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Structural and optical properties of GaN thin films grown on Al_2O_3 substrates by MOCVD at different reactor pressures

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ABSTRACT

GaN thin films grown by MOCVD on (0 0 0 1) Al_2O_3 substrates at different growth pressures were characterized by field-emission scanning electron microscopy, atomic force microscopy, micro-Raman, and photoluminescence at room temperature. It was found that there is an optimum pressure of 76 Torr at which the structural and optical properties of the GaN samples are superior. On the other hand samples grown at higher pressure exhibited hexagonal surface pits and surface spirals. The results showed that the growth pressure strongly influences the morphology, and significantly affects the structural and optical properties of the GaN epilayers.

1. Introduction

In recent years, the GaN semiconductor and its III–V alloys have been considered promising materials for applications in optoelectronics [1–3] and high power electronic devices due to their unique properties like a wide direct bandgap capable of covering, for the case of photonic devices, from the visible to the ultra-violet region of the electromagnetic spectrum. As a matter of fact, short-wavelength light emitting diodes and lasers have been developed at commercial scale [1–5]. In spite the extensive research devoted to the III–V nitrides, many problems concerning structural and crystalline aspects must be solved in order to target the next-generation devices. In this work we realized a study on GaN films grown by MOCVD on sapphire substrates as a function of the reactor pressure. We found the MOCVD reactor pressure strongly influences surface morphology as long as the structural and optical properties of the GaN samples.

2. Experiment

GaN epilayers were grown on (0 0 0 1) c-plane of Al_2O_3 substrates at different reactor pressures. Before the growth, sapphire substrates were degreased in organic solvents, blew dry with nitrogen and then transferred into the MOCVD reactor. The MOCVD system consists of a home-made quartz chamber with a graphite susceptor coated with SiC [6]. The susceptor is heated by an external high power lamps arrangement, the temperature of the samples was measured with a thermocouple in contact with the back of the substrate. Trimethylgallium (TMGa) and ammonia (NH_3) were used as precursors and high purity hydrogen (H_2), from a palladium membrane filter, as the carrier gas. The GaN films were grown with the conventional two steps mode. The first step consisted of a GaN buffer layer at low temperature of 530 °C for 15 min; the second stage was the growth of a GaN film at 900 °C [7,8]. The thickness of the buffer layer was determined accurately as 20 nm.

During the experiments the fluxes were 3.7 lpm (litters per minute) for the H_2 carrier gas, 0.5 lpm for the NH_3 and 0.5 ccm (cubic centimeters per minute) for the TMGa (equivalent to 6 μmol/min). Three samples grown at different reactor pressures were studied: M435 (95 Torr), M427 (80 Torr) and MFSC88 (76 Torr). The thickness of the samples was estimated by scanning electron microscopy in about 1 μm.

The field-emission scanning electron microscopy (FE-SEM) images were taken in a Jeol equipment model JSM-7401F with a resolution of 1.5 nm. The atomic force microscopy (AFM) characterization was performed in a non-contact Veeco microscope model Rtespa employing silicon tips with a curvature radii less than 10 nm, the AFM images were taken with a resolution of 250 × 250 pixels at

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a frequency of 2 Hz. The micro-Raman analysis was performed in the backscattering configuration in an equipment model LabRAM from Jobin Yvon with the 632.8 nm line. Photoluminescence spectra were measured with the 325 nm line at room temperature employing a 4 mW He–Cd laser from Kimmon Electric model IK3101R-D. Transmission electron microscopy (TEM) was performed in a FIA microscope model TITAN at 300 kV. Plan view samples were prepared with the standard mechanical procedure and ion milling with Ar^+ ions until electron transparent specimens were obtained.

3. Results and discussion

Fig. 1a shows a FE-SEM image of sample M435 which consists of a GaN film grown by MOCVD on an Al_2O_3 substrate at 95 Torr. Wide terraces surrounded by hexagons are observed on the surface of the film. In the image the large terraces correspond to the GaN epilayer deposited on the sapphire substrate and the polygons are surface pits with a density of $5.7 \times 10^7/cm^2$. The surface pits have hexagonal shape although those with smaller dimensions are circular in shape (the density was estimated considering both small circular and hexagonal-shaped surface pits). Larger hexagons have 1 μm from opposite sides.

In Fig. 1b the FE-SEM picture of the sample M427 grown at 80 Torr is depicted. The topography of this GaN sample appears with ridges arranged in spirals and the hexagons, seen in the previous figure, in less quantity. The surface hexagons density was $2.5 \times 10^6/cm^2$. The morphology of the sample MFSC88 grown at 76 Torr is shown in Fig. 1c. The surface is completely different for this epilayer. Truncated pyramids can be observed on a flat GaN surface.

AFM pictures are presented in Fig. 2a–c, the area under analysis was 100 μm^2. In Fig. 2a, a plan view of the GaN film corresponding to sample M435 shows the small circular and hexagonal-shaped pits, this image is consistent with its FE-SEM counterpart, hexagons seem to be immersed on the GaN regions. In the inset of Fig. 2a, a height profile demonstrates that surface hexagons have depths between 0.1 μm and 0.3 μm, this last value is around one third of the GaN film thickness.

The GaN ridges that form the spirals on the surface of the sample M427 are imaged by AFM in Fig. 2b. Ridges form helical paths on the surface of the film. Fig. 2c is the AFM picture of the sample MFSC88. The surface differs from the other samples. GaN crystals were formed at this reactor pressure (76 Torr) during the growth. The surface hexagons seen in sample M435 and the surface spirals imaged in sample M427 were not detected.

The growth conditions were the same for the three samples the only parameter that changed was the reactor pressure. These images indicate that the MOCVD reactor pressure strongly modifies the morphology of the GaN epilayers.

The presence of surface pits as a result of the GaN growth on Al_2O_3 substrates has been observed in samples grown by MOCVD [9], MBE [10] and HVPE [11]. In these papers surface pits were related to dislocations. Threading dislocations (TDs) in GaN films grown by MOCVD are of three types depending on the Burgers (b) vector [12]. Pure edge dislocations or a-type have a $b = 1/3\langle 1\,1\,{-2}\,0\rangle$, b-type or pure screw dislocations with $b = \langle 0\,0\,0\,1\rangle$ and mixed TDs or (a + b)-type with a Burgers vector of $b = 1/3\langle 1\,1\,{-2}\,3\rangle$. In particular screw and mixed TDs are associated to GaN steps terminations observed in AFM images [13,14]. In Fig. 1 small circular pits can be found in the division line between GaN steps. Even larger hexagons are in the trajectory of the border lines and presumably if the GaN growth will continue they were filled by the material ending with circular shape as the case of smaller pits. The height profile shown in the inset of Fig. 2a indicates depths of surface pits are a fraction of the film thickness so the growth filled them up. Therefore

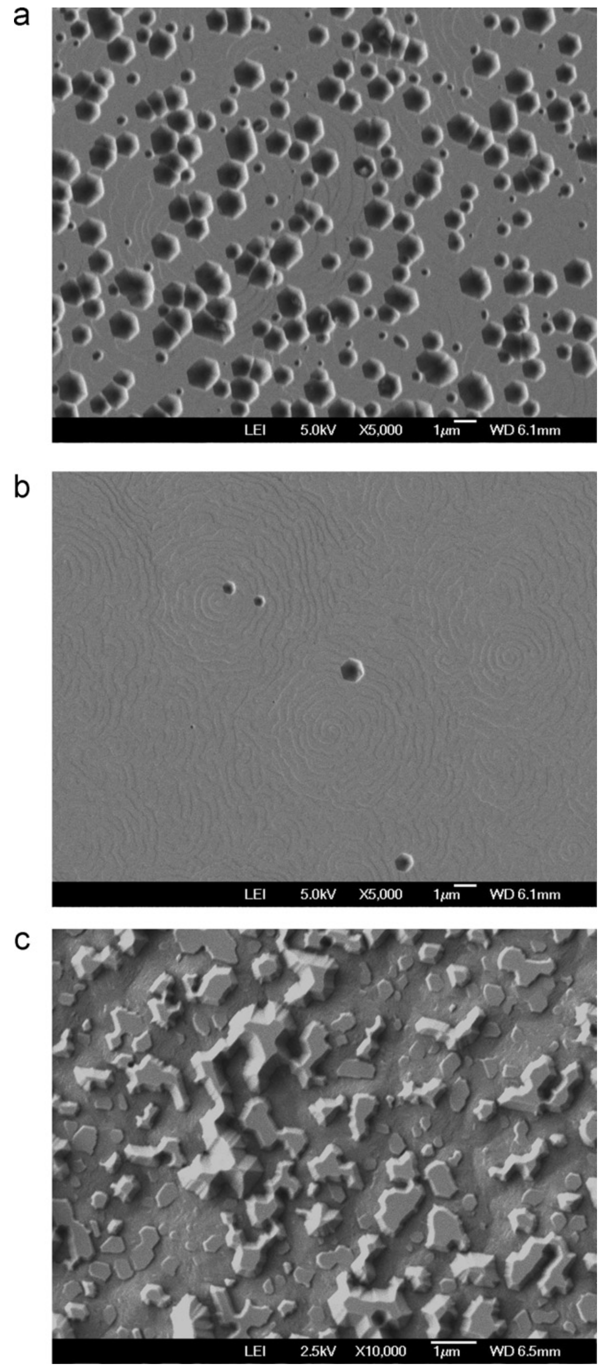

Fig. 1. FE-SEM images of GaN films grown on sapphire substrates at different MOCVD reactor pressures. (a) The surface of M435 grown at 95 Torr. Note the small circular pits and the large hexagonal-shaped surface pits. The micrograph of M427, grown at 80 Torr, is shown in (b). Surface spirals with helical ridges can be distinguished. In (c), the image of sample MFSC88 grown at 76 Torr shows a different morphology where the surface pits and spiral were not imaged.

we conclude that circular and hexagonal-shaped surface pits in the samples of Figs. 1 and 2 could be related to screw/mixed TDs. In literature there have been reports of GaN surface pits after etching [9,13,14] in higher density and with different dimensions than those presented in this work; we believe this could be due to different thicknesses of the samples and the etching mechanism as compared with the morphology of the as-grown GaN films in this paper.

On the other hand, surface spirals have been reported for many materials, moreover there is a model due to Frank et al. [15], which