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High-resolution emission spectroscopy of random lasing in GaN films pumped by UV-pulsed laser

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ABSTRACT

We report on room temperature photoluminescence on GaN films grown by metal organic chemical vapor deposition (MOCVD). A NdYAG pulsed-laser at 266 nm illuminates the films. Two components, at 363 nm and 370 nm, are identified in the near band edge structure on the spectra. A laser threshold of 700 ± 150 kW cm^{-2} is evidenced and corresponds to random lasing in the GaN film. A drastic narrowing of the spectral bandwidth from 5.2 to 1.8 nm is observed at 370 nm. High-resolution spectroscopy measurements show laser mode widths thinner than 50 pm leading to a high quality factor $Q=7750$. Low-resolution measurements show redshift from 370.0 to 373.1 nm for one component and from 363.1 nm to 363.9 nm for the other. Interpretation of this redshift is discussed.

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1. Introduction

Random lasing (RL) [1,2] is a good example that the disorder in a photonic material may lead to optical structures inducing interesting applications [3,4]. Random lasing in semiconductors was clearly identified since the pioneer work of Cao et al. [5] on powders where the coherent emission of light in ZnO was interpreted as resulting from the formation of closed-loop path for light through recurrent scattering in the medium. Many experimental and theoretical works have then been realized on random lasing [6,7]. Among all semiconductor materials, ZnO [8] and GaN [9] have drawn particular attention due to their unique wide gap and their potential applications in UV optoelectronics devices [10]. Many studies on RL have been done in ZnO structure: films, [11–16], powders [13,17,18], nano-wires [19–21], rods [8,22,23], nano-walls [24] and disks [25,26]. Comparing with this abundant literature on optically pumped RL in ZnO, reports on RL in GaN [27–31] appear to be more limited, since most of the studies were focused on complex electrically excited GaN-based hetero-structure [32,33].

Pumped by intense laser beam, such semiconductor films present optical interesting properties. The photo-induced carrier density may easily reach the Mott density ($n=1 \times 10^{19}$ cm^{-3}, in GaN at 300 K [34]) and even exceed it. At such high carrier concentrations, and at room temperature, the fraction of excitons, corresponding to an isolated electronhole pair, is so small that the properties of the semiconductor are well described by the electron–hole plasma (EHP) theory [35]. According to this interpretation, the exciton is screened by the Coulomb interaction with all the surrounding charges [36,37]. One of the most noticeable effects is the modification of the UV emission associated with the recombination of the electron–hole pairs. The well-defined and well-structured emission, observed at low temperatures and low carrier densities, and corresponding to the exciton recombination, disappears completely. A broad spectral band emerges in the vicinity of the band gap absorption (Fig. 4 in Ref. [38]). This band is commonly referred as the near band edge (NBE) and extends in GaN from 350 nm to more than 400 nm.

As observed in ZnO semiconductor, and above some threshold under high excitation, coherent emission can take place since the chemical potential of the electron hole pair becomes positive [37]. It leads to a spectrally limited region where a positive gain allows amplification of stimulated emissions in the UV spectral domain.

In this work, we report on the amplification of light by stimulated emission or by amplified spontaneous emission (ASE) from optically pumped GaN films. We study the photoluminescence (PL) spectra by emission spectroscopy when illuminated by a pulsed UV laser. It is shown that effective RL takes place in our sample. High-resolution spectroscopy allows us to determine, for the first time in GaN, a maximal spectral linewidth of 50 pm for the RL lasing modes. A significant redshift of the lasing component is observed.

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2. Experimental setup

The experimental set up has been described in a previous work [12]. The illuminating source was a frequency quadruped NdYAG laser (Brillant from Quantel) at 266 nm, with a repetition rate of 10 Hz. To prevent damages on films, we limited the beam energy to less than 1500 μJ. Dichroic mirrors and band pass filter were used to reject both infrared at 1064 nm and green at 532 nm primary beams. The laser beam impacts the films without further focusing. We measured on sample a quasi-circular spot size of 8 mm in diameters. An example of the temporal evolution of a single laser pulse is given on Fig. 1. This measurement was obtained using a phototube module (Hamamatsu H8496-11) with a resolution of 400 ps connected to a digital oscilloscope. Typical laser pulse width is measured around 2 ns FWHM.

GaN films were grown on a c-cut sapphire substrate by metal organic chemical vapor deposition (MOCVD) at $T=1150\ ^\circ C$. The sample was a thick GaN hetero-structure: (6 μm) n^--doped GaN/(3 μm) n^+-doped GaN/(4 μm) AlN/Sapphire. The scanning electron microscopy (SEM) cross section shows a dense, compact, and homogeneous structure without any evidencing of grain boundaries between crystallites (Fig. 2).

Considering the high value of the absorption coefficient of GaN at 266 nm [39], energy deposition in these films occurs close to the surface since the penetration depth of the beam is limited to the first hundred of nanometer. Thus, only the very first micrometer GaN of the hetero-structure is responsible for the photoluminescence reported in this experiment.

Emission spectroscopy measurements were carried out by two spectrometers. The spectrometer Ocean Optics USB2000 (spectral range 200–850 nm, slit 25 μm) had a low-resolution of 200 around 370 nm. It was coupled to an optical fiber on the extremity of which a short focal lens collected the light from the sample. Such an easy to handle configuration was suitable for low-resolution spectroscopy over large spectral range. To record spectra with a higher resolution, we used an ECHELLE technology spectrometer (Andor MECHELLE 5000, 195 mm focal length) with a resolution of 8000 corresponding to 50 pm around 400 nm. An ICCD gated camera (Istar DH334T Andor) was mounted on this spectrometer as detector. We used the same optical fiber as the one used for low resolution to couple this spectrometer with the sample. It could be noticed that ECHELLE technology is not so relevant to record broad bands because of an intrinsic long period structure superimposed on each spectra. All the spectra presented here are raw data and are uncorrected from this residual periodic structure.

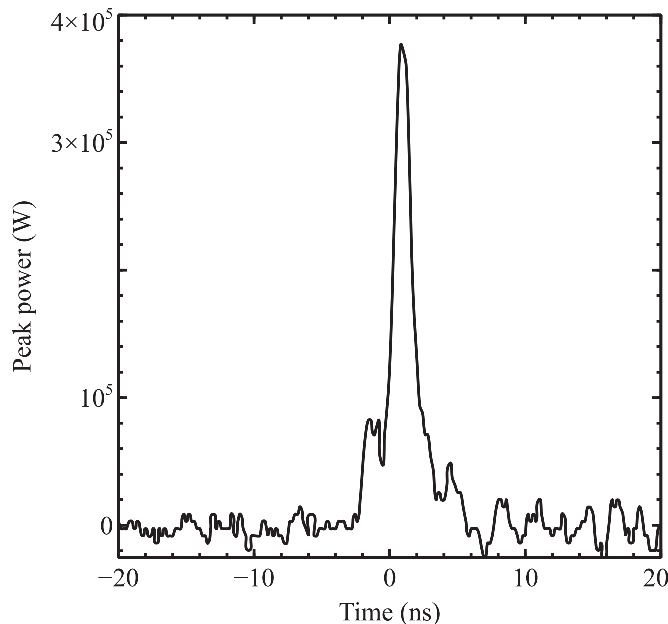

Fig. 1. Typical time evolution of the laser pulse (laser energy at 266 nm:800 μJ).

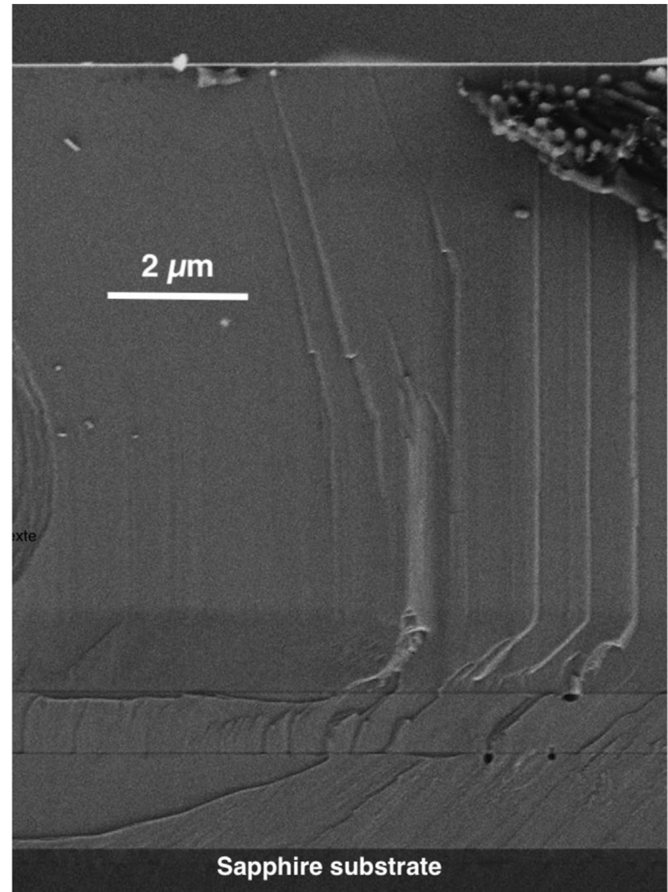

Fig. 2. SEM cross-section image of the GaN hetero-structure.

3. Results

In the UV–visible wavelength range, from 200 to 800 nm, the spectrum is strongly dominated by the NBE [25] of GaN that extends from 350 nm to 400 nm. PL measurements of this band are displayed on Fig. 3. The NBE is classically attributed to optical transition occurring through electron–hole recombination [40]. The energy of the emitted photons corresponds to the energy band gap of the semiconductor. At high temperatures and high carrier densities, when excitons do not exist any more, this NBE can be very broad compared with its resolved structure observed at low temperatures and low carrier densities [38]. For longer wavelength in the visible domain, the spectra present a weakand extremely broad band that extends from 450 to 650 nm. This green band is attributed to defects and vacancies in the films.

In our experimental conditions, we observe on this band two distinct spectral components, labeled A and B on Fig. 3. Both of them are well fitted by simple lorentzian profiles. The component A, at 363 nm, is 5.0 ± 0.2 nm wide and presents a weak redshift of 0.8 ± 0.2 nm of its maximum (from 363.1 ± 0.1 nm to 363.9 ± 0.1 nm) when the pumping power is increased. The second spectral component B, at longer wavelength, appears at low energy pumping on the red wing of the component A at around 370 nm. As the pumping energy is gradually increased, the component B is modified in three ways: first, its intensity increases

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