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# Creation of effective sources of white radiation based on GaP(As,N) on silicon substrates

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Abstract. The article is devoted to the study of light-emitting heterostructures based on GaP(As,N) dilute nitrides, monolithically grown on silicon substrates by plasma-assisted molecular beam epitaxy. Current-voltage characteristics and electroluminescence spectra of the grown heterostructures are obtained. For the first time, a unique effect is observed in GaP(As,N) dilute nitrides - the appearance of white electroluminescence when a reverse bias is applied. The result was obtained due to the original design of the light-emitting heterostructure and the unique properties of dilute nitride solid solutions.

#### 1. Introduction

An important task of modern physics is to find new ways to create sources of white radiation that can be used as effective light sources not only in display systems, backlighting or signaling equipment, but also for general lighting. Nowadays there are several approaches for creating white light sources. The first approach is a combination of an aluminum gallium nitride (AlGaN) semiconductor LED that emits blue light and a YAG luminophore that absorbs blue light and emits yellow. Despite the fact that the described technology has revolutionized the lighting industry, the materials used, for example, yttrium in the luminophore, are quite rare and expensive. In addition, the absorption of shortwave radiation by a fluorescent material and conversation it into longer wavelength radiation in the visible range leads to energy losses associated with a small Stokes shift of the luminophore [1].

The second approach involves placing three separate red, green and blue light-emitting diodes (LEDs) dies in a common enclosure. Combining these three colors results in a white glow. To date, this method is not effective for a number of applications due to the spectral temporal instability of white radiation associated with different degradation rate of each source in the common enclosure [2], insufficiently good thermal stability, which is an extremely important parameter for power applications [3]. In addition, due to the different characteristics of each radiation source, it is necessary to create separate control circuits for each color, which invariably leads to an increase in economic, time and technological costs for their creation [4].

The third approach involves manufacturing a white LED from a variable-gap material, i.e. containing at least two band gaps with different energies. A structure, containing a multiple quantum wells (MQWs), emitting at different wavelengths in the visible range was implemented [5]. Unfortunately, this approach also has disadvantages - this structure does not emit at all wavelengths of the visible range and does not demonstrate required emission spectrum.

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In addition to the above problems, there is a significant problem for all of the above approaches associated with the need to create a balance between the emission intensities of the red, blue and green emission bands. Recently [6] a new hybrid design of core-shell InGaN/GaN nanorods was demonstrated, which solves the problem of insufficiently intense of red emission band. The hybrid design of the InGaN/GaN nanorods allows to convert the excess blue emission to red emission.

Of great interest is the creation of sources of white radiation specifically on silicon. This is due to the low cost of silicon substrates, advanced silicon technology, and good thermal and mechanical properties. Many attempts have been made to create silicon-based white LEDs, for example, using silicon nanoparticles [7,8] and quantum dots [9], as well as by combining silicon with organic materials [9]. However, the result was either no white radiation, or very low efficiency, or short service life. Thus, in the world community, there is an increasing interest in the described topic.

# 2. Experiment

The article is devoted to the study of light-emitting heterostructures based on GaP(As,N) superlattices (SLs). Heterostructures were monolithically grown on silicon substrates using unique technology – plasma-assisted molecular beam epitaxy (PA-MBE). The advantages of the MBE method for growing heterostructures, containing superlattices are, the use of high purity materials, the availability of precision growth control methods, and ultrahigh vacuum during synthesis, which ensures high quality structures. In addition, the possibility of a sharp interruption and subsequent resumption of molecular beam arrival at the substrate makes it possible to create sharp heterointerfaces, which is important for the creation of nanoheterostructures, and the high temperature stability of the sources ensures the constancy of the composition.

Epitaxial growth on silicon is associated with a number of problems. One of them is the formation of antiphase domains due to the growth of the GaP(As,N) polar material on the nonpolar Si substrate. To solve this problem silicon substrates with  $4^0$  misorientated relative to the (001) surface in the [110] direction are used.

Preliminary cleaning of silicon substrates is one of the most important stages in the MBE technology for the formation of silicon heterostructures. Before epitaxial growth, Si substrates underwent a cycle of chemical treatment according to method [10].

In the in-situ procedure of pre-epitaxial preparation, the stage of low-temperature degassing is mandatory, since, in this case, a significant part of the physisorbed impurities fly away from the surface as a result of thermal desorption and do not form hard-to-remove compounds due to chemical interaction with silicon. For this the substrates were first heated in a high vacuum up to a temperature of  $350\pm50^{\circ}$ C in a pre-growth chamber, and then to a temperature of  $850\pm50^{\circ}$ C in the MBE epitaxial reactor.

The samples contained GaP and GaP/GaPN buffer layers to improve the layer morphology and transition from the silicon substrate to the active layers of the heterostructure. The active region of the light-emitting heterostructure consisted of 15 periods of superlattice (GaAsPN/GaPN). The period thickness was 14 nm. The GaP contact layers were doped with silicon and beryllium [11].

Metal contacts were formed on the surface of the heterostructure to obtain current-voltage (I-V) characteristics and electroluminescence (EL) spectra

## 3. Results and discussion

The current-voltage characteristics were obtained at room temperature. The reverse brunch of the I-V characteristic at room temperature is shown in figure 1.

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**Figure 1.** Reverse branch of the *I-V* characteristic of the light-emitting heterostructure obtained at 300 K

Up to voltage  $U_0 \sim -14$ V, no breakdown of the structure is observed. However, with a further increase in the reverse voltage, the I-V characteristic of the LED structure resembles the I-V characteristic of a Zener diode, which operates in a stable breakdown mode under conditions of the reverse bias of the p-n junction. By the high value of  $U_0$  it can be concluded that the avalanche mechanism of an increase in free charge carriers prevails. Thermal breakdown of the diode occurs at  $U_{BR} \sim -40$ V.

Further, the EL spectra were obtained at a temperature of 133 K. The studies were carried out at various values of voltage and current flowing through the heterostructure. The obtained data are shown in figure 2.



**Figure 2.** Electroluminescence spectra of a light-emitting heterostructure obtained at various voltages and currents flowing through the heterostructure at a temperature of 133 K

At a voltage lower than  $U_0$ , EL is not observed. However, as the reverse bias increases to -14.5 V, an EL peak with a length of 600 nm appears. With a further increase in the reverse bias, the EL peak broadens and additional peaks appear in the region of 500 nm and 700 nm. Thus, the electroluminescence spectrum covers the entire visible wavelength range and white EL is observed. However, it should be noted that there is an imbalance between the emission components with a predominance of narrow emission at 600 nm. As the reverse bias increases to more than -18 V, the intensity of the 600 nm peak decreases, while the intensity of 500 nm peak increases. However, the imbalance is still observed. This problem will need to be solved in the future by optimizing the LED design.

Previously, such an unusual effect of the appearance of white EL under reverse bias was observed in nitride LEDs and was explained by transitions between impurity levels [12,13], however, it was not used in devices because of its low efficiency compared to interband EL.

In dilute nitrides this effect is observed for the first time and can be is explained by the emergence of defects in dilute nitrides associated with the presence of nitrogen. The appearance of defects in dilute nitrides is associated with two main features. The first - is the use of plasma source of nitrogen during MBE. Molecular nitrogen is supplied to the source input, inside the source part of the  $N_2$  molecules is converted into atomic nitrogen, and a set of different particles is obtained at the source output: molecular nitrogen, atomic nitrogen and nitrogen ions. Molecular nitrogen does not interact with the epitaxial surface, atomic nitrogen is incorporated into the semiconductor layer being grown, and nitrogen ions "bombard" the surface of the growing film and create defects in it. To improve the crystalline and optical quality of the growing GaP(As,N) layer, it is necessary to choose the optimal operating conditions for the plasma source. With a decrease in the flow of molecular nitrogen entering the tube and an increase in the power supplied to the gas discharge region, the intensity of radiative recombination of the resulting heterostructure increases [14]. These process conditions were used for the epitaxial growth of the studied heterostructures.

The second feature is associated with the high electronegativity of nitrogen atoms and the small size of atoms compared to other elements of the V group. Thus, nitrogen atoms are built unevenly into the crystal lattice, nitrogen pairs of various configurations, triplets and chains of nitrogen atoms are formed. This leads to a disturbance of the translational symmetry of the crystal, as well as to the appearance of localized states. The broadening of the emission band can be associated with the contribution of the nitrogen pairs from the side of higher energies and their phonon replicas from the side of lower energies.

## 4. Conclusion

The article is devoted to the study of light-emitting heterostructures based on GaP(As,N) dilute nitrides, monolithically grown on silicon substrates by PA-MBE. *I-V* characteristics and EL spectra of the grown heterostructures are obtained. For the first time, a unique effect is observed in dilute nitrides - the appearance of white EL when a reverse bias is applied. This effect is associated with the presence of defects in the heterostructure, which arise due to the peculiarities of the technological process and the nonequilibrium properties of nitrogen atoms.

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