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Competing processes in nitride alloys in MQWs of LEDs

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Abstract The nature of competing processes leading to narrowing and broadening of the electroluminescence spectra width at half maximum under the injection current in nitride LEDs emitting at wavelengths of 270–280 nm and 530–540 nm has been studied. It was found out that there may be local regions with disturbed stoichiometry of random alloy fluctuations, enriched in excited defects ("deep center + local vibrations") in nitride MQWs with random alloy fluctuations. The capture of injected charge carriers by such defects in MQWs located in the space charge region of the pn junction was shown to lead to their coordination rearrangement in the lattice. This also results in a more equilibrium state of random alloy fluctuations and is accompanied by a narrowing of the full width at half maximum electroluminescence spectrum. However, this mechanism is a source of carrier loss that reduces the external quantum efficiency of LEDs at the maximum. It was shown experimentally that the higher the level of disorder in random alloy fluctuations, the lower the external quantum efficiency values. The non-equilibrium filling of the lateral random allow fluctuation by charge carriers in MQWs located outside the space charge region causes the broadening of the full width at half maximum electroluminescence spectrum in LEDs. This mechanism leads to a drop in external quantum efficiency at current densities $j > 30$ A/cm^2.

Keywords: MQW LEDs, alloy level disorder, FWHM, random alloy fluctuation

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Конкурирующие процессы в нитридных твёрдых растворах светодиодов с множественными квантовыми ямами

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Аннотация. Исследована природа конкурирующих процессов, приводящих к сужению и уширению ширины спектров электролюминесценции на полувысоте под действием тока в нитридных светодиодах, излучающих на длинах волн 270–280 нм и 530–540 нм. Выяснено, что в нитридных с множественных квантовых ямах (МКЯ), наряду со случайными флуктуациями состава, могут присутствовать локальные области с нарушенной стехиометрией состава твердого раствора, обогащенные возбужденными дефектами. Показано, что захват инжектируемых носителей заряда такими дефектами в МКЯ, находящихся в области объемного заряда *p–n* перехода, приводит к их координационной перестройке в решетке, а также к более равновесному состоянию твердого раствора и сопровождается сужением ширины спектра на половине высоты. Однако этот процесс является источником потерь носителей заряда, снижающим внешнюю квантовую эффективность светодиодов в максимуме. Экспериментально показано, что чем выше уровень разупорядоченности твердого раствора, тем ниже значения внешней квантовой эффективности. Увеличение ширины спектра светодиодов на половине высоты вызвано неравновесным заполнением латеральных неоднородностей состава твердого раствора носителями заряда в МКЯ, находящихся вне области объемного заряда. Этот процесс вызывает падение внешней квантовой эффективности при плотностях тока $j > 30$ А/см^2.

Ключевые слова: светодиоды с квантово-размерной активной областью, разупорядоченность твердого раствора, ширина спектра на полувысоте, случайные флуктуации состава

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Introduction

It is mentioned in the latest review [1] that insufficiently studied features of nitride-based alloy disorder prevent the realization of potential in the creation of emitting devices with high external quantum efficiency (EQE) in ultraviolet and green spectral range. The disorder can lead to the existence of local areas with impaired stoichiometry. According to [2], such phenomena as a narrowing spectra full width at half maximum (FWHM), as well as a FWHM broadening in electroluminescence (EL) spectra in QWs of LEDs under the injection current can also decrease EQE values [2]. However, the mechanisms causing these phenomena in MQWs and alloy disorder are understood relatively poorly [1, 3]. Our study is aimed at revealing reasons for these phenomena.

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Materials and Methods

In this work, we study commercial green and UV LEDs. We also investigated blue LEDs as a reference. These LEDs have 1 mmI active area and flip-chip design. The external quantum efficiency (EQE) of green LEDs is EQE ranging from 13% to 30% at wavelengths 530 nm. The lowest EQE values (3–7%) are observed in UV LEDs at 270–280 nm. Referent blue LEDs emitting at 450 nm exhibit EQE ranging from 50% to 70%. Current-voltage (*I–V*) characteristics of LEDs were obtained using a KEITHLEY 6487 power source. EL spectra and EQE measurements were conducted on LEDs at 300K, employing an Optronic Laboratories OL770-LED system with an OL ISA-670 integrating sphere. We carried out a joint analysis of changes in FWHM values and the dependence of EQE versus the injection current of these LEDs, with additional near-field distribution of EL parameters. Near-field was monitored by Mitutoyo optical microscope with Avantes AvaSpec-2048. The field of view of the optical system was 536×357 μm^2 with the best spatial resolution of 25 μm.

To assess the level of disorder in nitride alloy of LEDs (alloy LD), we used the value Δ_V which evaluates the difference between the values of the threshold voltage (u_{th}) determined from the direct branch of the *I–V* characteristics and the voltage corresponding to the peak wavelength of the study LEDs at the maximum (u_0), $\Delta_V = u_{th}$-u_0 [3, 4]. Previous studies have shown a good correspondence of this value with other techniques to estimate the alloy LD in LEDs [3]. Moreover, they found a correlation between a decrease in the values of Δ_V and the improvement in ordering. All the LEDs under the study were classified according to the parameter $\Delta_{V.}$

Results and discussion

A study of the spectrum of green (λ_{peak} = 530 nm) LEDs with 30% EQE in the near field at 0.2 mA and 50 mA showed that at a low injection level, local areas intensively emit with a different peak wavelength with exceed the maximum on 4 nm of LED radiation. As the injection current increases, the radiation intensity of these regions does not increase, which suggests that there is some disturbed of the stoichiometry of nitride allow in these areas. At the same time, the value of Δ_V in these LEDs significantly exceeds the value of Δ_V in blue LEDs, with a relatively low EQE of 50%. Fig. 1 shows the experimental dependencies of FWHM and EQE versus the injection current in green LEDs with different alloy LD and referent blue LEDs.

There is a correlation between decreasing EQE at maximum in MQWs located in the space charge region (SCR) around *p–n* junction (curves 1 on all dependencies with decreasing values of Δ_V). It should be noted that in blue LEDs with 70% EQE, Δ_V does not exceed 0.1 V, while in green LEDs with 13% EQE, Δ_V is 1 V. At the same time, there is a narrowing of the FWHM EL spectra under the injection current in MQWs, located in the SCR of the *p–n* junction, of all LEDs (Fig. 1, curves *2*). This effect is most pronounced for LEDs with a maximum value of Δ_V and a minimum EQE (Fig. 1, *a*). It is clearly visible in Fig. 1, *a* that this process is accompanied by an increase in EQE under injection current curve *2*, which indicates a transition to a more equilibrium composition in the solid solution. Narrowing of the FWHM EL spectra under the injection current reveals complex mechanisms of capture and recombination of charge carriers in MQWs based on nitrides [5, 6]. Such complex processes have been studied in gallium arsenide-based solid solutions by representations of excited defects, such as "deep center + local vibrations" [5].

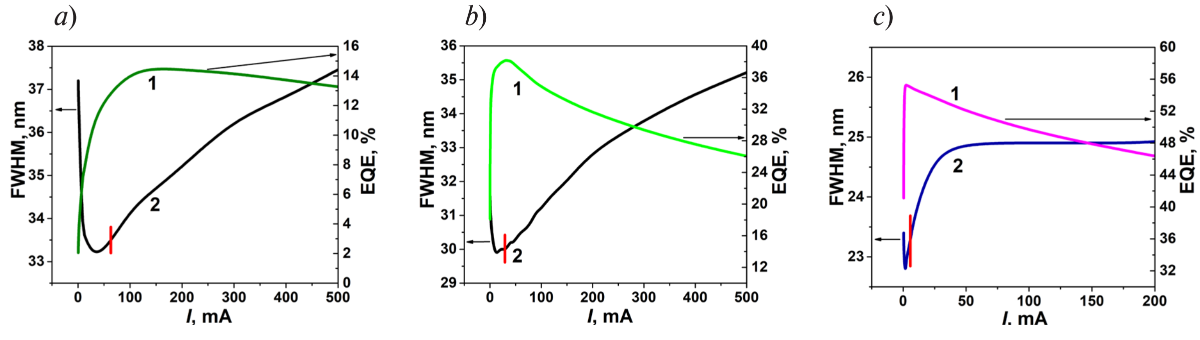

Fig. 1. Dependences of EQE (*1*) and FWHM (*2*) versus injection current in LEDs with different alloy LD and EQE: green 0.72V, 14% (*a*); green 0.46V, 30% (*b*); 0.13 V, 50% (*c*)
Red lines mark the FWHM corresponding the current when the *p–n* junction is full open

The possibility of the existence of such defects is due to fundamental properties of A3B5 such as the excess elastic energy of deformed bonds of main components in the lattice, which are characterized by the Huang-Rhys factor. It should be noted that the value of the Huang-Rhys factor is higher in nitrides than in arsenides [6]. According to theoretical concepts [6], the capture of injected charge carriers by such defects in MQWs located in the SCR region around pn junction leads to their coordination rearrangement in the lattice and multiphonon defect recombination. It seems that local areas with impaired stoichiometry are enriched by such defects. The observed narrowing of the FWHM EL spectra under the injection current in MQWs reflects the mechanism of transition of a nitride alloy in such areas to a more equilibrium state. However, this process is a source of carrier loss, reducing the EQE of LEDs to the maximum. The results presented in Fig. 1 show that the higher the alloy LD, the lower the EQE values. Thus, the presence of local areas with impaired stoichiometry is one of the reasons behind the low EQE in green LEDs. It seems that this conclusion is confirmed by the study of the optical properties of UV LEDs. Indeed, the estimated Δ_V in UV LEDs shows values of 1.5 V, i.e., the highest among all the studied LEDs, and the lowest EQE ~ 4–7%.

Fig. 2 shows EQE (*1*) and FWHM (*2*) dependencies versus injection current (Fig. 2, *a*) and EL spectra (Fig. 2, *b*) in UV LEDs. A more complex FWHM dependence (curve *2*) versus injection current than the LEDs shown in Fig. 1 is observed. It consists of two regions: Δ_V = 0.13 V and Δ_V = 1.5 V. Analysis of the EL spectra in UV LEDs in Fig. 2, *b* allows us to understand the reasons behind this complexity. The typical EL spectrum in UV LEDs in Fig. 2, *b* (curve *1*) at 1 mA demonstrates the phase separation of the alloy in the region 240–350 nm. At the same time, a narrow peak with a maximum intensity in the region of 270–280 nm (Fig. 2, *b*, curve *2*) does not change the magnitude of FWHM over a current increase, while the appearance of the spectrum in areas with lower intensity around this peak changes with increasing current. This kind of change in the type of spectrum with the injection level suggests that a narrow peak in the spectrum is caused by radiative recombination in MQW regions with an equilibrium composition, and in the 200–270 nm and 280–350 nm ranges there are local regions with impaired stoichiometry. Moreover, the number of such regions may significantly exceed the number of regions with an equilibrium composition. The FWHM broadening (Fig. 1, curves *2*) in MQWs located outside of SCR is caused by the process of none-equilibrium filling of spatial random alloy fluctuations by injected charge carriers. This process leads to a drop in EQE at j > 30 A/cm^2.

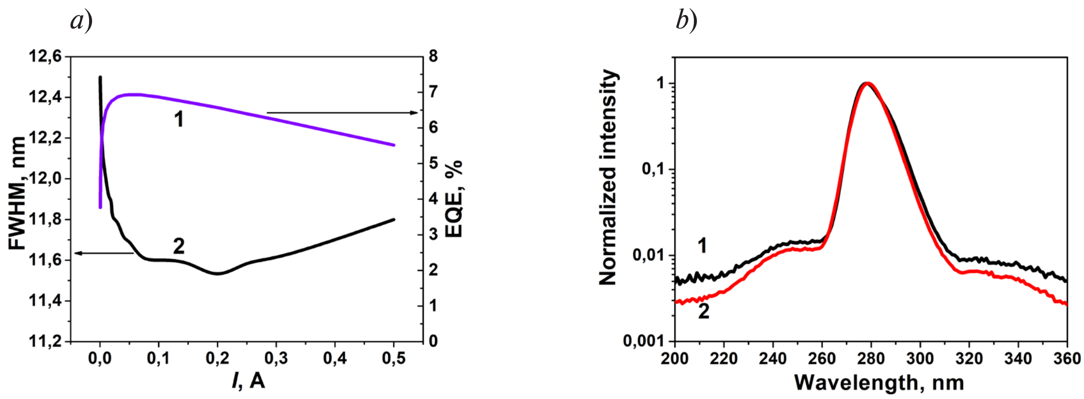

Fig. 2. Dependences of EQE (1) and FWHM (2) versus injection current of UV LEDs (λ_{peak} = 280 nm) (*a*) and EL spectra at 0.2 mA (*1*), 50 mA (*2*) (*b*)

Conclusion

The presence of local areas with impaired stoichiometry was revealed when studying the near-field distribution of self EL at small current of green LEDs with allow disorder. The processes occurring in such areas under the influence of injection current have been clarified. A study of competing processes leading to narrowing and broadening of FWHM in the EL spectra of green LEDs with different allow LD showed that both processes lead to loss of injected charge carriers and a decrease in EQE values. At the same time, the decrease in EQE values at the maximum is due to the presence in MQWs of the local regions with disturbed stoichiometry allow, enriched by excited defects. The source of charge carrier losses is the process of the capture of injected charge carriers by such defects in MQWs located in SCR of the *p–n* junction. UV LEDs, which have the lowest EQE values among all the studied LEDs, have the highest values of solid solution disorder. The results obtained suggest that one of the ways to increase the efficiency of LEDs is to reduce the level of disordered solids. A quantitative assessment of the level of disorder in terms of magnitude Δ_V can be used in the development of a technology for the growth of ordered solid solutions. The broadening of the FWHM EL spectrum of LEDs is caused by the nonequilibrium filling of the lateral random allow fluctuation by charge carriers in MQWs located outside the SCR. This process causes the beginning a drop in the EQE with the opening of *p–n* junction.

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