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Stimulated emission of AlGaN layers grown on sapphire substrates using ammonia molecular beam epitaxy

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***Abstract.* By optimising the growth temperature of the AlGaN layers and using high-temperature AlN buffer layers, high-quality $Al_xGa_{1-x}N$ layers (x = 0.15, 0.21, 0.26, and 0.3) were obtained, in which stimulated emission in the UV spectral range 330 – 297 nm was implemented with the threshold intensity of excitation $I_{th} \approx$ 0.7 – 1.4 MW cm^{-2}, respectively. It is found that the threshold value of the stimulated emission of AlGaN layers grown by molecular beam epitaxy is largely determined by the intensity of the process of thermal decomposition of GaN, which affects the surface morphology and, consequently, the amount of optical scattering loss. It is shown that no pronounced localisation of nonequilibrium charge carriers occurs in the AlGaN layers, which is manifested in the absence of a large Stokes shift and in the realisation of optical amplification at transitions in an electron-hole plasma, and also indicates a relatively homogeneous material composition.**

Keywords: *optical pumping, ultraviolet stimulated emission, AlGaN epitaxial layers, ammonia molecular beam epitaxy.*

1. Introduction

AlGaN-based solid solutions are the main material from which most modern optoelectronic devices are made that operate in the UV spectral range and are in demand for applications such as disinfection, spectral analysis in medicine, biology, and criminalistics, atmospheric monitoring, etc., as well as devices of power and high frequency electronics. Despite the impressive results achieved in recent years in the field of making light-emitting and photo-receiving devices based on AlGaN for the UV spectral region [1 – 4], their characteristics are still far from those of similar InGaN-based devices operating in the visible spectrum. One of the main problems with the growth of AlGaN epitaxial layers is the low surface mobility of aluminium atoms, due to the relatively high energy of the AlN bond [5]. As a result, the growth of AlN or AlGaN in comparison with GaN has a more pronounced three-dimensional character with the formation of a set of islands with a higher density of grain boundaries and, accordingly, dislocations. The AlGaN layers in the composition of the device heterostructures, whose characteristics depend on the efficiency of charge carrier transport (light emitting diodes, photodetectors, transistors with high electron mobility), are often required to ensure a high composition uniformity and smooth surface morphology, which is a difficult task because of the AlGaN tendency to form a three-dimensional surface during growth. In addition, the smoothness of the heterointerfaces of quantum wells can also affect the magnitude of the Auger recombination coefficient [6].

In this work, we studied the radiative properties of $Al_xGa_{1-x}N$ layers of different composition, grown by the method of ammonia molecular beam epitaxy (MBE), and the main factors that determine the magnitude of the excitation threshold of stimulated emission in them.

2. Experiment

The epitaxial structures with AlGaN layers studied in this work were grown by the ammonia MBE method in the STE3N setup (SemiTEq, Russia) on sapphire substrates with a diameter of 50.8 mm and a roughness no worse than 0.2 nm. The pre-growth preparation of the substrate included annealing at a temperature of 1000 °C for 30 min and nitridation in an ammonia flow of 30 cm^3 min^{-1} under standard conditions and a temperature of 850 °C. The design of the heterostructures studied is schematically shown in Fig. 1. The epitaxial process began with the growth of the buffer structure with a total thickness of 460 nm, which included the seed and buffer layers of AlN, as well as the matching $Al_xGa_{1-x}N$ layer of variable composition. The growth of the AlN seed layer began at a low (0.05 μm h^{-1}) growth rate, which by the time of the beginning of the buffer layer growth increased to 0.2 μm h^{-1}. The temperature of the substrate and the flow of ammonia with the growth of AlN layers was 1085 °C and 100 cm^3 min^{-1}. As shown in Refs [7, 8], such conditions for the growth of AlN layers make it possible to ensure their good structural properties and a smooth surface morphology. The growth of the matching and active $Al_xGa_{1-x}N$ layers in different samples was carried out at a fixed flow of ammonia of 100 cm^3 min^{-1}, different temperatures (T_{gr} = 860 – 935 °C) and flow ratios x = Al/(Al + Ga) = 0.15 – 0.3.

We studied photoluminescence (PL) and stimulated emission (SE) in grown AlGaN layers under excitation by the 5th or 4th harmonic of an Nd : YAG laser [λ_{exc} = 213 nm and I_{exc} $\approx$ 0.1 MW cm^{-2} (PL), λ_{exc} = 266 nm and I_{exc} $\approx$ 0.1 – 10 MW cm^{-2} (SE)]. When measuring the parameters of SE, the radiation of the exciting laser was focused on the surface of the structure in a stripe with a size of 100 μm × 2 mm, ori-

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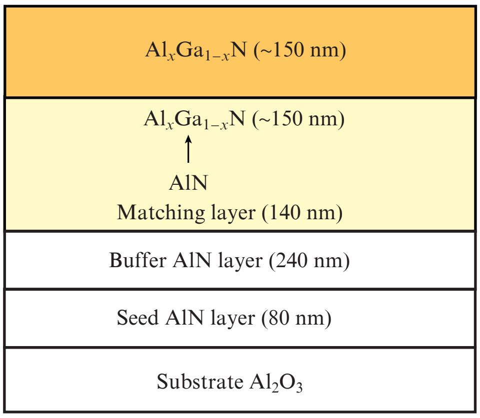


Figure 1. Construction of epitaxial structures with AlGaN layers.

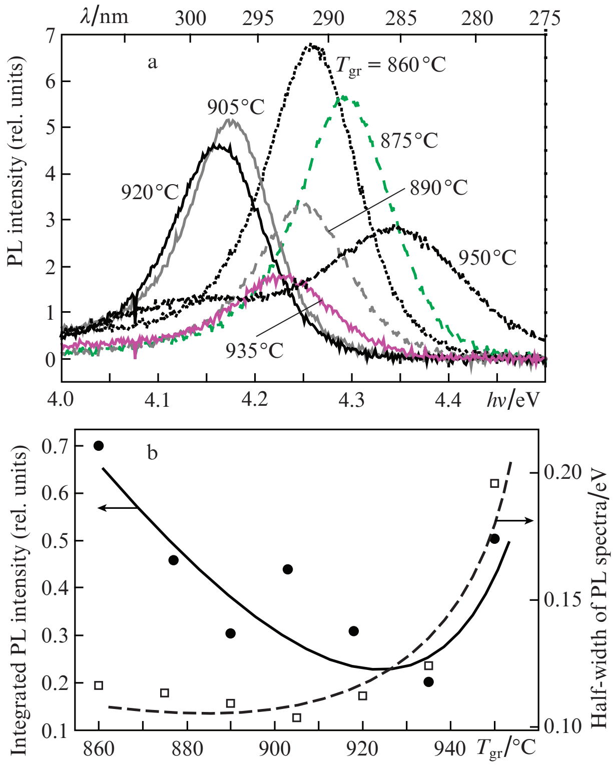


Figure 2. (a) PL spectra of the AlGaN layers grown at different temperatures and (b) dependences of the integrated intensity and half-width of the PL spectra on the growth temperature.

ented perpendicularly to the edge. The measurements were purposefully carried out without creating a Fabry–Perot resonator in order to exclude the accidental influence of the quality of the mirrors of the resonator formed by cleavages on the SE threshold. The PL and SE spectra were measured using a MayaPro spectrometer (Ocean Optics) from the surface and the end face of the AlGaN layer, respectively. To determine the absorption edge of AlGaN, the optical transmittance was measured using a Cary 500 spectrophotometer, and the surface morphology was studied using a Nanoflex atomic force microscope (AFM) (Solar LS).

3. Results and discussion

The SE threshold excitation intensity is a value sensitive to the quality of the waveguide boundaries. The scattering of radiation on the roughness of the boundaries can make a significant contribution to the amount of optical loss [9]. In lasers based on epitaxial structures, the waveguide boundaries are heterojunctions between layers of different composition and the semiconductor–air interface. Accordingly, to produce low-threshold semiconductor lasers, it is critical to control the influence of various growth parameters on the smoothness of heterointerfaces and surfaces of epitaxial structures.

To determine the radiative characteristics of AlGaN layers as functions of the temperature of their growth T_{gr}, we studied the PL in a series of AlGaN layers grown at T_{gr} in the range of 860–935 °C. The ratio of fluxes Al/(Al + Ga) for this series of layers was $x = 0.3$. The measurement results are shown in Fig. 2.

An increase in temperature from 920 to 935 °C leads to a significant increase in the half-width of the spectrum, which indicates the formation of inhomogeneities in the AlGaN composition. The non-monotonic nature of the PL intensity dependence on the growth temperature is obviously the result of the competition of two processes associated with an increase in the heterogeneity of the AlGaN composition. On the one hand, an increase in the inhomogeneity leads to an increase in the rate of radiative recombination due to an increase in the localisation of nonequilibrium charge carriers and the suppression of their transport to the nonradiative recombination centres. On the other hand, the formation of inhomogeneities can lead to a deterioration in the quality of the structure and, accordingly, to an increase in the concentration of centres of nonradiative recombination of the material due to the creation of local regions with high mechanical stresses.

For layers grown at T_{gr} = 860, 875 and 905 °C and having a small half-width of the PL spectra, the SE was obtained at $\lambda \approx 300$ nm at the threshold excitation intensities $I_{th} \approx 2$, 1.4, and 9 MW cm^{-2}, respectively. The SE spectrum and the dependence of its intensity on the intensity of exciting radiation for a layer grown at T_{gr} = 875 °C are shown in Fig. 3.

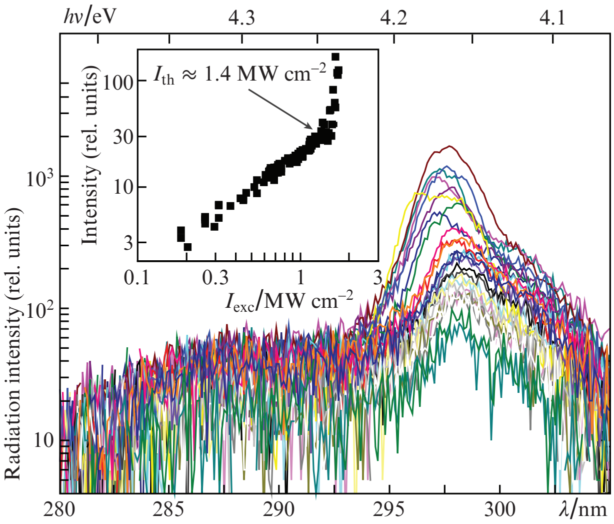
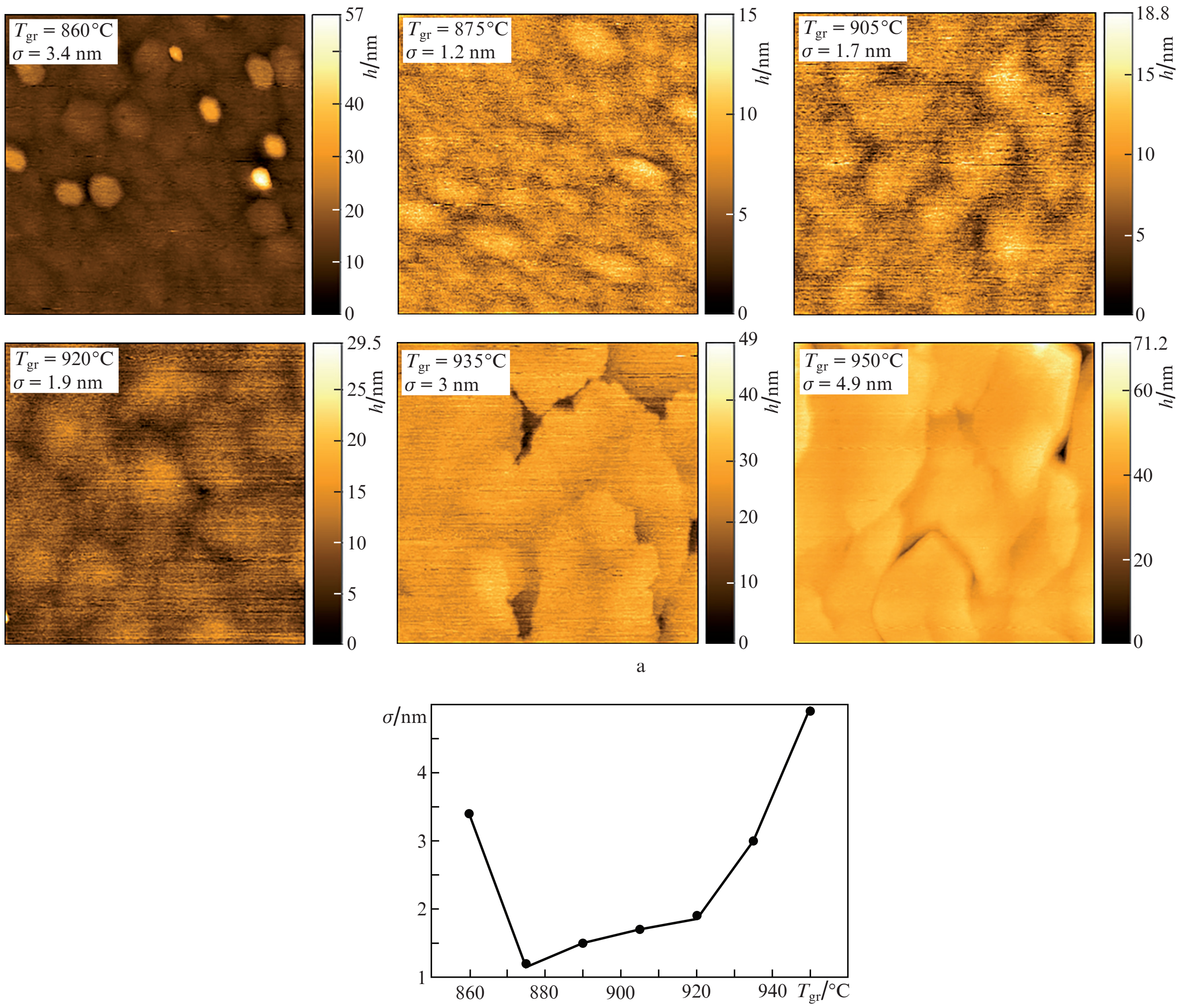


Figure 3. Spectra of emission from the end face of AlGaN grown at T_{gr} = 875 °C (the inset shows the dependence of the integrated radiation intensity on the intensity of the exciting radiation).



a

b

Figure 4. (a) AFM images of the surface areas of AlGaN layers with a size of 5×5 µm and (b) dependence of the mean square roughness σ on T_{gr}.

To assess the influence of the surface morphology of the layers on the SE, measurements were made using the AFM, the results of which are shown in Fig. 4. Relatively low values of mean square surface roughness σ are obtained for the range T_{gr} = 875–920 °C (the minimum σ = 1.2 nm is obtained for T_{gr} = 875 °C), which corresponds to the relatively small half-width of the PL spectra (see Fig. 2). The reason for the more pronounced formation of inhomogeneities of the AlGaN composition at high T_{gr} is the process of thermal decomposition of GaN, which at typical pressures in the growth chamber (10^{-4} – 10^{-3} Pa) turns out to be quite intense [10, 11]. This process leads to the formation of a rough surface of the growing layer with the formation of composition inhomogeneities and other structural defects in AlGaN, which can significantly impair its laser properties. It should be noted that for a layer with a minimum value of σ the threshold intensity of excitation of SE is minimal, and the reason for the absence of SE in layers grown at high temperatures is the formation of a rough surface of the layers. One of the reasons for its formation in the case of a layer grown at a low temperature (T_{gr} = 860 °C) may be an insufficiently high mobility of surface atoms during growth, contributing to the realisation of a three-dimensional growth mode.

Thus, we can conclude that the threshold excitation intensity of SE in AlGaN layers is largely determined by the surface morphology and, consequently, the magnitude of optical loss due to scattering, which depends on the intensity of thermal decomposition of GaN during growth.

The radiation properties of $Al_xGa_{1-x}N$ layers of different composition, grown under optimal conditions (T_{gr} = 875 °C, ammonia flow 100 cm^3 min^{-1}) were investigated with the values of the metal flux ratio Al/(Al + Ga)=0.3, 0.26, 0.21, and 0.15, which determined the molar fraction x of AlN. Figure 5 shows the PL characteristics of the obtained $Al_xGa_{1-x}N$ layers as functions of the composition. With an increase in the molar fraction of AlN, the half-width of the spectrum increases, which indicates a more pronounced formation of inhomogeneities in the AlGaN composition. According to the integrated intensity PL, the layers differ slightly, however, in the corresponding dependence, we can distinguish a minimum at x = 0.21. The increase in PL intensity with decreasing x from 0.21 to 0.15 is obviously associated with the improved material quality due to its composition approaching that of the GaN binary compound. One more reason is an increase in the coefficient of luminescence output from the layer due to an increase in the oscillator strengths of optical transitions with TE polarisation [12, 13]. An increase in the PL intensity with increasing x from 0.21 to 0.3 may be due to the increased localisation of non-equilibrium charge carriers and, accordingly, the inhibition of their transport to the nonradiative recombination centres due to composition inhomogeneities.

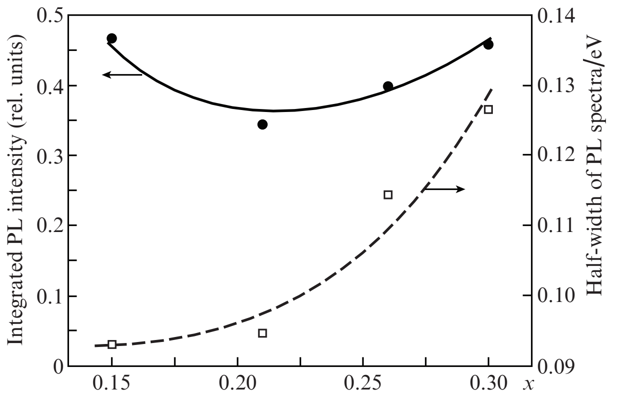


Figure 5. Integrated intensity and half-width of the PL spectra of $Al_xGa_{1-x}N$ layers grown at T_{gr} = 875 °C and an ammonia flow of 100 cm^3 min^{-1} as functions of the AlN mole fraction.

In all layers grown at T_{gr} = 875 °C, we obtained SE with I_{th} ≈ 0.7, 1.1, 1.4, and 1.4 MW cm^{-2} for x = 0.15, 0.21, 0.26 and 0.3, respectively. As one can see from Fig. 5, for the $Al_{0.15}Ga_{0.85}N$ layer with the smallest value I_{th} ≈ 0.7 MW cm^{-2}, the ratio of the integrated intensity to the half-width of the PL spectrum is maximal. The spectra of the SE and the threshold characteristic for this layer are shown in Fig. 6.

Figure 7 shows the PL and SE spectra for all layers of the series, as well as the energy values corresponding to the absorption edge determined from the transmission spectra [14]. The Stokes shift for all layers does not exceed 20 meV, which indicates a relatively weak localisation of non-equilibrium charge carriers. Moreover, for each of the samples, the SE band is shifted from the PL maximum to the long-wavelength side, which indicates the absence of pronounced band tails [15]. The main mechanism of stimulated emission in the layers under study appears to be optical gain at transitions in an electron-hole plasma, since the stimulated emission spectra are rather strongly shifted to the long-wavelength region relative to the absorption edge. Thus, we can conclude that the grown AlGaN layers have a relatively high homogeneity in composition, which is crucial for applications in heterostructures requiring efficient carrier transport (conductive layers of light-emitting and photo-receiving devices, barrier layers of transistors with high electron mobility, etc.) The relatively low SE threshold in AlGaN epitaxial layers without pronounced composition fluctuations (localised states) indicates their high quality.

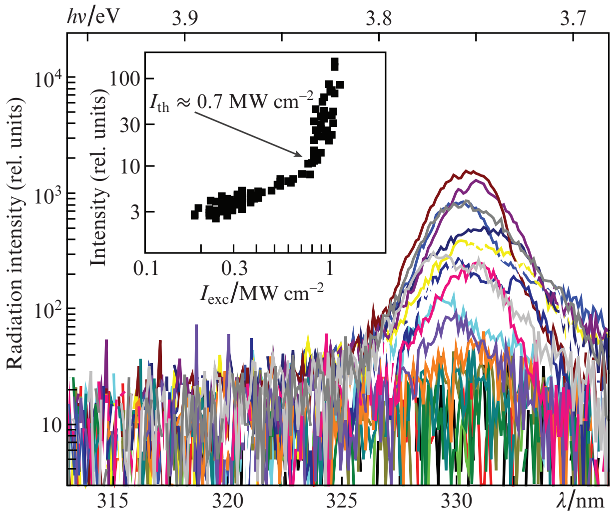


Figure 6. SE spectra in the $Al_{0.15}Ga_{0.85}N$ layer (the inset shows the dependence of the integrated radiation intensity on the intensity of the exciting radiation).

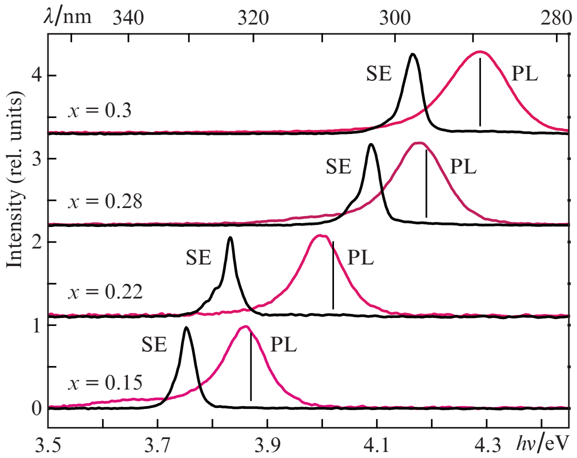


Figure 7. Normalised PL and SE spectra in $Al_xGa_{1-x}N$ layers of different composition. The vertical lines mark the energy positions of the absorption edge.

4. Conclusions

It was shown that the threshold of SE excitation in AlGaN layers grown using the ammonia MBE method is largely determined by the intensity of the process of thermal decomposition of GaN, which affects the surface morphology and, therefore, the optical scattering loss. The growth conditions were determined, which allowed fabrication of a series of $Al_xGa_{1-x}N$ layers with a thickness of 150 nm with different composition (x = 0.15, 0.21, 0.26, and 0.3), in which stimulated emission in the range λ = 330 – 297 nm with relatively low values of threshold excitation intensity I_{th} ≈ 0.7 – 1.4 MW cm^{-2} was obtained. It is shown that in these $Al_xGa_{1-x}N$ layers there is no strongly pronounced localisation of nonequilibrium charge carriers, which indicates homogeneity of their composition.

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