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Effect of growth temperature on photoluminescence properties of NH_3-MBE grown GaN-on-Si layers

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Abstract. In this paper, we present the results of the investigation of GaN-on-Si layers grown by the ammonia MBE technique within the technologically acceptable temperature range (775 °C–825 °C) by photoluminescence technique. The lowest value of the concentration of defects was obtained at a growth temperature of 825 °C. The increase in the concentration of defects in the film with decreasing growth temperature can be explained by the deviation from the optimum growth temperature and consequently by the deterioration of the crystalline perfection of the GaN layers.

Keywords: GaN, ammonia-MBE, photoluminescence, heterostructures, point defects

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Влияние температуры роста на фотолюминесцентные свойства слоев GaN-on-Si, выращенных методом NH_3-МЛЭ

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Аннотация. В данной работе представлены результаты исследования слоев GaN-on-Si, выращенных методом аммиачной МЛЭ в диапазоне температур 775–825 °C методом фотолюминесценции. Наименьшее значение концентрации дефектов было получено при температуре роста 825 °C. Увеличение концентрации дефектов в пленке с понижением температуры роста можно объяснить отклонением от оптимальной температуры роста и, как следствие, ухудшением кристаллического совершенства слоев GaN.

Ключевые слова: GaN, аммиачная МЛЭ, фотолюминесценция, гетероструктуры, точечные дефекты

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Introduction

In recent years, the market for high-power, ultrahigh-frequency electronics has seen an increase in the proportion of high electron mobility transistors (HEMTs) based on AlGaN/GaN heterostructures, which are successfully competing with similar devices based on Si and GaAs due to the large band gap [1]. High values of spontaneous and piezoelectric polarization in AlGaN/GaN heterostructures provide a high concentration of two-dimensional electron gas (2DEG) near the heterojunction $n_e > 10^{13}$ cm^{-2}. The high carrier density in HEMTs enables high current densities, which in combination with high breakdown voltage and high thermal conductivity contribute to increased device performance. In addition, AlGaN/GaN HEMTs can operate at higher temperatures, and the high binding energy of the gallium and nitrogen atoms significantly increases resistance to ionizing radiation. Despite the significant achievements of III-nitride technology, it should be noted that the problems of reducing power dissipation and improving the stability and reliability of devices are still relevant. High parameters of devices based on III-nitride materials have been predicted theoretically, but the practical implementation of AlGaN/GaN transistor and diode structures reveals a number of problems: high threshold voltage, current non-linearity, the manifestation of current saturation at unexpectedly low voltages and the limitation of the voltage variation range, the effect of current collapse. In particular, the problems of increasing breakdown voltages and reducing various types of leakage currents are acute [2, 3], so the possibilities of overcoming them are being actively investigated.

It is well known that leakage currents in the undoped GaN layer are caused by a high background concentration of free carriers (mainly electrons). The main background impurities from the residual atmosphere are oxygen and carbon, which lead to unintentional doping of the GaN buffer layer and the appearance of n-type and p-type conductivity, respectively [4] Intentional doping of GaN with acceptors (C, Fe) is a common method to compensate for the uncontrolled incorporation of donors to obtain high resistivity GaN layers [5, 6]. However, donor compensation with carbon significantly degrades the structural properties of GaN, in particular the surface morphology of GaN layers [7], which negatively affects the electrophysical parameters of 2DEG. When GaN buffer layers are doped with iron, a ‘memory effect’ is observed [8]. This effect leads to unintentional doping of all layers of the heterostructure, including the channel of the transistor structure. Another ambiguous approach, often implemented in the metal organic chemical vapor deposition (MOCVD) technique, is based on the intentional reduction of the structural perfection of the initial GaN buffer layers. On the one hand, this approach leads to an increase in the number of edge dislocations in the GaN layers, which results in an increase in the number of deep acceptor levels [9] to which electrons from donor levels are trapped. However, an increase in dislocation density inevitably leads to deterioration in the electrophysical properties of 2DEG and, as mentioned above, threading dislocations can be sources of electrons [10]. A number of papers report the possibility of controlling the background doping with carbon and oxygen [11, 12] by changing the growth conditions. Obviously, the growth conditions also influence both the concentration of point defects in the GaN layers and the dislocation density in the grown heterostructures. From a technological perspective, it is important to achieve a high resistivity of the GaN buffer layer in order to reduce leakage currents. Ammonia molecular beam epitaxy (NH_3-MBE) technique is characterized by lower growth temperatures compared MOCVD and allows obtaining sharp heterojunctions that is important at growth of heterostructures with binary AlGaN barrier [13, 14]. Plasma MBE technique needs to maintain a slight metal enrichment (2 monolayers) on the surface of the growing film during the growth, which inevitably leads to a local violation of the stoichiometric conditions and formation of metal drops on the surface, this effect is not present during ammonia MBE growth.The aim of this work is to investigate the

effect of growth temperature on the concentration of defects, including background impurities and donor-like defects, in GaN-on-Si layers grown by NH_3-MBE technique.

Materials and Methods

The investigated samples were grown on the NH_3-MBE Compact-21N machine on two-inch high-resistance Si (111) wafers. After the nitridation process, a 50 nm thick AlN nucleation layer was grown, followed by a buffer layer consisting of 160 nm AlN, 160 nm $Al_{0.6}Ga_{0.4}N$ and 160 nm $Al_{0.3}Ga_{0.7}N$. In the final stage of growth of GaN-on-Si heterostructures, 1000 nm thick GaN layers were grown at selected temperatures (775 °C, 800 °C and 825 °C) in a 200 sccm ammonia flow at a growth rate of about 600 nm/h (*BEEP*(Ga) = $1.3 \cdot 10^{-6}$ Torr). In the following text the samples of the investigated series are referred to as 'GaN-775', 'GaN-800', 'GaN-825'. For steady-state PL excitation, the He-Cd laser with λ_{las} = 325 nm was used. The output excitation power of the He-Cd laser was about of 10 mW. The N_2 laser with λ_{las} = 337 nm, the repetition frequency 1 kHz and the pulse duration 7 ns was used to measure PL decay curves. The output average excitation power of the N_2 laser was about of 27 mW. PL was investigated by double monochromator SDL-1 with a cooled FEU-79 multiplier operating in photon counting mode. The time-correlated photon counting method was used to measure the PL decay curves in the range of delay times 0—100 μs. The measurements were performed at room temperature.

Results and Discussion

Fig. 1, *a* shows the steady-state photoluminescence spectra of GaN layers at room temperature. For GaN-on-Si, due to the large difference between the refractive indices of GaN and Si, the effect of light interference on the recorded spectrum leads to a periodic modulation of the emission intensity. The measured PL spectra consist of two bands: thenear-band-edge PL band in the UV spectral region with an intensity maximum position of about 3.4 eV (NBE) and the yellow defect-related band (YB) with an intensity maximum position of about 2.2 eV. In the spectra, there is a weak blue band at 2.8—3.0 eV, also due to defects. Lateral tensile stresses of GaN-on-Si thin epitaxial layers in the (0001) plane leads to a decrease of the band gap and as a consequence to the NBE redshift compared to undeformed GaN. TheNBE band for GaN-800 has the highest red shift that indicates the largest value of tensile stresses in this structure. The shape of the edge PL band of the studied structures is asymmetric, and contains two additional peaks at a distance of ~70 meV and ~140 from the main peak, which are associated with LO phonon replicas of the free exciton (FE) band [15]. For all investigated structures, the intensities of the first and second LO phonon replicas are 5 and 20 times lower than the intensity of the FE peak, respectively. The presence of lines associated with LO phonon replicas shows crystalline perfection of GaN-on-Si layers [16]. Fig. 1, *b* shows the decay curves of the yellow PL band after pulse excitation, measured at room temperature at registration energy of 2.175 eV, close to the position of the yellow PL band intensity maximum. The intense peak in the 0.12—0.2 μs time range corresponds to the registration of the laser pulse. Therefore, t_0 = 0.2 μs should be taken as the starting point of the decay time. The peak in the 25—40 μs time range is due to inductions in the PL photon registration system. The PL intensity of all structures is decreased for more than 100 μs, approximation by the function $I(t) \sim t^{\gamma}$ gives an exponent of degree $\gamma < 1$. The value of $\gamma = 0.55$ is highest for the GaN-775 structure, average $\gamma = 0.46$ for the GaN-825 structure and lowest $\gamma = 0.35$ for the GaN-800 structure. Exponential PL decay law is expected for the electron transition from the conduction band to the acceptor level, while long non-exponential PL decay is indicative of donor-acceptor type transitions.

The yellow and blue bands are usually associated with the carbon impurity (C_N) [17, 18]: the yellow YL_1 band with a maximum position at 2.17 eV is due to the electron transition from the conduction band to the (−1/0) CN level, the blue BLC band with a maximum position at 2.85 eV is due to the electron transition from the conduction band to the (0/+1) C_N level. It is believed that PL bands caused by defects provide information about crystal defects and are actively used to improve the technology of their growth. Since the ratio of the intensities of these bands to the intensity of the NBE in the PL spectrum of GaN can be proportional to the defect concentration, it can be used as a criterion for the crystalline quality of the GaN. The intensity of the yellow band is approximately the same for all structures of the investigated series, with the maximum value of the yellow band intensity for the structure GaN-775 and the minimum value for the

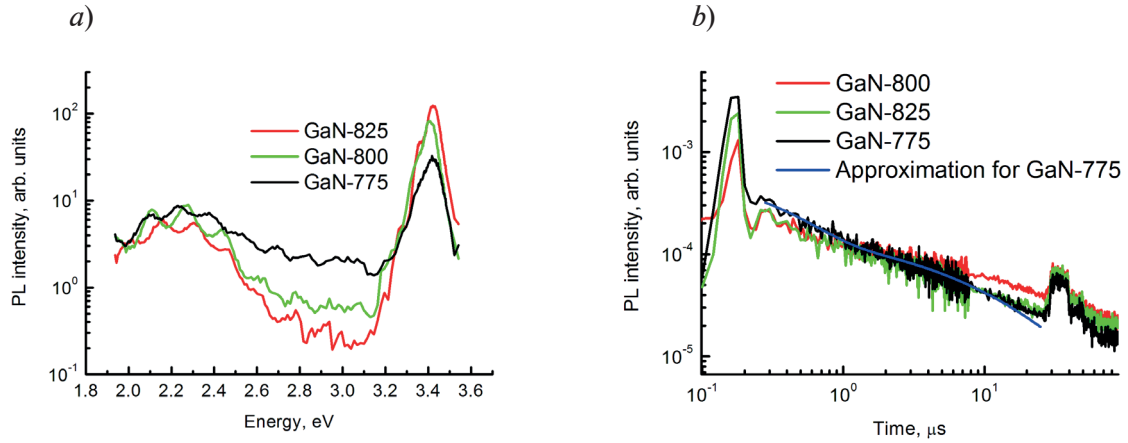

Fig. 1. Photoluminescence spectra (*a*) and decay curves at registration energy of 2.175 eV with the example of approximation (see the text for details) (*b*) of GaN layers measured at room temperature

structure GaN-825. A similar ratio is observed for the blue band: the GaN-775 structure has the maximum intensity of the blue band; the GaN-800 structure has a much lower intensity of the blue band, and in the spectrum of the GaN-825 structure the blue band almost merges with the background. The intensity of the NBE band is inversely correlated with the intensity of the defect bands: it is minimal for the GaN-775 structure and maximal for the GaN-825 structure. However the intensity of the NBE photoluminescence at a given temperature and excitation power depends on the concentrations and capture cross sections not only of the radiative recombination centers but also on the different centers of non-radiative recombination and their charge states, which are determined by the position of the Fermi level. In other words, at high acceptor doping levels, the ratio of defect PL band intensities to NBE PL intensities is very low, which is explained by the low concentration of non-radiative recombination centers. Consequently, among the structures studied in this work, the GaN-825 structure has the lowest concentration of non-radiative recombination centers, while the GaN-775 structure has the highest concentration of non-radiative recombination centers.

The dependence of the non-exponential PL intensity on time (t) and radiation energy (E) is well described by the deep center donor-acceptor recombination model developed in [19]:

$$I(E,t)=\int_0^{\infty} I_E(t)\exp\left(\frac{E'-E-E_{em}}{2\Delta^2}\right)dE', \tag{1}$$

$$I_E(t)=\frac{4\pi N_D}{E^4}\left(\frac{e^2}{\varepsilon}\right)^3 W_{\max}\exp\left(-\frac{2e^2}{\varepsilon E a_d}-W_{\max}t\exp\left(-\frac{2e^2}{\varepsilon E a_d}\right)\right)\times$$
$$\times\exp\left(4\pi N_D\int_0^{\infty}\left(\exp\left(-W(r)t\right)-1\right)r^2dr\right), \tag{2}$$

$$W_R(r)=W_{\max}\exp\left(-2\frac{r}{a_d}\right), \tag{3}$$

here the shape of the PL band of a single donor-acceptor pair is assumed to be Gaussian which is true for the configuration coordinate model for values of the Huang-Rhys parameter $S >> 1$, ad is the donor's Bohr radius, the value of the effective electron mass in GaN is $m^* = 0.2$ and the dielectric permittivity is $\varepsilon = 9.5$, r is the distance between donor and acceptor, $W_R(r)$ is the probability of radiative transition per unit time, W_{max} is the pre-exponential factor, N_D is the concentration of the neutral donors (the dominant defect component).

The approximation of the experimentally measured decay curves by the theoretical dependence at a fixed value $W_{max} = 2\cdot10^6$ s^{-1} [20] allows to estimate the N_D values for all the structures studied. The concentration of neutral donors of about $6\cdot10^{17}$ cm^{-3} is highest for the GaN-775 structure, average of about $3\cdot10^{17}$ cm^{-3} for the GaN-825 structure and lowest for the GaN-800 structure of

about $1 \cdot 10^{17}$ cm^{-3}. Since the grown structures are significantly compensated by radiative and non-radiative defects, the obtained value is much lower than the total donor concentration (N_D^{total}). According to the well-known expression

$$N_D / N_D^{total} = \left[1 + \frac{g_1}{g_0} \exp\left(- \frac{F - E_d}{kT} \right) \right]^{-1}, \quad (7)$$

Here F is the position of the Fermi level below the conduction band, g_0 and g_1 are degeneracy multiplicities of empty and filled charge states. At sufficiently high temperature and high defect concentration, the Fermi level is below the donor level, so the ratio is $N_D/N_D^{total} << 1$. Since the GaN-775 structureis characterized by a highest defect concentration, and consequently the lowest F the ratio N_D/N_D^{total} is expected to be the smallest for this structure. Therefore, the highest N_D value means the highest N_D^{total} value for the GaN-775 structure. At the same time, it cannot be stated that the GaN-800 structure has the lowest donor concentration since the GaN-825 structure is characterized by a lowest defect concentration and has the highest ratio N_D/N_D^{total}. So the lowest N_D^{total} can be in the GaN-825 structure.

Conclusion

Based on the above results of the study by the PL method of a series of GaN-on-Si samples grown by the NH_3-MBE technique at different growth temperatures a number of conclusions can be drawn. The growth of GaN-on-Si layers at 825 °C allows obtaining GaN layers with the lowest concentration of non-radiative defects as evidenced by the lowest intensity of the defect-related PL bands, the concentration of donors is also low. Decreasing the growth temperature of GaN-on-Si layers leads to an increase in the concentration of non-radiative defects. The increase in the number of defects in the film with decreasing growth temperature can be explained by the deviation from the optimum growth temperature and consequently by the deterioration of the crystalline perfection of the GaN layers. The growth of GaN-on-Si layers at 775 °C is characterizes by the highest concentration of donors.

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