Thin Solid Films 519 (2011) 2516–2519

Contents lists available at ScienceDirect

Thin Solid Films

journal homepage: www.elsevier.com/locate/tsf

Photoluminescence and spin relaxation of MnZnO/GaN-based light-emitting diodes

Lung-Chien Chen *, Ching-Ho Tien, Yi-Min Luo, Chien-Sheng Mu

Department of Electro-optical Engineering, National Taipei University of Technology, Taipei 106, Taiwan, ROC

ARTICLE INFO

Article history:
Received 30 April 2010
Received in revised form 23 November 2010
Accepted 3 December 2010
Available online 10 December 2010

Keywords:
Photoluminescence
Spin relaxation
Manganese zinc oxide
Gallium nitride
Light-emitting diodes
Metal-organic chemical vapor deposition

ABSTRACT

This study investigates the spin relaxation of GaN-based light-emitting diodes with an MnZnO film by examining its photoluminescence (PL) and time-resolved magnetization modulation photoluminescence. PL measurements reveal that the application of a magnetic field produced a clear difference between the intensities of the right (σ^+) and left (σ^-) circular polarization components. The circular polarization was identified as $P_{circ} = [I(\sigma^+) - I(\sigma^-)]/[I(\sigma^+) + I(\sigma^-)]$, where $I(\sigma^+)$ and $I(\sigma^-)$ are the intensities of the σ^+ and σ^- components, respectively. The PL polarization was 3.6% in a 0.5 T magnetic field. In a magnetic field, the photo-ionized lifetime and spin-polarized lifetime values were approximately 13.64 and 54.54 ns, respectively. The right-circular-spin-polarization lifetime and the left-circular-spin-polarization lifetime values were about 39.09 and 40.01 ns, respectively.

1. Introduction

Spin injection using spin-polarized light-emitting diodes (spin-LED) has been recently reported [1–3]. Spin-polarized carrier injection and transport are the basis of spintronics, and have been used to develop devices whose quantum states can be controlled and operated using spin-polarized carriers [4–6]. The process of producing gallium nitride (GaN)-based LEDs by electrical injection has attracted a lot of interest for important components in solid-state lighting applications, including street lighting and indoor lighting. Spin-polarized GaN-based LEDs are a method for improving the light-extraction efficiency of their owing to magneto-optical effects [7]. The carrier relaxation processes in spin-LEDs have been widely investigated because they significantly affect optoelectronic characteristics [8–11]. The dynamics of carrier recombination for InGaN/GaN or GaN/AlGaN quantum wells or GaN-based LEDs have been investigated using time-resolved photoluminescence measurements [12–15]. Our previous study shows that the application of a magnetic field increases the optical output power of GaN-based LEDs with an MnZnO film by about 60% and 50% at injection currents of 20 and 100 mA, respectively [7]. The magneto-optical current, comprising a spin-polarized current and a photo-ionized current, increases exponentially with the forward bias. The spin-polarized current component dominates in the magneto-optical current. However, the spin dynamics of the MnZnO/GaN-based LEDs remain unclear.

To understand spin dynamics in a spin-LED, this study extends our previous study by discussing how the photoluminescence (PL) and time-resolved magnetization modulation photoluminescence (TRMMPL) vary in GaN-based LEDs with ZnO:Mn (MnZnO) films in a magnetic field [7]. This study also examines the spin-polarized properties of GaN-based LEDs with and without MnZnO films, including the difference between the intensities of right circular polarization (σ^+) and left circular polarization (σ^-).

2. Experimental details

In this study, low-pressure metal-organic chemical vapor deposition is utilized to grow a GaN-based LED structure. First, a 30 nm-thick GaN nucleation layer was grown on a (0001)-oriented sapphire substrate at a reactor temperature of 550 °C. The reactor temperature was then increased to 1030 °C to grow a 4 μm-thick Si-doped n-type GaN layer. The reactor temperature was then decreased to 860 °C to grow five pairs of InGaN (250 Å)/GaN (40 Å) multiple-quantum-wells (MQWs), which formed the active region. The reactor temperature was then increased to 1050 °C to grow a 0.1 μm-thick Mg-doped p-type $Al_{0.01}Ga_{0.99}N$ cladding and electron-reflective layer and a 0.4 μm-thick Mg-doped p-type GaN contact layer. Heat treatment was carried out at 650 °C for 10 min in ambient nitrogen to activate the p-type dopant. The carrier concentration of the n-type and p-type GaN were 3×10^{17} cm^{-3} and 1×10^{18} cm^{-3}, respectively. The 2350 Å-thick ITO spacer and the 4000 Å-thick MnZnO film were then formed sequentially on top of the p-type GaN layer by evaporation and spray pyrolysis, respectively. Fig. 1(a) depicts a cross-section of the completed structure in a magnetic field. All measurements were made at room temperature.

* Corresponding author. Tel.: +886 2 2771 2171; fax: +886 2 8773 3216.
E-mail address: ocean@ntut.edu.tw (L.-C. Chen).

0040-6090/$ – see front matter © 2010 Elsevier B.V. All rights reserved.
doi:10.1016/j.tsf.2010.12.014

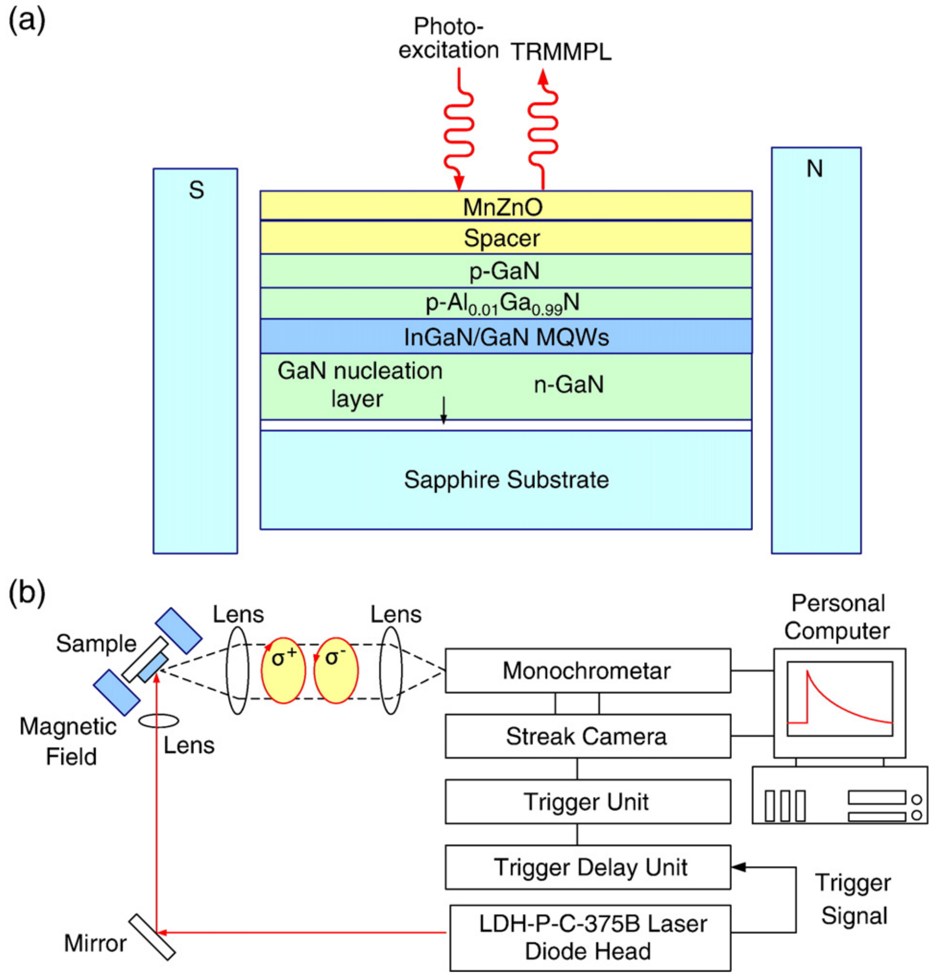

Fig. 1. (a) MnZnO/GaN-based LED in a magnetic field. (b) Experimental apparatus for TRMMPL measurement.

PL spectra were measured with a picosecond laser diode head (PicoQuant GmbH, LDH-P-C-375B, Germany) with a pulse duration of 50 ps and an excitation wavelength of 375 nm as the excitation source. Because the energy bandgaps of MnZnO, ITO, and GaN are about 3.2, 3.5, and 3.4 eV, respectively, an LDH-P-C-375B laser diode can be adopted as the excitation source for an MnZnO/GaN-based LED. Fig. 1(b) shows the experimental apparatus for measuring TRMMPL. This experimental setup was based on a laser diode at a repetition rate of 80 MHz. Detection was performed using a fast scan streak camera in conjunction with a monochromator using gratings with grooves of 100, 300 or 1200 lines/grooves. The time-resolution for detection was approximately 500 ps. The circular polarization was analyzed by passing PL light through a $\lambda/4$ wave plate and a linear polarizer. The high-intensity pump circular polarization laser pulse was used to spin-selectively excite electrons in the spin-LED [10]. The TRMMPL of the sample was then measured using a detection wavelength of $\lambda = 460$ nm.

3. Results and discussion

Fig. 2 shows the room temperature (RT) PL spectra of a GaN-based LED without and with an MnZnO film under applied magnetic fields of 0 and 0.5 T, analyzed to elucidate for right (σ^+) and left (σ^-) circular polarizations. Circular polarization was analyzed by passing PL light through a photoelastic modulator and a linear polarizer. The peak wavelength of the PL spectra of a GaN-based LED without MnZnO film was approximately 460.5 nm, as presented in Fig. 2(a). The nature of the emission line was associated with the near band-to-band (B–B) recombination. No PL polarization was observed in zero fields (0 T) in a GaN-based LED without an MnZnO film. The difference between the PL intensities in and out of a magnetic field may be attributed to the photoionization process, which is a modification of the photo-induced current because of the magnetic field in the active area of the LED. However, in a GaN-based LED with an MnZnO film, a magnetic field produced a clear difference between the intensities of the σ^+ and σ^- components, and the σ^- component with the low energy part in the PL spectrum dominates. The peak wavelengths of the σ^+ and σ^- components of the PL spectra of a GaN-based LED with an MnZnO film were approximately 457 and 455 nm, respectively, as displayed in Fig. 2(b). The difference between the peak positions may be attributed to the Zeeman splitting of the quantum wells of the LED coursed by magnetic field. The large difference between the intensities of these components reveals a large carrier spin polarization in the MnZnO film. Circular polarization is determined as $P_{circ} = [I(\sigma^+) - I(\sigma^-)] / [I(\sigma^+) + I(\sigma^-)]$, where $I(\sigma^+)$ and $I(\sigma^-)$ represent the intensities of the σ^+ and σ^- components, respectively. The PL polarization was 3.6% in a 0.5 T magnetic field. This result is consistent with our previous research, which revealed EL polarization (2.9%) and a spin-polarized current-to-total current ratio of 2.77% at a forward bias of 3.4 V [7].

Time-resolved spectroscopy is a useful technique for evaluating the optical properties of semiconductors. For ease of explanation and understanding, two words “photo-ionized lifetime” and “spin-polarized lifetime” are defined here. The first is linked to the modification of the intensity lifetime due to the magnetic field. The second concerns the modification of the intensity lifetime under a magnetic field when the MnZnO layer is inserted. Fig. 3 plots the transient