Nonradiative recombination processes of carriers in InGaN/GaN probed by the microscopic transient lens spectroscopy

Koichi Okamoto,a) Kenichi Inoue, Yoichi Kawakami, and Shigeo Fujita
Department of Electronic Science and Engineering, Kyoto University, Kyoto 606-8501, Japan
Masahide Terazima
Department of Chemistry, Graduate School of Science, Kyoto University, Kyoto 606-8502, Japan
Ayumu Tsujimura and Isao Kidoguchi
Advanced Technology Research Labs., Matsushita Electric Industrial Co., Ltd., Osaka 570-8501, Japan

(Received on 25 June 2002)

Temporally and spatially resolved observations of the nonradiative recombination (NR) processes of carriers in low dislocated GaN and InGaN/GaN were successfully obtained by using microscopic transient lens spectroscopy. The heat generations and conductivities of NR processes were detected by the signal intensities and the time profiles. We found that the thermal conductivities were not so different at the seed region (threading dislocation density (TDD) = $1 - 2 \times 10^8$ cm$^{-2}$) and the wing region (TDD = $1 - 2 \times 10^6$ cm$^{-2}$) of air-bridged lateral epitaxially grown GaN and InGaN/GaN, but the amount of heat generated at the wing regions was much smaller than that at the seed regions.

I. INTRODUCTION

GaN-based optical devices such as light-emitting diode and laser diode have very strong emissions in spite of high threading dislocation densities ($\text{TDD} = 10^8 - 10^{10} \text{ cm}^{-2}$). For a wider application of GaN-based optical devices, the development of the emission efficiency has been expected. A nonradiative recombination (NR) process of carrier in a material is one of the most important processes to control the optical property because a great numbers of carriers decay by the NR process at room temperature. It had been reported that the threading dislocations (TD) in GaN might act as the nonradiative recombination centers (NRC). In order to reduce the TDD in GaN and InGaN, the growth technique has been developed rapidly. Recently, low dislocated GaN ($\text{TDD} = 10^6 \text{ cm}^{-2}$) have been achieved by using the epitaxial lateral over growth technique. However, remarkable enhancement of emission efficiencies of low dislocated GaN and InGaN has so far not been found. The actual correlation between the NR process and the TDD is still unknown because the direct observation of the NR process has been very difficult. Recently, we succeeded in the direct detection of the heat generation of the NR process probed by the transient grating (TG) technique. In this article, we developed the microscopic transient lens (MTL) technique for the selective time-resolved measurement of the heat generation and conduction of the NR processes.

II. EXPERIMENTAL SETUP

A frequency-tripled beam of a Nd:YAG laser (355 nm) and a He-Ne laser (633 nm) were used as pump and probe beams, respectively. Both beams were focused at a sample by an objective lens ($\times 100$). Spot sizes of both beams were about 3 μm in diameter. By the excitation with a Gaussian spatially distributed pump beam, refractive index ($n$) in the sample is also spatially modulated by the modulation of the carrier density change ($\delta N$) and the temperature change ($\delta T$). Such modulations act as transient lenses. Focus/defocus of the probe beam at the transient lens was detected by a photomultiplier tube with an optical fiber. All measurements were at room temperature (23 °C).

The samples used in this study were grown by metal-organic chemical vapor deposition. Recently, Kidoguchi et al. accomplished air-bridged lateral epitaxial growth (ABLEG). ABLEG-GaN has two regions, one is a seed region having high TDD ($1 - 2 \times 10^9 \text{ cm}^{-2}$) and another is a wing region having low TDD ($1 - 2 \times 10^8 \text{ cm}^{-2}$). The sample structure of ABLEG-GaN was shown in Fig. 1. We used three samples; bulk-GaN, ABLEG-GaN, and InGaN (35 Å)/GaN(105 Å), a three-quantum wells (3QW) structure grown on ABLEG-GaN.

III. RESULTS AND DISCUSSION

The time profile of the MTL signal taken for the bulk-GaN was shown in Fig. 2. This signal has two components; the spikelike dip component just after photoexcitation and the slow decay component. The fast component and the slow component are due to the defocus and focus of the probe beam at the transient concave and convex lens effects in the sample, respectively. From ($\partial n/\partial N$)$>0$ and ($\partial n/\partial T$)$<0$, we assigned the fast and slow components to the modulations of the carrier density and the temperature, respectively. The signal intensity and decay of the fast component represents the created carrier density and the carrier recombination/diffusion, respectively. The signal intensity and decay of the slow component represent the amount of the generated heat by the NR process and the thermal conductivity of materials.
Time and spatial dynamics of $\delta n$ and $\delta T$ can be described by the rate equation including the diffusion/recombination of carriers and the generation/conduction of heat with cylindrical coordinates. By solving this rate equations, thermal conductivity $k$ of GaN can be obtained by $k = D_0 n \rho C_p$, with the density ($\rho = 6.095 \text{ g cm}^{-3}$) and the heat capacity ($C_p = 9.745 \text{ cal mol}^{-1} \text{ K}^{-1}$). $\tau_{\text{rad}}$ and $\tau_{\text{nonrad}}$ are the radiative and nonradiative recombination lifetime, respectively. $Q$ is the heat amount generated by the NR process. The optical pass of the probe beam at the transient lens given by the ABCD low of Gaussian beam. In this experimental condition, the time profile of the signal intensity of MTL $[S(t)]$ is proportional to $\delta T(t, r = 0)$. Therefore, the decay component of the thermal signal can be described as

$$S(t) = \frac{I(t) - I(0)}{I(0)} \propto -\frac{w_0^2}{4D_{\text{th}}^2 + w_0^2}.$$  \hspace{1cm} (2)

We could fit the slow decay component of Fig. 2 by using this equation and a value for $D_\text{th}/w_0^2$ was obtained. To obtain the $D_\text{th}$ value, a value of $w_0^2$ is needed. We estimated $w_0 = 2.6 \mu m$ by using the $D_\text{th}$ value for this sample obtained by the TG measurement $[D_\text{th} = 0.77 \text{ cm}^2 \text{s}^{-1} (\kappa = 2.3 \text{ W cm}^{-1} \text{ K}^{-1})]$. We can use this $w_0$ value for the measurement of ABLEG-GaN. The time profiles of the selective MTL signals at seed and wing regions in ABLEG-GaN were shown in Fig. 3. We fitted these decays by using Eq. (2) and obtained the $D_\text{th}$ values at each region. The calculated $\kappa$ obtained from $D_\text{th}$ were $\kappa = 2.2 \text{ W cm}^{-1} \text{ K}^{-1}$ at seed region and $\kappa = 2.0 \text{ W cm}^{-1} \text{ K}^{-1}$ at wing region. $\kappa$ values of each region were not so different and close to $\kappa = 2.3 \text{ W cm}^{-1} \text{ K}^{-1}$ of bulk-GaN. $\kappa$ of GaN had been measured as $1.3 \text{ W cm}^{-1} \text{ K}^{-1}$ by Sichel and Pankove in 1977. Recently, Luo et al. reported that $\kappa$ of GaN grown by the lateral epitaxial overgrowth (LEO) with TDD $= 5 \times 10^6 \text{ cm}^{-2}$ is $1.55 \text{ W cm}^{-1} \text{ K}^{-1}$ though $\kappa$ of GaN with TDD $= 10^{10} \text{ cm}^{-2}$ is $1.35 \text{ W cm}^{-1} \text{ K}^{-1}$ as probed by the scanning thermal microscope (STM). In a similar way, Asnin et al. reported $\kappa$ of GaN-LEO with TDD $> 10^5 \text{ cm}^{-2}$ as $1.7 - 1.8 \text{ W cm}^{-1} \text{ K}^{-1}$. Florescu and co-workers reported that the $\kappa$ values of GaN depend on the carrier concentration and thickness between 0.5 and 1.95 $\text{ W cm}^{-1} \text{ K}^{-1}$. They also developed the STM with spatial resolutions and reported that the $\kappa$ values of GaN is $2.00 \text{ W cm} < \kappa < 2.10 \text{ W cm} K$ on the partially overgrown regions. Obtained $\kappa$ values of bulk-GaN and ABLEG-GaN in this study are close to the maximum value which has ever been measured. This fact suggests that both our bulk-GaN and our ABLEG-GaN should have the highest crystallinity.

Although we could not find the difference of the $\kappa$ values at each region, we found a remarkable difference of the thermal signal intensity at each regions. We could compare the thermal signal intensities by the NR processes of unit carrier densities at each region by normalization of the carrier signal intensities. It was found that the heat generation at the wing region was much smaller than that at the seed region. At the wing region, low TDD should reduce the NR processes of carriers. A similar result was obtained for the InGaN/GaN 3QW structure grown on ABLEG-GaN (Fig. 4). This fact suggests that the TD in GaN and InGaN must act as the NRC. For the first time, we have succeeded in detecting the heat generation from the TD and in clarifying the relationship between the NR process and TD in GaN and InGaN by
InGaN/GaN though it acts as the NRC. It is not effective for the emission efficiency of GaN and energy level in the crystals. This is the possible reason that TD tons to electrons and carriers, or trapping into the deep en-

domalization processes after the thermal dissociation of exci-

dominant thermalization path way, for example, slow ther-

GaN and InGaN. We believe that another process must be the

should not be a dominant path way of the thermalization in

region. We considered that the NR processes of excitons

efficiencies are decided only by the ratio between the radia-

tons contribute to the heat generation. If internal emission

spectra, we knew that the emissions of these samples are

dominantly attributed to free excitons and the internal effi-

ciency of emission of each region is 0.6% at room

temperature.10 This fact means that the 99.4% of free exci-
tons contribute to the heat generation. If internal emission
efficiencies are decided only by the ratio between the radia-
tive and nonradiative recombination processes of excitons,
emission efficiency should be dramatically different at each

region. We considered that the NR processes of excitons

should not be a dominant path way of the thermalization in

GaN and InGaN. We believe that another process must be the

dominant thermalization path way, for example, slow ther-

malization processes after the thermal dissociation of exci-
tons to electrons and carriers, or trapping into the deep energy
level in the crystals. This is the possible reason that TD is not effective for the emission efficiency of GaN and InGaN/GaN though it acts as the NRC.

IV. CONCLUSION

Microscopic transient lens (MTL) spectroscopy is a powerful tool for the time and spatially resolved detection of the nonradiative recombination (NR) processes of carriers in GaN and InGaN/GaN. We found that the threading disloca-
tions (TD) in GaN actually act as the nonradiative recombi-
nation centers (NRC) of created free excitons. However, TD

should not be effective for the emission efficiency because

another slow thermalization processes due to the thermal disso-
ciation or the trapping in the deep level of excitons should be

the dominant process for thermalization in GaN and

InGaN/GaN.

ACKNOWLEDGMENTS

This work was partly supported by the Japan Society for the Promotion of Science (No. 12002454) and the Kyoto University-Venteure Business Laboratory Project.