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On a successful experience of homoepitaxy of β-Ga_2O_3 layers on native substrates

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The paper proposes a method for producing substrates from bulk crystals of the β-Ga_2O_3 gallium oxide by cleaving. With an example of growing the β-Ga_2O_3 and β-$(Al_xGa_{1-x})_2O_3$ layers on the fabricated substrates by epitaxy from metal-organic compounds, the possibility of using these substrates for homoepitaxy is shown. An analysis of the surface morphology and structural quality of the obtained layers has been carried out.

Keyword: gallium oxide, substrates, metal-organic vapor phase epitaxy, homoepitaxy.

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Unique properties of gallium oxide (primarily, of the most stable β-phase), such as a wide band gap (4.5 to 5.4 eV as per various estimates [1]) and high breakdown fields (up to 8 MV/cm as per calculations [2], 5.3 MV/cm in the device [1]), make this material promising for manufacturing highvoltage devices and ultraviolet sensors. The gallium oxide β-phase possesses also other advantages: controllable doping with *n*-type impurities, relatively high electron mobility (up to 196 $cm^2/(V \cdot s)$ [3]), radiation resistance [4].

To produce high-quality semiconductor devices, especially power-electronics devices, epitaxial layers with a minimum number of defects are necessary. This largely depends on the choice of the substrate material. For epitaxial growth of different Ga_2O_3 polytypes, different substrate materials are used. For instance, α-Ga_2O_3 and κ-Ga_2O_3 are often grown on artificial sapphire substrates [5,6]. There exist reports on successful growth of α-Ga_2O_3 layers on the diamond surface [7]. However, the most efficient way to obtain low-defect layers is to use homoepitaxy (namely, growing structures on „native“ substrates). Here another advantage of gallium oxide manifests itself, i. e. the possibility of fabricating bulk crystals by relatively cheap methods of growing from a melt (first of all, the Czochralski or Stepanov method).

This work presents the results of developing a method for growing low-defect bulk β-Ga_2O_3 crystals, preparing substrates from the obtained crystals, and also growing β-Ga_2O_3 and β-$(Al_xGa_{1-x})_2O_3$) layers on the prepared substrates by using metal-organic vapor phase epitaxy (MOVPE). The main goal of the layer growth experiments was to demonstrate the fundamental possibility of using the substrates for subsequent homoepitaxy.

Bulk β-Ga_2O_3 crystals were grown using the NIKA-3 machine (FSUE EZAN, Russia). The melt was obtained using an iridium crucible 26 mm in height and 40 mm in diameter which was placed in the zirconium-dioxide thermal zone. The crucible was heated by induction. To grow bulk β-Ga_2O_3 crystals, fragments of previously grown β-Ga_2O_3 crystals were used as seeds. The initial raw material for the melt was Ga_2O_3 powder 99.999% pure. The crystal pulling speed was about 0.15 mm/min.

The conventional approach to fabricating substrates from bulk crystalline material implies cutting, chemical and mechanical polishing, and cleaning. However, in the case of the β-Ga_2O_3 single crystal, another method for preparing substrates is available, which employs the cleavage nature of the β-Ga_2O_3 crystals: thin plates easily get peeled from the bulk crystal along the cleavage plane (100), and the surface of the resulting plates (substrates) appears to be quite smooth and does not require additional processing. In this way, substrates were obtained in the form of plane-parallel plates about 10×15 mm in size (about 1 mm in thickness).

Epitaxial growth of the β-Ga_2O_3 and β-$(Al_xGa_{1-x})_2O_3$ layers on the obtained substrates was performed using an Epiquip VP-50 MOVPE growing machine upgraded for growing oxides. The machine was equipped with a horizontal reactor with induction heating. The growth was performed at the temperature of 750°C and pressure of 100 mbar. The carrier gas (nitrogen) flow was 4.5 slm (liter per minute under standard conditions), the oxygen flow was 1 slm, the trimethylgallium and trimethylaluminum flows were 21 and 6 μmol/min, respectively. The growth rate was about 750 nm/h.

Morphology of the layer surfaces was studied by atomic force microscopy (AFM) with an NTEGRA Aura microscope (NT-MDT, Russia). Images of cleavages, as well as layer thicknesses, were obtained using a scanning electron microscope (Tescan MIRA-3, Czech Republic). The elemental composition was determined by energy-

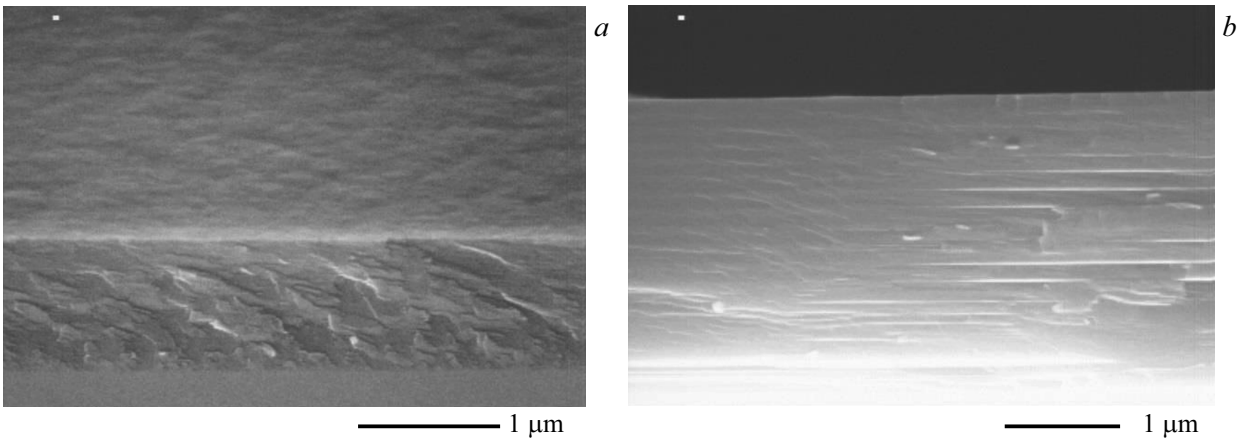

Figure 1. Images of layers on the structure cleavages obtained with a scanning electron microscope. *a* — sample 1, the β-Ga_2O_3 layer about 0.9 μm thick; *b* — sample 3, a system consisting of 11pairs of β-Ga_2O_3/β-$(Al_xGa_{1-x})_2O_3$ layers (each pair about 0.16 μm thick), and the lower β-Ga_2O_3 layer about 0.9 μm thick, which is adjacent to the substrate.

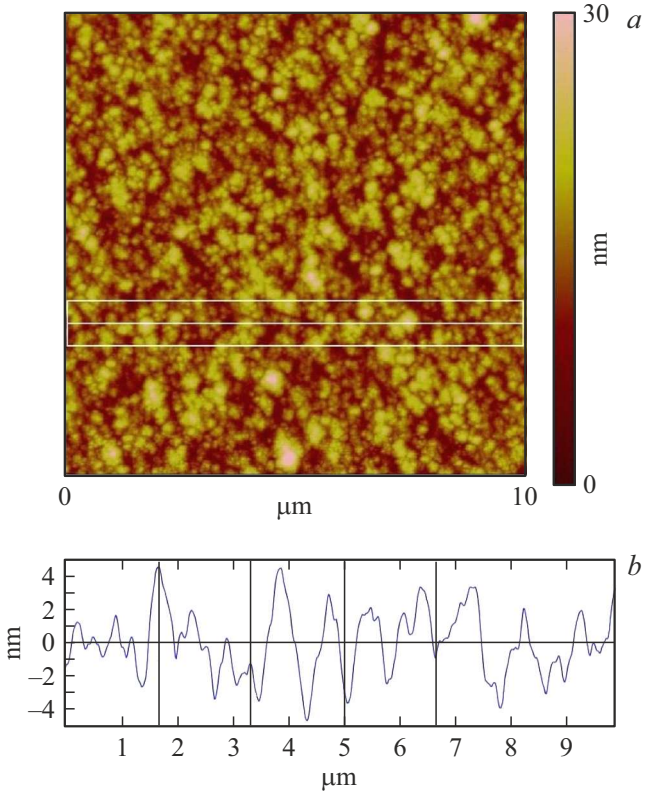

Figure 2. AFM image of surface morphology of the β-Ga_2O_3 epitaxial layer. *a* — 2D surface relief map; *b* — surface relief profile.

dispersive X-ray spectroscopy (EDX). Analysis of the phase composition and crystalline quality of the layers was performed using a DRON-8 X-ray diffractometer (NPO „Burevestnik“, Russia).

As shown in [10], the height of steps on the surface of the substrate fabricated by the described method was about 3–4 nm. This is comparable to the roughness of commercially produced substrates and is satisfactory for epitaxial growth. On the obtained substrates, there were grown an β-Ga_2O_3 layer (sample 1), β-$(Al_xGa_{1-x})_2O_3$ layer (sample 2), and also a system of 11 pairs of alternating layers β-Ga_2O_3/β-$(Al_xGa_{1-x})_2O_3$ grown on a thicker β-Ga_2O_3 layer (sample 3). The technique of using such a system of alternating layers is characteristic of epitaxial technologies and is applied when it is necessary to ensure a defect-free transition between layers of significantly different compositions. According to EDX data, the aluminum content in sample 2 was about 4 at.%, that in sample 3 was about 2 at.%. The low content of aluminum was caused by the desire to avoid growing of highly-stressed layers. Fig. 1 presents the images of layers on the structure cleavages.

Surface morphology of the β-Ga_2O_3 layer (sample 1) is shown in Fig. 2. One can see that the layer has an island-type surface characteristic of layers grown on non-disoriented substrates. The surface of gallium oxide layers obtained in [11] for the substrate disorientation angles less than 0.1° has a similar appearance.

The cleavage planes in β-Ga_2O_3 significantly affect the steps formation. The adatom-substrate binding energy on terraces of (100) planes is less than binding energy of adatoms to vertical walls of the steps; this is expected to ensure the predominance of stepwise growth. For example, when direction of the step vertical wall coincides with the [001] or $[00\bar{1}]$ direction, weak bonds on the (001) planes promote formation of stepwise walls consisting of (001) cleavage planes. If the rate of adsorption on the step wall and that of adatom diffusion are approximately the same along the terraces, relatively straight steps may be formed.

Another important factor determining the mode of epitaxial film growth is the growth temperature that governs the surface diffusion. As shown in [12], surface morphology of the β-Ga_2O_3 epitaxial layer improves as the growth temperature increases from 800 to 1000°C despite the

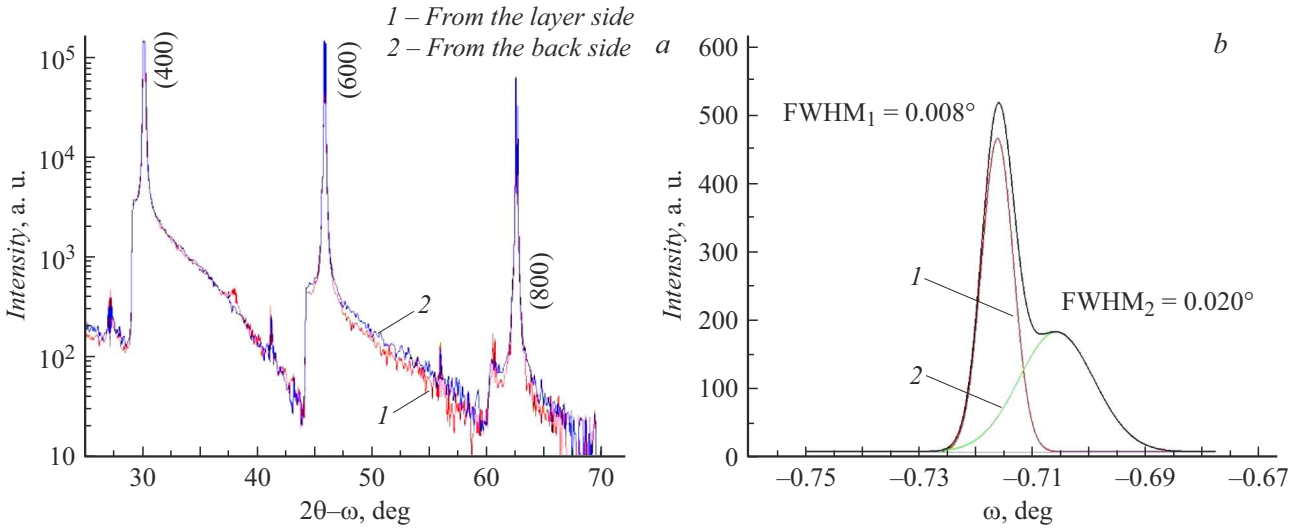

Figure 3. Results of X-ray diffraction analysis of sample 3. *a* — spectra of $2\theta-\omega$-scanning from the layer side (*1*) and from the substrate side (*2*); *b* — rocking curve for a system of 11 pairs of epitaxial layers β-Ga_2O_3/β-$(Al_xGa_{1-x})_2O_3$.

experiments show that the growth rate decreases in this case [13].

In our case, the growth surface remains parallel to the (100) plane. And, despite the (100) plane generally promotes the layer-by-layer growth (the so-called step-flow growth), the absence of substrate disorientation (typically by $0.2-0.3^\circ$ with respect to the growth surface normal) makes the density of seed steps on the initial substrate low, due to which the substrate surface consists mainly of flat step-free areas. In addition, the chosen growth temperature (750°C) is probably insufficient to ensure extensive surface diffusion. Due to these factors, the layer growth in our experiments was not of the layer-by-layer character but of the island-like one.

Fig. 3 presents the X-ray diffraction $2\theta-\omega$-spectra and rocking curve for sample 3 which is the most interesting structure. Fig. 3, *a* clearly demonstrates that the grown layers contain only the β-phase of gallium oxide, which is desired for homoepitaxy. The aluminum content in the epitaxial layers (2 at.%) is too small to noticeably deform the crystal lattice and insufficient to form the α- phase. The sample 3 rocking curve (Fig. 3, *b*) splits into two clearly distinguishable individual peaks. The more intense and narrow peak (curve *1*) is a reflection from the substrate and coincides with the rocking curve measured from the sample back side, i.e. from the substrate side (not shown in the figure). This confirms the previously studied [8] high crystalline quality of the used substrates. The wider, less intense and somewhat asymmetrical peak (curve *2*) relates probably to the system of layers. This indicates that the system of layers is not monocrystalline but consists of slightly disoriented mosaic blocks.

Thus, the work describes a method for preparing substrates from bulk β-Ga_2O_3 crystals by cleaving along cleavage plane (100) and demonstrates the fundamental possibility of growing epitaxial layers on the surfaces of such substrates without additional surface treatment. To improve the quality of grown layers, it is necessary to optimize the growth process, primarily in terms of growth temperature and ratio of the precursor flows. To the authors' knowledge, this is the first case of successful epitaxy on gallium oxide substrates manufactured in Russia.

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Conflict of interests

The authors declare that they have no conflict of interests.

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