

PACS numbers: 61.05.cp, 61.72.Cc, 61.72.Mm, 68.35.Ct, 68.55.J-, 81.07.Bc, 81.15.Cd

## Synthesis and Structure of Thin GaN Films by Radio-Frequency Sputtering

O. M. Bordun<sup>1</sup>, I. Yo. Kukharsky<sup>1</sup>, M. V. Protsak<sup>1</sup>, I. I. Medvid<sup>1</sup>,  
I. M. Kofliuk<sup>1</sup>, Zh. Ya. Tsapovska<sup>1</sup>, and D. S. Leonov<sup>2</sup>

<sup>1</sup>*Ivan Franko National University of Lviv,  
50, Drahomanov Str.,  
UA-79005 Lviv, Ukraine*

<sup>2</sup>*Technical Centre, N.A.S. of Ukraine,  
13, Pokrovska Str.,  
UA-04070 Kyiv, Ukraine*

The structure and features of thin GaN films deposited by means of the radio-frequency (RF) ion-plasma sputtering are investigated. As shown, thin GaN films are formed from nanocrystallites with average dimensions of 14.3 nm and rather low crystal-lattice stresses. The influence of both the working-gas N<sub>2</sub> pressure and the substrate temperature on the film deposition rate is investigated.

Досліджено структуру й особливості нанесення тонких плівок GaN способом високочастотного (ВЧ) йонно-плазмового розпорошення. Показано, що тонкі плівки GaN формуються з нанокристалітів, середні розміри яких становлять 14,3 нм із достатньо низькими значеннями напружень кристалічної ґратниці. Досліджено вплив тиску робочого газу N<sub>2</sub> та температури підкладки на швидкість нанесення плівок.

**Key words:** gallium nitride, thin films, RF sputtering, structure.

**Ключові слова:** нітрид Галію, тонкі плівки, високочастотне напорошення, структура.

(Received 23 March, 2024)

## 1. INTRODUCTION

Thin films of gallium nitrides and oxides are the objects of intensive research due to the prospects of their use in the creation of effective sources of visible and near ultraviolet (UV) radiation, elec-

troluminescent displays, and receivers of ionizing and UV radiations [1–6]. In addition, possessing unique parameters such as a large band gap, high charge-carrier drift velocity, high breakdown voltage, and high chemical and thermal stability, GaN is one of the most promising materials for creating new generation microelectronic and nanoelectronic devices [7–9].

In general, the question of the physical properties of thin films is complicated by the fact that films do not always have a perfect structure and can be polycrystalline, amorphous, or contain inclusions of other phases. Obtaining stable, reproducible properties of polycrystalline films is further complicated by the presence of intergranular boundaries (IGB). The physical properties of polycrystalline thin films are largely determined not only by the material properties but also by the energy levels arising from the presence of the IGB. Such levels are also determined by the size of the crystallites that form the thin films. Therefore, the problem of analysing the size of crystallites and the influence of IGB is an important task, when investigating the possibility of using thin films in optoelectronic devices, in particular, light-emitting structures, or studying electrical conductivity processes. This has led to the structural studies of thin GaN films reported in this paper. The films were obtained by the method of RF ion-plasma sputtering, which is optimal for obtaining homogeneous semiconductor and dielectric films [10].

## 2. EXPERIMENTAL TECHNIQUE

Thin GaN films with a thickness of 0.3–1  $\mu\text{m}$  were obtained by RF ion-plasma sputtering on sapphire substrates ( $\text{Al}_2\text{O}_3$ ). The RF sputtering was carried out in a nitrogen atmosphere at pressures from  $5 \cdot 10^{-3}$  to  $5 \cdot 10^{-2}$  Torr. The target for sputtering was metallic Ga. The temperature of the substrates during sputtering varied from 400 to 650°C, and the RF discharge power was from 100 to 150 W.

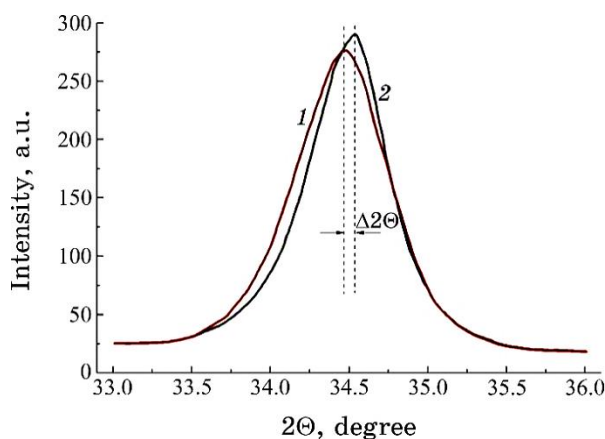
The structure and phase composition of the obtained films were studied by x-ray diffraction analysis (Shimadzu XDR-600). X-ray diffraction studies showed the presence of a polycrystalline structure with a predominant orientation in the (002) plane. The structure of the films is close to the ideal wurtzite structure. The thicknesses of the films were determined based on the interference pattern in the transmission spectra according to the method [11, 12].

## 3. RESULTS AND DISCUSSION

One of the most reliable methods for determining the composition and structure of the substance under study is x-ray phase analysis.

The characteristic x-ray diffractograms of thin GaN films obtained by RF ion-plasma sputtering in a nitrogen atmosphere are shown in Fig. 1. The analysis of the diffractogram shows that it contains a dominant, relatively wide reflection band in the region  $2\Theta \approx 33.5\text{--}35.5^\circ$ , which is caused by the reflection reflex from the (002) plane in GaN with a hexagonal wurtzite structure. It should be noted that this situation is in a good agreement with the results of some studies of GaN films obtained by RF sputtering [13, 14]. A similar situation is observed in the RF sputtering of oxide compounds [15].

For the ordered wurtzite structure, the maximum for the reflection from the (002) plane has a value of  $2\Theta \approx 34.4^\circ$ . A precise comparison of the diffraction band due to reflection from the (002) plane in the obtained GaN films and the ideal wurtzite structure revealed some shifts of the  $2\Theta$  maximum that exceed the hardware error of the experiment ( $\Delta 2\Theta > 0.07^\circ$ ). The half-width of this band also slightly increases. The shift of the maximum of this reflection band toward lower values of  $2\Theta$  may be due to the partial increase in interplanar distances in GaN films relative to single-crystal samples. This situation is in good agreement with the characteristic lower packing density of thin films relative to single crystal samples [16]. In addition, during RF sputtering, the structure of the resulting films depends on such factors as RF-radiation power, substrate temperature, pressure in the working chamber, energy, and composition of the bombarding ions. Considering that the sputtering atmosphere consists of nitrogen, the formation of defects containing excess nitrogen due to the reactivity of the sputtering and the interaction of nitrogen contained in the plasma with the formed GaN film is crucial for the formation of films.



**Fig. 1.** X-ray diffractograms of thin GaN films obtained by RF sputtering inside the  $N_2$  atmosphere (1) and ordered wurtzite structure (2) (reflex (002)).

It is known that the width of the diffraction bands depends on the size of the coherent scattering regions (the size of the nanocrystallites) and the presence of defects and mechanical stresses in the sample. Thus, the data on the widths of diffraction bands on x-ray diffraction patterns can be used to determine the size of the particles, which form the sample and to analyse the mechanical stresses in the samples [17].

According to Debye and Scherrer, the relationship between the diffraction bandwidth and the crystallite size  $d$  is given by equation

$$d = \frac{0.94}{b \cos \Theta}, \quad (1)$$

where  $\lambda$  is the diffracting wavelength, which in our case is of 0.15418 nm ( $\text{CuK}_\alpha$ -radiation),  $\beta$  is the half-width of the band (width at half height), and  $\Theta$  is the diffraction angle.

The second reason for the broadening of the bands in the diffractograms may be imperfections in the crystallite structure and defects that cause stresses in the crystals [18]. The band broadening associated with stresses is described by equation

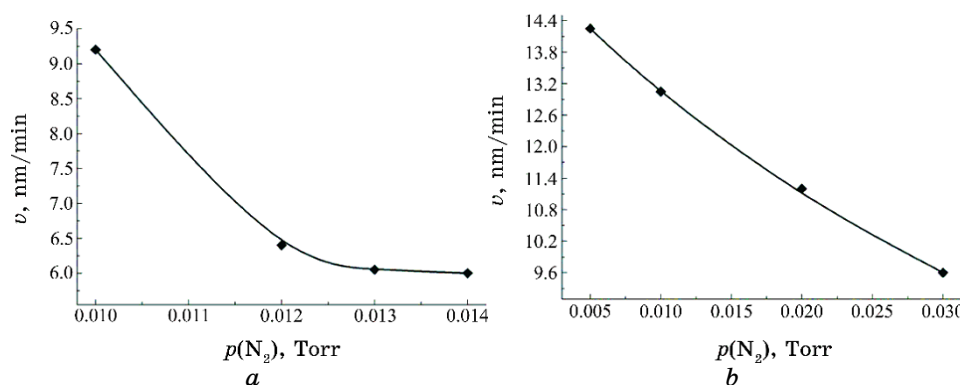
$$\varepsilon = \frac{\beta}{4 \operatorname{tg} \Theta}, \quad (2)$$

where  $\varepsilon$  is the average stress value of the crystal lattice.

The calculations for the main characteristic diffraction band of reflection from the (002) plane in thin GaN films show that the average size of nanocrystallites forming a thin film is  $d = 14.3$  nm and the average stress is  $\varepsilon = 0.0045$ . The obtained values of  $\varepsilon$  indicate that GaN films obtained by RF sputtering in a nitrogen atmosphere are characterized by rather low average lattice stresses. For comparison, for example, when RF sputtering thin films based on  $\text{Y}_2\text{O}_3$ , depending on the sputtering atmosphere, the average stresses are  $\varepsilon = 0.0125\text{--}0.0148$  [19].

Our studies show a significant effect on the rate of deposition of thin GaN films of the  $\text{N}_2$  pressure in the sputtering chamber and the temperature of the substrate, on which the films are deposited.

The dependence of the thin GaN film deposition rate on the pressure of the working gas  $\text{N}_2$  at different RF radiation powers is shown in Fig. 2. As can be seen from the results, the sputtering rate is decreased with increasing the  $\text{N}_2$  pressure. At the same time, for the same pressure values, an increase in power leads to an increase in the speed of film deposition. For example, as can be seen in Fig. 2, *a*, *b*, at an  $\text{N}_2$  pressure of 0.01 Torr at a power of 100 W, the film deposition rate averages to 9.2 nm/min, and at a power of 140 W, this value reaches 13 nm/min. In addition, it was found



**Fig. 2.** Dependence of the deposition rate of thin GaN films during RF sputtering on the pressure of the working gas ( $\text{N}_2$ ) at a power of 100 W (a) and 140 W (b). The substrate temperature is  $600^\circ\text{C}$ .

that with a decrease in the power of RF radiation, the rate of deposition of GaN films with increasing working gas pressure begins to decrease more and more slowly.

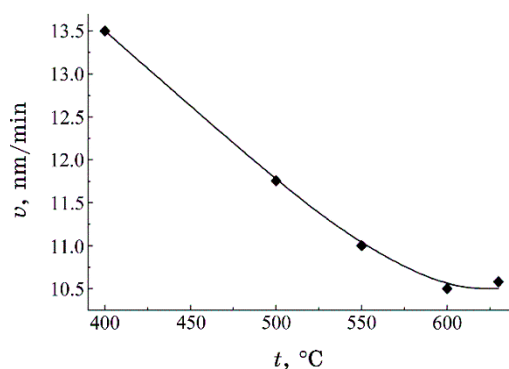
The decrease in the sputtering coefficient, which is estimated by the rate of deposition of GaN onto the sapphire substrate with the increasing pressure of the working gas  $\text{N}_2$ , can be explained based on the phenomena of backward diffusion and backscattering.

During backward diffusion, the diffuse return of sputtered atoms with an average kinetic energy  $E_a$  approximately equal to the average kinetic energy of the working-gas atoms  $E_N$  ( $E_a \approx E_N$ ) falling on the target is observed. In backscattering, the atoms, which have been atomized, return to the target due to their scattering on the working gas atoms. The observed phenomenon of a decrease in the rate of deposition of thin GaN films depending on the substrate temperature can also be explained based on the backward diffusion phenomenon (Fig. 3).

As can be seen from the above results, with an increase in the substrate temperature from  $400$  to  $600^\circ\text{C}$ , a rather noticeable decrease in the rate of deposition of thin GaN films is observed. Within the temperature region around  $600^\circ\text{C}$  and higher, this dependence reaches saturation. This may indicate that starting from a substrate temperature of about  $600^\circ\text{C}$  and above, there is certain equilibrium between the number of deposited atoms and the number of diffusely returned atoms.

#### 4. CONCLUSIONS

Our studies show that the RF ion-plasma sputtering in a nitrogen



**Fig. 3.** Dependence of the deposition rate of thin GaN films during RF sputtering on the substrate temperature at a power of 100 W and a chamber pressure of 0.01 Torr.

atmosphere produces thin GaN films with average crystallite sizes of 14.3 nm and rather low lattice stresses. The decrease in the film deposition rate with increasing working gas pressure and increasing substrate temperature, which are associated with the processes of backward diffusion and backscattering, was found.

## REFERENCES

1. C. M. Furqan, Jacob Y. L. Ho, H. S. Kwok, *Surfaces and Interfaces*, **26**: 101364 (2021); <https://doi.org/10.1016/j.surfin.2021.101364>
2. L. Srinivasan, C. Jadaud, F. Silva, J.-Ch. Vanel, J.-L. Maurice, E. Johnson, P. Roca i Cabarrocas, and K. Ouaras, *J. Vac. Sci. Technol. A*, **41**: 053407 (2023); <https://doi.org/10.1116/6.0002718>
3. F. Roccaforte and M. Leszczynski, *Nitride Semiconductor Technology. Power Electronics and Optoelectronic Devices* (Wiley–VCH Verlag GmbH & Co. KGaA: 2020).
4. M. Higashiwaki, *AAPPS Bull.*, **32**: 3 (2022); <https://doi.org/10.1007/s43673-021-00033-0>
5. O. M. Bordun, B. O. Bordun, I. Yo. Kukharskyy, and I. I. Medvid, *J. Appl. Spectrosc.*, **86**, No. 6: 1010 (2020); <https://doi.org/10.1007/s10812-020-00932-4>
6. O. M. Bordun, B. O. Bordun, I. Yo. Kukharskyy, and I. I. Medvid, *J. Appl. Spectrosc.*, **84**, No. 1: 46 (2017); <https://doi.org/10.1007/s10812-017-0425-3>
7. A. Zhong, L. Wang, Y. Tang, Yo. Yang, J. Wang, H. Zhu, Zh. Wu, W. Tang, and B. Li, *Chin. Phys. B*, **32**: 076102 (2023); <https://doi.org/10.1088/1674-1056/acb8a>
8. M. Monish, Sh. Mohan, D. S. Sutar, and S. S. Major, *Semicond. Sci. Technol.*, **35**, No. 4: 045011 (2020); <https://doi.org/10.1088/1361-6641/ab73ec>
9. V. Bondar, I. Kucharsky, B. Simkiv, L. Akselrud, V. Davydov, Yu. Dubov, and S. Popovich, *phys. stat. sol. (a)*, **176**, No. 1: 329 (1999);

- [https://doi.org/10.1002/\(SICI\)1521-396X\(199911\)176:1<329::AID-PSSA329>3.0.CO;2-E](https://doi.org/10.1002/(SICI)1521-396X(199911)176:1<329::AID-PSSA329>3.0.CO;2-E)
10. Kiyotaka Wasa, Makoto Kitabatake, and Hideaki Adachi, *Thin Film Materials Technology: Sputtering of Compound Materials* (William Andrew: 2004).
  11. R. Swanepoel, *J. Phys. E: Sci. Instrum.*, **16**, No. 12: 1214 (1983);  
<https://doi.org/10.1088/0022-3735/16/12/023>
  12. A. P. Caricato, A. Fazzi, and G. Leggieri, *Applied Surface Science*, **248**: 440 (2005); <https://doi.org/10.1016/j.apsusc.2005.03.069>
  13. T. Maruyama and H. Miyake, *J. Vac. Sci. Technol. A*, **24**, No. 4: 1096 (2006);  
<https://doi.org/10.1116/1.2208988>
  14. R. S. de Oliveira, H. A. Folli, C. Stegemann, I. M. Horta, B. S. Damasceno, W. Miyakawa, A. L. J. Pereira, M. Massi, A. S. da Silva Sobrinho, and D. M. G. Leite, *Materials Research.*, **25**: e20210432 (2022);  
<https://doi.org/10.1590/1980-5373-MR-2021-0432>
  15. O. M. Bordun, I. O. Bordun, and I. Yo. Kukharskyy, *J. Appl. Spectrosc.*, **82**, No. 3: 390 (2015); <https://doi.org/10.1007/s10812-015-0118-8>
  16. O. M. Bordun and L. M. Lymarenko, *Ukr. J. of Physics*, **42**, Nos. 11–12: 1390 (1997) (in Ukrainian).
  17. S. L. Morelhão, *Computer Simulation Tools for X-Ray Analysis. Scattering and Diffraction Methods* (Cham, Switzerland: Springer International Publishing: 2016).
  18. U. Welzel, J. Ligot, P. Lamparter, A. C. Vermeulen, and E. J. Mittemeijer, *J. Appl. Cryst.*, **38**: 1 (2005); <https://doi.org/10.1107/S0021889804029516>
  19. O. M. Bordun, I. O. Bordun, I. M. Kofliuk, I. Yo. Kukharskyy, I. I. Medvid, O. Ya. Mylyo, and D. S. Leonov, *Nanosistemi, Nanomateriali, Nanotekhnologii*, **17**, Iss. 4: 711 (2019); <https://doi.org/10.15407/nnn.17.04.711>