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Carrier Dynamics in InGaN/GaN SQW Structure Probed by the Transient Grating Method with Subpicosecond Pulsed Laser

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Carrier dynamics in GaN and InGaN/GaN SQW structures were observed by using the transient grating (TG) method with sub-picosecond pulsed laser at room temperature. The diffusion coefficients (D) of photo-created carriers were estimated by the decay rate time of TG signals and the photoluminescence (PL) lifetime. It was found that D depends on the emission wavelength (In composition). The relationship between the emission efficiencies and carrier diffusion was considered in terms of the spatial inhomogeneity of In composition.

Recently, InGaN/GaN-based light emitting diodes (LEDs) have been commercialized in ultraviolet (UV), blue, green, and amber spectral region [1, 2]. In particular, the external quantum efficiency (η_{ext}) of about 20% is now achieved in blue (450 nm) LEDs. However, η_{ext} values are still lower for LEDs out of this blue spectral range, but detailed reasons for the reduction of η_{ext} have so far not been elucidated. In this study, we try to elucidate this reason by the viewpoint of carrier dynamics. The transient grating (TG) method which is one of third-order nonlinear spectroscopy, has been used for GaN to detect the nonlinear susceptibility [3], exciton dephasing time [4], time response of scattering [5], or quantum beat [6], etc. It is also a powerful tool to directly detect diffusion processes. By using this method, Haag et al. [7] have measured and reported the carrier diffusion in GaN. We observed the diffusion of heat energy generated by the nonradiative recombination of carriers in GaN and ZnSe by the TG method with nano-second pulsed laser [8, 9]. In this work, we observed carrier diffusion in InGaN/GaN-based SQW structures in UV, blue, green, and amber spectral region by using the TG method with sub-picosecond pulsed laser.

The samples used in this study were grown on a (0001) oriented sapphire (Al_2O_3) substrate by a two-flow metalorganic chemical vapor deposition (MOCVD) technique [10]. The thickness of GaN bulk layer is 4 μm. GaN/InGaN SQW structure is composed of a GaN (1.5 μm), an n-GaN:Si (2.3 μm), an InGaN SQW (3 nm) and a GaN cap (5 nm) layer. LED structure of GaN/InGaN SQW is composed of GaN (30 nm), n-type

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GaN:Si (5 μm), InGaN SQW (3 nm), p-type AlGaN:Mg (60 nm) and p-type GaN:Mg (150 nm).

For the TG measurement, a mode-locked fiber laser and regenerative amplifier system (Clark) was used. The frequency doubled beam (388 nm) was used as pump beam. Pulse width, power, and repetition rate were 500 fs, 1 mW and 1 Hz, respectively. The pump beam was split into two coherence beams and crossed again in the sample with $\theta = 30°$ and 120°. The modulation of carrier density (grating) is created in the sample by the interference pattern. The fundamental beam (775 nm) was used as a probe beam with the optical delay unit. The probe beam was partly diffracted, which was detected by a photomultiplier tube (Hamamatsu) and averaged with a boxcar integrator. For the time-resolved photoluminescence (TRPL) measurements, the frequency doubled beam of a mode-locked Ti:sapphire laser (Spectra-Physics; 370 nm) was used for excitation. For the scanning confocal laser microscopy (Tokyo Instruments) was used with the 488 nm line of Ar^+ laser. The whole measurements have been performed at room temperature (23 °C).

Figure 1a shows the time profile of the TRPL measurement (A) and the TG measurement with $\theta = 30°$ (B) and 120° (C) taken for bulk GaN. These profiles could be fitted by a single exponential function. The PL lifetime (τ_{PL}) and the TG decay time (τ_{TG}) were obtained as τ_{PL} = 50 ps, $\tau_{TG}(30°)$ = 43 ps, and $\tau_{TG}(120°)$ = 16 ps. The time and spatial behavior of the TG signal intensities were described by the diffraction theory [11] and the diffusion-recombination coupled rate equation of carriers [9]. By solving these equations, the time profile of the TG signals is given by a single exponential function and D is obtained by $1/\tau_{TG} = Dq^2 + 1/\tau_{PL}$, where q is the wave number of grating ($q = 2\pi/\Lambda$). Figure 1b shows the relationship between τ_{TG} and q^2. This plot shows a good linear relationship and D was obtained by the slope as 0.54 cm^2s^{-1}. The diffusion length (Λ) of carriers is estimated as Λ = 0.05 μm by $\Lambda = (D\tau_{PL})^{1/2}$. This value is close to the reported values by cathodoluminescence, Λ = 0.1 [12], 0.25 [13], or 0.05 μm [14].

In a similar way, τ_{PL} and D values of other samples were measured and plotted against the emission wavelength (In composition) in Fig. 2. It was found that τ_{PL} becomes lager with increasing In composition. A drastic change was observed in the region larger than 450 nm, which is the highest η_{ext} point. D also becomes gradually

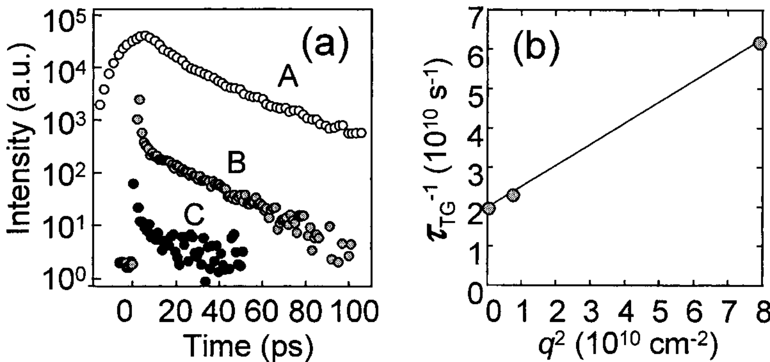

Fig. 1. a) Time profile of the TRPL measurement (A) and TG measurements with $\theta = 30°$ (B) and 120° (C) taken for GaN at room temperature. b) Relationship between the TG decay time (τ_{TG}) and the grating constant (q^2) taken for GaN

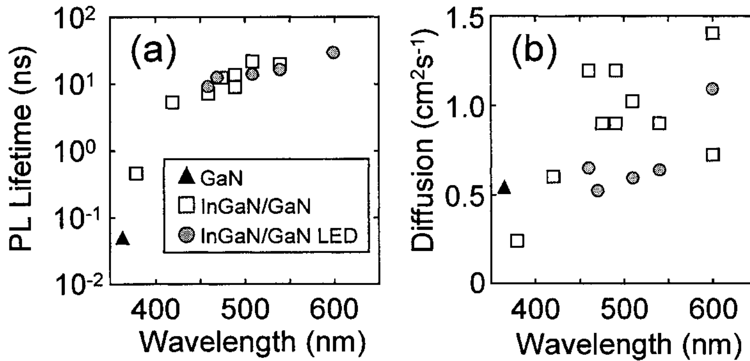

Fig. 2. a) PL lifetimes and b) diffusion coefficients of GaN bulk layer, InGaN/GaN SQW, and InGaN SQW LED structures plotted against the emission wavelength (In composition)

larger with increasing In composition expect lowest value of SQW (380 nm). At the room temperature, τ_{PL} should be nearly equal to the nonradiative recombination lifetime ($\tau_{non\text{-}rad}$) of carriers, because nonradiative recombination is the dominant process of carriers. $\tau_{non\text{-}rad}$ can be described by the product of thermal velocity of carriers (v_{th}), the capture cross section (σ) to the nonradiative recombination center (NRC), and the density (N_{NRC}) of NRC. The increase of $\tau_{non\text{-}rad}$ suggests a decrease of $v_{th}\sigma N_{NRC}$. This fact should be due to the slow carrier diffusion by In fluctuation. In fact, D of SWQ at 380 nm was very small. In this region, carriers should be localized in the fluctuation of In composition, which acts as radiative center (RC) of carriers. This localization contributes to the high η_{ext} value. On the other hand, η_{ext} was reduced for a large amount of In. This fact is often interpreted as the increment of NRC by the degrading crystal properties for a large amount of In. However, Fig. 2 shows that both $\tau_{PL}(\tau_{non\text{-}rad})$ and $D(v_{th})$ become larger with In composition. Thus, $N_{NRC}\sigma$ must become smaller with increasing In content. This fact suggests that the NRC should not increase and so it should not be the origin of the reduction of η_{ext}. Otherwise, the fast diffusion of carriers should be due to the delocalization of carriers, which is a negative factor for η_{ext}.

D should depend on the spatial inhomogeneity of In composition. By using scanning near-field optical microscopy (SNOM), we have been observed the spatial inhomogeneity of PL intensity (In fluctuation) of the InGaN/GaN SQW [15]. Similar inhomogeneity of PL was observed by using scanning confocal laser microscopy (SCLM). Figure 3 shows the SCLM image and the spectrum for LED (540 nm). A PL inhomogeneity within micron scale was observed. This scale of In fluctuation is as long as the diffusion length of carriers. In the scale of recombination pathways, In composition may be homogeneous. Such microscopic homogeneity should be the reason of the fast diffusion of carriers. The PL spectra for each region were also shown in Fig. 3 (right part). It was found that the PL intensities were smaller in the In rich region (longer PL wavelength). This behavior is opposite to the case of UV-blue region and suggests that the In rich region should not act as RC. Another reason of the fast diffusion of carriers in this region may be the effect of a piezoelectric field (PEF). PEF is induced by the large amount of In and fluctuates along the In fluctuation within micron scale. Such a fluctuation of PED causes carrier acceleration in lateral direction.

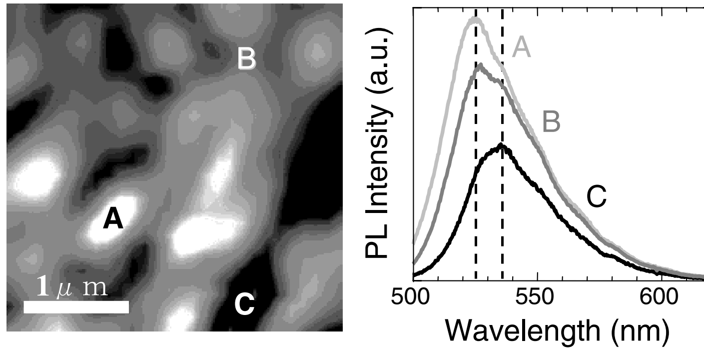

Fig. 3. Image (left part) and PL spectrum (right part) of the scanning confocal laser microscope taken for InGaN/GaN LED structure with emission at 540 nm at room temperature

In conclusion, we propose that the main reason of the reduction of η_{ext} for a large amount of In is not the increment of NRC, but the delocalization of carriers due to fast diffusion. To develop new devices with higher η_{ext} RC in the active layers must be effective.

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References

[1] T. MUKAI and S. NAKAMURA, Jpn. J. Appl. Phys. **38**, 5735 (1999).
[2] T. MUKAI, M. YAMADA, and S. NAKAMURA, Jpn. J. Appl. Phys. **37**, L1358 (1998).
[3] H. HAAG, P. GILLIOT, R. LÉVY, B. HÖNERLAGE, O. BRIOT, S. RUFFENACH-CLUR, and R. L. AULOMBARD, Appl. Phys. Lett. **74**, 1464 (1999).
[4] S. PAU, J. KUHL, F. SCHOLZ, V. HAERLE, M. A. KHAN, and C. J. SUN, Appl. Phys. Lett. **72**, 557 (1998).
[5] BAHMAN TAHERI, J. HAYS, J. J. SONG, and B. GOLDENBERG, Appl. Phys. Lett. **68**, 587 (1996).
[6] T. AOKI, G. MOHS, M. KUWATA-GONOKAMI, and A. A. YAMAGUCHI, Phys. Rev. Lett. **82**, 3108 (1999).
[7] H. HAAG, B. HÖNERLAGE, O. BRIOT, and R. L. AULOMBARD, Phys. Rev. B **60**, 11624 (1999).
[8] K. OKAMOTO, Y. KAWAKAMI, SG. FUJITA, and M. TERAZIMA, Anal. Sci. **17**, 312 (2001).
[9] K. OKAMOTO, Y. KAWAKAMI, SG. FUJITA, M. TERAZIMA, and S. NAKAMURA, Proc. Internat. Workshop Nitride Semiconductors, Nagoya (Japan), IPAP Conf. Ser. **1**, 540 (2000).
[10] S. NAKAMURA, Jpn. J. Appl. Phys. **30**, L1705 (1991).
[11] H. KOGELMIK, Bell. Syst. Tech. J. **48**, 2909 (1969).
[12] X. ZHANG, P. KUNG, D. WALKER, J. PIOTROWSKI, A. ROGALSKI, A. SAXLER, and M. RAZEGHI, Appl. Phys. Lett. **67**, 2028 (1995).
[13] S. J. ROSNER, E. C. CARR, M. J. LUDOWISE, G. GIROLAMI, and H. I. ERIKSON, Appl. Phys. Lett. **70**, 420 (1997).
[14] T. SUGAHARA, H. SATO, M. HAO, Y. NAOI, S. KURAI, S. TOTTORI, K. YAMASHITA, K. NISHINO, L. T. ROMANO, and S. SAKAI, Jpn. J. Appl. Phys. **37**, L398 (1998).
[15] A. KANETA, T. IZUMI, K. OKAMOTO, Y. KAWAKAMI, SG. FUJITA, Y. NARITA, T. INOUE, and T. MUKAI, Jpn. J. Appl. Phys. **40**, 102 (2001).