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# Electron microscopy determination of crystallographic polarity of aluminum nitride thin films

Noriyuki Kuwano[a], Jesbains Kaur[b,*], Siti Rahmah[a]

[a] Microscopy Laboratory, Malaysia-Japan International Institute of Technology, Universiti Teknologi Malaysia, Jalan Sultan Yahya Petra, 54100 Kuala Lumpur, Malaysia
[b] Research Centre for Nano-Materials and Energy Technology, School of Science and Technology, Sunway University, Bandar Sunway, 47500 Subang Jaya, Selangor, Malaysia

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ABSTRACT

Aluminum nitride (AlN) crystallizes usually in the wurtzite structure ($P6_3mc$) and it has a crystallographic polarity. In this work, the polarity in AlN was characterized by using several methods of transmission electron microscopy (TEM) in order to examine their applicability. AlN was deposited by metalorganic vapor phase epitaxy (MOVPE), followed by annealing at 1550 °C. TEM samples were prepared by using a focused ion beam (FIB) mill. Observation was performed with microscopes of JEM-2100, JEM-ARM200 F and FEI Titan Cubed G2 at Kyushu University (Japan), and the following results were obtained. (1) Conventional TEM imaging: Under a diffraction condition with $hkil$ = 0002, inversion domains or an inversion domain boundary (IDB) was observed. (2) Scanning TEM (STEM) High-Angle Annular Dark Field (HAADF) imaging: Even when atomic column images of Al and N are not resolved completely from each other, the polarity was determined from the shape of atomic column images. (3) Scanning moire fringe imaging: The moire fringe pattern indicated the position of IDB and determine the direction of polarity. (4) Convergent beam electron diffraction (CBED): CBED was applicable for determination of the polarity in AlN at the acceleration voltage of 120 kV. Hence the polarity, direction of polarity and inversion domain boundary was determined using advanced TEM methods.

## 1. Introduction

Aluminum nitride (AlN) is a direct transition semiconductor with a wide band gap of 6.2 eV. AlN can be alloyed with GaN and/or InN so that the band gap energy can be controlled very widely from 6.2 eV to 0.5 eV. (Moses et al., 2011) Therefore, (Al, Ga, In)N system is very promising as basic material applicable to optoelectronics. (Al,Ga,In)N usually crystalizes in the wurtzite structure ($P6_3mc$) which does not have inversion symmetry as shown in Fig. 1. Instead it has a crystallographic polarity. The directions of [0001] and [000$\bar{1}$] can be distinguished and the planes of (0001) and (000$\bar{1}$) are also different from each other. The polarity is very crucial since the physical and chemical properties are different depending on the polarity accordingly. For example, the polarity causes for piezo-electric effect which introduces the quantum-confined Stark effect. (Chichibu et al., 1998; Chen et al., 2007). Atomic species on the top surface is different with the polarity; III-group element on (0001) plane and nitrogen on (000$\bar{1}$) plane. Therefore the behaviour of crystal growth and the absorption of impurity atoms are different, depending on the polarity of planes. There are several methods to determine the polarity; e.g., chemical etching, x-ray abnormal absorption, etc. Among these, TEM is a powerful tool as the microstructure can be analyzed together in a good spatial resolution. In case of GaN, the polarity can be determined easily and there have been many studies on this up to date. However, there are some difficulties practically in determination of polarity by TEM. (Imura et al., 2013) In this work, several methods of TEM were applied for analysis of polarity by using an identical sample in order to examine the applicability of the methods for AlN.

## 2. Methods

TEM experiments were performed at The Ultramicroscopy Research Center of Kyushu University, Japan. (Jesbains et al., 2016a) Thin foil samples for cross sectional TEM observation were prepared by using a focused ion beam (FIB) mill (Hitachi MI4000 L). (Jesbains et al., 2016b). The thickness of thin foil sample was thought to be 60–100 nm. Conventional TEM (CTEM) images and convergent beam electron beam diffraction (CBED) patterns with microscopes (JEOL, JEM2100) and (JEOL, ARM200 F) at the accelerating voltage $E_a$ = 200 and 120 kV, respectively, and high-angle annular dark-field (HAADF) images were

* Corresponding author.
*E-mail address:* jesbainss@sunway.edu.my (J. Kaur).

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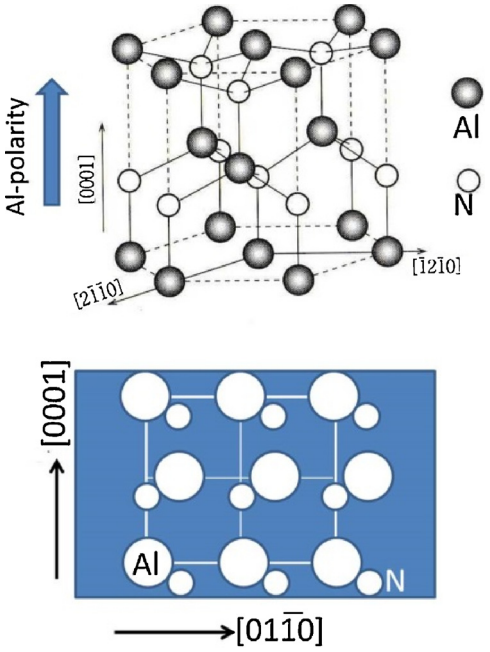


**Fig. 1.** Crystal structure of AlN in wurtzite type and its projected structure in **[2$\bar{1}\bar{1}$0]** direction.

observed with a scanning TEM (FEI, Titan Cubed G2 60–300 with an aberration corrector for probe-forming lens) at $E_a$ = 300 kV.

## 3. Materials

The samples of AlN thin layers which is about 200–300 nm in thickness were offered by Hiramatsu and Miyake research group at Mie University and was prepared as following method. (Miyake et al., 2016; Jesbains et al., 2016a) AlN was grown epitaxially onto a (0001) sapphire substrate at 1150 °C by the metal-organic vapor phase epitaxy (MOVPE) with trimethyl-Al and ammonia as sources of Al and N, respectively. Hidrogen ($H_2$) was used as carrier gas. Thereafter the AlN/sapphire sample was annealed at 1550 °C for 2 h under a controlled atmosphere of nitrogen and carbon monoxide ($N_2$ + CO).

## 4. Results and discussion

Fig. 2 shows a low magnification bright-field image of AlN/sapphire observed under the condition of systematic diffraction with $\boldsymbol{g}$ = 0002,

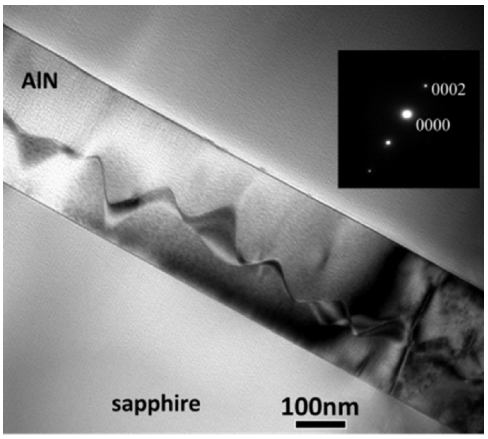


**Fig. 2.** Low magnification BF image of AlN/sapphire taken at $E_a$ = 200 kV under the condition of systematic diffraction along $\boldsymbol{g}$ = 0002. A corresponding electron diffraction pattern for this image is inserted.

where $\boldsymbol{g}$ is the diffraction vector. It is clearly seen that the AlN layer is divided into an upper and a lower layers by a single inversion domain boundary (IDB) of a zig-zag shape. (Jesbains et al., 2016a): i.e., the upper and lower layers are inversion domains. It is noted in Fig. 2 that the IDB has an image contrast which is quite similar to that of a stacking fault (SF). The crystal structure of AlN is considered to be composed with (0001) atomic layers of Al and N, and the positions of Al atomic layer and N atomic layer are different by $c/8$ in $c$-axis direction. The positions of Al and N are exchanged with each other at an IDB. If just Al atomic layers are taken into account in the AlN structure, IDB can be considered as a kind of SF with a shift vector of $\boldsymbol{B} = \boldsymbol{c}/8$. This can explain well the characteristics of image contrast which is similar to the ones of a normal SF.

When the kinematical diffraction approximation is held, the intensities of diffraction with $\boldsymbol{g}$ are equal to those of $-\boldsymbol{g}$ due to Friedel's law. However, inversion domains sometimes have different brightness in the TEM image due to the dynamical diffraction effect. The phenomenon is used for observation of inversion domains in conventional TEM images. Even in the case that the inversion domains have almost the same brightness in image contrast, IDBs are visible as in the case of SFs.

Fig. 3 shows high resolution HAADF images of the upper layer and the lower layer of AlN. It is known that in normal HAADF images, atom column sites appear with brightness almost proportional to $Z^{1.7}$, where Z is the atomic number of atom column site (Rouviere et al., 2008). The method of HAADF images has a merit that the image contrast is

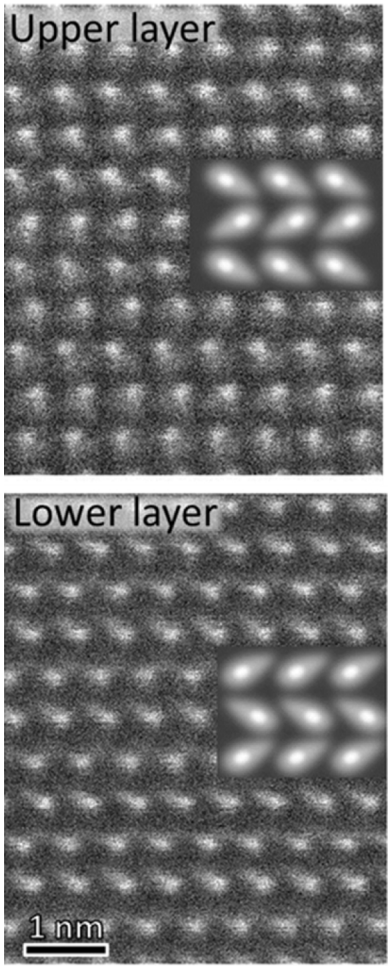


**Fig. 3.** HAADF images of the upper and the lower AlN layer regions taken at $E_a$ = 300 kV.
Illustrates of expected images are inserted for guidance (Jesbains et al., 2016a)