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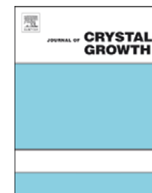

Homoepitaxy on bulk ammonothermal GaN

R. Dwilinski *, R. Doradzinski, J. Garczynski, L.P. Sierzputowski, M. Zajac, M. Rudzinski

AMMONO Sp. z o.o., Czerwonego Krzyża 2/31, Warszawa 00-377, Poland

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ABSTRACT

In this work results of extensive characterization of homoepitaxial layers grown on truly bulk ammonothermal gallium nitride (GaN) substrates are presented. The 2-μm-thick layers were deposited using metalorganic chemical vapor deposition. The photoluminescence (PL) and reflectance results show very intensive, perfectly resolved excitonic structure in range of band-edge emission of gallium nitride. This structure consists of both lines related to free excitons emission and very narrow lines (full-width at half-maximum (FWHM) value of the order of 0.3 meV) related with excitons bound to neutral acceptor and different neutral donors. In high excitation condition the biexciton emission was observed. The luminescence is uniform in the whole sample surface range. High PL homogeneity corresponds with structural and microscopic measurements performed on these layers. It proves that ammonothermal GaN substrates with perfect crystalline properties enable to grow excellent quality, strain-free homoepitaxial layers.

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1. Introduction

Gallium nitride (GaN) has attracted a great attention for its material properties that are useful for applications in short-wavelength optoelectronic and high-power electronic devices [1,2]. Unfortunately, these devices are manufactured mainly by heteroepitaxial methods. The well-known disadvantages of heteroepitaxy are: lattice mismatch, thermal-expansion-coefficient difference and chemical incompatibility, which in effect lead to highly stressed GaN epilayers with high dislocation density and mosaic crystal structure. This limits seriously the life time and power of the aforementioned devices. Under such circumstances the growth of bulk GaN, which can be used as a substrate for homoepitaxial growth, is highly demanded.

During the past decade, an enormous effort was undertaken to find the appropriate technique for bulk GaN production. So far, various approaches to achieve truly bulk or quasi-bulk GaN substrates have been proposed [3–6]. At present, the hydride vapor phase epitaxy (HVPE) technique is the most common method, mainly due to the high growth rate. However, this method suffers from all disadvantages resulting from the necessity of the use of non-native substrates (sapphire). On the other hand, the ammonothermal method boosts a lot of new hope to overcome this obstacle because of its fundamental advantages, such as the possibility of growing high-diameter truly bulk monocrystals with excellent structural properties, low operation costs and very good scalability desired for commercial applications. In this communication we present the results of extensive (optical, structural and microscopic) studies of homoepitaxial layers grown on truly bulk ammonothermal GaN substrates. We show that ammonothermal GaN grown at AMMONO company (A-GaN) can be successfully used as substrates for homoepitaxy.

2. Sample preparation

Truly bulk GaN substrates for homoepitaxy were grown by the ammonothermal method at AMMONO company. This technique can be regarded as an analogue of commercially available hydrothermal technology of α-quartz production in which the supercritical aqueous solution can be used for chemical transport and re-crystallization of silicone dioxide (SiO_2). In the ammonothermal method supercritical ammonia plays a role of water, and nitrides can be grown instead of oxides. It is based on chemical transport of GaN from low-temperature dissolution zone of the autoclave to the high-temperature crystallization zone [6,7] in the presence of a mineralizer—the ionic substance used to increase a reversible dissolution of ammonia. The obtained crystals have excellent crystalline quality—full-width at half-maximum (FWHM) value of X-ray rocking curve is at the order of 10 arcsec, the radius of curvature of the crystal lattice of the order 100–1000 m and dislocation density of about $5\times10^3 cm^{-2}$. The carrier concentration of the resulting crystals can be

*Corresponding author. Tel.: +48 601 387 901; fax: +48 22 814 02 07.
E-mail address: dwilinski@ammono.com (R. Dwilinski).

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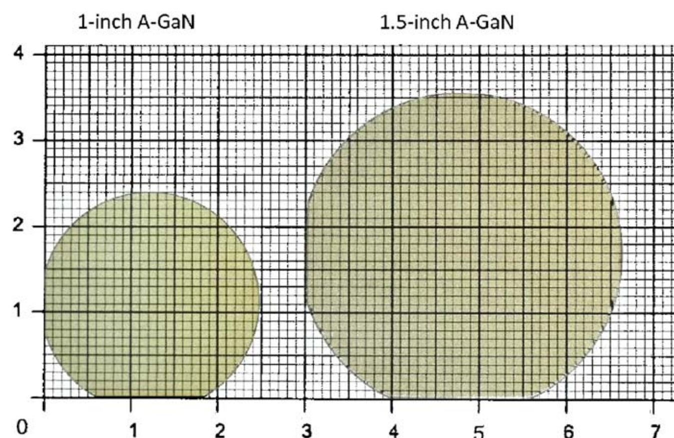

Fig. 1. Photograph of 1 and 1.5-in A-GaN wafer manufactured at AMMONO company.

controlled by appropriate doping [8]. Both n-type ($n\sim10^{19}\,cm^{-3}$) and semi-insulating ($\rho\sim10^{10}\,\Omega\,cm$) substrates can be grown via the ammonothermal method. Recently, we have succeeded in manufacturing 1-in A-GaN substrate—a *c*-plane oriented and polished wafer sliced from a larger A-GaN crystal [7]. In this paper, we present for the first time the 1.5-in A-GaN substrate (Fig. 1). The enlargement of size is possible due to perfect scalability of the ammonothermal method. We performed homoepitaxy on Ga-face of such polished and oriented substrates without any buffer layer.

The epilayers were deposited by metalorganic vapor phase epitaxy (MOVPE) using a RF-heated AIXTRON AIX-200 low-pressure horizontal reactor on Ga-face of the polar direction (0 0 0 1) of GaN crystal. Trimethylgallium and ammonia were used as Ga and N precursors, respectively. The growth procedure was started by annealing the substrate for 5 min at high temperature (1100 °C) in a H_2+NH_3 atmosphere at a total reactor pressure of 350 mbar. For all samples grown on A-GaN substrates the ammonia (NH_3) was kept open from the moment when the temperature in the reactor reached 500 °C till the end of the process when the grown sample was cooled down to 500 °C. This ammonia gas flow was kept constant using 2 standard liters per minute (slm) till the end of the growth process. This step was used to prevent first GaN substrate and then epitaxial layers from decomposition [9]. After the annealing, the growth time was set to give 1- and 2-μm-thick GaN layer. The growth temperature was 1170 °C for the whole process and was carried out at a total reactor pressure of 50 mbar. The epilayers were characterized by various techniques: X-ray diffraction, defect selective etching (DSE), microphotoluminescence (μ-PL), photoluminescence (PL) and reflectance.

3. Structural study

In order to check the overall structural quality of GaN epilayers grown on A-GaN substrates, the X-ray rocking curves mode measurements were performed on samples by means of a Panalytical X'Pert Pro MRD high-resolution diffractometer with the following set-up: CuKα1 line, U=40 kV, 0.1 mm × 0.1 mm slit for the incident beam and an open-detector mode for the diffracted beam. Representative rocking curves measured on (0 0 0 2) plane of epilayer grown on n-type and SI substrate are shown on Fig. 2. The resulting FWHM values (22–24 arcsec) are, to our knowledge, outstanding results that strongly indicate excellent crystalline properties of the studied material. This

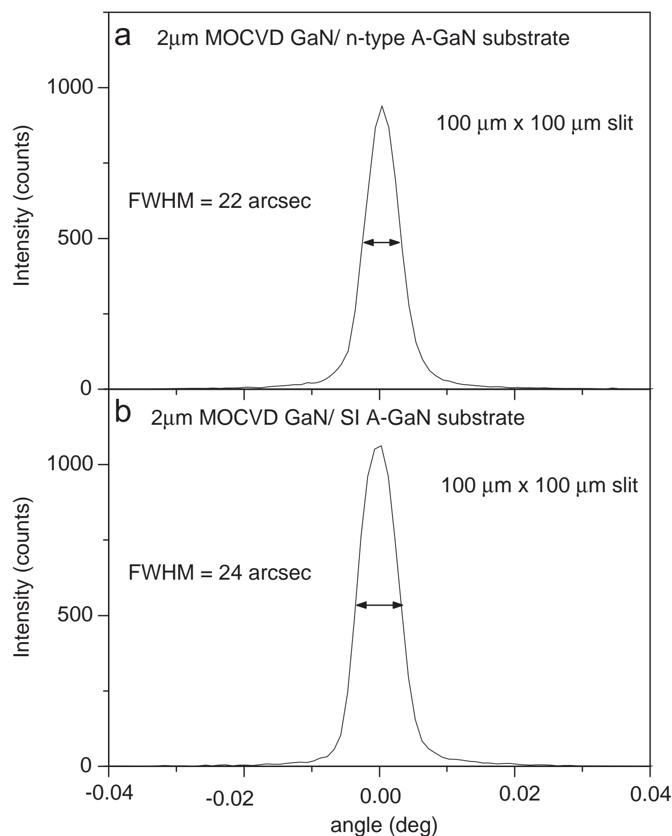

Fig. 2. X-ray rocking curves of the GaN epilayers grown on n-type (a) and SI (b) A-GaN substrates for the symmetric (0 0 0 2) reflections.

result is a direct consequence of high crystalline quality of A-GaN substrates adapted to MOVPE growth.

In the next step, for the purpose of determining the type and density of dislocations propagating through the layer, defect selective etching in a molten eutectic mixture of KOH–NaOH (E) [10] was applied. In order to reveal all types of threading dislocations, the temperature was set to 420 °C and etching time was 5–30 min. Due to low number of defects, the etching process was few times longer than typical etching time used so far [11]. In this way the size of etch pits is increased, therefore, dislocation density can be revealed and calculated using differential interference contrast (DIC) optical microscopy. In some cases, where local dislocation density was higher and then additional scanning electron microscopy (SEM) observations were performed. Fig. 3 presents SEM images taken at smaller (five-hundredth—Fig. 3b) and larger (five-thousandth—Fig. 3c) magnifications.

In general, three types of dislocations (edge, screw and mixed) can be primarily distinguished after selective etching of the GaN layer grown on the *c*-plane-oriented substrate (Fig. 3a). They are visualized by different patterns of hexagonal pits, occurring in three grades of size. In the first approach, the size and morphology of etch pits formed on dislocations in GaN during etching depend mainly on the energy of dislocations represented by their Burgers vector [11,12]. The dislocations in the wurtzite GaN lattice are characterized by large difference in the magnitude of the Burgers vector: edge dislocations (b_e)=a, screw dislocations (b_s)=2.66a, and mixed dislocations (b_m)=3.66a, where a is the lattice constant. The thermodynamics of the etching process predicts that the change of the chemical potential during formation of the etch pits is inversely proportional to the energy of the dislocations. Therefore, one expects that dislocations of larger energy will be etched easier

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