

# A Study of the Formation Modes of Nanosized Oxide Structures of Gallium Arsenide by Local Anodic Oxidation

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**Abstract**—Results obtained in a study of the influence exerted by the technological modes of local anodic oxidation (LAO) on the process in which nanosized oxide structures (NOSs) are formed on the surface of gallium arsenide are reported. The effect of the amplitude and duration of voltage pulses applied to the probe–substrate system, relative air humidity in the technological chamber, and amplitude of cantilever oscillations on the geometric parameters of gallium arsenide NOSs has been examined. It was found that raising the relative humidity from 60 to 90% results in a decrease in the threshold LAO voltage from 7.0 to 6.0 V. It is shown that raising the oscillation amplitude from 0.1 to 2.8 nm leads to a decrease in the NOS height from  $3.20 \pm 0.34$  to  $1.10 \pm 0.13$  nm and in the NOS diameter from  $218.4 \pm 29.5$  to  $78.1 \pm 10.3$  nm.

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## 1. INTRODUCTION

Local anodic oxidation (LAO) using a scanning probe microscope (SPM) is a promising method of nanolithography, which has a number of advantages over optical lithography and makes it possible to obtain nanosized structures on the surface of a wide variety of materials [1–4].

Nanosized oxide structures (NOSs) are formed on the substrate surface by the LAO method upon the application of voltage pulses to the probe–substrate system. In this case, an electrochemical reaction accompanied by the charge-and-mass transfer of ionized substrate atoms and hydroxyl groups occurs in the adsorbate meniscus between the substrate and the probe to yield oxides and hydrogen [3].

Gallium arsenide GaAs and solid solutions based on this compound (AlGaAs, InGaAs, etc.) are promising materials for nanoelectronics, widely used to manufacture integrated circuits operating at frequencies higher than 10 GHz and low-noise high-power microwave devices. The wide range in which the parameters of the solution can be varied because of the use of an extended set of initial materials, direct-band structure, high radiation hardness, and high carrier mobility can be attributed as the main advantages of compounds based on gallium arsenide [2]. In contrast/ whereas, low heat conductivity, incompatibility with silicon micromachining technology, and the high cost of gallium arsenide substrates are disadvantages of compounds based on gallium arsenide [2].

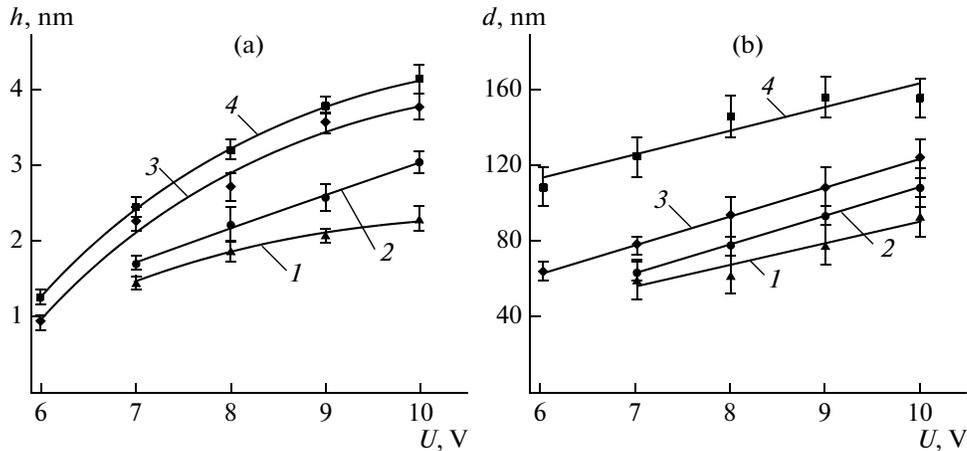
The goal of our study was to examine how the modes of local anodic oxidation of gallium arsenide epitaxial structures (amplitude and width of voltage pulses applied to the probe–substrate system, cantile-

ver oscillation amplitude, and relative humidity within the technological chamber) affect the specific features of the formation of nanosized oxide structures of gallium arsenide.

## 2. EXPERIMENTAL PROCEDURE

The experimental gallium arsenide structures were fabricated by molecular-beam epitaxy in the modular system of a growth cluster in the NANOFAB NTK-9 nanotechnological complex (NT-MDT Private Company, Zelenograd) [5]. The structures were grown on “epi-ready” semi-insulating GaAs (100) wafers. After the passivating oxide layer was removed, a 0.8- $\mu\text{m}$ -thick buffer layer of undoped GaAs was formed on the wafer surface. Then, a 2- $\mu\text{m}$ -thick layer of silicon-doped GaAs was grown. According to the results of measurements of the Hall carrier mobility using an Ecopis HMS-3000 installation, the background impurity concentration in the buffer layer was  $7 \times 10^{12} \text{ cm}^{-3}$ , and the impurity concentration in the silicon-doped layer was  $5 \times 10^{17} \text{ cm}^{-3}$ . The average surface roughness of the structures was determined by atomic-force microscopy (AFM) to be  $0.76 \pm 0.15$  nm.

The formation modes of nanosized oxide structures of gallium arsenide were studied by the LAO method, using a Solver P47 Pro SPM with DSP-20 cantilevers that have a conducting carbon coating [5]. Nanolithography was performed in the tapping AFM mode with a Nova 1.0.26.1511 applied-software package. The relative humidity within the technological chamber was monitored with an Oregon Scientific ETHG913R digital humidity meter.



**Fig. 1.** (a) Height and (b) diameter of gallium arsenide NOSs vs. the amplitude of the applied voltage at relative humidities of (1)  $60 \pm 1$ , (2)  $70 \pm 1$ , (3)  $80 \pm 1$ , and (4)  $90 \pm 1\%$ .

The effect of the amplitude of the applied-voltage pulse on the geometric parameters of the nanosized oxide structures formed by the LAO method was studied at relative humidities within the SPM technological chamber of  $60 \pm 1$ ,  $70 \pm 1$ ,  $80 \pm 1$ , and  $90 \pm 1\%$ . Upon the application of 1000-ms voltage pulses with amplitudes of 5.0 to 10.0 V, arrays of nanosized oxide structures were formed on the gallium arsenide surface at a feedback current of 0.1 nA (*SetPoint* parameter) in the SPM control system.

To study the influence exerted by the width of the applied-voltage pulse on the geometric parameters of the nanosized oxide structures, we performed local anodic oxidation of the gallium arsenide surface by applying voltage pulses with an amplitude of 10.0 V and widths in the range from 10 to 500 ms at a *SetPoint* parameter of 0.1 nA and relative humidity of  $90 \pm 1\%$ .

When studying the effect of the cantilever-oscillation amplitude on the geometric parameters of NOSs, we performed probe nanolithography on the gallium arsenide surface by applying voltage pulses with an amplitude of 10.0 V and width of 100 ms. At a constant relative humidity of  $80 \pm 1\%$ , NOS arrays were formed in the tapping AFM mode with DSP-20 cantilevers and the *SetPoint* parameter varied within the range from 0.01 to 0.3 nA. Then, upon performing AFM spectroscopy, we obtained the dependences of the *Mag* signal on the *SetPoint* parameter, which makes it possible to determine how the cantilever-oscillation amplitude depends on the *SetPoint* parameter for a prescribed value of the feedback current in the SPM control system.

We examined the electrical properties of NOSs on the gallium arsenide surface by means of nanolithography of a square-shaped test structure in the scanning mode with a special template (graphics file of \*.bmp format). Nanolithography was performed in

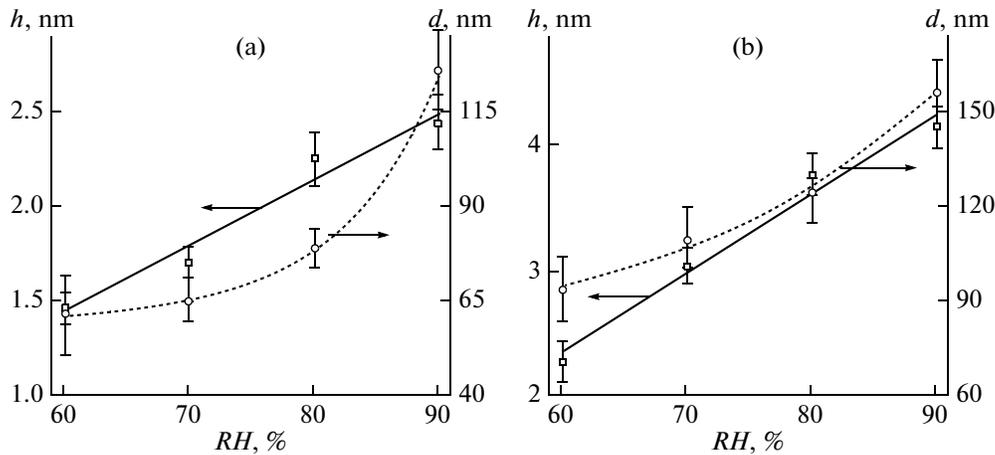
the tapping AFM mode with 10.0-V voltage pulses at a sweep rate of  $0.6 \mu\text{m/s}$ , a relative humidity of  $80 \pm 1\%$ , and a *SetPoint* parameter equal to 0.1 nA. Further, the test structure of gallium arsenide NOSs was scanned in the spreading-resistance-imaging mode by contact AFM upon the application of a voltage of 4.0 V. The current–voltage characteristics on the NOS and gallium arsenide surface were obtained.

We found the geometric parameters of the NOSs (height and diameter) by statistical processing of the resulting AFM images of arrays constituted by 49 gallium arsenide NOSs using the Image Analysis 3.5 software package by our measurement procedure certified in conformity with GOST (State Standard) R8.563–96 [6, 7]. The use of arrays with a large number of NOSs improves the reliability of statistical data processing.

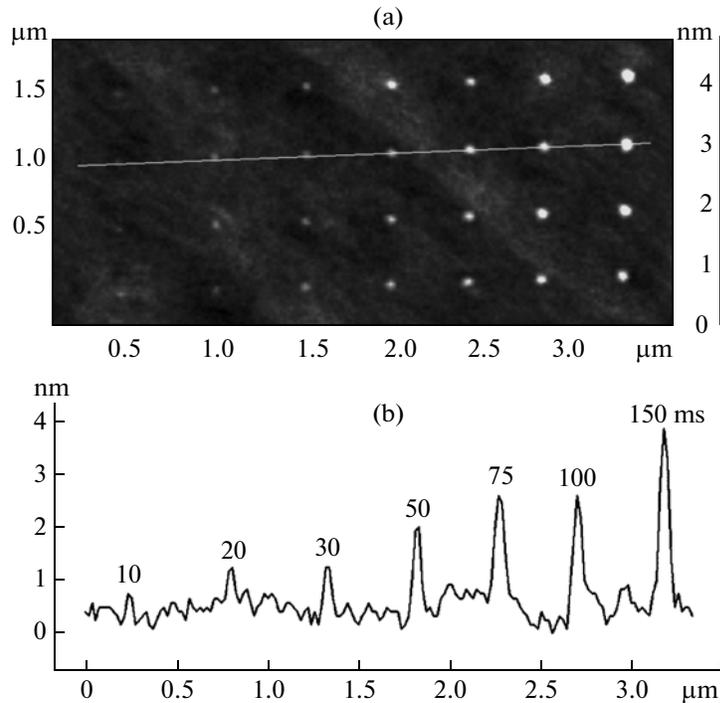
### 3. RESULTS AND DISCUSSION

Using the statistical data obtained, we constructed the dependence of the average values of the NOS height  $h$  and diameter  $d$  on the amplitude of the applied-voltage pulse at various relative humidities (Fig. 1).

An analysis of these dependences demonstrated that raising the voltage-pulse amplitude from 6.0 to 10.0 V at a relative humidity of 90% results in an increase in the NOS height from  $1.24 \pm 0.11$  to  $4.13 \pm 0.20$  nm, and in the NOS diameter, from  $109.8 \pm 9.8$  to  $156.9 \pm 10.5$  nm. No nanosized oxide structures were observed on performing LAO at a voltage amplitude of 5.0 V. This effect of the voltage amplitude between the probe and substrate can be attributed to two main factors. First, an increase in the strength of the electric field increases the number of active species (oxygen ions and hydroxyl groups) formed upon the decomposition of water molecules in the electric field. Second, the strength of the electric field directly



**Fig. 2.** Height and diameter of gallium arsenide NOSs vs. the relative humidity at voltages of (a) 7 and (b) 10 V.



**Fig. 3.** (a) AFM image and (b) profilogram of gallium arsenide NOSs obtained at various voltage-pulse widths.

affects the diffusion flux of these species into the LAO zone, which leads to an increase in the NOS growth rate.

In addition, an analysis of Fig. 1 demonstrated that, on raising the relative humidity from  $60 \pm 1$  to  $90 \pm 1\%$ , the voltage amplitude at which the LAO process is observed decreases from 7.0 to 6.0 V and both the height and diameter of the gallium arsenide NOS being formed grow. This effect may be due to an increase in the number of water molecules in the probe–substrate system. At low relative humidity, the number of adsorbed water molecules is insufficient for providing the substrate-surface-oxidation reaction.

Therefore, the voltage at which the oxidation process is observed will decrease with increasing relative humidity within the SPM technological chamber.

Using the statistical data obtained, we constructed the dependence of the height and diameter of gallium arsenide NOSs on the relative humidity  $RH$  for NOSs formed at voltage amplitudes of 7.0 and 10.0 V (Fig. 2). The larger geometric parameters of the NOSs formed at a higher relative humidity can be attributed to an increase in the diameter of the water meniscus formed.

On processing the AFM images partly shown in Fig. 3, we constructed dependences of the average

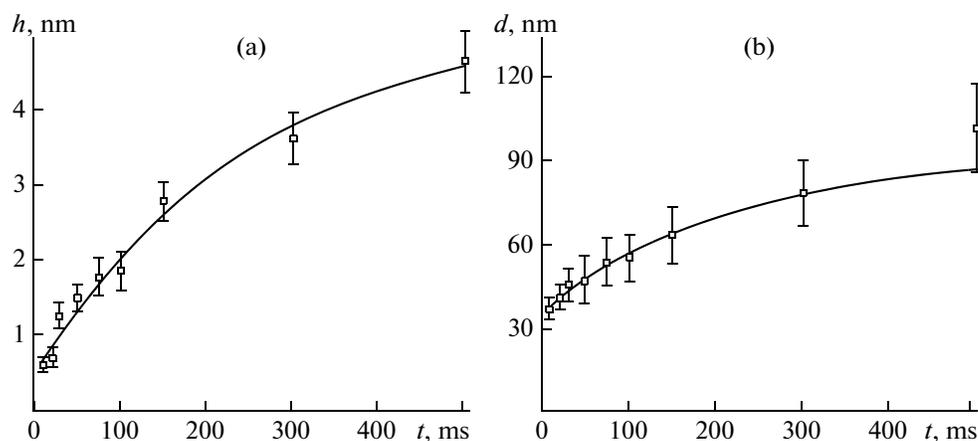


Fig. 4. (a) Height and (b) diameter of gallium arsenide NOSs vs. the voltage-pulse width.

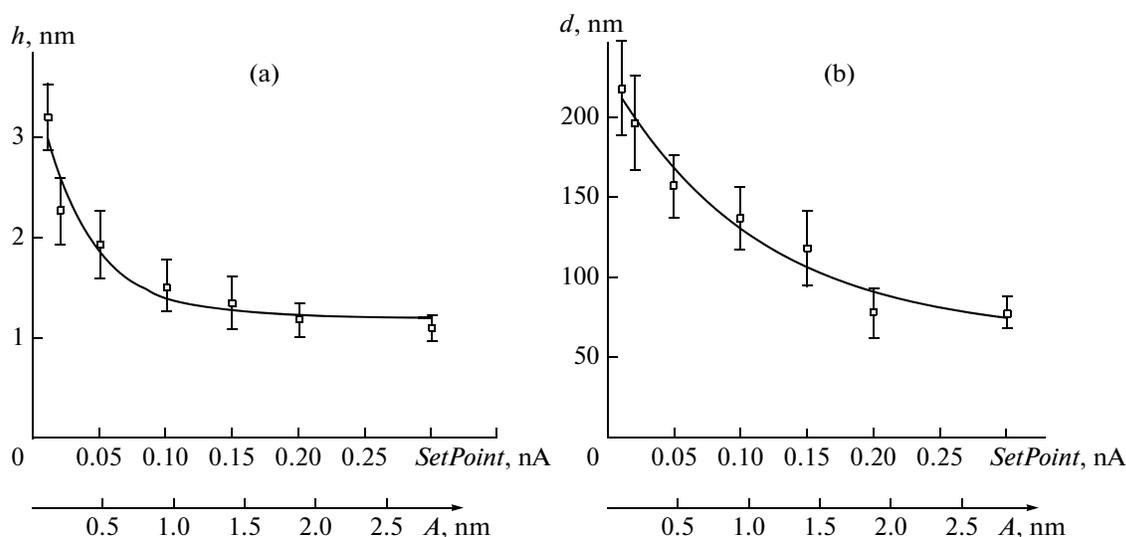


Fig. 5. (a) Height and (b) diameter of gallium arsenide NOSs vs. the cantilever-oscillation amplitude.

height and diameter of the gallium arsenide NOSs on the width of the voltages pulses (Fig. 4). Analysis of these dependences demonstrated that, as the voltage-pulse width is raised from 10 to 500 ms, the NOS height grows from  $0.59 \pm 0.08$  to  $4.64 \pm 0.42$  nm, and the NOS diameter, from  $37.1 \pm 4.3$  to  $101.3 \pm 15.5$  nm.

The decrease in the NOS growth rate can be attributed to the effect of the time in which the active species diffuse across the oxide layer into the LAO zone. As the oxidation duration is increased, the NOS growth rate decreases because the electric-field strength falls due to an increase in the oxide thickness. As a consequence, the number of active species decreases and the drift component of their flux decreases.

Statistical processing of the AFM images and spectroscopic data yielded dependences of the NOS geo-

metric parameters on the cantilever-oscillation amplitude  $A$  (Fig. 5).

It follows from the results we obtained that, as the oscillation amplitude is raised from 0.1 to 2.8 nm, the NOS height decreases from  $3.20 \pm 0.34$  to  $1.10 \pm 0.13$  nm, and the NOS diameter, from  $218.4 \pm 29.5$  to  $78.1 \pm 10.3$  nm. A possible reason for this dependence of the geometric parameters of gallium arsenide NOSs in LAO in the tapping AFM mode is that cantilever oscillations cause the periodic formation and disintegration of water meniscus, during whose existence oxidation occurs [8]. Figure 6 shows schematically the LAO process in the tapping AFM mode. Analysis of this scheme shows that, at an adsorbate film thickness  $D$ , determined by the relative humidity, an increase in the cantilever-oscillation amplitude from  $A_1$  to  $A_3$  results in a decrease in the duration of meniscus existence from  $dt_1$  to  $dt_3$ , and, consequently, a decrease in the

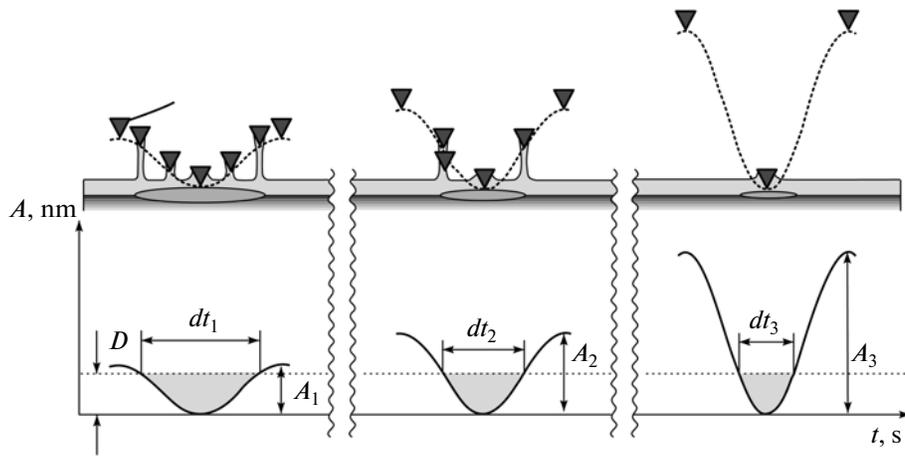


Fig. 6. Schematic of the LAO process in the tapping AFM mode.

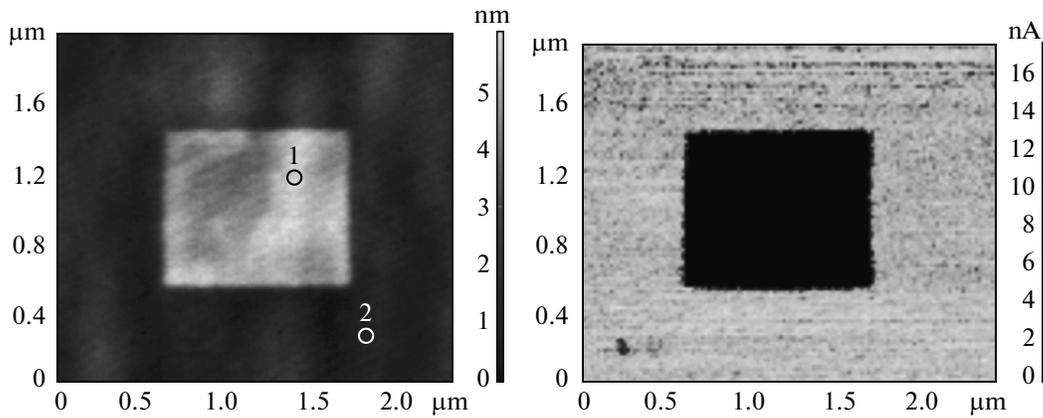


Fig. 7. AFM images of a test gallium arsenide NOS structure: (a) topology and (b) distribution map of the spreading current.

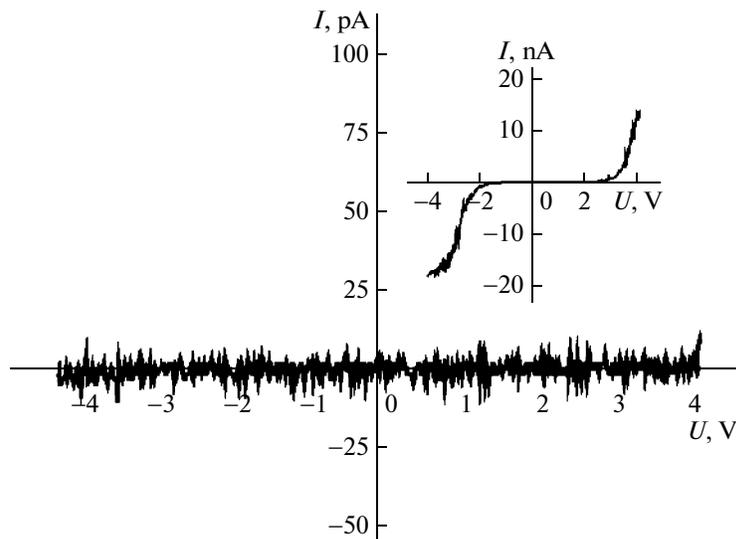


Fig. 8. Current–voltage characteristic measured at point 1. The inset shows the characteristic at point 2 (see Fig. 7a).

oxidation time over an oscillation period. In this case, the surface is oxidized during discrete time intervals,  $dt$ , dependent on the cantilever-oscillation amplitude, rather than during the whole applied-voltage pulse.

Analysis of the AFM images of the test NOS structure in Fig. 7 demonstrated that the  $1 \times 1 \mu\text{m}$  test structure we fabricated has a higher resistance than the initial gallium arsenide. When a bias voltage of 4.0 V is applied to the probe–substrate system, the spreading current over the gallium arsenide surface is  $15.4 \pm 1.5 \text{ nA}$ , and over the surface of the test gallium arsenide NOS structure, it is  $9.7 \pm 1.9 \text{ pA}$ . These results are in good correlation with the current–voltage characteristics in Fig. 8, measured on the surface of our test NOS structure and gallium arsenide (points 1 and 2 in Fig. 7a, respectively), which is indicative of the dielectric nature of gallium arsenide NOSs. Consequently, oxide films formed by the LAO method can be used to form insulating layers in the fabrication of GaAs-based elements for micro- and nanoelectronics.

The study yielded the dependences of the geometric parameters of gallium arsenide nanosized oxide structures on the amplitude and width of voltage pulses applied to the probe–substrate system, the relative humidity in the technological chamber, and the cantilever-oscillation amplitude in the tapping LAO mode. It was found that raising the amplitude and width of the voltage pulses leads to an increase in the height and diameter of gallium arsenide NOSs. Increasing the relative humidity within the technological chamber results in a decrease in the voltage at which the LAO process starts to be observed, and also in an increase in the geometric parameters of gallium

arsenide NOSs. It was shown that the cantilever-oscillation amplitude is also a controlling parameter of the LAO process: as this amplitude is increased, the height and diameter of the nanosized oxide structures being formed decrease. It was found that, in the structures under study, the spreading currents in NOSs are three orders of magnitude smaller than those in gallium arsenide.

Thus, local anodic oxidation is a promising method for nanometer-resolution lithography. The results obtained can be used to develop technological processes for the fabrication of elements for nanoelectronics.

## REFERENCES

1. *Nanotechnologies in Electronics*, Ed. by Yu. A. Chaplygin (Tekhnosfera, Moscow, 2005) [in Russian].
2. *Nanotechnologies in Semiconductor Electronics*, Ed. by A. L. Aseev (SO RAN, Novosibirsk, 2004) [in Russian].
3. R. Garcia and V. Ramses, *Chem. Soc. Rev.* **35**, 29 (2006).
4. V. I. Shevyakov, *Izv. YuFU, Tekh. Nauki* **117** (4), 35 (2011).
5. B. G. Konoplev and O. A. Ageev, *Izv. YuFU, Tekh. Nauki* **89** (12), 165 (2008).
6. O. A. Ageev, B. G. Konoplev, and V. A. Smirnov, *Semiconductors* **44**, 1703 (2010).
7. O. A. Ageev and V. A. Smirnov, *MVI 14-2009* (2009).
8. S. Nishimura, T. Ogino, and Y. Takemura, *J. Phys.* **100**, 2 (2008).

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