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Characterization of GaN thick layer grown by the HVPE: Comparison of horizontal with vertical growth

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Abstract GaN films were grown on the vertical and horizontal reactors by the hydride vapour phase epitaxy (HVPE). The structural and optical characteristics of the GaN films were investigated depending on the reactor-type. GaN epilayers were characterized by double crystal X-ray diffraction (DC-XRD), transmission electron microscopy (TEM) and photoluminescence (PL). Surface defects of two kinds of the GaN films were revealed by the wet chemical etching method, using H_3PO_4 acid at 200°C for 8 minutes. Hexagonal etch pits were analyzed by optical microscopy and SEM. Etch pit densities were calculated to be approximately 1.4×10^7 and $1.2\times10^6\ cm^{-2}$ for GaN layers grown on horizontal and vertical reactors, respectively. Those results show GaN grown in the vertical reactor having a better quality of optical properties and crystallinity than that in the horizontal reactor.

Key words Gallium nitride, Hydride vapor phase epitaxy, Photoluminescence

1. Introduction

Single crystalline group III nitride semiconductors have attracted huge research interest during last two decades due to their unique properties and potential applications in short-wavelength light source/detector and high temperature/frequency devices [1, 2]. Taking advantage of the direct wide bandgap of GaN (3.44 eV), blue and green light emitting diodes (LEDs) have been commercialized [3, 4]. The ability of GaN to form solid solutions with AlN and InN, making bandgap engineering possible, is essential for defining the emission wavelength of the LEDs. The high thermal conductivities of GaN and AlN make them suitable for high power applications, where the heat generated by devices must be efficiently dissipated.

However, achievement of its full potential has still been limited by a dramatic lack of suitable GaN bulk single crystals. Most of the current device structures are grown on sapphire or 6H-SiC. However, since their lattice parameters and thermal expansion coefficients are not well-matched to GaN, the growth generates huge densities of defects, with thread dislocation (TDs) being the most prevalent [5]. This leads to deterioration in the electrical and optical character of the device produced from the III-nitrides. Some techniques developed such as lateral epitaxial overgrowth (LEO) have recently been developed to reduce the number of threading dislocations that originate from the interface with the lattice-mismatched substrates. Therefore, this work is aimed for improving crystal quality and optical properties of GaN. GaN films were grown in the horizontal and vertical reactors by the hydride vapor phase epitaxy (HVPE). Characteristics of two-types of GaN samples were compared with each other.

2. Experimental

GaN layers were grown on a sapphire (0001) subtrate using the HVPE with the horizontal and vertical reactors. Melted gallium source for gallium and ammonia (NH_3) source for nitrogen were used as precursors for the growth of GaN layers. A 50~100 nm thick AlN buffer layer fabricated by the nitration of sapphire substrate prior to the GaN growth. This step is an important one for reducing the defect density and yellow luminescence (YL). The thickness of substrate is about

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350 μm and the growth temperature is approximately 1000°C. The growth condition were maintained in the same for the comparision.

The surface of GaN samples was mechanically polished to remove surface features such as hillocks and pits from as well as mechanical bowing and thermal expansion stresses. Then they were etched by the H_3PO_4 acid at 200°C at 8 minutes.

The surface morphology of the etched GaN films was observed using an optical microscope (Olympus, Japan) and a scanning electron microscope (SEM, JSM-5900, Japan). Microstructural characteristics of GaN layer was investigated by a transmission electron microscope (TEM, JEOL 2010, Japan). The crystallinity was examined by an double-crystal X-ray diffractometer using Cu Kα1 radiation. The room-temperature photoluminescence were analysed using a 325 nm He-Cd laser at 17 mW as excitation power.

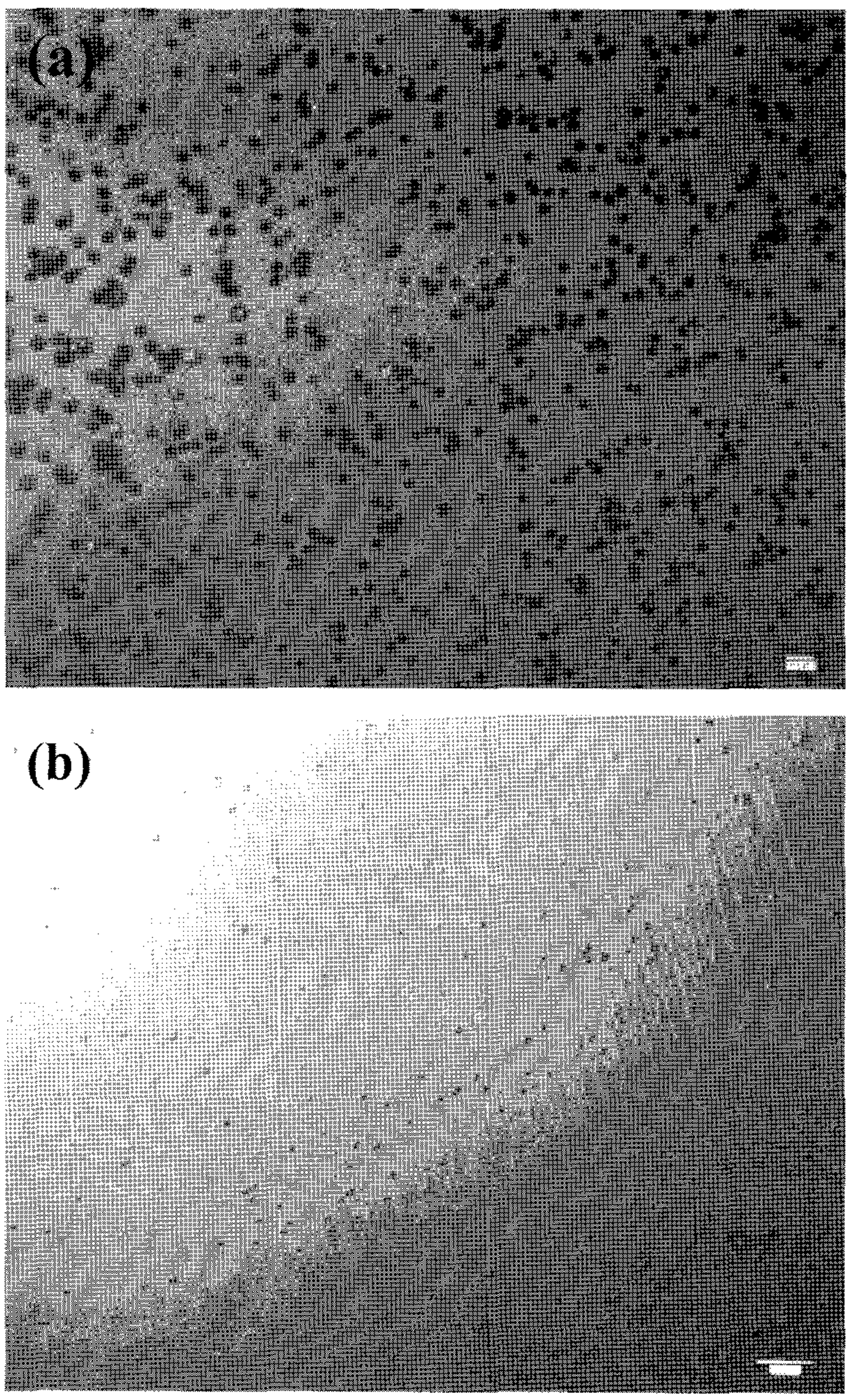

Fig. 1. Etched GaN surface in (a) the horizontal reactor and (b) the vertical reactor.

3. Results and Discussion

Figure 1 shows the etched GaN surfaces. The appropriate etching time depends on defect density and type of defects; long times are needed for lower defect density material. The etched pit density was calculated to be approximately 1.4×10^7 and $1.2\times10^6\,cm^{-2}$ for the specimens from horizontal and vertical reactors, respectively.

Using SEM etched pits shapes were examined deeply and represented in Fig. 2. Ga-polar GaN films (0001) are selectively attacked and show hexagonal etch pits after etching in hot phosphoric acid [6, 7]. The etching process revealed the hexagonal etch pits reflecting the crystal symmetry of GaN [8]. Hexagonal etch pits associated with defects formed on Ga-polar films, leaving the defect-free GaN areas intact and the morphology unchanged. Ideally, one would expect that a specific etch pit shape and size would be associated with specific defects.

Microstructural characteristics of GaN layers was

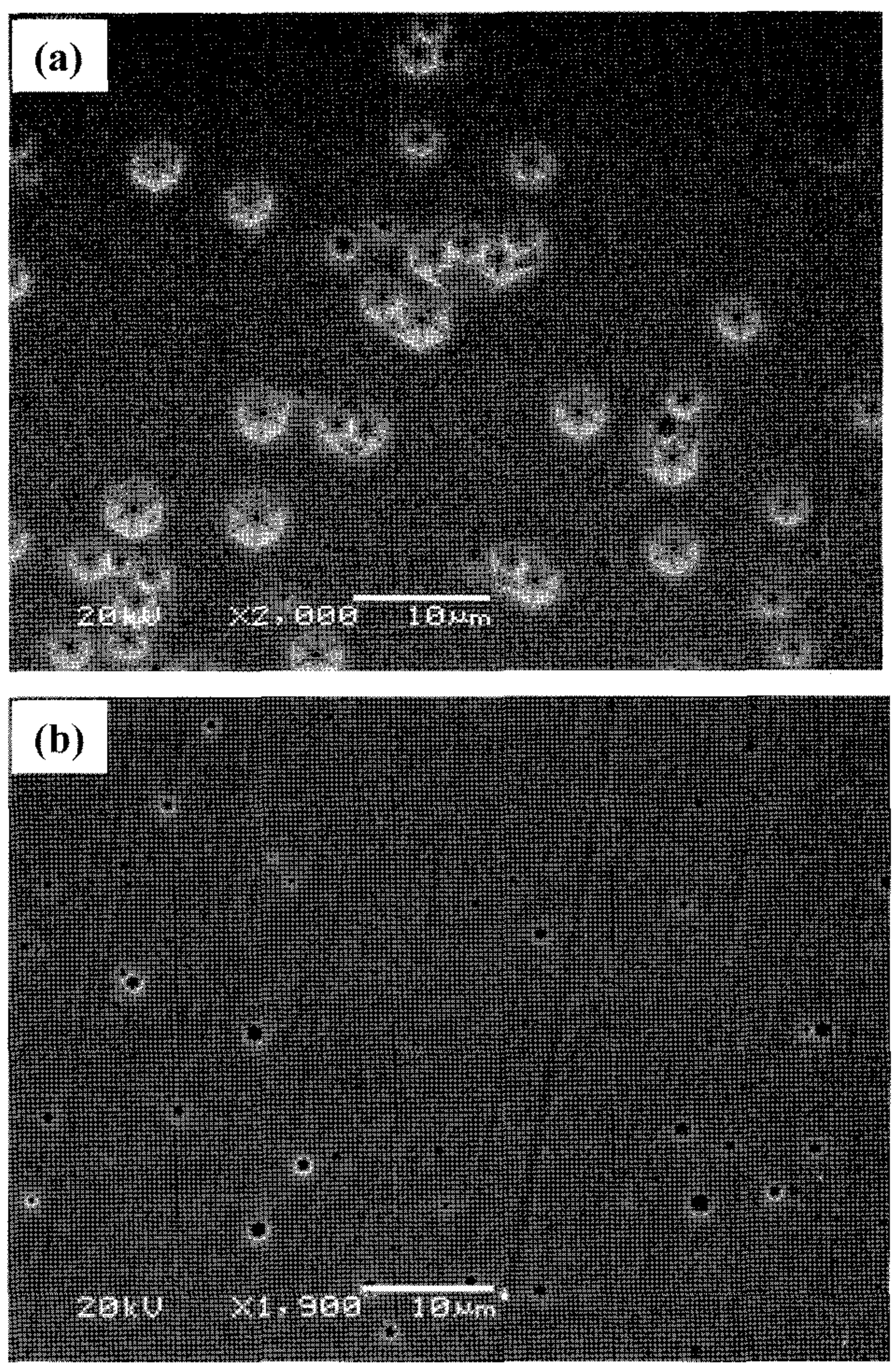

Fig. 2. EM images of etched GaN in (a) the horizontal reactor and (b) the vertical reactor.

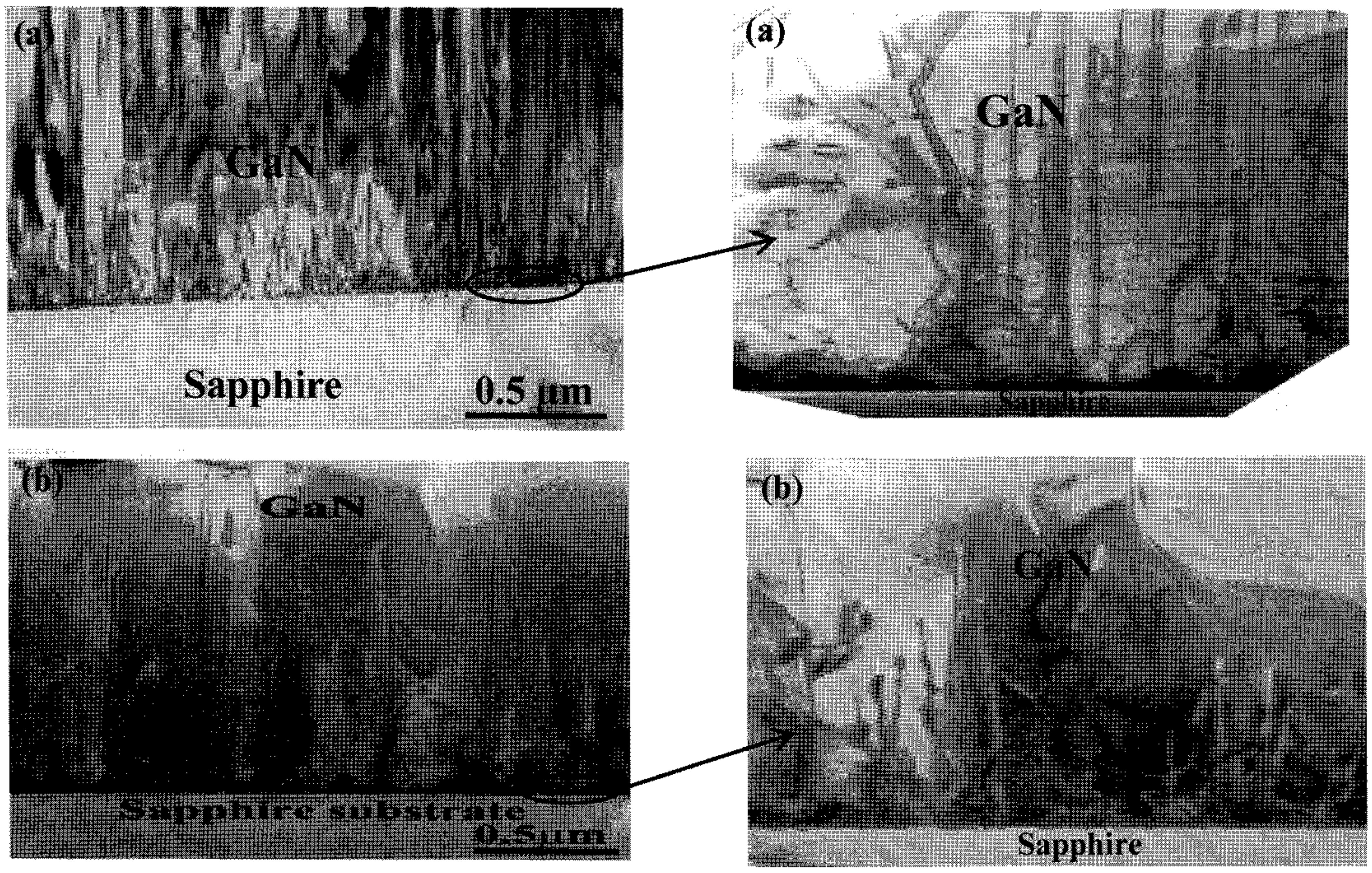

Fig. 3. Cross sectional TEM images of unetched GaN/sapphire in (a) the horizontal reactor and (b) the vertical reactor.

analysed by a cross-section TEM. In both of cases, the dislocations were originated from the substrate to GaN and dissipate most of the strain which was created during the growth due to the large difference between the lattice constants of the two materials. While the dislocation in GaN grown in horizontal reactor grows to GaN layer, there is dislocation interactions and annihilation leading to defect-free regions (Fig. 3) in GaN of the vertical growth. This will lead to a higher optical properties and crystal quality of GaN grown in the vertical reactor.

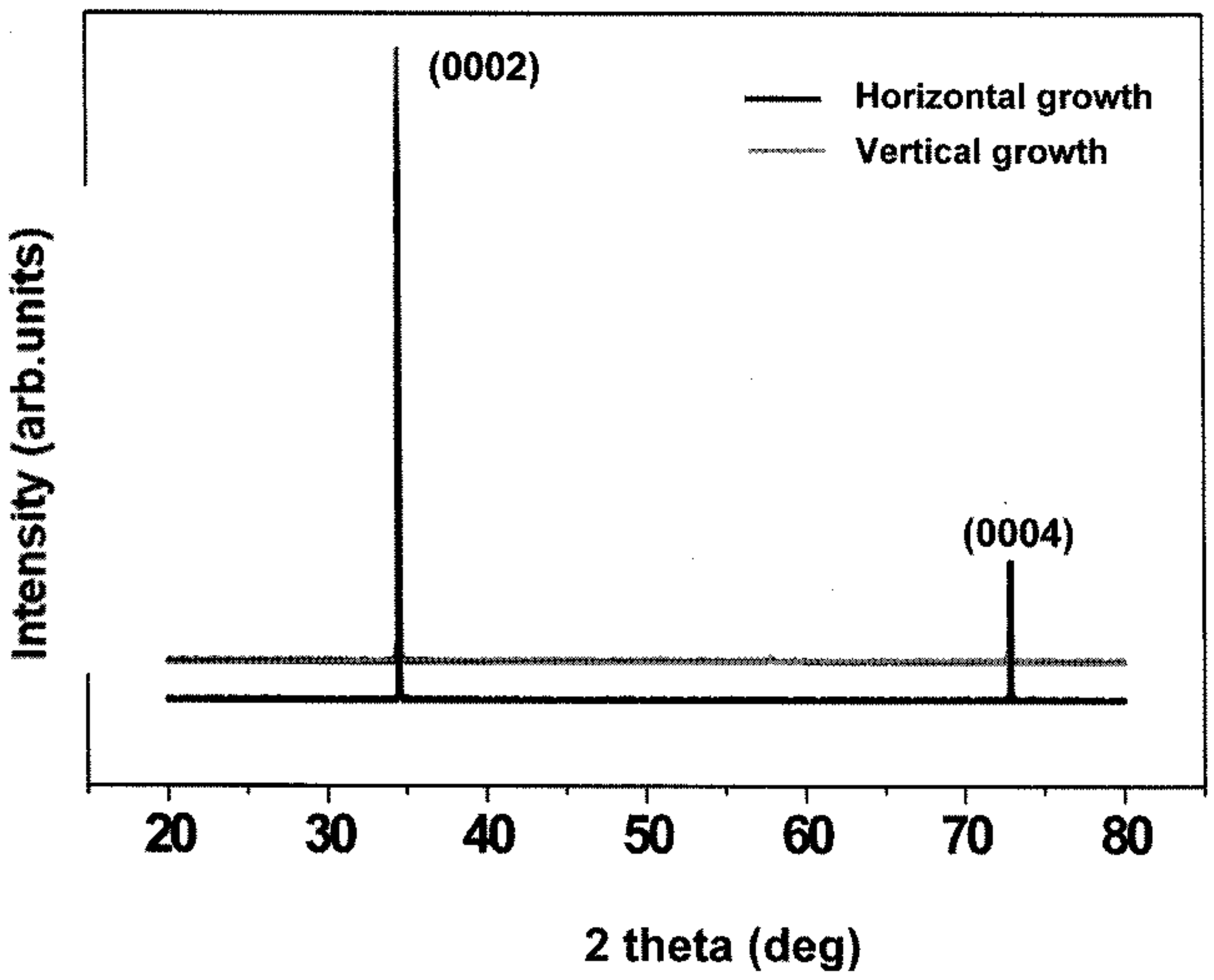

Fig. 4. XRD of GaN samples grown in the horizontal and vertical reactors.

Figure 4 shows the XRD pattern of the GaN film deposited on C-plane sapphire. There is a pronounced (0002) and (0004) peaks of the GaN. The epitaxial relationship is GaN <0001>// sapphire <0001>. That C-plane GaN was grown on C-plane sapphire by the HVPE and the orientation relationship between the GaN and the sapphire substrate could be established. The double-crystal x-ray rocking curve of the (0002) diffraction of the GaN film is found to have a full width at half maximum (FWHM) of 262 and 187 acrsec (Fig. 5). The about 100 arcsec broader FWHM in GaN grown in the horizontal reactor indicates a higher defect density which agrees with above EPD and TEM results.

The variation of PL spectra and intensity are shown in Fig. 6. Generally room-temperature PL spectra of GaN crystal are dominated by the band-edge transition at 3.39 eV. From the relative intensity of the band-edge PL peaks, it is recognized that GaN grown in the vertical reactor showed a higher PL intensity. In addition, defect-related YL bands are seen.

4. Conclusions

Thick c-plane GaN films were grown on (0001) sapphire in the vertical reactor and the horizontal reactor by the HVPE. It was found that the GaN film produced by

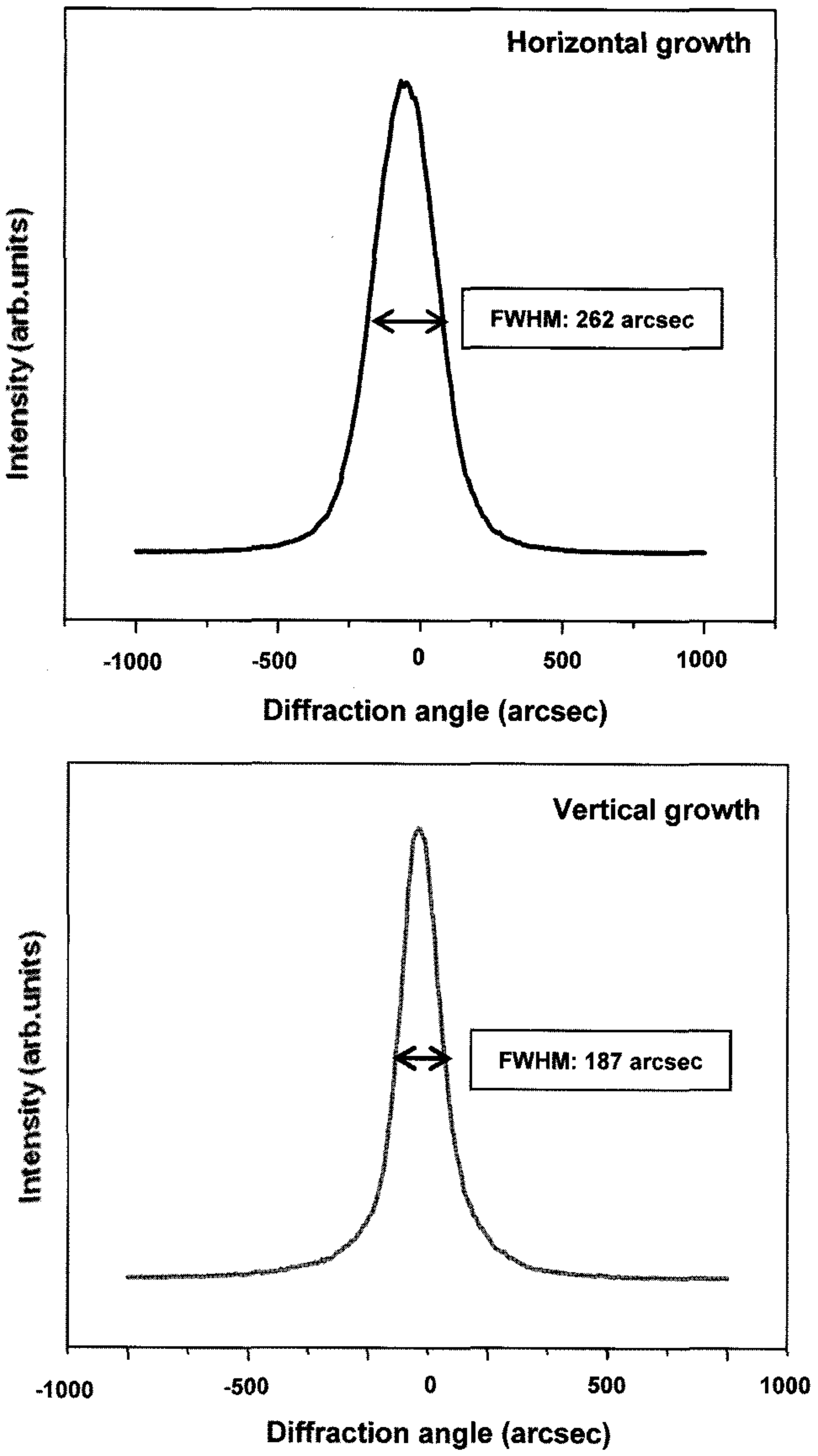

Fig. 5. XRD rocking curve of GaN samples grown in the horizontal and vertical reactors.

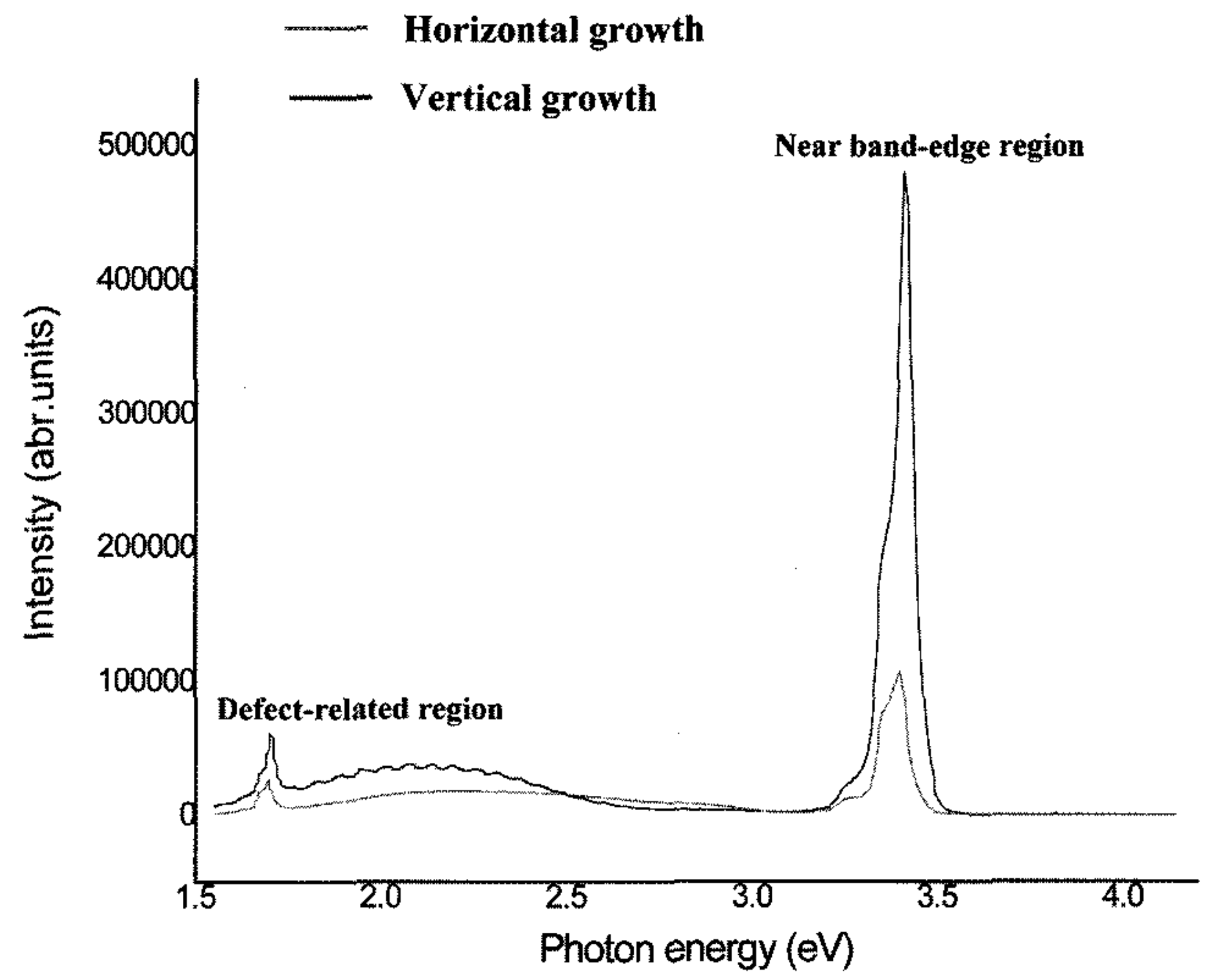

Fig. 6. PL spectra of GaN samples of the horizontal and vertical growth.

the vertical reactor had a better quality with optical properties and lower etch pits density. Ga-polar GaN films (0001) are selectively etched to show hexagonal etch pits reflecting the crystal symmetry of GaN. Moreover, the vertical grown epilayer etched by hot H_3PO_4 showed lower etch pits density which agreed with much higher PL intensity, narrower FWHM of DC-XRD and investigation of TEM compared with the horizontal grown GaN layer.

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