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Impact of carrier localization and diffusion on photoluminescence in highly excited cyan and green InGaN LED structures

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ABSTRACT

Localization of charge carriers is of crucial importance in InGaN light emitting devices since it governs carrier transport and ensures high radiative efficiency. In this work, we observe the dynamics of carrier localization from an untypical redshift of photoluminescence spectra. We investigate two light emitting diode structures grown by MOCVD on c-plane sapphire and emitting at 500 nm (cyan) or 530 nm (green). For the study, we employ photoluminescence, differential transmission, and light induced transient gratings techniques. We observe non-monotonous dynamics of photoluminescence peak position: a blueshift at short delay times (< 10 ns) is later replaced by a redshift, which increases both with time and excitation. The redshift values as large as 30 meV in the cyan and 20 meV in the green structure were observed at 40 ns delay. We attribute this redshift to density-dependent and diffusion-driven carrier redistribution between the shallower and deeper localized states. The carrier delocalization with increasing density is confirmed by growing diffusivity of carriers with excitation. We also demonstrate a correlation between the growth of diffusion coefficient and the onset of efficiency droop.

1. Introduction

InGaN/GaN quantum structures are attractive for various lighting applications due to high internal quantum efficiency (IQE) reaching 90% for blue and 30% for green light emitting diodes (LEDs) [1]. High IQE values are obtained even in the heteroepitaxial structures despite the high density of threading dislocations exceeding 10^8–10^9 cm^{-2}. Localization of free carriers in the local potential minima is believed to be responsible for low nonradiative recombination rate at dislocations [2]. Quantum dot-like indium clusters were believed to be the main source of local potential minima [3], but later it has been shown that both random indium density fluctuations and monoatomic variations in quantum well thickness can result in carrier localization even at room temperature [4]. Recently, it was demonstrated that localization is much stronger for holes than electrons due to higher effective mass of the former [5–7].

Role of carrier localization becomes stronger with the increasing indium content since it is accompanied by a larger disorder in distribution of indium atoms [8]. On the other hand, higher indium content also results in stronger internal electric fields, higher strains, and larger defect densities. Stronger carrier localization can further separate the carriers from the defect states, but it also results in the decrease of radiative recombination coefficient [8,9] due to non-correlated potential fluctuations of valence and conduction bands and a smaller overlap of electron and hole wavefunctions [10,11]. Carrier localization can also alter the nonradiative recombination (SRH and Auger) rates [9,12]. An opposite process of carrier delocalization takes place at high temperatures and/or carrier densities, which increases carrier mobility and nonradiative recombination rate leading to the onset of efficiency droop at carrier densities that are lower than those required for Auger recombination [13–15].

In this paper, we study the impact of localization to carrier dynamics by analyzing the time and excitation-dependent spectra of photoluminescence and absorption, as well as diffusion coefficient. For this, we use several optical techniques: time-resolved and time-integrated photoluminescence (TRPL and TIPL), differential transmission (DT), and light induced transient gratings (LITG). Concerted application of PL and DT enables monitoring the electronic transitions in both the localized and extended states, while LITG provides a unique possibility to directly measure and separate the carrier lifetime and diffusivity. This allows for overcoming a certain limitation of SNOM and confocal microscopy, were only a product of carrier diffusion coefficient and

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carrier lifetime – carrier diffusion length – can be measured [16–18]. As a result, we reveal an untypical redshift in PL peak position, which increases with time and carrier density and cannot be related neither to band gap renormalization nor to electric field dynamics. Based on the analysis of DT and PL data, we attribute this shift to a slow relocalization of carriers from shallower to deeper potential minima. We show that carrier diffusivity increases with density due to delocalization and discuss the correlation between the growth of diffusivity and the onset of efficiency droop.

2. Samples and techniques

We investigate two complete InGaN/GaN multiple quantum well (MQW) LED structures, grown by metalorganic chemical vapor deposition on c-plane sapphire substrates. They emit light in the cyan and green spectral ranges with characteristic peak wavelengths of 500 nm and 530 nm (hereafter, the structures are referred to as "cyan" and "green", respectively). The samples consist of a standard sequence of LED epilayers: a buffer layer (unintentionally doped GaN), a moderately n-doped GaN:Si current spreading layer followed by a highly n-doped GaN:Si contact layer, an active layer consisting of a stack of five InGaN QWs with 3 nm well width (with an indium content of ~26% for cyan and ~30% for green emission), separated by GaN barriers, and a p-doped AlGaN:Mg electron blocking layer as well as a p-type GaN:Mg contact layer on top. Since the growth processes and layer structures of both samples are basically identical, we estimate that the In distributions are fairly similar, although the amplitude of variations might be a bit larger in the sample with higher In content [19].

TRPL, TIPL, DT, and LITG measurements were carried out using 250 fs duration pulses generated by a PHAROS (Light Conversion) laser (1030 nm, 30 kHz repetition rate) and spectrally tuned with an ORPHEUS (Light Conversion) optical parametric amplifier. Pump wavelength was set at 392 nm (DT, TIPL, LITG) or 405 nm (TRPL) to selectively excite the quantum wells. Pump energy fluence (further, we will be using a term "excitation") as high as 1 mJ/cm^2 per pulse could be achieved. A Hamamatsu C10627 streak camera along with an Acton SP2300 spectrometer was used for the measurements of TRPL spectra and kinetics. TIPL spectra were measured using a fiber spectrometer AvaSpec ULS 2048 (300 mm^{-1}).

To calibrate and compare the quantum efficiency in the structures, an integrating sphere and a three-measurement approach [20] were employed instead of a temperature-dependent PL method. The latter is based on an assumption that nonradiative recombination is thermally activated and vanishes at low temperatures. Recently, it has been shown that this assumption lacks a fundamental support [21]. To estimate the defect-related absorption coefficient at 392 nm in GaN buffer, the test samples were produced by removing the active LED structures by reactive ion etching. The measured absorption coefficient values in quantum wells and GaN buffer were equal to 8×10^4 cm^{-1} and 2×10^2 cm^{-1}, correspondingly. These values were used for estimation of photogenerated carrier density and quantum efficiency. We assume that light extraction coefficient is the same in both samples since they have the same structure.

White light continuum pulses within a 380–550 nm spectral range were created in a sapphire window and used as a probe in DT. Transmittance with and without pump ($T_p(\lambda, t)$ and $T_0(\lambda)$, correspondingly) at various probe delays and spectral positions was recorded using a Harpia system (Light Conversion) with a spectrograph with 300 lines/mm grating and a CCD camera. Differential transmittance is further presented in a scale of mOD as:

$$T(\lambda, t) = 1000\times\log\left(\frac{T_0(\lambda)}{T_p(\lambda, t)}\right). \quad (1)$$

In LITG measurements, the sample was excited by a spatially-modulated interference field of two coherent pump beams, which created a transient spatially modulated free carrier pattern

$$N(x) = N_0+\Delta N\left(1+\cos\left[\frac{2\pi x}{\Lambda}\right]\right) \quad (2)$$

with the grating spacing Λ [22]. For probing, the pulses at 1030 nm were used, since the sample is transparent for this wavelength. Diffraction efficiency $\eta(t)$ of a transient grating is given as

$$\frac{I_{diffracted}(t)}{I_{transmitted}(t)} = \eta(t)\propto\exp\left(\frac{-2t}{\tau_g}\right), \quad (3)$$

where the grating decay time τ_g consists of recombination and diffusion times τ_R and τ_D as

$$\frac{1}{\tau_g} = \frac{1}{\tau_R}+\frac{1}{\tau_D}, \quad (4)$$

where $\tau_D = \Lambda^2/4\pi D$. Since τ_D depends on Λ, the bipolar diffusion coefficient D can be determined by performing measurements with different induced grating periods. LITG technique has been successfully used previously for lifetime and diffusivity measurements in GaN and InGaN layers and structures [23–25].

All measurements were performed at room temperature.

3. Results and discussion

Fig. 1 shows the TIPL (top pictures) and DT (bottom pictures) spectra in the cyan (a) and green (b) QW samples recorded at various excitation levels. At low excitation (10 μJ/cm^2), the PL lines are centered at 2.53 eV (490 nm, cyan) and 2.36 eV (525 nm, green) and have full width at half maxima (FWHM) around 140 meV for both samples. The QE curves (see Fig. 4(b)) are typical for the optically pumped InGaN QWs [26], with the onset of efficiency droop at ~60–70 μJ/cm^2 (this corresponds to the carrier density of ~10^{19} cm^{-3}). The measured peak QE values reach 18 ± 3% in the cyan and 19 ± 3% in the green structures; we note that light extraction was not optimized in the structures. Since the PL linewidth is determined by the fluctuations of hole localization energy due to random alloy disorder [27] and a direct link was shown to exist between the disorder and luminosity of polar InGaN layers [28], the similar FWHM and QE of PL emission indicate similar structural quality of investigated structures, despite higher indium content in the green QWs. DT signal has a negative sign (bottom pictures in Fig. 1) indicating the absorption bleaching due to the state filling effect [29]. DT spectra are shifted towards the higher photon energies with respect to the PL peak by ~230 meV (cyan) and ~360 meV (green). Stokes shift is considerably larger in the green sample, which points out to stronger carrier localization in this structure [30].

Two trends are visible in TIPL spectra with increasing excitation: (i) the width of PL spectra increases and (ii) a redshift in PL peak position at low excitations is followed by a blueshift at high excitations; this is especially noticeable in the cyan sample. The PL line broadening with increasing carrier density has been attributed to gradual saturation of shallow localized states and the consequent population of both higher and lower energy states [31,32]. The behavior of PL line position, however, is untypical: in polar QWs, one expects a PL blueshift with excitation due to screening of the internal electrical field and state filling [33]. Only at very high carrier densities, a redshift of PL band due to band gap renormalization has been observed [34].

To investigate the temporal evolution of PL peak position, TRPL and DT spectra were recorded at various excitations. Fig. 2 displays the PL and DT peak positions for several delay times as a function of excitation in the cyan sample. It is seen from Fig. 2(a) that the PL redshift at high excitations evolves in time span of tens of nanoseconds. Thus, within first 5 ns only a blueshift in PL peak position is seen. A closer look into the early stage of spectral dynamics is provided by DT measurements with better time resolution (Fig. 2(b)). Only the blueshift of DT spectra