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Surface Morphology of Thin β-Ga_2O_3 Films Obtained by Radio-Frequency Sputtering

O. M. Bordun[1], B. O. Bordun[1], I. Yo. Kukharskyy[1], I. I. Medvid[1],
I. I. Polovynko[1], Zh. Ya. Tsapovska[1], and D. S. Leonov[2]

[1]*Ivan Franko National University of Lviv,*
50, Drahomanov Str.,
UA-79005 Lviv, Ukraine
[2]*Technical Centre, N.A.S. of Ukraine,*
13, Pokrovska Str.,
UA-04070 Kyiv, Ukraine

Thin films of β-Ga_2O_3 formed from nanocrystalline grains are obtained by the method of radio-frequency (RF) ion-plasma sputtering. Investigations of the surface morphology of thin films by atomic force microscopy (AFM) have shown that the average size of nanocrystalline grains, which form films, increases during thermal treatment. Based on the analysis of the results of the distribution of grain diameters, it is found that, regardless of fulfilment of thermal treatment, for thin films of β-Ga_2O_3, there is a trimodal distribution, which is formed during the deposition of films. The fulfilment of thermal treatment does not change the shape of this distribution and leads to a uniform increase of the grain sizes throughout the entire distribution.

Методою високочастотного (ВЧ) йонно-плазмового розпорошення одержано тонкі плівки β-Ga_2O_3, які формуються з нанокристалічних зерен. Дослідження морфології поверхні тонких плівок методою атомно-силової мікроскопії (АСМ) показали, що при проведенні термооброблення зростає середній розмір нанокристалічних зерен, які формують плівки. На основі аналізи результатів розподілу розмірів діяметрів зерен встановлено, що, незалежно від здійснення термооброблення, в тонких плівках β-Ga_2O_3 спостерігається тримодальний розподіл, який формується у процесі нанесення плівок. Здійснення термооброблення не змінює форму даного розподілу і приводить до рівномірного зростання розмірів зерен вздовж всього розподілу.

Key words: gallium oxide, thin films, nanocrystallites, trimodal distribution.

Ключові слова: оксид ґалію, тонкі плівки, нанокристаліти, тримодальний розподіл.

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1. INTRODUCTION

Among the large number of materials for optoelectronics, luminescent materials used in the creation of displays, scintillators, devices for recording and visualization of information occupy a special place. In this regard, the films based on β-Ga_2O_3 are widely used as thin film materials, which are promising for field-effect transistors (FET) [1], gas sensors [2] and electrodes, transparent in the UV region [3, 4]. Depending on the method of obtaining and the dopant, such thin films are used as photoluminophors [5, 6], cathodoluminophors or electroluminophors [7, 8]. In general, the optical and electrical properties of thin β-Ga_2O_3 films are determined by the methods they were obtained, the regimes of deposition, and subsequent technological methods, which create different degrees of perfection of the obtained samples. Taking into account that the luminescence efficiency, electrical conductivity, and photoelectric properties of thin β-Ga_2O_3 films are largely determined by the dimensional, morphological, and structural properties of the nanoparticles, which form these films, the effect of thermal treatment on the surface morphology of thin films is investigated in this article. Thin films are obtained by radio-frequency (RF) ion-plasma sputtering. The application of this method is considered as optimal for the deposition of semiconductor and dielectric thin films and allows controlling the structure and stoichiometry of the obtained thin films [9]. Among the high-precision methods in determining the size and study of the morphology of nanoparticles, there is atomic force microscopy (AFM), which is used in this work.

2. EXPERIMENTAL TECHNIQUE

Thin films of β-Ga_2O_3 with a thickness of 0.2–0.8 μm were obtained by RF ion-plasma sputtering on substrates of υ-SiO_2 fused quartz. RF sputtering was carried out in an atmosphere of argon in the system using the magnetic field of external solenoids for compression and additional ionization of the plasma column. After deposition of the films, the heat treatment in argon atmosphere at 1000–1100°C was held. X-ray diffraction studies showed the presence of a polycrystalline structure of films with a predominant orientation in the (400), (002), (111), and (512) planes. The diffraction patterns for β-

Ga_2O_3 films were described by us earlier [10] in more details.

The surface morphology of films was investigated using atomic force microscope (AFM) 'Solver P47 PRO'. Processing of experimental data and calculation of surface morphology parameters were carried out, using the Image Analysis 2 software package.

3. RESULTS AND DISCUSSION

Figure 1 shows the microphotographs of the surface of thin β-Ga_2O_3 films, obtained by radio-frequency (RF) ion-plasma sputtering, without thermal treatment and after thermal treatment in an argon atmosphere. The topography of the samples was quantitatively characterized by standard parameters: root mean square roughness, maximum height of grains, mean grains' diameter, which were calculated according to AFM data for sections of the same size (1000×1000 nm). The characteristic parameters of thin β-Ga_2O_3 films without thermal treatment and after thermal treatment in ar-

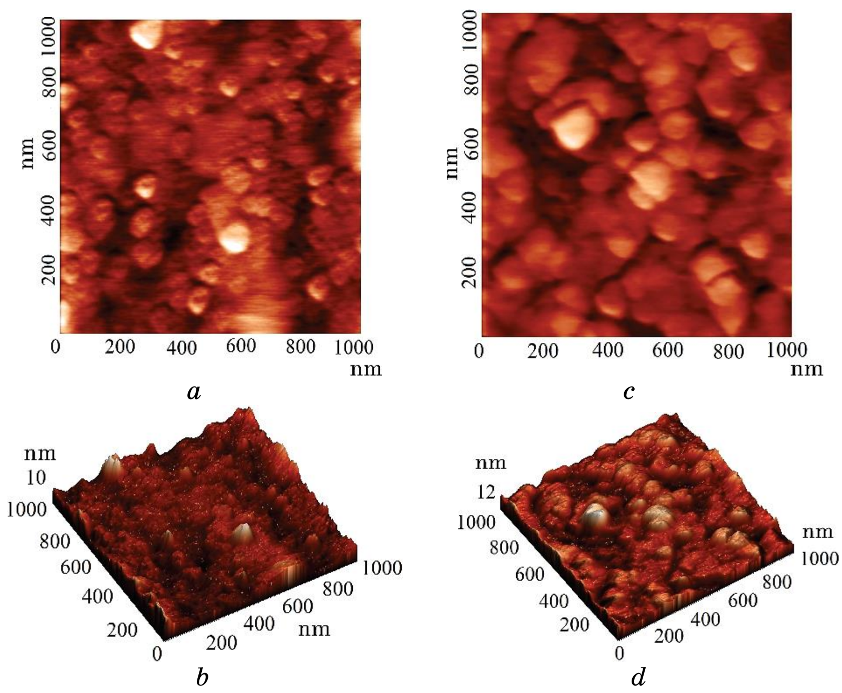

Fig. 1. Images of the surface morphology of thin β-Ga_2O_3 film obtained by RF sputtering: (*a*, *b*) without thermal treatment and (*c*, *d*) after thermal treatment in an argon atmosphere. Images *a* and *c* are two-dimensional; images *b* and *d* are three-dimensional.

gon atmosphere are listed in Table 1. As can be seen from the obtained results, the presence of thermal treatment has a significant effect on the size of the crystal grains and the surface roughness of the films.

The analysis of AFM images (Fig. 1) and parameters of crystalline grains (Table 1) of the surface of thin β-Ga_2O_3 films shows that, after thermal treatment of these thin films, the sizes of nanocrystalline grains, which form these thin films, increase. The increase in the size of the crystal grains and, in particular, the increase in the values of the root mean square roughness and the average diameter of the grains indicate a complication of the surface structure.

A comparison of the histograms of the distribution of grains' heights (Fig. 2) shows that thermal treatment of thin β-Ga_2O_3 films leads to the formation of higher peaks on the film surface and the increase of the interval of scatter of peaks in height.

The increase in the size of crystal grains and the simultaneous

TABLE 1. Parameters of crystallite grains of thin β-Ga_2O_3 films.

Parameter	Without thermal treatment	Thermal treatment in argon atmosphere
Mean grains' diameter, nm	19.6	62.7
Root mean square roughness, nm	8.5	10.6
Max height grains, nm	9.1	12.0
Mean grains' area, nm^2	692	6121
Mean grains' volume, nm^3	6572	70312

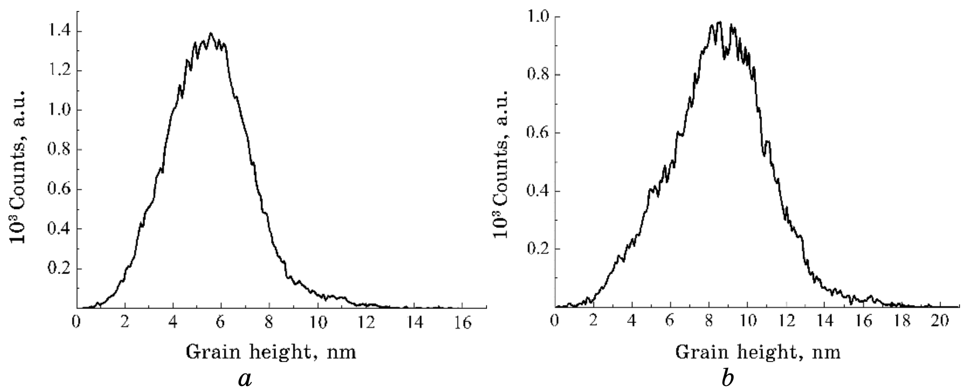

Fig. 2. Grain-height distribution on the AFM image of thin β-Ga_2O_3 films obtained by RF sputtering: (*a*) without heat treatment, and (*b*) after thermal treatment in argon atmosphere.

decrease in the concentration of grains in thin β-Ga_2O_3 films after thermal treatment (Fig. 1) indicate the possibility of the transition of the surface of thin β-Ga_2O_3 films after thermal treatment in a more nanostructured state due to the crystallization of the surface layer.

The characteristic size distributions of grain diameters of thin β-Ga_2O_3 films depending on the thermal treatment are shown in Fig. 3.

In a thorough review [11], the growth of crystal grains and the evolution of crystal structures are analyzed, and it is shown that polycrystalline thin films with a thickness of about 1 μm or less, that is characteristic for our studied β-Ga_2O_3 films, often have 2*D*-like structures. In such structures, the most grains' boundaries are perpendicular to the film surface. Most materials analyzed in [11] form films with nonequilibrium grains, the size of which is smaller than the film thickness, and they form two-dimensional structures only after thermal treatment. Based on numerous results in [11], it was also concluded that the formation of grains in thin films is difficult-to-describe accurately on the basis of model representations or using comparisons with experimental analyzes used for the study of foam or monolayers. In the general, the grain sizes in polycrystalline films are lognormally distributed by size.

In some cases, further grain growth occurs due to 'abnormal' growth or predominant growth of several grains, which usually have specific crystallographic orientation ratios relative to the substrate surface plane. This situation is most likely characteristic of thin β-Ga_2O_3 films, which are obtained by us and characterized by a well-developed structure of the x-ray diffraction spectrum [10].

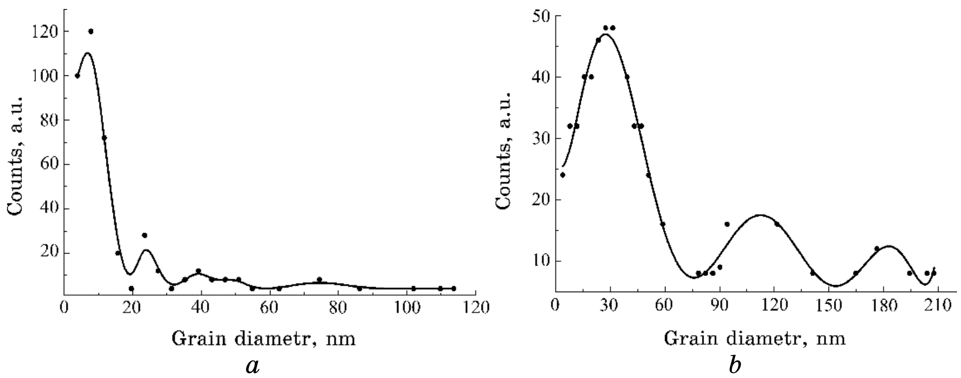

Fig. 3. Distribution of grain-diameter sizes and calculated approximation of the diameter distribution on AFM images of thin β-Ga_2O_3 films: (*a*) without thermal treatment, and (*b*) after thermal treatment in argon atmosphere.

TABLE 2. Maxima on the approximation of the distribution of grain-diameter sizes of thin β-Ga_2O_3 films.

Film of β-Ga_2O_3	d_1, nm	d_2, nm	d_3, nm	d_2/d_1	d_3/d_2
Without thermal treatment (I)	7	24	39	3.43	1.63
Thermal treatment in argon atmosphere (II)	30	108	178	3.60	1.65
d_i(II)/d_i(I)	4.29	4.50	4.56		

When the number of growing grains leads to a 'matrix' of grains outside the static boundaries, a bimodal size distribution of grains develops, that is called the growth of secondary grains [12]. The grains, which grow abnormally, often have a limited or uniform texture. The growth of secondary grains in thin films usually involves evolution in the distribution of grain textures as well as evolution in the distribution of grains by size.

Our results of grain-size diameter distribution for thin β-Ga_2O_3 films (Fig. 3) show that, regardless of the presence of thermal treatment of these films, at least, a trimodal distribution is observed, that is most likely characterized by growth of not only secondary grains but also tertiary ones. Since this situation is typical for freshly applied films without thermal treatment, the growth of secondary and tertiary grains occurs in the process of RF spraying of thin films. The characteristic maxima on the approximation of the distribution of grain-diameter sizes of thin β-Ga_2O_3 films and some relations between them are given in Table 2.

As can be seen from Table 2, the presence of thermal treatment leads to an increase in grain size in the trimodal distribution, but does not change the shape of the distribution. This is confirmed by fairly close values of the ratios of primary, secondary and tertiary maxima in the distribution of grain-diameter sizes in films both without thermal treatment and in the presence of thermal treatment. In addition, the close values of the ratio d_i(II)/d_i(I) indicate that, in the process of thermal treatment, there is an almost uniform increase in the size of the grain diameters of the primary, secondary and tertiary maxima distribution of grain diameters.

4. CONCLUSIONS

It has been established that, during RF ion-plasma sputtering, thin β-Ga_2O_3 films consisting of nanometer grains are formed. According to AFM data, it is shown that the presence of thermal treatment in an argon atmosphere leads to an increase in the average diameters of nanocrystalline grains from 19.6 to 67.2 nm. Based on the analysis of grain-diameter size distribution results, it was found that, regardless of the presence of thermal treatment, the trimodal distribution is observed in thin β-Ga_2O_3 films due to the growth of secondary and tertiary grains, which occur in the process of RF deposition of thin films. The presence of thermal treatment does not change the shape of the established trimodal distribution and leads to a uniform increase in grain diameters of the primary, secondary and tertiary maxima of the distribution of grains' diameters.

REFERENCES

1. K. Matsuzaki, H. Yanagi, T. Kamiya, H. Hiramatsu, K. Nomura, M. Hirano, and H. Hosono, *Appl. Phys. Lett.*, **88**, No. 9: 092106 (2006).
2. N. D. Cuong, Y. W. Park, and S. G. Yoon, *Sensors and Actuators B: Chemical*, **140**, No. 1: 240 (2009).
3. M. Orita, H. Ohta, M. Hirano, and H. Hosono, *Appl. Phys. Lett.*, **77**, No. 6: 4166 (2000).
4. O. M. Bordun, B. O. Bordun, I. Yo. Kukharskyy, I. I. Medvid, I. S. Zvizlo, and D. S. Leonov, *Nanosistemi, Nanomateriali, Nanotehnologii*, **17**, No. 3: 483 (2019); https://doi.org/10.15407/nnn.17.03.483
5. J.-G. Zhao, Zh.-X. Zhang, Z.-W. Ma, H.-G. Duan, X.-S. Guo, and E.-Q. Xie, *Chinese Phys. Lett.*, **25**, No. 10: 3787 (2008).
6. O. M. Bordun, B. O. Bordun, I. Yo. Kukharskyy, and I. I. Medvid, *J. Appl. Specrosc.*, **84**, No. 1: 46 (2017).
7. P. Wellenius, A. Suresh, J. V. Foreman, H. O. Everitt, and J. F. Muth, *Mater. Sci. Eng. B*, **146**, Nos. 1–3: 252 (2008).
8. T. Minami, T. Shirai, T. Nakatani, and T. Miyata, *Jpn. J. Appl. Phys.*, **39**, No. 6A: L524 (2000).
9. E. V. Berlin and L. A. Seydman, *Ionno-Plazmennyie Protsessy v Tonkoplenochnoy Tekhnologii* [Ion-Plasma Processes in Thin-Film Technology] (Moscow: Tekhnosfera: 2010) (in Russian).
10. O. M. Bordun, I. Yo. Kukharskyy, B. O. Bordun, and V. B. Lushchanets, *J. Appl. Spectrosc.*, **81**, No. 5: 771 (2014).
11. C. V. Thompson, *Sol. State Phys.*, **55**: 270 (2001).
12. C. V. Thompson, *J. Appl. Phys.*, **58**: 763 (1985).