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Luminescence properties of heavily doped $Al_xGa_{1\text{-}x}N/AlN$ films grown on sapphire substrate

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ABSTRACT

Luminescent properties of the $Al_xGa_{1\text{-}x}N$ films with Si dopant concentration more than 10^{20} cm^{-3} grown by molecular beam epitaxy on sapphire substrates with AlN buffer film were investigate at room temperature. Time-resolved photoluminescence spectroscopy was employed to study the donor-acceptor pair transitions in the $Al_xGa_{1\text{-}x}N/AlN/Al_2O_3$ structures with Al mole fraction x from 0.47 to 1 under action of Nd:YAG (λ = 266 nm) laser radiation. The radiation inside planar waveguide consists of spontaneous emission and amplified spontaneous emission. The spontaneous emission spectra demonstrated inhomogeneous broadening with FWHM of 0.58 eV covers full visible range and propagate randomly in all directions. The amplified spontaneous emission propagate at near the critical angle of incidence along zigzag path under total internal reflection conditions at the interfaces of the waveguide. Its spectrum consists of several TE_m and TM_m modes, which have mutually orthogonal polarizations. The optical measured gains are equals to $g \approx 58\ cm^{-1}$ for $Al_{0.65}Ga_{0.35}N$ at 510 nm and $g \approx 20\ cm^{-1}$ for $Al_{0.74}Ga_{0.26}N$ films at λ = 528 nm. The measured values of quantum yields of spontaneous emission for $Al_xGa_{1\text{-}x}N$ films grow monotonically from 0.11 to 0.79 with increasing x from 0.47 to 0.74. Estimated emission cross-sections for the donor-acceptor pair transitions are more than 10^{-18} cm^2.

1. Introduction

Semiconductor laser sources for blue-green and ultraviolet (UV) spectral ranges are very useful in many fields of science and technology due to the wide range of available wavelengths, compactness, efficiency, reliability, low cost and ease of use. $Al_xGa_{1\text{-}x}N$ alloys with a bandgap in the range of 3.4–6.2 eV has emerged as perspective semiconductor materials with applications to laser sources [1–3]. At present, the shortest wavelength of UV laser diodes at 336 nm based on GaN have reported [4]. For the lasing on shorter wavelength, optical pumping is typically used [5–8]. The shortest wavelength of stimulated emission with λ = 214 nm has been realized under the optical excitation of an AlN layer on a c-Al_2O_3 substrate with a threshold pumping power density of 9 MW/cm^2 [5]. It was reported about the excitation of stimulated emission in the range of 235–250 nm in $Al_{0.7}Ga_{0.3}N/AlN$ heterostructures with multiple quantum wells when used femtosecond optical pumping [6].

The luminescence characteristics of heavily silicon doped $Al_xGa_{1\text{-}x}N$ films with a different Al content (x) were studied under the excitation by a low-energy (< 20 keV) electron beam [9]. The samples were grown by molecular beam epitaxy on sapphire substrates. The luminescence spectra of the $Al_xGa_{1\text{-}x}N$ films with $x > 0.42$ exhibited a broadband emission covering the entire visible spectral range from 1.72 to 2.7 eV (400–720 nm), while the band-edge luminescence is negligibly weak. Super-radiance within the broad band was obtained under pumping the samples by pulsed electron beam with a power up to 200 kW. It was later determined that wide band emission in the visible region is attributed to donor-acceptor and free electron-acceptor transition involving the same acceptor [10]. As a result, light–emitting structures of coherent and non-coherent natures from blue-green to far–red parts of the spectrum, including the structures with a broad emission band in a single emitting element, and tunable lasers in a large wavelength range can be created. Moreover, the wideband emission spectrum of heavy doped AlGaN samples gives premises for developing lasers with ultra-short pulses of femtosecond duration.

In this study, the spectral, temporal and polarization characteristics of spontaneous emission and amplified spontaneous emission of heavily silicon doped $Al_xGa_{1\text{-}x}N$ films were experimentally investigated under

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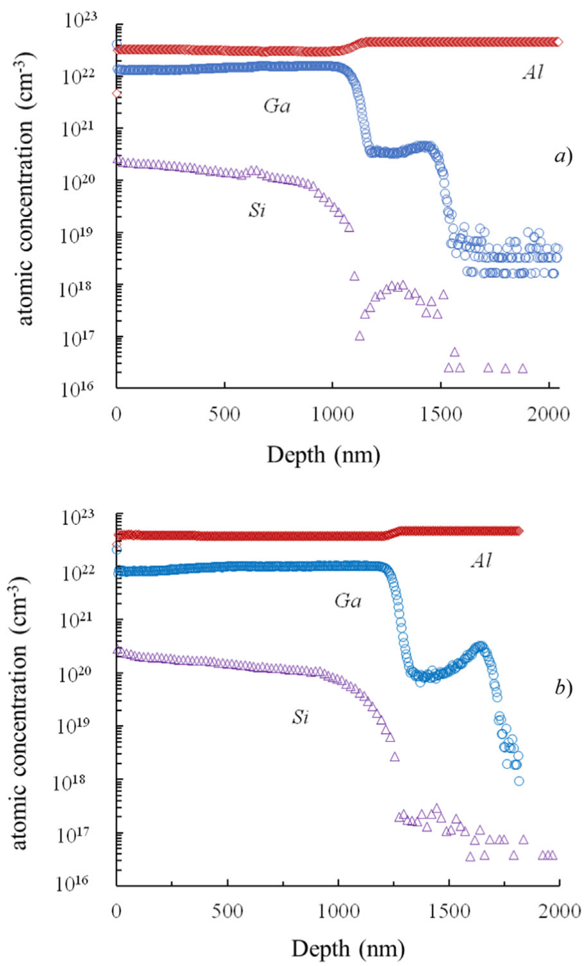

Fig. 1. The SIMS analysis of the heavily doped $Al_{0.65}Ga_{0.35}N$ (*a*) *and* $Al_{0.74}Ga_{0.26}N$ (*b*) films. The Al, Ga, Si atomic concentrations versus the depth from outer surface.

action by laser radiation with λ = 266 nm (E_L = 4.66 eV). The most experiments reported here were devoted to studies of the $Al_xGa_{1-x}N$ films with high luminescence quantum yields. Applying the stripe excitation method, we have measured optical gain in the $Al_{0.74}Ga_{0.26}N$ and $Al_{0.65}Ga_{0.35}N$ films under optical pumping. This method provides a means for investigations in a wider range of conditions than under the electron beam pumping and can be used for excitation thick films with populating inversion.

2. Experimental setup

The $Al_xGa_{1-x}N$ films studied were grown by molecular beam epitaxy in a modified Riber CBE-32 machine. Ammonia was used as a source of active nitrogen, the mixture of silane (SiH_4) with nitrogen (N_2) was used as a source of impurity atoms (Si). The gas mixture flow was 3 sccm; the silane content was 0.7%. The AlGaN films with a thickness h_0 ≈ 1200 nm were grown at a temperature of 860 °C and an ammonia flow of 130 sccm at total mixture pressure 5 × 10^{-5} Torr on (0001) – oriented 430 – µm – thick nitridated sapphire substrates. The AlN buffer film were grown on this sapphire substrate with the thickness h_p ≈ 350 nm (see Fig. 1). The synthesis technology and the parameter measurement for AlGaN films are described previously in more detail [10,11].

The secondary ion mass spectroscopy (SIMS) was used to measure the Si dopant concentration n_{Si}. For the $Al_{0.74}Ga_{0.26}N$ and $Al_{0.65}Ga_{0.35}N$ films n_{Si} varied from 8 × 10^{19} to 2 × 10^{20} cm^{-3} (see Fig. 1). The geometrical dimensions of the samples about (15 × 10) mm^2 were

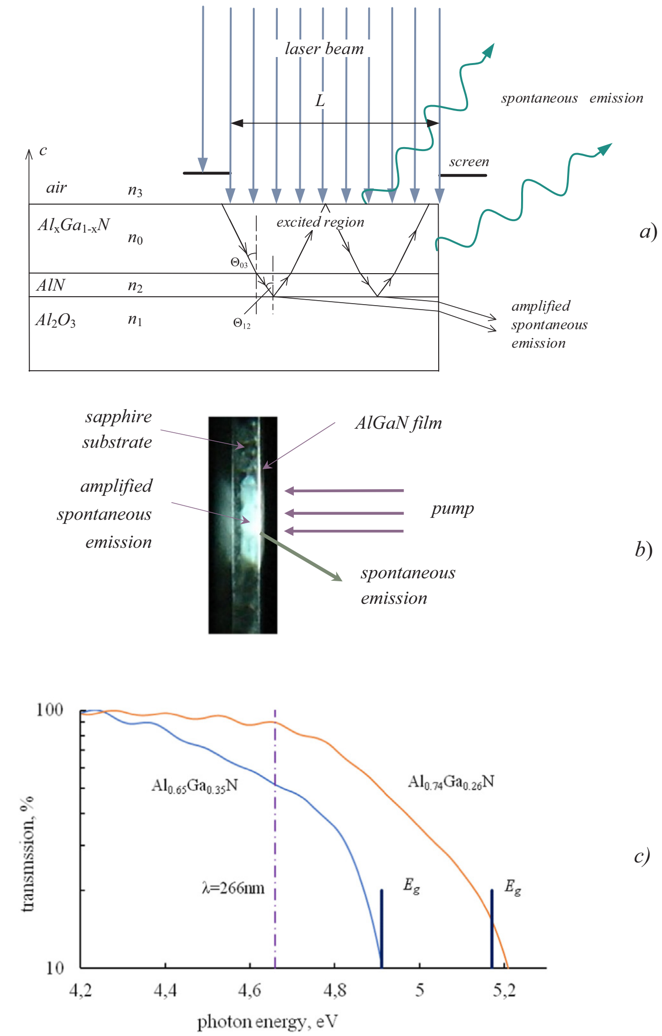

Fig. 2. Experimental setup for investigation of the luminescence properties of $Al_xGa_{1-x}N/AlN/Al_2O_3$ structures under optical pumping. *a*) Outline of the experimental setup and the scheme of the $Al_xGa_{1-x}N/AlN/Al_2O_3$ waveguide structure; *b*) Photo of the cleaved edge of $Al_{0.65}Ga_{0.35}N/AlN/Al_2O_3$ structure during pumping by laser radiation at λ = 266 nm; *c*) Absorption spectrum of $Al_{0.74}Ga_{0.26}N$ and $Al_{0.65}Ga_{0.35}N$ films near bandgap level E_g.

prepared by cleaving without edges polishing.

Fig. 2a shows the experimental setup and the scheme of the AlGaN/AlN/Al_2O_3 waveguide structure. The fourth harmonic (λ = 266 nm) of the Nd:YAG laser radiation with pulse duration of 8 ns FWHM at a repetition rate of 10 Hz was used to excite the photoluminescence. The laser beam was directed perpendicular to the AlGaN film surface, into a homogeneous stripe with the width about 0.4 mm. To measure net optical gain, the variable-stripe-length method was used. The length of the stripe (excitation length) *L* is determined by the position of a screen and was varied using microscrews with increments of 10 µm. The maximum pulse power density on the surface of the sample was P ≈ 8 MW/cm^2. Spontaneous emission (SE) spectra were recorded at an angle of 45° with respect to the sample surface normal. Simultaneously, amplified spontaneous emission (ASE) spectra were collected from cleaved edges of the sapphire substrates at a small angle (several degrees) to the plane of the waveguide (see Fig. 2b).

Due to effective transitions between the valence and conduction bands at x < 0.56, the pump radiation is effectively absorbed in the $Al_xGa_{1-x}N$ films. In the other case at x > 0.56 the pump photon energy of 4.66 eV becomes smaller than the bandgap width for $Al_xGa_{1-x}N$ films. Therefore, for these structures, the pump absorption is due to