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Influence of the aluminum content on the luminescent and electronic properties of β -Ga₂O₃

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Abstract. In this work, we investigated bulk β -Ga₂O₃ samples grown by the Czochralski method on Al₂O₃ and Ga₂O₃ seeds. The elemental composition of the samples and its effect on the luminescent and electrophysical properties of the samples were determined. The Kelvin probe microscopy was used to study the processes of localization and dissipation of charges in the samples. It was shown that in a β -Ga₂O₃ sample grown on an Al₂O₃ seed, the characteristic charge dissipation time is 10 times longer.

1. Introduction

Gallium oxide is a wide-gap semiconductor with intense intrinsic luminescence and, therefore, is a promising material for optoelectronics. The band gap of gallium oxide is 4.8–4.9 eV at room temperature (depending on the composition and structural modification of the crystal). Particular interest in this material is associated with the possibility of obtaining bulk gallium oxide by the methods of Czochralski and Stepanov [1]. In this regard, quite a few papers have recently been published devoted to the study of the luminescent and electrical properties of bulk crystals, films, and nanostructures of gallium oxide [2–4]. Researches are carried out for both pure and alloyed materials [2].

In this work, we study bulk β -Ga₂O₃ samples grown by the Czochralski method on Al₂O₃ and Ga₂O₃ seeds. The main goal of this work is to determine the effect of aluminum impurity on the luminescent and electrophysical properties of the samples.

2. Growth method

The studied β -Ga₂O₃ single crystals were obtained by pulling from the melt according to the Czochralski method in a Nika-3 growth setup with induction heating (manufactured by FGUP EZAN, Chernogolovka). Powdered Ga₂O₃ was used as the initial material, the reagent purity was 99.99% (4N), and the crucible was made of Ir. The pulling of crystals was carried out in an atmosphere of carbon dioxide CO₂ at a pressure of about 1 Bar on various seeds: sapphire (Al₂O₃) seed and β -Ga₂O₃ seed. The details of the technological process were described earlier elsewhere [5, 6]. The grown crystals had a cylindrical shape with a diameter of ~ 20 mm and a length of ~ 15 mm. Samples for investigation were obtained from crystals by cutting and cleaving along sliding planes. The samples grown on various seeds were cleaved along surface (100).



3. Research methods

The composition of samples was determined by X-ray spectral microanalysis using an electron probe microanalyzer CAMEBAX (Cameca). Cathodoluminescence (CL) studies were carried out on the same setup using optical spectrometer for studying cathodoluminescence spectra [7], with the possibility of obtaining cathodoluminescence images and measuring the dynamics of the absorbed current [8]. This approach made it possible to determine whether the inhomogeneities of cathodoluminescence are related to the inhomogeneity of the impurity distribution. Cathodoluminescence images were obtained with an electron beam diameter of 200 μm , an electron beam energy of 10 keV, and a current of 30 nA. The cathodoluminescence spectra and the dynamics of the absorbed current were recorded at the following parameters of the electron beam: electron beam energy 5 keV, electron beam current 5 nA, and diameter 10 μm .

The topography of the surface of the samples under study, as well as the charge localization and dissipation processes were studied using an atomic force microscope (AFM) Ntegra-Aura (NT-MDT, Zelenograd, Moscow). Investigations of the electrophysical properties of the samples were carried out by the Kelvin probe microscopy (KPM) method. In this work, charges were injected into the samples along the line. When the size of the studied area was $2.5 \times 2.5 \mu\text{m}$, the line length was 5 μm , and the charging time was ~ 10 seconds. The choice of this method of charges injection is associated with rather short characteristic times of charges dissipation. After the creation of the charge lines, the surface potential maps were recorded. The analysis of the profiles of the obtained distributions was used to compare the samples under study.

4. Results and discussion

4.1. Determination of aluminum content in samples

It was shown that the aluminum content in the sample grown on a sapphire seed was about 0.01 wt%. In the sample grown on a $\beta\text{-Ga}_2\text{O}_3$ seed, the aluminum content is less than 0.001 wt%.

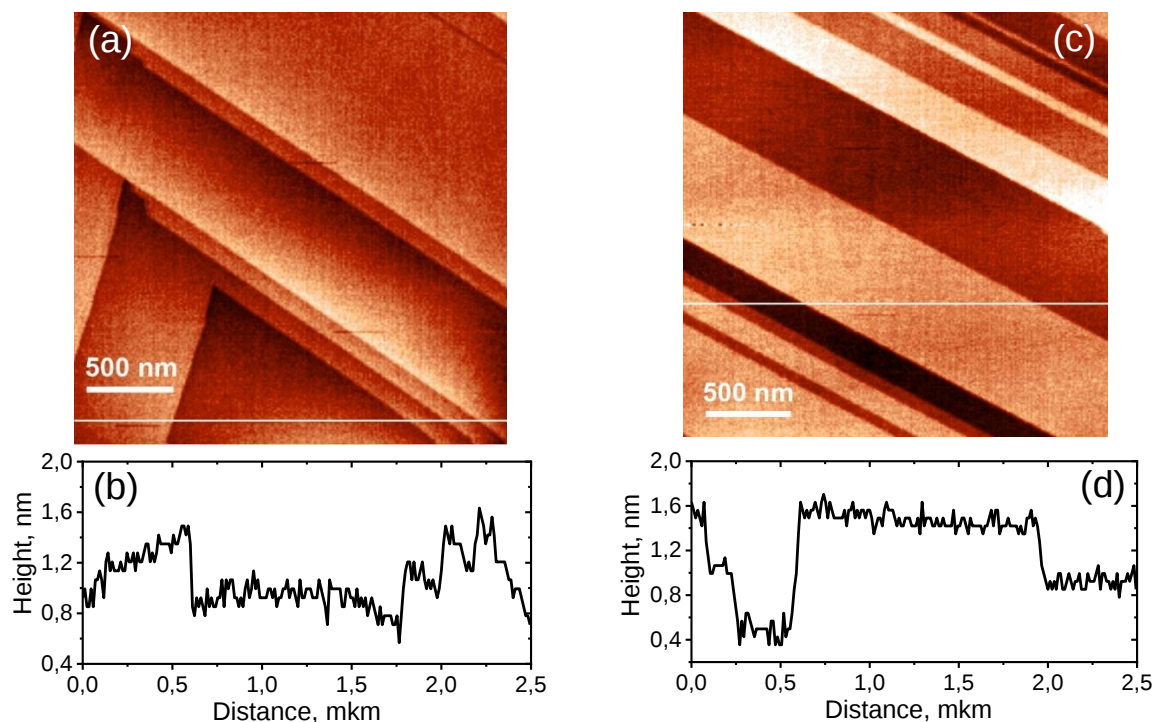


Figure 1. Surface topography and the corresponding height profile along white horizontal lines for (100) surfaces of $\beta\text{-Ga}_2\text{O}_3$ samples grown: (a, b) - on a $\beta\text{-Ga}_2\text{O}_3$ seed, (c, d) - on an Al_2O_3 seed.

4.2. β -Ga₂O₃ surface (100) topography and cathodoluminescence features

The topography of the (100) surface was investigated by the AFM method (Figure 1). AFM studies have shown that steps of various heights are present on the sample surface. The sample grown on a β -Ga₂O₃ seed exhibits steps along the cleavage planes described in [9]. The height of the steps is about 4 Å. The observed differences in topography between samples grown on natural and Al₂O₃ seeds require further investigation.

CL images of β -Ga₂O₃ samples were obtained from the (100) surface (Figure 2). Contrast stripes are visible on the CL images; the distance between the stripes is different in different regions of the sample. Most likely, these stripes are associated with the crystallization plane. The direction of stripes coincides with the directions of the steps observed by the AFM method. It was shown earlier [10] that a change in the cathodoluminescence intensity is observed in these stripes, that is, a change in the content of point defects is observed. It was shown by XRD that the inhomogeneity of the luminescence is not associated with the inhomogeneity of the distribution of aluminum. Interestingly, the CL images show steps in only one, namely (001), crystallographic direction.

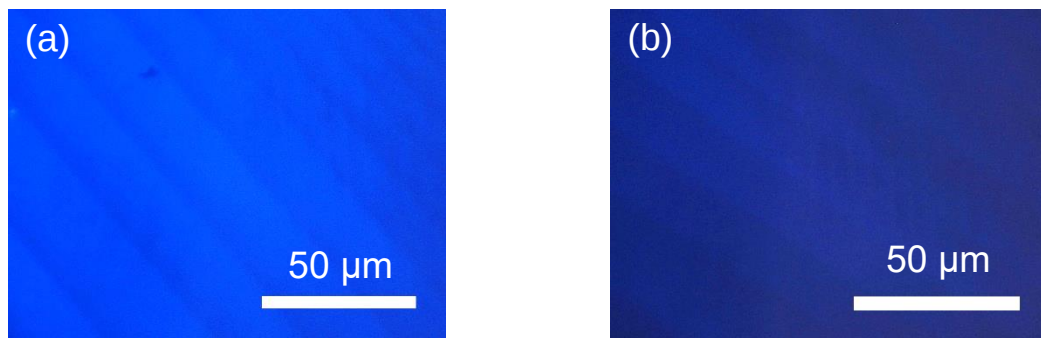


Figure 2. CL images of the (100) surface of β -Ga₂O₃ samples at a temperature of 300 K, grown: (a) – on a β -Ga₂O₃ seed, (b) – on an Al₂O₃ seed

Figure 3 shows the CL spectra of the samples grown on various seeds. The spectra were approximated by the sum of two Gaussian bands with maxima at 2.8 eV and 3.2 eV. It can be seen that, in the sample grown on a native seed, the CL intensity is approximately 40% higher, and the intensity of both CL bands increases. The position and half-width of the CL bands are the same for all studied samples. Accordingly, it can be argued that the aluminum impurity does not lead to the appearance of additional bands or to a change in the half-width and position of the bands, as compared to pure Ga₂O₃.

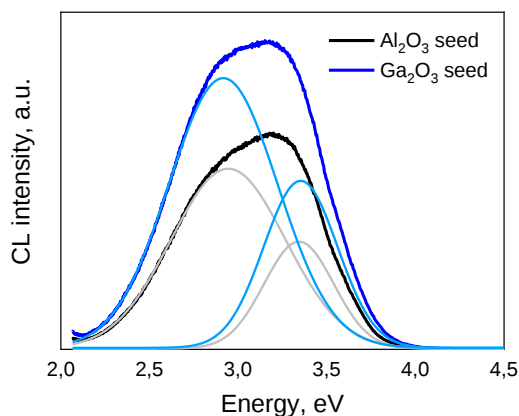


Figure 3. CL spectra obtained at a temperature of 300 K from the surface (100). Blue spectrum – sample grown on β -Ga₂O₃ seed, black spectrum – on Al₂O₃ seed. Thin lines represent an approximation of the spectra by the sum of Gaussian bands.

4.3. Study of the electrophysical properties of samples

It was shown in [10] that the process of electron localization prevails in bulk β -Ga₂O₃ at room temperature. A study of the processes of localization and dissipation of a negative charge was carried out by the method of KPM according to the technique proposed in [8]. The surface of the samples was charged in the contact mode along the line (white dashed line in figure 4a, figure 4b), then the change in the surface potential was recorded. It was found that the charge states of the traps change in all samples. Each horizontal scan line characterizes the distribution of the potential and, accordingly, the localized charge over the surface at a particular moment in time. In figures 4c, 4d the potential distributions after 10 seconds after the end of charging are shown. In figures 4e and 4f the changes in the amplitude of the potential versus the time elapsed after charging the surface are shown; such dynamics makes it possible to compare the processes of charge dissipation in the samples under study.

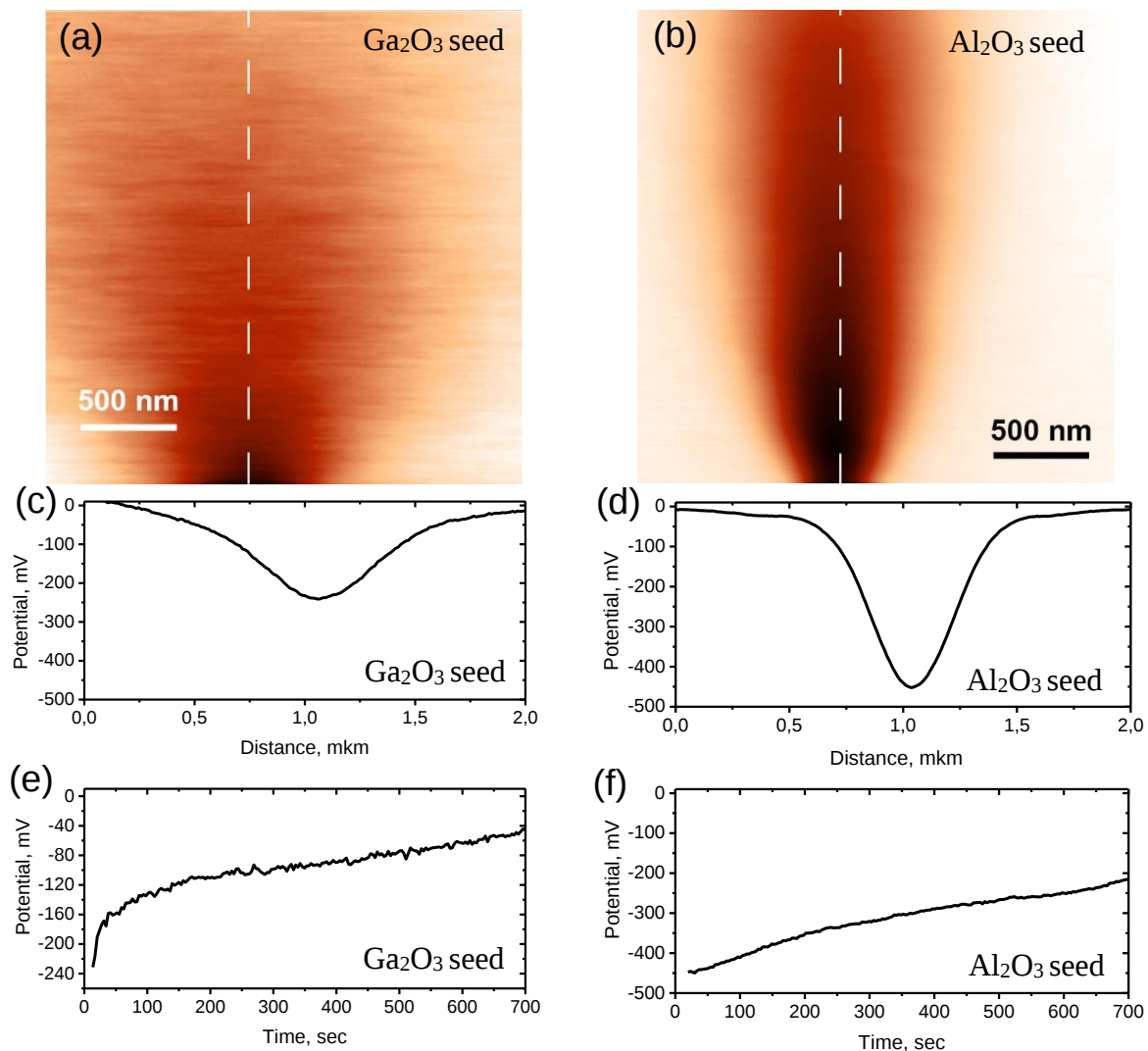


Figure 4. (a, b) Image of the change in the surface potential distribution in time after the injection of charges from the AFM tip into bulk β -Ga₂O₃ along the vertical line; (c, d) - potential distribution profile 10 seconds after the end of charging, (e, f) - dependence of the potential amplitude on the time elapsed after the end of charging. The figures show the seeds on which the samples under study were grown.

It can be seen that in the sample grown on a β -Ga₂O₃ seed, the potential amplitude is significantly lower and the charge dissipation proceeds much faster than in the sample grown on an Al₂O₃ seed. Since the amplitude is measured 10 seconds after the end of charging, it is possible that its significant drop occurs already during this time, and the values of the initial amplitude and, accordingly, the injected charge cannot be compared on different samples. Nevertheless, it was determined that the charge dissipation time in a sample grown on an Al₂O₃ seed is 10 times larger than in a sample grown on a Ga₂O₃ seed. It can be assumed that aluminum in β -Ga₂O₃ contributes to the formation of deeper traps.

5. Conclusions

We studied β -Ga₂O₃ samples grown by the Czochralski method on Al₂O₃ and Ga₂O₃ seeds. The aluminum content in the samples was determined. It is shown that an aluminum content of the order of 0.01 wt% leads to a decrease in the luminescence intensity in comparison with a pure material, but does not affect the shape of the spectrum. It was also shown that in a sample with an aluminum content of about 0.01 wt%, the charge dissipation process proceeds more slowly.

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