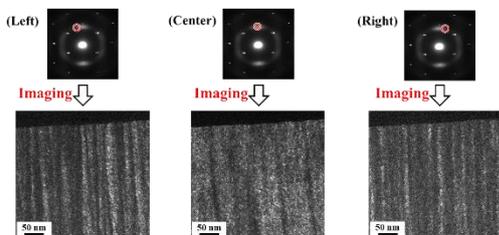


Fig. 3 Dark-field images of red-circle regions near film surface of CAAC-IGZO cross-sectional sample.

As seen from the results, in the dark-field images of the left and right sides of the arched (009) spot, string-like contrasts that are slightly tilted with respect to the direction perpendicular to the substrate surface were observed. On the other hand, a distinct orientation state was not observed in the dark-field image of the center of the (009) spot. This is probably because the thickness of the cross-sectional sample made by FIB is large, and thus in the cross-sectional observation of the center portion, multiple string-like microcrystals overlap each other in the thickness direction to form an unclear image. In the left and right portions, crystals are separately observed because the number of crystals is small. To observe each crystal more clearly, dark-field image observation was performed with use of a cross-sectional sample processed to be as thin as possible (Fig. 4).

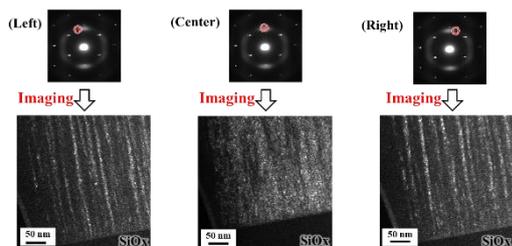


Fig. 4 Dark-field images of red-circle regions near interface between CAAC-IGZO and thermal oxide. The cross-sectional sample is thinned in the depth direction.

As a result, the string-like contrast that shows crystals with a width of approx. 10 nm extending in the direction perpendicular to the substrate was obtained. A region consisting of high density of IGZO crystals oriented nearly normal to the Si substrate. We call this region as “highly oriented region”. Furthermore, it can be demonstrated that texture growth in the CAAC-IGZO film starts at a point several angstroms apart from the interface with the base layer, and is tilted several degrees with respect to the direction perpendicular to the substrate.

Dark-field TEM observation of plan-view sample

To observe distribution of crystals each having a long-range order which is seen as the string-like contrast observed in the cross-sectional sample, a plan-view sample was also made by the FIB system, and dark-field image observation was performed focusing on the (100) diffraction spots having the equivalent d-spacing. Fig. 5 shows the results.

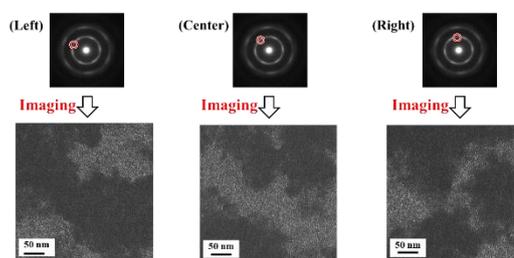


Fig. 5 Dark-field images of red-circle regions of CAAC-IGZO plan-view sample.

The dark-field images of the portion with the highest spot intensity were observed. The contrast in the dark-field image shows that highly oriented regions having a width of approx. 100 nm exist, and are connected to highly oriented regions tilted several degrees therefrom. Therefore, in the CAAC-IGZO, crystal orientations are continuously connected even in the mesoscopic region.

4. Conclusions

To understand correlation between the CAAC-IGZO structure and device electric characteristics, dark-field TEM observations of cross-sectional and plan-view samples have been performed. It was demonstrated that the CAAC-IGZO has continuous crystal orientation connection even in the mesoscopic region. From these results, one can conclude that the CAAC-IGZO has few defects which adversely affect electric characteristics of minute field-effect transistors.

References

- [1] N. Kimizuka *et al.*, J. Solid State Chem. **60**, 382 (1985).
- [2] N. Kimizuka *et al.*, Japanese Published Patent S63-239117.
- [3] S. Yamazaki *et al.*, Jpn. J. Appl. Phys. **53**, 04ED18 (2014).
- [4] N. Sorida *et al.*, Jpn. J. Appl. Phys. **53**, 115501 (2014).
- [5] K. Nomura *et al.*, Nature **432**, 488 (2004).
- [6] T. Kamiya *et al.*, Proc. IDW'13, 280 (2013).
- [7] Y. Waseda, *et al.*, Mater. Trans. **11**, 1691 (2018).
- [8] S. Yamazaki, *et al.*, J. Soc. Info. Disp. **22**, 5567 (2014).
- [9] S. Yamazaki, ECS Trans. **67**(1), 29 (2015).