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Direct Evidence of Nanoscale Carrier Localization in InGaN/GaN Structures Grown on Si Substrates

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InGaN insertions in a GaN matrix provide efficient lateral carrier localization on a nanoscale level as established by nonresonant and resonant time-resolved photoluminescence studies. In the case of resonant excitation the line width is defined by the excitation laser pulse only, while the decay characteristic remains the same as that in the case of nonresonant excitation. No spectral shift of the resonant photoluminescence peak which could be caused by reducing the piezoelectric screening of nonequilibrium carriers was found. This demonstrates that only one electron-hole pair may be generated in a compositional domain, which must have a strong localization and wide energy spacing. [DOI: 10.1143/JJAP.42.L1057]

KEYWORDS: time-resolved photoluminescence, quantum dots, InGaN, Si substrate

There exists a strong continuous interest in studying the optical properties of InGaN/GaN heterostructures which has been motivated by the successful fabrication of InGaN/GaN high-brightness light emitting diodes (LEDs)[1)] and laser diodes[2,3)] operating in the violet and blue-green spectral ranges. Many research groups are active in this field (see[4)] for example and references therein), however, to date there is still a widely discussed controversy regarding the understanding of the basic processes responsible for light emission in these structures. On the one hand, wurtzite GaN-based semiconductors generate strong piezoelectric fields due to the high piezoelectric constants in these materials. As a result, a spectral shift of the bandgap transition energy is expected due to a quantum confinement Stark effect in the thin layers.[5–7)] On the other hand, similar effects can be explained by the saturation of compositional fluctuations in the InGaN layers having a limited density of states,[8,9)] or by the quantum dot (QD) nature of the InGaN insertions.[10,11)]

Currently available commercial blue and green GaN-based light emitters are solely grown on sapphire and SiC substrates by metal-organic vapor phase epitaxy (MOVPE). This technology suffers from several disadvantages such as the lack of large-area high-quality substrates, the low thermal conductivity of sapphire, problems with processing Al_2O_3, and the need for elaborate contacting and etching. The conducting SiC substrates are more expensive and are limited in size. Thus, heteroepitaxial growth of GaN on silicon substrates seems to be a promising alternative solution, which also allows the monolithic integration of light emitters on a Si chip. Recently, several groups have reported on the successful growth of GaN-based devices on Si substrates (see ref. 12 for example and references therein). In this paper we present data of resonant photoluminescence (PL) and time-resolved (TR) PL studies which evidences the lateral localization of nonequilibrium carriers in our InGaN-GaN insertions on Si substrates.

The structure investigated in this paper were grown on a Si(111) substrate by metal-organic vapor phase epitaxy (MOVPE).[13)] Trimethylgallium (TMG), trimethylaluminum (TMA), trimethylindium (TMI) and ammonia were used as precursors. The structure consists of a 1.2-μm-thick GaN buffer layer grown on an AlN nucleation layer obtained by nitridation of a previously deposited AlAs layer. The active region consists of 5 periods of 1.5-nm-thick InGaN/3-nm-thick GaN superlattice and a 50-nm-thick GaN cap layer. The GaN buffer and cap layers were grown utilizing H_2 as a carrier gas at the temperature of 1150°C. During growth of the active region, N_2 was used as carrier the gas and the formation of the InGaN/GaN superlattice was performed while the temperature was periodical changed from 750°C to 1100°C. The PL was measured at low temperature (2 K) and samples were excited either by a doubled Ti-sapphire laser (for resonant PL investigations) or by tunable dye lasers with a repetition rate of 3.79 MHz at wavelengths of 291 nm (doubled 582 nm) and 427 nm with an excitation density of 600 W/cm^2 (for TRPL investigations). In order to pump the dye lasers the second and third harmonics of a Nd-YAG laser were used. The TRPL measurements were carried out at 2 K using a single photon counting technique. The excitation was performed by using the above described laser systems with typical pulse widths of 2 ps, either excited above the GaN band gap or resonantly via excited states transitions. The time resolution of the detection system was 30 ps. In order to optimize a dynamic range of PL signals at several wavelengths calibrated gray filters were used.

In Fig. 1(a) a typical PL spectrum of the InGaN-GaN/Si sample recorded under nonresonant excitation is presented. One can clearly see a broad band between 2.6 eV and 3.3 eV originating from the active InGaN region. This PL line is spectrally modulated due to a light interference effect within the epitaxial slab. Si/GaN and air/GaN interfaces provide large refractive index steps causing a strong reflection at these interfaces. On the basis of the period of oscillation we experimentally obtained a structure thickness (~1.6 μm), which agrees fairly well with the thickness expected on the basis of the growth rate calibration. Resonant excitation (Fig. 1(b)) does not affect the PL line shape except for the vanishing of the defect "yellow" line originating from the GaN buffer layer. The change of the excitation wavelength results only in a change of the PL intensity. A PL excitation (PLE) spectrum is shown in Fig. 1(b) by solid squares. This spectrum was obtained by plotting the intensity of the resonant PL spectra at 2.76 eV as a function of the excitation

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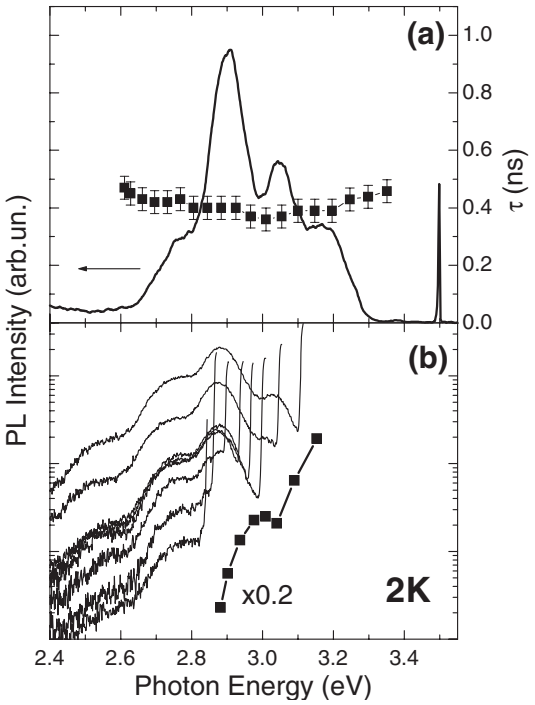

Fig. 1. (a) Time-integrated photoluminescence (PL) spectrum measured under nonresonant excitation (solid line) as well as spectral dependencies of initial time constant. (b) Time-integrated PL spectra measured under resonant excitation (with excitation energies of 3.153, 3.090, 3.040, 3.009, 2.976, 2.936, 2.902, and 2.881 eV). Solid squares present a PL excitation spectrum which is obtained by plotting the intensity of resonant PL spectra at 2.76 eV as function of excitation energy.

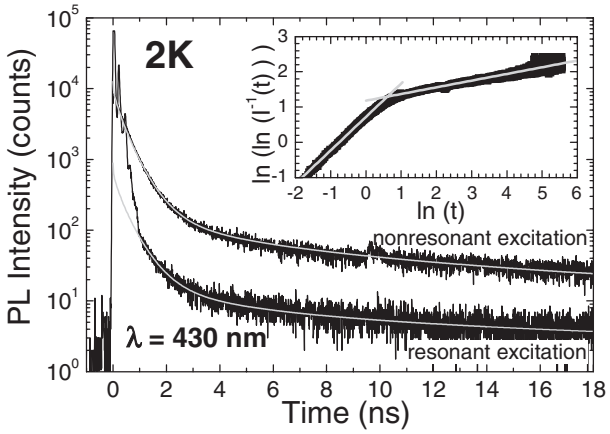

Fig. 2. Time-resolved PL under nonresonant and resonant excitation at 430 nm. Gray lines correspond to the fitting by eq. (1). Insert shows double logarithm of signal *versus* logarithm of time.

photon energy. One can observe that there is a peculiarity in the PLE spectrum at about 3.0 eV. The maximum of the PL spectrum (~2.9 eV) and the peculiarity on the PLE spectrum (~3.0 eV) are separated by more than 100 meV. This behavior can be explained within the framework of the higher oscillator strength of excited subband transitions in a quantum well (QW) subjected to a high piezoelectric field, by the existence of mesoscopic localized states due to long-range disorder in the quantum well, or by the peculiarities of PLE spectra in QD structures.[14] In the last case, the PLE maximum corresponds to the first QD exciton excited state. This effect is well documented for 3D InAs QDs in a GaAs matrix which are proven to have a quasi-ideal δ-function-like density of states.[14,15] Among the three possible scenarios, the correct explanation can be chosen on the basis of resonant time-resolved studies. In all possible "QW" cases resonant excitation within the continuum of the PL spectrum results in a rapid disappearance of the resonant PL signal. In QDs, the ground state which fits the detection energy may be populated either resonantly or via the excitation in the nearest excited state which can be shifted significantly to higher energies from the ground state absorption energy. For predominantly small QDs having a large energy separation between ground and excited QD exciton states, resonant excitation results in a resonant population of QD ground states with a slow PL decay time similar to that observed under nonresonant excitation.

The results of the TR PL studies are presented in Fig. 2. The decay PL processes under nonresonant and resonant excitation have the same highly nonexponential character and the same time constants. (The rapid decay at near zero moment (even with few maximums) under resonant excitation is the response of the detection system to the bright and fast excitation laser pulse.) The fitting law for this behavior is a stretched exponent,[16,17] which is typical of disordered media.[18] In order to determine the fitting parameters we plotted on the insert in Fig. 2 the double logarithm of the signal *versus* the logarithm of the time. On this insert the PL behavior can be easily fitted by two straight lines with slopes of 1.0 and 0.2. Thus, we used the following equation for the fitting procedure:

$$I(t) = A_1 \exp[-(t/\tau_1)]^{\beta_1} + A_2 \exp[-(t/\tau_2)]^{\beta_2} \qquad (1)$$

where $\beta_1 = 1.0$ and $\beta_2 = 0.2$. Time constants (τ_1 and τ_2) have values of approximately 0.4 ns and are practically independent from the registration energy. In order to illustrate this we plot in Fig. 1(a) a dependence of the initial time constant on the registration energy. This behavior is typical for InAs/GaAs QDs,[19] while for the QW system, the time constant should drop practically to zero (<0.1–1 ps) on the high-energy side of the PL peak.[20] Even in a model of highly disordered InGaN QWs a continuum of the states exists if the characteristic scale of disorder is greater than few tens of nanometers. Moreover, the carrier hopping to lower energy states due to small barrier heights between the neighboring localized states must occur on a relatively slow time scale, causing a significant delay in the evolution of the longer wavelength PL emission. In our case the PL signal appears with an onset time faster than 7 ps, which is very close to our time resolution (5 ps), at all of the wavelengths under nonresonant excitation, as one can expect for QDs with fast carrier relaxation from the excited to the ground state.

In order to confirm our suggestion regarding the QD nature of the InGaN-GaN structures on Si, we performed resonant TR PL investigations in a manner similar to the one reported for InGaN-GaN multilayers on sapphire.[21] As follows from the continuum nature of the density of states in QWs, resonant excitation at energies significantly above the PL onset energy results in fast carrier relaxation towards lower energy states accompanied by significant spectral diffusion of the remaining resonant PL peak towards a lower

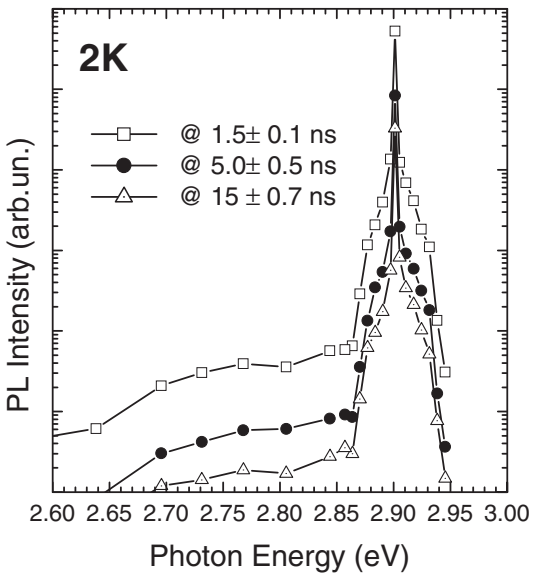

Fig. 3. Time-delayed PL spectra at several time delay after laser pulse (□-1.5 ns, ●-5.0 ns and △-15 ns) measured under resonant excitations.

energy as well as its broadening. In addition, in the case of wurzite-type strained QWs, reduced carrier concentration upon the radiative recombination of nonequilibrium carriers results in reduced screening of the piezoelectric potential, which further shifts PL towards the longer wavelength side of the spectrum.

In opposition to this prediction, the effect was not revealed in our case. Most of the PL intensity, after the exciting laser light is gone, exactly resembles the spectral shape of the excitation pulse, while the decay time of the resonant PL corresponds to the radiative decay behavior and the time scale similar to that under nonresonant excitation (Fig. 2). The PL peak position does not shift in time and no spectral broadening is observed (Fig. 3). This behavior clearly evidences that the electron hole-pair is laterally localized, that most of the domains have a very small size, and that only the ground state is populated. Additionally, a complete lack of the spectral shift indicates that no screening of the piezoelectric field takes place in the nanodomain during the PL decay process, evidencing that only one electron-hole pair is generated upon excitation. Thus, the InN-rich compositional domains apparently have a predominantly nanoscale size and wide energy spacing between the energy levels, representing a QD. Assuming the energy spacing of approximately 100 meV, one may expect lateral size of approximately 3–4 nm.

An additional weak shoulder is revealed on the longer wavelength side of the PL spectrum. The shoulder appears on a time scale shorter than 7 ps after the laser pulse and can be attributed to the fast energy transport from the QD exciton excited state towards its ground state. Due to the significant size dispersion of the QDs, the larger QDs may have an excited state energy which corresponds to the ground state energy of most other QDs.

In order to explain the two-exponential behavior of the PL decay process one may suppose that two different types of nanodomains take part in recombination processes, in opposition to the case of InGaN-GaN QDs on sapphire substrates, where no such behavior was observed. The two types of localized states can originate, for example, from flat InGaN insertions having a higher InN composition and from higher three-dimensional islands having a stronger piezoelectric impact on the electron and hole wavefunction overlap and, thus, a longer radiative lifetime. In the case of flat domains, the piezoelectricity effects must play a minor role and the overlap of electron and hole wavefunction (and, consequently, decay time) is weakly dependent on the domain nonuniformity in agreement with a simple exponential decay with $\beta_1 = 1.0$. In 3D domains piezoelectric fields strongly depend on the size and the shape of QDs. Thus, QDs having a very different wavefunction overlap may coexist at the same transition energy. This results in a highly nonexponential decay with $\beta_1 = 0.2$. It is important to note that these values are in good agreement with our previous results,[22] obtained for InGaN-GaN structures on sapphire substrates. Higher nonuniformity revealed by a wider linewidth of the PL line under nonresonant excitation correlates with lower values of β, and thus evidences a stronger deviation from simple exponential behavior.

In conclusion, resonant and nonresonant TRPL studies of InGaN/GaN structures grown on Si substrate evidence an inhibition of the relaxation processes. The observations are very similar to those reported for well documented and extensively studied InAs/GaAs QDs except for the strongly non-exponential resonant PL decay process. The latter observation is in full agreement with the expected far stronger impact of piezoelectric fields on the electron and hole wavefunction overlap in InGaN-GaN-based QD structures. The two-exponential behavior of the PL decay process may be explained by two different types of nanodomains taking part in recombination processes. This behavior strongly differs from the case of InGaN-GaN QDs on sapphire substrates, where no such effects were observed.

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1) S. Nakamura, T. Mukai and M. Senoh: Jpn. J. Appl. Phys. **32** (1993) L8.
2) S. Nakamura, M. Senoh, S. Nagahama, N. Iwasa, T. Matsushita, H. Kioku and Y. Sugimoto: Jpn. J. Appl. Phys. **35** (1996) L74.
3) I. Akasaki, S. Sota, H. Sakai, T. Tanaka, M. Koike and H. Amano: Electron. Lett. **32** (1996) 1105.
4) H. Morkoc: *Nitride Semiconductors and Device* (Springer-Verlag, Berlin, Heidelberg, 1999).
5) T. Takeuchi, Sh. Sota, M. Katsuragawa, M. Komori, H. Takeuchi, H. Amano and I. Akasaki: Jpn. J. Appl. Phys. **36** (1997) L382.
6) Ch. Wetzel, T. Takeuchi, H. Amano and I. Akasaki: Jpn. J. Appl. Phys. **38** (1999) L163.
7) L.-H. Peng, C.-W. Chuang and L.-H. Lou: Appl. Phys. Lett. **74** (1999) 795.
8) S. Chichibu, T. Azuhata, T. Sota and S. Nakamura: Appl. Phys. Lett. **69** (1996) 4188.
9) O. Moriwaki, T. Someya, K. Tachibana, S. Ishida and Y. Arakawa: Appl. Phys. Lett. **76** (2000) 2361.
10) N. N. Ledentsov, Zh. I. Alferov, I. L. Krestnikov, W. V. Lundin, A. V. Sakharov, I. P. Soshnikov, A. F. Tsatsul'nikov, D. Bimberg and A. Hoffmann: Compd. Semicond. **5** (1999) 61.
11) A. V. Sakharov, W. V. Lundin, I. L. Krestnikov, V. A. Semenov, A. S.

Usikov, A. F. Tsatsul'nikov, Yu. G. Musikhin, M. V. Baidakova, Zh. I. Alferov, N. N. Ledentsov, J. Holst, A. Hoffmann, D. Bimberg, I. P. Soshnikov and D. Gerthsen: Phys. Status Solidi (b) **216** (1999) 435.
12) A. Dadgar, M. Poschenrider, O. Contreras, J. Christen, K. Fehse, J. Blasing, A. Diez, F. Schulze, T. Riemann, F. A. Ponce and A. Krost: Phys. Status Solidi (a) **192** (2002) 308.
13) A. Strittmatter, A. Krost, J. Blasing and D. Bimberg: Phys. Status Solidi (a) **176** (1999) 611.
14) N. N. Ledentsov, M. Grundmann, N. Kirstaedter, O. Schmidt, R. Heitz, J. Böhrer, D. Bimberg, V. M. Ustinov, V. A. Shchukin, P. S. Kop'ev, Zh. I. Alferov, S. S. Ruvimov, A. O. Kosogov, P. Werner, U. Richter, U. Gösele and J. Heydenreich: Solid State Electron. **40** (1996) 785.
15) D. Bimberg, M. Grundmann and N. N. Ledentsov: *Quantum Dot Heterostructures* (Wiley & Sons, Berlin, 1999).
16) M. Pophristic, F. H. Long, C. Tran, I. T. Ferguson and R. F. Karlicek, Jr.: Appl. Phys. Lett. **73** (1998) 3550.
17) M. Pophristic, F. H. Long, C. Tran, I. T. Ferguson and R. F. Karlicek, Jr.: J. Appl. Phys. **86** (1999) 1114.
18) H. Scher, M. F. Shlesinger and J. T. Bender: Phys. Today **26** (1991) 24.
19) R. Heitz, M. Veit, N. N. Ledentsov, A. Hoffmann, D. Bimberg, V. M. Ustinov, P. S. Kop'ev and Zh. I. Alferov: Phys. Rev. B **56** (1997) 10435.
20) C. Gourdon and P. Lavallard: Phys. Status Solidi (b) **153** (1989) 641.
21) I. L. Krestnikov, N. N. Ledentsov, A. Hoffmann, D. Bimberg A. V. Sakharov, W. V. Lundin, A. F. Tsatsul'nikov, A. S. Usikov, Zh. I. Alferov, Yu. G. Musikhin and D. Gerthsen: Phys. Rev. B **66** (2002) 155310.
22) I. L. Krestnikov, A. V. Sakharov, W. V. Lundin, A. S. Usikov, A. F. Tsatsul'nikov, Yu. G. Musikhin, D. Gerthsen, N. N. Ledentsov, A. Hoffmann and D. Bimberg: Phys. Status Solidi (a) **192** (2002) 49.