

On the Growth of High-Temperature Epitaxial AlN (AlGaN) Layers on Sapphire Substrates by Ammonia Molecular Beam Epitaxy

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Received March 14, 2017

Abstract—The growth of high-temperature AlN and AlGaN layers on (0001) sapphire substrates by ammonia based molecular-beam epitaxy is studied. Factors affecting the formation of inverted domains in high-temperature AlN films are examined. The density of inverted domains is found to correlate with the density of nucleation islands during the initial stages of growth. The denser coverage of a surface by nucleation islands suppresses the formation of inverted domains. It is possible to increase the density of surface coating at the nucleation growth stage by increasing the degree of substrate nitriding, reducing the deposition temperature, and using intense ammonia fluxes during deposition of the initial layers. The kinetic model in the mean field approximation is developed to explain the observed effects of growth parameters on the density of nucleation islands. The growth features of AlN and its structure are taken into account. The obtained results are used to grow AlN/AlGaN layers with improved structural quality. The grown films have a root-mean-square surface roughness of 2 Å and 120 arc s FWHM of X-ray diffraction peaks for the AlN 0002 reflection. The density of inverted domains is decreased to below 10^5 cm^{-2} . Improvement in the quality of the AlN films is achieved by using two-step growth and by the application of gallium as a surfactant.

Keywords: aluminum nitride, ammonia-based molecular beam epitaxy, nucleation islands, inverted domains, surfactant

DOI: 10.1134/S1027451017060167

INTRODUCTION

Gallium, aluminum and indium nitrides, and also their solid solutions are widely applied in micro- and optoelectronics due to their unique properties. In microelectronics nitride semiconductors are applied for the creation of high-performance microwave devices on the basis of high electron mobility transistors (HEMT) [1]. Despite the remarkable progress of the last two decades, the developed morphology of nitride films and the high density of crystal defects in them remain the main factors limiting the characteristics of devices on the basis of nitrides. These problems are the consequence of the necessary usage of substrates mismatched with nitrides in terms of lattice parameters and temperature expansion coefficients [2].

For high-performance devices of the millimeter range on the basis of GaN-HEMT, heterostructures with the lowest possible thickness of the barrier layer and sharp heteroboundaries are required. Fulfillment of these conditions when using the method of metalorganic chemical vapor deposition (MOCVD) is difficult due to the impossibility of instant replacement of the composition of the gas mixture in the growth reactor. For this reason the growth of heterostructures for a high-performance HEMT is per-

formed using molecular-beam epitaxy (MBE), the version of which is ammonia-based MBE. In the ammonia-based MBE, ammonia is used as a source of active nitrogen through the decomposition of ammonia molecules into nitrogen and hydrogen at a heated substrate.

Due to the later appearance in comparison with MOCVD and plasma MBE, ammonia-based MBE to date is one of the least studied methods of nitride epitaxy. In only 2012 was a paper published, in which data about different regimes of GaN deposition in the case of homoepitaxial growth on MOCVD-templates GaN were generalized as a function of the substrate temperature, and the values of the ammonia and gallium fluxes [3]. This work supplemented the earlier study [4], in which the connection between the deposition temperature of GaN films and their insulating properties were studied. On the other hand, the necessity of the use of nucleation and buffer layers grown by other methods is being mentioned since the earliest studies on growth by ammonia MBE, since the growth of GaN on buffer layers prepared by the method of ammonia-based MBE led to the formation of GaN of extremely poor quality [5]. Even to date the preparation of heterostructures for HEMT with a low rough-

ness of the surface and interfaces of layers exclusively by means of ammonia-based MBE remains a complicated problem [4]. For this reason the use of such structures is limited to high-power electronics in the range up to 18 GHz [6]. When preparing structures suitable for use in millimeter range devices, it is considered necessary to perform growth on templates preliminarily manufactured using gas-phase epitaxy [7]. Only in this case it was possible to prepare heterostructures, which in terms of morphological and electrophysical properties are not inferior to structures grown by the method of plasma MBE.

Thus, quality heterostructures for microwave applications can be prepared by the method of ammonia-based MBE, if before the deposition of functional GaN layers it is possible to form rather qualitative buffer layers. This will make it possible to avoid the necessity of the enlistment of additional growth methods and exclude the destructive interaction of template surface with the atmosphere. The application of high-temperature AlN films as buffer layers for ammonia-based MBE to date seems to be the most promising, since AlN, unlike GaN, does not decompose under high vacuum conditions at temperatures higher than 1000°C, and high temperatures favor an improvement in the morphology and structure of epitaxial films [8]. The use of high-temperature AlN films is also promising due to the possibility of the application of gallium as a surfactant [9, 10], which improves the morphology and structure of AlN layers. For implementation of these prospects it is necessary to study the effect of the growth parameters on the properties of high-temperature AlN films. In addition to insufficient knowledge of the formation mechanisms of such films, there is an obvious need to reduce the density of inverted polarity domains in high temperature AlN films on sapphire [11, 12], the presence of which leads to the development of the relief of films and the appearance of leaks in heterostructures [13].

In this work we present the results of studies of the growth of high-temperature AlN layers by ammonia-based MBE on sapphire substrates. The effect of conditions of AlN formation, including the use of gallium as surfactant, on the density of inverted polarity domains and the structural perfection of layers is studied.

EXPERIMENTAL

Samples were grown on sapphire (0001) substrates with a bevel angle of the surface of 0.2° in the direction of the *m* axis using a Semiteq STE3N* setup for ammonia-based molecular-beam epitaxy. To provide heating, the rear side of the substrates was coated with a titanium layer with a thickness of 300 nm. The substrate temperature was measured using an infrared pyrometer calibrated to the melting temperature of aluminum (660°C). Substrates were annealed under high vacuum conditions at a temperature of 850°C and

subsequent nitridation in an ammonia flux of 30 sccm for 15 min, if not specified otherwise. The thicknesses of films during the process of growth were measured using a laser interferometer. Al and Ga fluxes are given in units of the growth rate of films (0001) AlN and (0001) GaN in μm/h, as in [14].

The deposited AlN and AlGa_xN films had a thickness of 300 nm, if not specified otherwise. In the process of growth, the film surface was observed using the reflection high energy electron diffraction (RHEED) at an electron-beam accelerating voltage of 20 keV. The topography of the surface of the obtained samples was studied by the method of atomic force microscopy (AFM) in the tapping mode on an NTEGRA Aura scanning probe microscope (NT-MDT). Observation of the surface reconstructions using RHEED showed that all grown films had metal polarity (Al-polarity) [15]. The structural perfection of the obtained films was estimated by X-ray diffraction [16] obtained using a Rigaku SmartLab diffractometer (CuK_α-radiation, wavelength of 1.54 Å). Samples for study by the method of transmission electron microscopy (TEM) on a Titan 80–300 setup (FEI) were prepared via mechanical polishing with subsequent etching with argon ions.

GROWTH OF HIGH-TEMPERATURE ALN FILMS

A series of AlN films of six samples was prepared. Before the beginning of deposition all films were subjected to nitridation in an ammonia flux of 30 sccm for 15 min. Three samples were grown in an ammonia flux of 60 sccm and at substrate temperatures of 1020, 1050, and 1100°C; the other three samples were deposited in an ammonia flux of 200 sccm at the same values of the substrate temperature. For all six samples the aluminum flux was 0.2 μm/h. The thickness of all films was 300 nm. In the process of growth the electron diffraction pattern was observed. According to laser interferometry data, the growth rate for the samples was the same and was 0.2 μm/h, i.e., corresponded to the aluminum flux.

An example of the change in the diffraction pattern during the process of growth of the AlN film is shown in Fig. 1. The observed parallel lines correspond to the normal direction to the substrate plane. For all six samples in the first 1.5–3 minutes of growth the diffraction pattern was a set of diffraction spots. The position of spots corresponded to sites of the reciprocal AlN lattice. Further, linear vertical reflections appeared in the diffraction patterns of all samples against the background of spots. During the subsequent growth of all samples, except for the film deposited at a temperature of 1100 °C in an ammonia flux of 60 sccm, complete transition from a mixed diffraction pattern to a linear one was observed, which corresponds to a transition from the three-dimensional

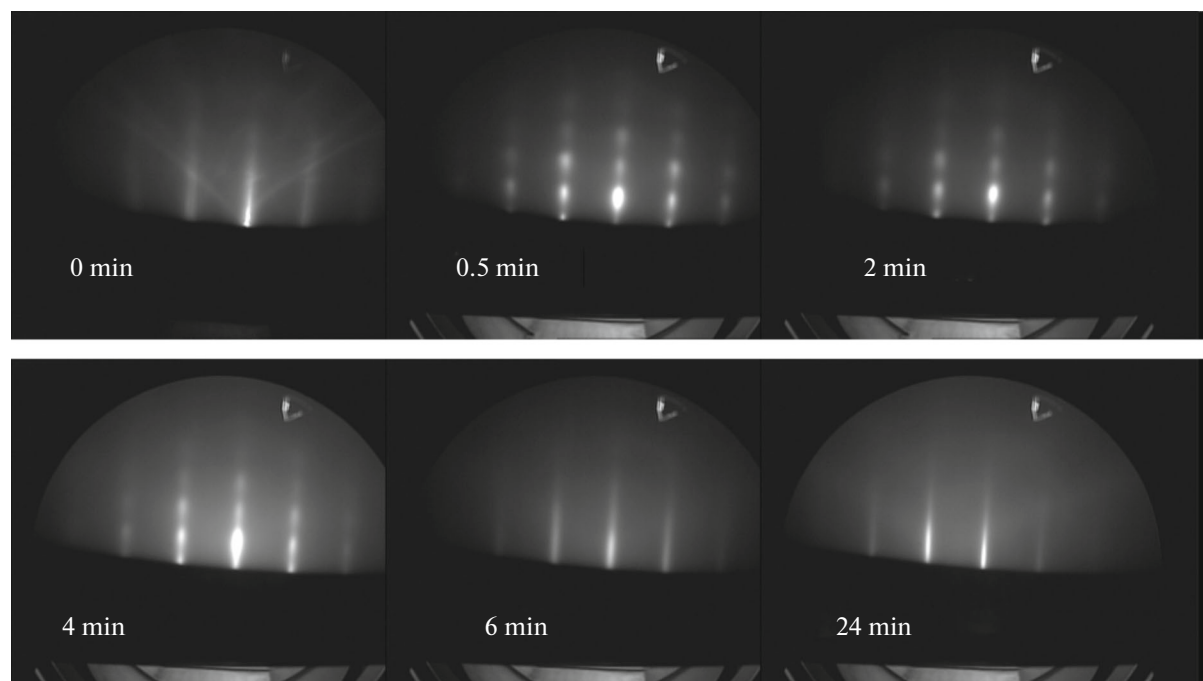


Fig. 1. Evolution of the RHEED pattern in the AlN sample grown at a substrate temperature of 1100°C in an ammonia flux of 200 cm³/min.

mode of growth of an island nucleation layer to the two-dimensional growth of a continuous film (the 3D–2D transition) [10]. Data on the transition time are given in Table 1. Table 1 also shows the AlN thicknesses corresponding to the transition time calculated according to the known AlN deposition rate. The diffraction pattern of the sample deposited at the temperature of 1100°C in an ammonia flux of 60 cm³/min remained a superposition of bright point and linear reflections until the termination of the growth process (Fig. 2).

AFM images of the surface regions of the obtained AlN films with a size of 10 × 10 μm are given in Fig. 3. Hillocks with a diameter of ~200 nm were observed on the surface of all samples. It is seen from the presented images that the density of hillocks, i.e., the amount per unit area of the surface depends both on the depo-

sition temperature and the value of the ammonia flux supplied during the process of growth.

The hillocks observed on the AlN surface using AFM were described in earlier works [11, 12] and were found to be the surface terminations of inverted polarity domains. In [11] this was confirmed via the etching of structures in an alkali solution and by observations of diffraction from these regions in a convergent electron beam. An additional experiment on etching of one of the six obtained samples in KOH solution showed that pits were formed in the place of hillocks, while the relief of the surrounding regions did not change significantly. This result confirms that the found formations are surface terminations of inverted domains.

Table 1. Transition time from three-dimensional to two-dimensional growth at different ammonia fluxes and AlN growth temperatures

Temperature, °C	Ammonia flux, cm ³ /min	Transition time, min	Film thickness at the time of the transition, nm
1020	60	16	53
1050	60	20	67
1020	200	12	40
1050	200	14	46
1100	200	13	43

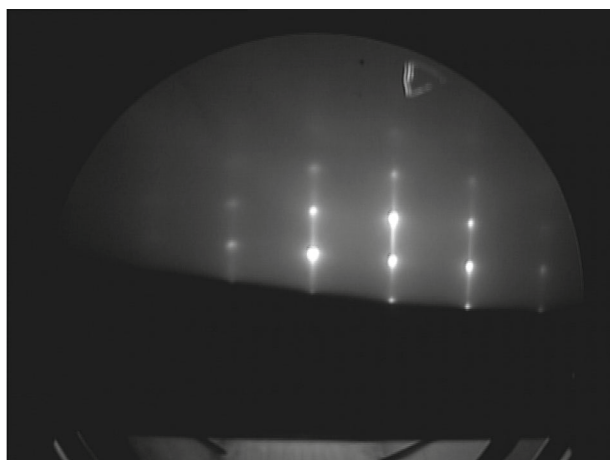


Fig. 2. RHEED pattern in the AlN film deposited at 1100°C in an ammonia flux of 60 sccm at the time of growth termination.

The plot of the dependence of the hillock density on the conditions of film deposition is given in Fig. 4a. It is seen from the presented plot that the hillock density increases with increasing deposition temperature. In turn, the more intense ammonia flux favors the formation of a smaller number of hills. The minimal density of such objects is observed upon AlN film grown at a temperature of 1020°C in an ammonia flux of 200 cm³/min, and is 2×10^7 cm⁻².

The more intense ammonia flux and the decrease in temperature favor accelerated transition from the three-dimensional stage of nucleation growth to two-dimensional growth. Figure 4b shows a plot of the dependence of the inverted domain density on the duration of the 3D–2D transition. Referring to AFM data, it is possible to conclude that the absence of the transition to a linear diffraction pattern in the sample grown at 1100°C in an ammonia flux of 200 cm³/min is associated with a high density of hillocks, which are the origin of the spotty RHEDD pattern (Fig. 2).

Terraces with stepwise boundaries (Fig. 3) were distinctly visible on the surface of all samples, except for that grown in an ammonia flux of 60 sccm and at a temperature of 1100°C, and at higher deposition temperatures terraces with more clearly defined boundaries were formed. Steps on the surface of samples were oriented perpendicularly to the bevel direction of the surface of substrates with respect to the sapphire (0001) plane. The width of terraces was about 0.5 μm, and the height was 2–3 nm. The ratio of the height of the steps to the length of the terraces corresponds to the bevel angle of the substrate of 0.2°. Thus, the planes of terraces were oriented parallel to the basal (0001) plane of the sapphire substrate.

High-resolution AFM images were also obtained; the scanning region was 1×1 μm. On the surface of all obtained AlN films pits were found, the density of

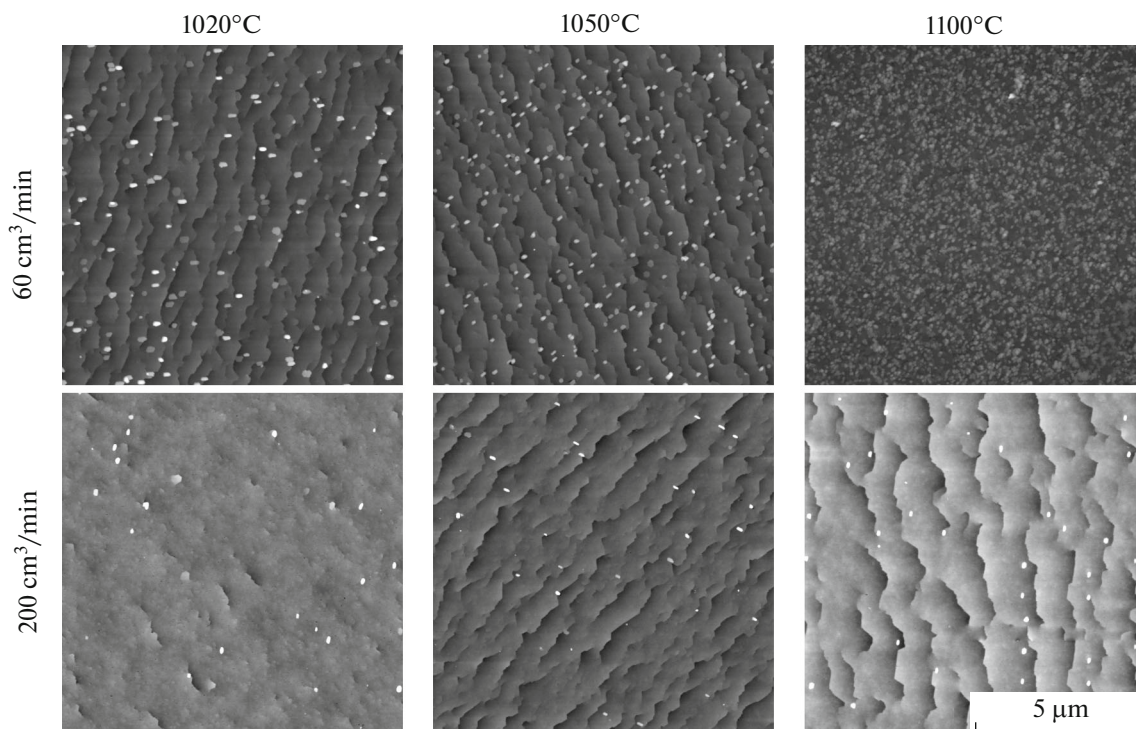


Fig. 3. AFM images of regions of the surface of AlN films grown at different temperatures and ammonia fluxes, size of 10×10 μm.

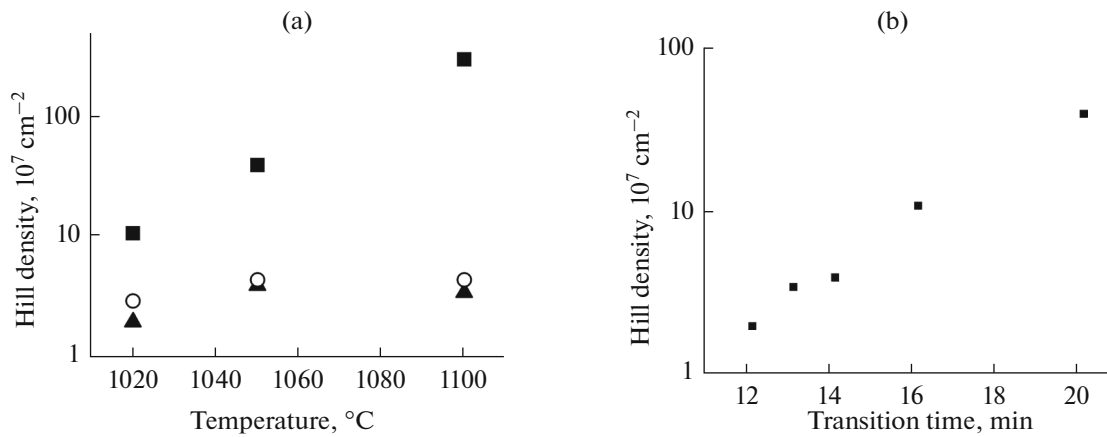


Fig. 4. Dependence of the density of hills on the surface of the AlN film: (a) on the temperature in the process of growth in an ammonia flux of 60 (squares), 200 cm^3/min (triangles); (b) on the transition time from three-dimensional growth of the nucleation layer to two-dimensional growth. Circles denote data for the AlGaIn film.

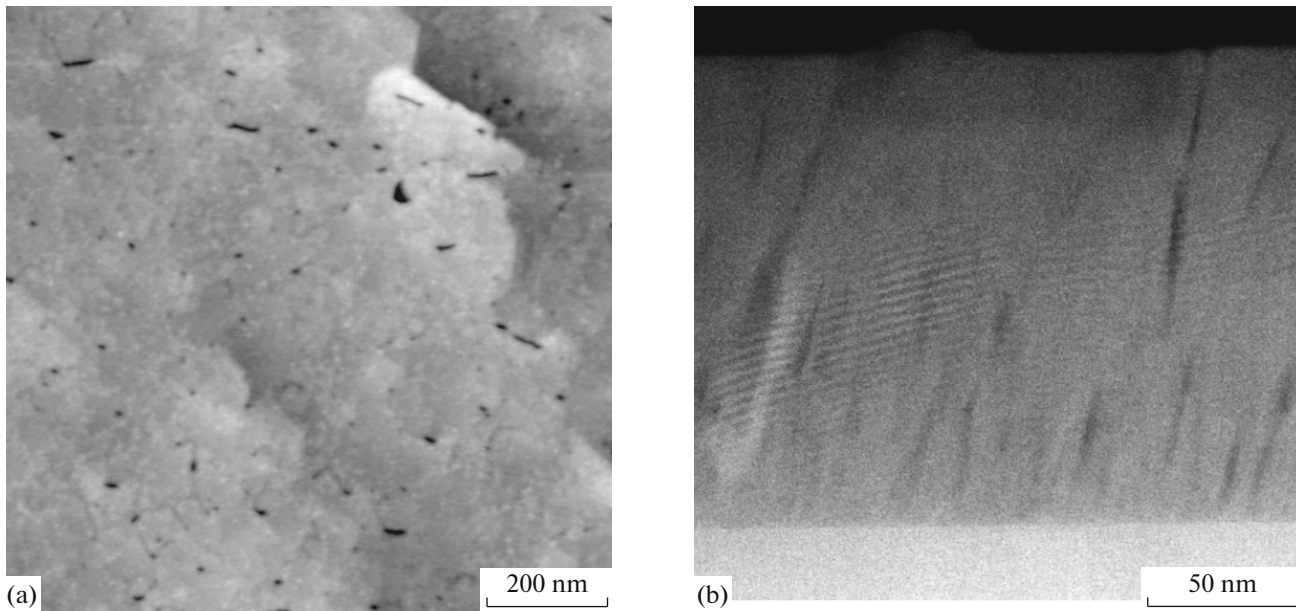


Fig. 5. Penetrating pores in AlN films: (a) AFM image of the surface of an AlN film with an area of $1 \times 1 \mu\text{m}$; (b) TEM image of the cut of the AlN film.

which was higher than 10^9 cm^{-2} and did not depend on the deposition conditions (Fig. 5a).

For TEM study of the films we manufactured samples, on which the cut plane was perpendicular to the substrate plane. One of the obtained images is shown in Fig. 5b. It is seen that the total thickness of the AlN film is permeated with pores. In the surface terminations of pores pits are observed. Thus, pits observed using AFM are the surface terminations of penetrating pores.

Diffraction peaks of the 0002 reflection were recorded for AlN films obtained in an ammonia flux of 60 sccm. The widths of the diffraction peaks at half-height are given in Table 2. It is seen from the pre-

sented data that the width decreases with increasing temperature which indicates a decrease in the density of penetrating screw dislocations with increasing deposition temperature. It is important that the density of penetrating screw dislocations decreases with

Table 2. Dependence of the widths of the rocking curves at the half-height of reflections 0002 for AlN films grown in an ammonia flux of 60 sccm at different temperatures

Temperature, $^{\circ}\text{C}$	Width, arc s
1020	257
1050	248
1100	217

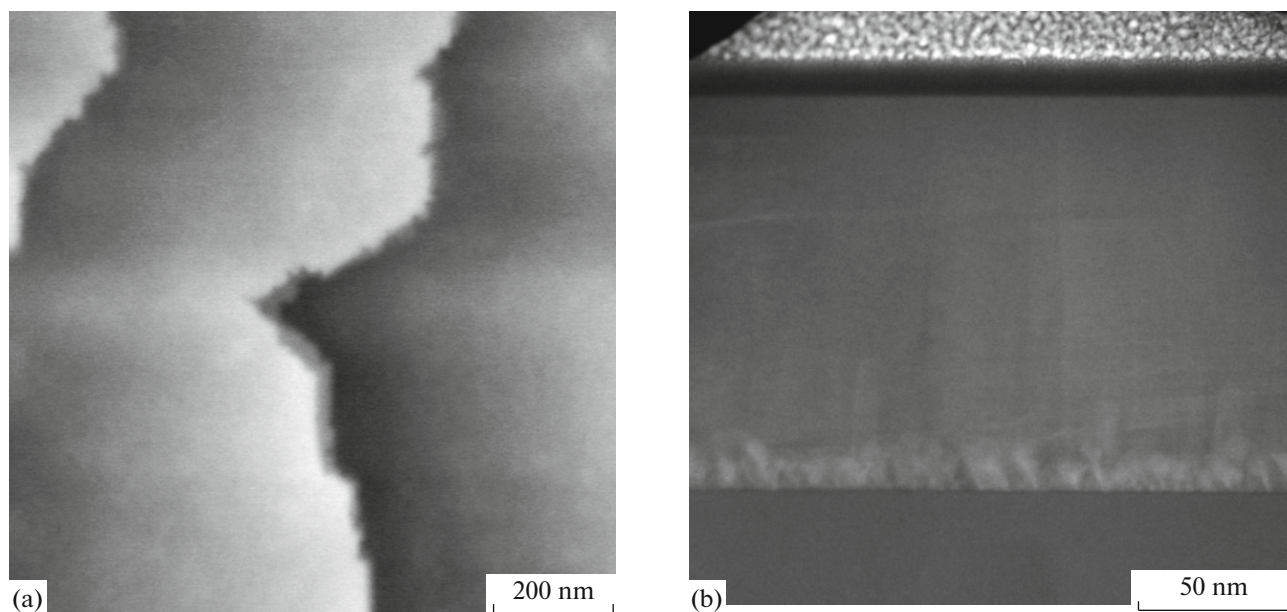


Fig. 6. Morphology of the AlGaIn film: (a) AFM image of the surface; (b) TEM image of the film in the dark-field mode. No penetrating pores are seen on the surface and in the volume of the film.

increasing temperature, in spite of an increase in the inverted domain density.

THE EFFECT OF A GALLIUM ADDITIVE ON THE INVERTED DOMAIN DENSITY AND THE MORPHOLOGY OF THE FILMS

To compare with AlN samples, three AlGaIn samples were grown with a thickness of 300 nm in an ammonia flux of 200 cm³/min at temperatures of 1020, 1050, and 1100°C. A flux of 200 sccm was chosen, because it favors the formation of a lower amount of hillocks. The growth rates at the corresponding temperatures were 330, 245, and 205 nm/h, i.e., the deposition rate decreased with increasing temperature due to an increase in gallium desorption [10]. Observation of the surface during the growth using RHEED showed that in AlGaIn films, similar to AlN, the transition from three-dimensional to two-dimensional growth is implemented.

AFM analysis of the surface showed that the presence of gallium did not lead to considerable changes in the hillock density (Fig. 4a), however their mean lateral sizes increased to 400 nm. This increase in sizes can be associated with the mechanism of selective enhancement of the lateral growth component upon AlGaIn formation under conditions of strong gallium desorption [10]. The surface of the AlGaIn films also had a terrace-stepwise morphology; however, the average width of the terraces increased twofold and was 1 μm on average at a step height of 4–5 nm (Fig. 6a). The terraces had an atomically smooth surface, and pits present on AlN films were not observed on their surface.

The sample grown at 1050°C was studied using TEM. To this end, a cut was made perpendicularly to the film surface. Figure 6b shows the dark field image. No traces of penetrating pores present in AlN were observed either on the surface or in the volume of the obtained films.

Thus, the addition of gallium upon film growth under conditions of strong desorption makes it possible to eliminate penetrating pores in the films, it does not greatly affect the inverted domain density. The elimination of penetrating pores is an important effect. These defects are apparently penetrating dislocations with an open core. It was established that similar defects can form channels of parasitic conductivity [17], which negatively affects the characteristics of devices.

THE EFFECT OF THE INITIAL STAGES OF AlN GROWTH ON THE INVERTED POLARITY DOMAIN DENSITY

The generalization of experimental data on the correlation of the inverted domain density with the growth conditions makes it possible to conclude the following. The domain density decreases with an increase in the ammonia flux and the substrate temperature during AlN growth. In addition, an increase in the ammonia flux and a decrease in the substrate temperature lead to a decrease in the 3D–2D-transition time and, respectively, a decrease in the 3D-layer thickness. It is important to note that the transition time to 2D growth is also reduced at an increase in the time of substrate nitriding. Upon nitridation of the

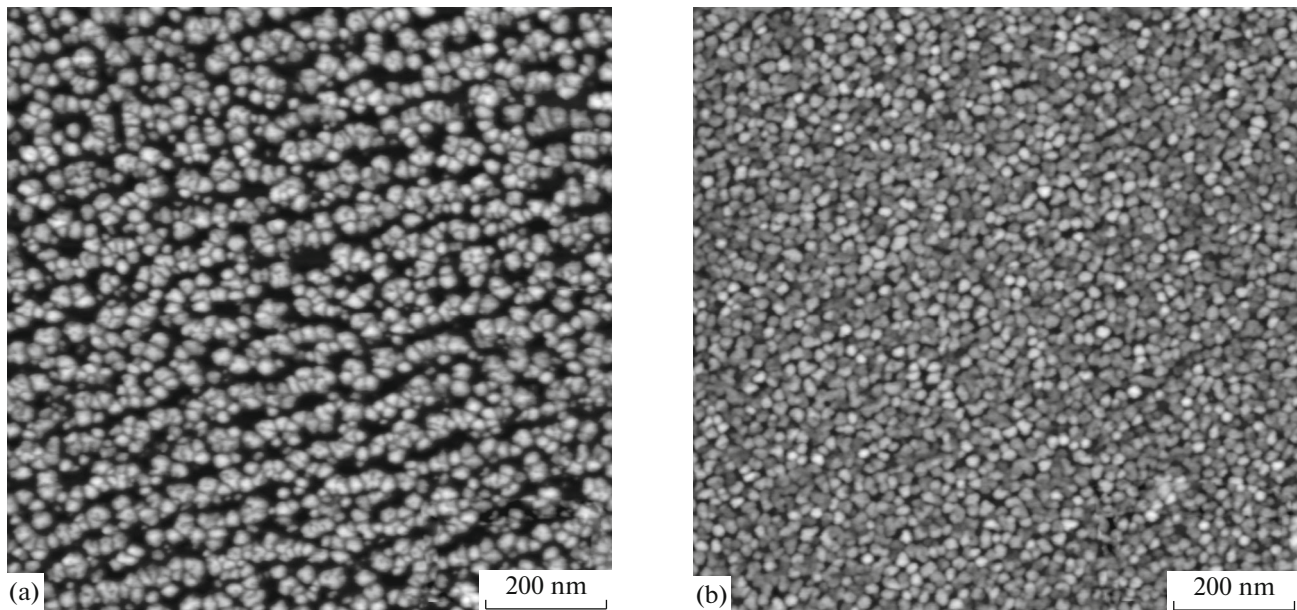


Fig. 7. AFM images of a region with a size of $1 \times 1 \mu\text{m}$. AlN nucleation layers on the sapphire surface prepared under different conditions: (a) the surface was not subjected to nitriding; ammonia flux during growth $30 \text{ cm}^3/\text{min}$; (b) nitriding for 60 min; ammonia flux $200 \text{ cm}^3/\text{min}$.

sapphire surface for 40 min instead of 15 min in an ammonia flux of $200 \text{ cm}^3/\text{min}$ the transition to 2D growth in the AlN film deposited at 1050°C and aluminum and ammonia fluxes of $0.2 \mu\text{m}/\text{h}$ and $200 \text{ cm}^3/\text{min}$, respectively, is reduced from 14 to 8 min. This corresponds to a decrease in the thickness of the 3D layer from 46 to 26 nm. The decrease in the transition time can be associated with the formation of a larger amount of nucleation islands. In this case the high concentration of nucleation centers on the surface leads to the faster merging (coalescence) of islands due to a smaller distance between them.

To verify the assumption that the decrease in the transition time is associated with an increase in the density of nucleation islands at the initial stages of the formation of AlN on sapphire, we manufactured two experimental samples. We deposited onto the surface of two sapphire plates the amount of aluminum corresponding to a thickness of 10 nm of the homogeneous epitaxial AlN film. The first sample was not subjected to nitridation, and Al deposition was carried out in a relatively low ammonia flux of 30 sccm and at a substrate temperature of 1100°C . The second sample was subjected to nitriding for 60 min, and the supply of aluminum was carried out at a maximal ammonia flux of 200 sccm and a substrate temperature of 1020°C . In this manner two extreme cases of nucleation layer growth were implemented: a low degree of nitriding of the substrate in combination with a low ammonia flux and a high substrate temperature for AlN formation and a high degree of nitriding in combination with a high ammonia flux and a lower substrate temperature.

According to fast electron diffraction RHEED data, on both samples three-dimensional AlN films of the island type were obtained. The results of AFM study of the surface of the obtained samples are given in Fig. 7. The first sample (Fig. 7a) demonstrates a smaller amount of nucleation islands, while the islands themselves have large sizes. The islands are not distributed uniformly over the film surface, and decorate the boundaries of vicinal steps: it is seen well in the AFM image (Fig. 7a) how the islands are organized in periodic rows. The second sample (Fig. 7b), on the contrary, is characterized by the uniform distribution of nucleation islands and a higher degree of coating of the surface. The obtained result confirms the assumption that the observed decrease in the time of the transition from 3D to 2D growth upon an increase in the ammonia flux during nucleation layer growth, a decrease in the deposition temperature and an increase in the degree of nitriding of the substrate, increases the concentration of nucleation centers on the substrate surface.

Though these data do not clarify the mechanism of the formation of inverted polarity domains, they show that the intensity of inversion domain formation depends on the concentration of nucleation centers during the initial stages of growth or, equivalently, on the thickness of the 3D layer and the transition time to two-dimensional growth. It is apparent that for the formation of nucleation centers of the N-polar phase of AlN, regions not occupied by nucleation centers of Al polarity are needed; therefore, the amount of inverted domains increases with increasing free area

on the sapphire surface and with increasing time, during which no AlN-metal polarity is formed on them. It follows that to reduce the inverted domain density it is necessary to provide the maximal density of nucleation islands of Al polarity and to reduce the transition time to the two-dimensional growth mode.

It follows from the performed experiments that the density of nucleation islands can be controlled by varying the degree of nitridation of the sapphire surface, and also setting the growth temperature and the value of the ammonia flux. The effect of the degree of nitriding on the density of nucleation centers has an obvious explanation—AlN crystallites are more actively formed on nitrided regions, which are 1–2 single layers of aluminum nitride formed by the substitution of oxygen atoms in the substrate for nitrogen atoms [18]. On a clean surface Al_2O_3 and AlN, and GaN are formed less actively which is noted in many works and was observed many times in experiments. The correlation between the formation of inverted domains and the nitridation of substrates was also noted earlier [19, 11]. To explain the effect, which is produced by the temperature and value of the ammonia flux on the density of nucleation centers, it is necessary to consider the kinetics of AlN nucleation in more detail.

MODEL OF THE FORMATION OF AlN NUCLEATION ISLANDS

For many single-component [20] and binary [21] compounds on a homogeneous surface the experimental results confirm the predictions of kinetic models, according to which the density of nucleation islands is proportional to: $(D/F)^{-i/(i+2)}$. Here D is the kinetic constant of the surface diffusion of adatoms, F is the value of the flux of atoms coming to the surface, and i is the critical size of nucleation centers. The diffusion of adatoms increases exponentially with increasing temperature, which explains the decrease in the density of nucleation islands. To explain the effect of the value of the ammonia flux and the degree of nitriding of the substrate on the density of nucleation centers, it is necessary to take into account that aluminum nitride (AlN), the same as and other nitrides of metals of the III group, have a series of unusual properties.

Nitrides have a layered structure, in which layers of metal atoms alternate with layers of nitrogen atoms. In bulk nitride crystals the exact stoichiometric ratio between amounts of metal and nitrogen atoms is maintained, and there is one nitrogen atom per aluminum atom in AlN, independent of the ratio of fluxes of the metal and nitrogen components. The aluminum layer is formed first on the nitrided surface. Nitrogen atoms, in turn, form a second atomic layer joining with aluminum. The direct formation of aluminum nitride

crystallites, which have much greater stability than clusters consisting of only aluminum atoms, occurs as a result of this reaction. Thus, the formation of N–Al bonds strengthens clusters and does not allow them to decompose later. It is also necessary to take into account that AlN nucleation centers are preferably formed on nitrided regions. Ammonia molecules interact with a sapphire surface less actively than with the AlN surface, and the nitridation process has a low intensity [18]. The latter means that a change in the degree of nitriding of the surface during formation of the nucleation layer can be ignored.

Let n be the concentration of free aluminum atoms on the surface, N be the concentration of nucleation islands, n_s be the concentration of clusters containing s aluminum atoms, D be the constant of the surface diffusion of aluminum atoms, F_{Al} and F_{NH_3} be specific fluxes of gallium atoms and ammonia molecules, respectively, σ_s be the cross section of capture of an aluminum atom at a cluster containing s aluminum atoms

Since a model is introduced for qualitative explanation of the effect of the degree of nitriding of the substrate and the value of the ammonia flux on the density of nucleation centers, we consider the simplest model of nucleation taking into account nitriding and the flux of the nitrogen component. In accordance with the simplified classical model of the growth of nucleation centers [20], we suppose that the islands containing $s \leq i$ aluminum atoms, are unstable and can dissociate, and the bond energy of atoms with the cluster can be ignored. The intensity of the process of the formation of clusters with the size $i + 1$ on the isotropic surface is $dN/dt = D\sigma_i n_i n$. We take into account that the islands of aluminum atoms with a size larger than i are also unstable (due to high-temperature surface conditions), if they do not form bonds with nitrogen, i.e., if the formation of AlN, which is stable at high temperatures does not occur. We consider that for the formation of a stable AlN island containing $i + 1$ aluminum atoms, no less than $p > 0$ nitrogen atoms are required. We also take into account the effect of nitridation on the formation of nucleation centers and consider that the cluster can be formed only on $m > 0$ neighboring nitrogen atoms substituting oxygen atoms in the substrate during nitridation. Thus, the intensity of the formation of nucleation centers is proportional to the density of nitrided cells of the surface to the power m and ammonia flux to the power p :

$$\frac{dN}{dt} = D\sigma_i n_i n (A\eta^m F_{\text{NH}_3}^p), \quad (1)$$

here A is the temperature-dependent proportionality coefficient, η is the degree of nitridation of the substrate expressed as the amount of nitrided cells per unit area of the surface.

The number of separate aluminum atoms n changes due to their arrival on the surface with the flux F_{Al} , the formation of nucleation centers and the attachment of atoms to nucleation islands:

$$\frac{dn}{dt} = F_{\text{Al}} - D\sigma_i n_i n (A\eta^m F_{\text{NH}_3}^p) - D\bar{\sigma} N n, \quad (2)$$

where $\bar{\sigma} = (1/N) \sum_{k>i} \sigma_k n_k$ is the average cross section of the capture of atoms at islands.

After some time from the beginning of deposition the amount of unstable nucleation centers with a size of $\leq i$ reaches quasi-equilibrium values due to the fact that their formation rate is balanced by the decay rate [22]. Under such conditions the amount of unstable clusters of aluminum atoms with a size of $s \leq i$ is proportional to n^s :

$$n_s \sim n^s. \quad (3)$$

In the quasisteady-state mode the number of single aluminum atoms n reaches the equilibrium value; therefore, in Eq. (2) $dn/dt = 0$, and the nucleation term is negligibly small [21] which yields

$$n \sim F_{\text{Al}} / (D\bar{\sigma} N). \quad (4)$$

Integrating Eq. (1) into $d\Theta = F_{\text{Al}} dt$, after substitutions (3) and (4) we obtain:

$$N \sim (\eta^m F_{\text{NH}_3}^p)^{1/(i+2)} (F_{\text{Al}} / D)^{i/(i+2)}. \quad (5)$$

Expression (5) and its derivation show qualitatively how the value of the ammonia flux affects the island density. At a small value of the nitrogen component flux clusters of critical size can dissociate before they form an AlN nucleation island. An increase in the ammonia flux increases the probability of the formation of a nucleation center before the decay of the cluster. This leads to the fact that in a strong ammonia flux a larger number of nucleation Al-polar AlN islands are formed. The effects of an increase in the degree of nitriding and an increase in the nitrogen-component flow at the stage of growth of nucleation layers multiply which makes their combined use for an increase in the density of nucleation centers more effective than a variation in only one of the parameters.

ULTRASMMOOTH ALN FILMS WITH IMPROVED STRUCTURE

The obtained results show that the formation of inverted polarity domains can be suppressed due to an increase in the degree of nitriding of the sapphire substrate, a decrease in the deposition temperature and an increase in the ammonia flux. The density of islands in the performed experiment was reduced to $2 \times 10^7 \text{ cm}^{-2}$ which is several times less than the density of inverted domains achieved in [12] due to the preliminary formation of a cold aluminum layer. The formation of penetrating pores, in turn, can be suppressed, if one

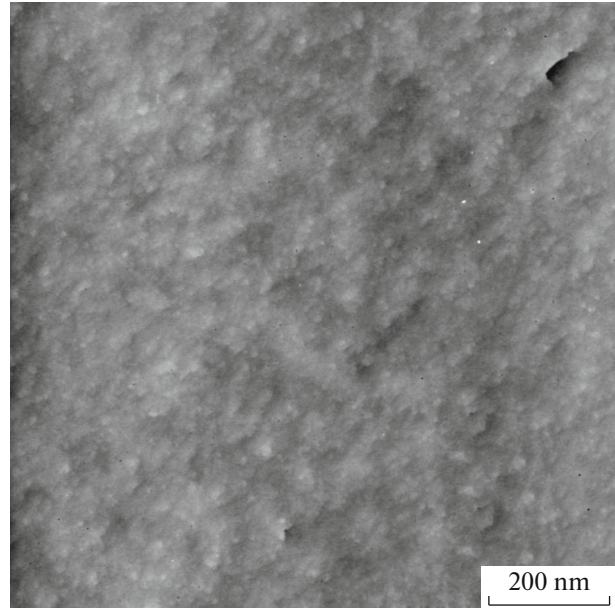


Fig. 8. AFM image of the surface of the AlGaN film grown by the two-step method (region of $10 \times 10 \mu\text{m}$). The root-mean-square roughness of the surface is 2 Å.

uses gallium as a surfactant. However, the problem is that the decrease in the growth temperature increases the density of screw dislocations. Thus, one AlN layer cannot simultaneously solve the tasks of reducing the density of inverted domains and improving crystal perfection. To separate these functions, we used a widely applied approach for splitting the growth process into two stages.

On the sapphire substrate, which was preliminarily nitrided for 40 min instead of 15 min, we grew an AlN film, the first 10 nm of which were formed at a temperature of 1050°C in an ammonia flux of 200 sccm. The second layer was deposited at a temperature of $\sim 1120^\circ\text{C}$ at a supply of the gallium flux as SAC for suppressing the process of the formation of penetrating pores. As a result, we managed to prepare an AlN film, the root-mean-square roughness of which is only 2 Å (Fig. 8). The inverted polarity domain density in these films is no higher than 10^5 cm^{-2} . The prepared AlN layers have a degree of crystal perfection high for ammonia-based MBE, which is characterized by the width of the diffraction peak at a half-height of 120 arc s for the 0002 reflection. The typical width of the rocking curve for AlN films (0002) on sapphire in the case of ammonia-based MBE, according to published data, is larger than 200 arc s. Such characteristics are record ones for AlN films grown on sapphire substrates by the method of ammonia-based MBE. It is important to note that the repetition of experiments on the growth of similar samples shows good reproducibility of the results.

CONCLUSIONS

Mechanisms of the growth of high-temperature AlN layers on sapphire substrates using the method of ammonia-based molecular-beam epitaxy were studied. Factors affecting the formation of inverted polarity domains, which are manifested in the form of island formations on the surface of the film in Al-polar epitaxial AlN layers on sapphire substrates were revealed. It was established that the inverted domain density correlates with the density of coating of substrates with nucleation islands at the initial growth stages, and the dense coating suppresses domain formation. It is shown that it is possible to increase the surface coating density at the nucleation growth stage by increasing the degree of nitridation of the substrate and by reducing the deposition temperature, and also using high ammonia fluxes during the deposition of the initial layers. A kinetic model is presented to explain the found regularities describing the formation of AlN nucleation centers in the self-consistent field approximation. According to the proposed model, the increase in the ammonia flux increases the probability of the formation of stable AlN nucleation centers from less stable aluminum clusters of critical size. The effect of nitridation and an increase in the ammonia flux at the growth stage of nucleation centers multiply.

A two-step approach to the preparation of AlN layers on sapphire substrates with improved structure and morphology is developed on the basis of the revealed growth regularities. The first layer performs the function of suppressing the process of the formation of inverted domains. The second layer is grown at a higher temperature which favors improvement in the crystal perfection of the film, and with the use of gallium as surfactant for suppressing the process of the formation of penetrating pores. As a result, we prepared AlN films with a thickness of 300 nm with a root-mean-square roughness of 2 Å and a reduced density of defects. The inverted domain density in these films does not exceed 10^5 cm^{-2} . The obtained characteristics are record ones for AlN layers grown on sapphire substrates by the method of ammonia-based molecular-beam epitaxy.

ACKNOWLEDGMENTS

Samples were studied by the method of transmission electron microscopy at the Resource center of probe and electron microscopy of the National Research Center “Kurchatov Institute”. X-ray structural analysis was carried out at the Resource center of

laboratory X-ray methods of the National Research Center “Kurchatov Institute”. The work was supported by the Ministry of Education and Science of the Russian Federation (agreement no. 14.607.21.0116 of October 14 2015, RFMEFI60715X0116).

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Translated by L. Mosina