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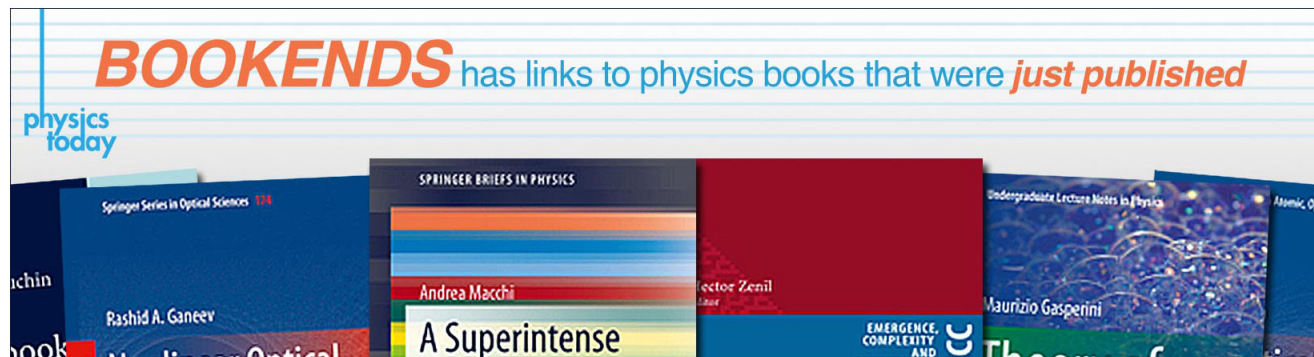
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# Time-resolved cathodoluminescence and photocurrent study of the yellow band in Si-doped GaN

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Time-resolved cathodoluminescence (TRCL) and photocurrent (PC) spectroscopies have been applied to the study of the yellow band of Si-doped GaN. Measurements carried out combining both techniques unambiguously reveal the complex nature of this broad emission and confirm that different deep defect levels are involved in the observed luminescence. Five emission bands centered at 1.89, 2.03, 2.16, 2.29, and 2.38 eV were found by steady state and time-resolved CL investigations, while PC spectra showed four transitions at about 2.01, 2.14, 2.28, and 2.43 eV. The behavior of the deep-level emissions intensity as a function of the excitation pulse width as well as their decay times were investigated by TRCL. A decay time of 245  $\mu$ s was measured for the 2.29 eV emission band, while longer decay times of 315 and 340  $\mu$ s were found, respectively, for the 2.16 and 2.38 eV bands, in agreement with TRCL spectra. The appearance of the 2.03, 2.16, 2.29 eV and 2.38–2.43 eV peaks both in PC and CL spectra suggests that these bands are related to deep acceptor to band transitions, as supported by the single exponential character of the corresponding decay transients. © 2003 American Institute of Physics. [DOI: 10.1063/1.1592296]

## I. INTRODUCTION

During recent years much effort has been devoted to the study of gallium nitride (GaN) and related compounds due to their multiple applications in optoelectronic devices and high-power/high-temperature electronics. However, the role of defects controlling the electrical and luminescence properties of these materials is still not fully understood. This is especially the case for deep level defects. In particular, a broad emission centered at about 2.2–2.3 eV, known as the yellow band, is frequently observed in GaN films grown by different methods. Yellow luminescence has been suggested<sup>1</sup> to be detrimental for GaN-based optoelectronic devices since it represents a competing recombination path that reduces the intensity of the near-band gap emission. The origin of this band remains unclear and different models, including transitions from a shallow donor or the conduction band to a deep acceptor,<sup>2–4</sup> from a shallow donor to a deep donor,<sup>5</sup> and from a deep donor to a shallow acceptor,<sup>6</sup> have been proposed to explain that emission. Whether this band is related to a point defect<sup>4</sup> or to a distribution of states inside the gap<sup>7</sup> is still an open question. The microscopic origin of the defects responsible for the yellow band is also under discussion and a concluding relation between such emission and the extended defects has not been established yet. Moreover, photoluminescence (PL) decay times have been measured by different research groups for the yellow band range from 1 ms to less than 1 ns.<sup>5,8,9</sup> Variations in peak position, shape,

and decay times reported in different studies suggest that several bands involving different defects could contribute to this emission.<sup>10–12</sup>

Various spectroscopic techniques have been applied to the study of gap states in GaN. Among these, cathodoluminescence (CL) in the scanning electron microscope (SEM) has been frequently used to obtain information on the spatial distribution of the different emission bands and their association to point and extended defects, e.g., Refs. 12–15. Electron beam excitation usually leads to light emission by all mechanisms of radiative recombination present in a semiconductor, while PL is a much more selective technique. Several investigations concerning the recombination kinetics of GaN defect centers have been carried out by time-resolved PL.<sup>5,8,9,16</sup> Time-resolved cathodoluminescence (TRCL) experiments represent a very powerful tool to gain information on charge carrier lifetimes and to distinguish different capture processes by separate measurements of the transient behavior of the various bands of luminescence spectrum. Principles and applications of TRCL are described in Ref. 17. Another spectroscopic technique, spectrally resolved photocurrent (PC), has also been recently applied to the study of trapping processes in GaN.<sup>18,19</sup> As compared with CL, which provides evidence of radiative internal and band-to-level transitions, PC allows detection of radiative and nonradiative level-to-band and band-to-band transitions.

In this work, the capabilities of TRCL spectroscopy combined with the high sensitivity of the PC technique have been used to investigate the yellow band in Si-doped GaN. Our measurements unambiguously reveal that this broad emission is actually composed of several bands with peak

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energies extending in the green and red ranges of the visible spectra. The decay time of each component has been determined by TRCL. Comparison of CL and PC spectra indicates that some of the observed emissions are due to deep levels to band transitions or transitions involving deep levels and very shallow levels with a maximum depth on the order of the thermal energy.

## II. EXPERIMENT

Si-doped GaN, 2  $\mu\text{m}$  thick, grown by molecular beam epitaxy (MBE) on sapphire was investigated in this work. Schottky diodes 200 Å thick were formed by gold evaporation, while ohmic contacts were prepared using soldered indium. Room temperature capacitance–voltage and Hall measurements indicate a free carrier concentration of  $n = 7 \times 10^{17} \text{ cm}^{-3}$  and a mobility of  $\mu_n = 177 \text{ cm}^2/\text{V s}$ .

CL observations were carried out in a Hitachi S-2500 SEM. Measurements were performed at accelerating voltages ranging from 5 to 20 kV and temperatures between 85 and 295 K. The experimental conditions were carefully established to avoid undesired electron beam irradiation effects. Steady state CL spectra were acquired using a charge coupled device camera with a built-in spectrograph (Hamamatsu PMA-11) providing a spectral resolution of 1 nm. In order to perform time-resolved measurements, a periodic beam was generated using a graphite chopper and beam-blanking electronics consisting of a HP 8131A function generator and a HP 6204B power supply. To record CL spectra at different delay times, the signal from a photomultiplier was collected by a boxcar integrator (PAR162) triggered by the pulse generator, and then fed to a computer (see Ref. 20 for further details). The time resolution of the TRCL setup is about 100 ns, while the spectral resolution is 2 nm. Time-resolved CL spectra were measured at delay times ranging from 500 ns to 5 ms with time windows between 50 ns and 500  $\mu\text{s}$ . The decay transients of the different CL emissions and their intensity behavior as a function of the excitation pulse width were monitored with the aid of a digital oscilloscope.

Spectral photocurrent analyses were carried out at room temperature. A white light source was focused on the entrance slit of a monochromator. From the exit slit the monochromatic beam was chopped at a 3–5 Hz frequency and focused onto the semitransparent Schottky barriers. The slit width was 500  $\mu\text{m}$ , providing a spectral resolution of 2 nm. No external bias was applied, so that charge collection was achieved by the built-in electric field of the Schottky junction. The light wavelength was changed from 300 to 620 nm (4.1–2.0 eV) and the corresponding photocurrent signal was collected by a lock-in amplifier. CL and PC spectra were corrected for the optical response of the systems used.

## III. RESULTS

CL microscopy investigations revealed quite a homogeneous luminescence distribution. Contrast observed in panchromatic images was very similar to that observed in monochromatic CL micrographs. On the other hand, CL spectra were found to depend on beam excitation conditions. In par-

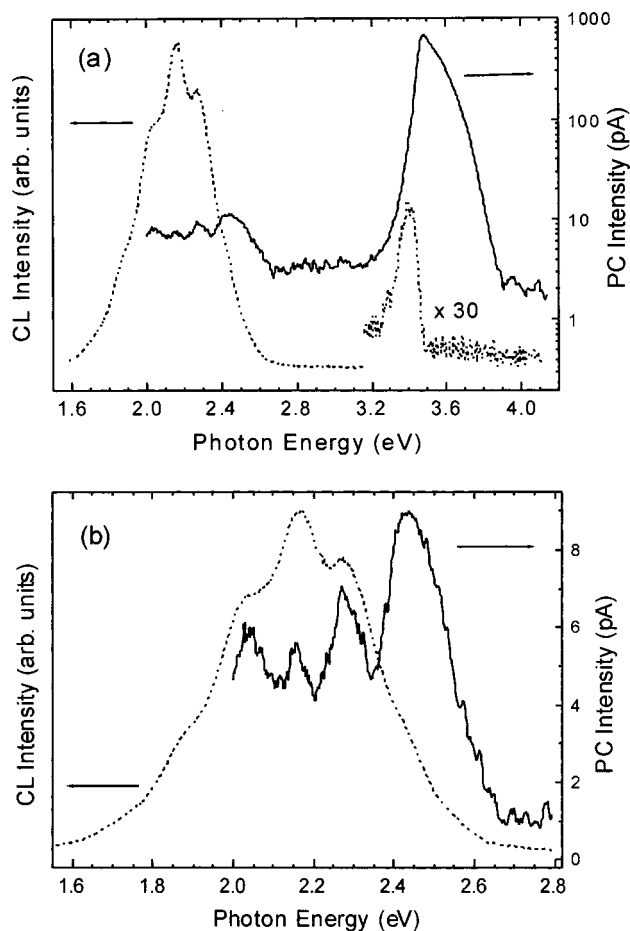


FIG. 1. (a) Room temperature PC spectrum (solid line, log. scale) and steady state CL spectrum (15 kV, 5 nA, dotted line) of a Si-doped GaN film. (b) Detail of the same spectra showing the different deep level related peaks.

ticular, a low current density favored a higher ratio of the yellow luminescence to near-band edge emission intensity, which agrees with previous CL observations performed in undoped GaN thin films.<sup>21,22</sup> Therefore, in order to enhance the yellow band emission, the CL measurements presented in this work were carried out using low beam currents ( $\sim 5 \text{ nA}$ ) or by defocusing the SEM electron beam.

Representative PC and CL spectra obtained at room temperature are shown in Fig. 1(a). In addition to the band gap excitation, the PC spectrum shows four other resolved peaks centered at 2.43, 2.28, 2.14 and 2.01 eV. Besides the near-band gap emission centered at 3.39 eV, the CL spectrum shows an intense broad yellow band centered near 2.16 eV. The complex nature of this emission can be better appreciated in Fig. 1(b), showing two well-resolved peaks at about 2.29 and 2.16 eV, plus other shoulders, respectively, centered at about 1.89, 2.03 and 2.38 eV. The correlation between the peaks appearing in the low energy range of the PC spectrum and those appearing in the CL spectrum is evident.

It should be mentioned that these peaks cannot be ascribed to the microcavity effect related to the GaN–vacuum and GaN–sapphire interface that sometimes affects PL experiments.<sup>23</sup> The mentioned set of bands was found in CL spectra collected under various excitation conditions, both in planar and cross-section configuration, although slight differ-

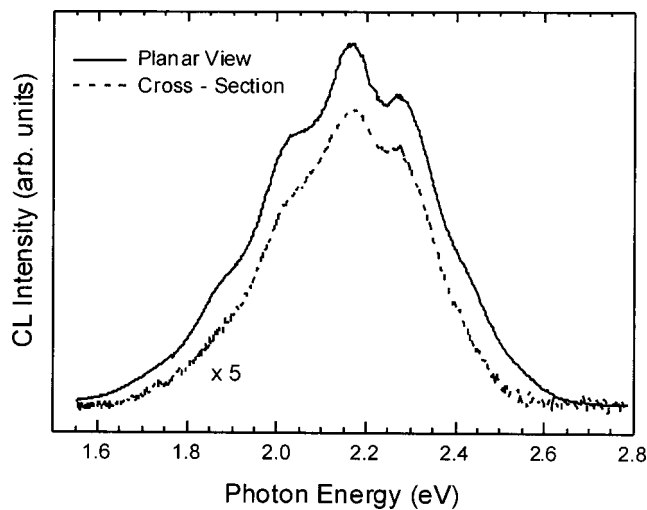


FIG. 2. Comparison of room temperature CL spectra (15 kV, 5 nA) from a Si-doped GaN film obtained in planar view (solid line) and cross-section (dashed line) configuration. The same emission bands appear in both spectra regardless of the geometry of the measurement.

ences were observed in the relative intensities of such emissions depending on the position considered. Figure 2 shows that the same CL bands are observed, regardless of the geometry of the measurement. TRCL results, that will be presented in the following, confirm that the peaks appearing in our CL spectra actually correspond to emission bands and are not caused by optical interference effects.

Figure 3 shows a low temperature (87 K) CL spectrum of a Si-doped GaN film. Near-band gap emission bands are observed at 3.477 and 3.410 eV. The peak at 3.477 eV is due to donor-bound excitons while the 3.410 eV peak has been attributed to excitons bound to extended defects.<sup>24</sup> Other peaks centered at 3.286, 3.195 and near 3.10 eV, usually attributed to shallow donor-acceptor pair (DAP) transitions, are also visible in the violet range of the spectrum. Thermal

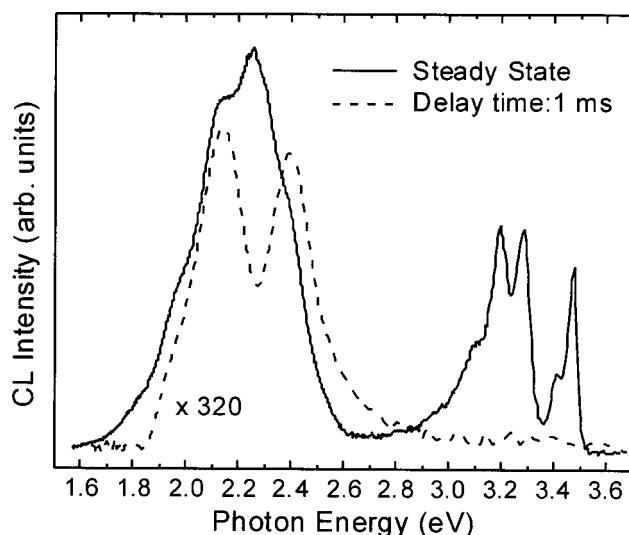


FIG. 3. CL spectra recorded at 87 K (15 kV, 5 nA). The solid line corresponds to a spectrum recorded under steady state conditions while the dashed line corresponds to a time-resolved spectrum recorded for a delay time of 1 ms and a 50  $\mu$ s time window.

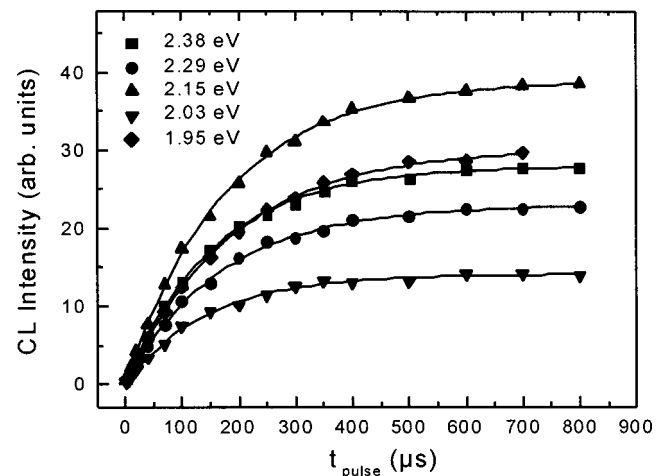


FIG. 4. CL intensity of the observed deep level related emissions as a function of the excitation pulse width ( $T=87$  K). The electron beam accelerating voltage used is 15 kV and the beam current 5 nA.

quenching of the latter bands was observed above 150 K. The yellow luminescence—centered at about 2.26 eV at this temperature—dominates the overall emission. All the deep level bands observed at room temperature are also found at 87 K, showing a slight intensity increase. Some of the emissions involved in the yellow band were resolved by TRCL due to their different decay times (Fig. 3). In particular, spectra recorded at delay times longer than approximately 400  $\mu$ s show a strong decrease of the 2.29 eV band that allows us to clearly resolve the 2.38 eV emission. No shift of the observed bands was found by increasing delay time.

Additional information on the deep-level traps can be obtained by observation of the onset of CL and its kinetics.<sup>25</sup> Figure 4 represents the CL intensity of each of the emissions involved in the yellow band as a function of the electron beam excitation pulse length. The experimental data are well fitted by the following empirical law:

$$I_{CL} = I_s [1 - \exp(-t/\tau_f)], \quad (1)$$

where  $I_s$  represents the CL intensity in the quasisteady state and  $\tau_f$ —the key parameter providing information on the trap filling kinetics—is the time for the CL intensity to reach a factor  $(1 - 1/e)$  of  $I_s$ . Results obtained after fitting our data to Eq. (1) are presented in Table I. It is worth noting that a steady state condition is not reached for any of the observed emissions with pulses shorter than about 400  $\mu$ s.

In order to extract information about the corresponding decay times, CL transients recorded at the peak energies of the bands observed in steady state and TRCL spectra were analyzed. These transients are well described by single exponential decays. Table II summarizes the results obtained at

TABLE I. Fitting parameters to Eq. (1) for the different CL emissions observed in the Si-doped GaN investigated ( $T=87$  K).

Energy (eV)	2.38	2.29	2.16	2.03	1.96
$I_{sat}$ (arb. units)	$28 \pm 1$	$23 \pm 1$	$39 \pm 2$	$30 \pm 2$	$14 \pm 1$
$\tau_f$ ( $\mu$ s)	$144 \pm 5$	$177 \pm 5$	$168 \pm 5$	$160 \pm 5$	$188 \pm 6$



TABLE II. Decay times measured at 87 K for the GaN:Si deep level CL emissions after excitation with a 20  $\mu$ s pulse.

Energy (eV)	2.38	2.29	2.16	2.03	1.96
Decay time ( $\mu$ s)	$340 \pm 5$	$245 \pm 5$	$315 \pm 5$	$295 \pm 5$	$280 \pm 5$

87 K after excitation with a 20  $\mu$ s electron pulse for the five deep level related emissions present in our Si-doped GaN. All the measured decay times lie in the  $10^2$   $\mu$ s range. The shorter decay time corresponds to the 2.29 eV emission band, while longer decay times are observed for the 2.16 and 2.38 eV bands, as would be expected from the TRCL spectrum shown in Fig. 3.

#### IV. DISCUSSION

Our CL measurements confirm the coexistence of several emissions in the so-called yellow band of GaN:Si, which indicates that great care should be taken when discussing the characteristics and temporal behavior of this luminescence due to its complex nature. As stated in the Introduction section, an electronic transition gives rise to a peak in a PC spectrum if charge carriers involved in the process reach the conduction or the valence band. A transition between two levels can be detected by PC only if the energy separation between at least one of the levels and a band is lower than the thermal energy. Simultaneous appearance of part of the mentioned components in PC and CL spectra (Fig. 1) indicates then that such emissions are related to deep level to band transitions or to transitions involving deep levels and very shallow levels with a maximum depth on the order of the thermal energy (25 meV in our case).

Present day views generally agree in the acceptor character of some of the deep levels involved in the GaN yellow luminescence, being gallium vacancies ( $V_{\text{Ga}}$ ) and its complexes with oxygen or carbon are often suggested candidates for these levels.<sup>4,26</sup> The influence of nitrogen vacancies on the yellow emission has been also reported.<sup>5,10</sup>

The existence of shallow acceptors in *n*-type GaN, with binding energies of 85 and 115 meV, was recently proposed,<sup>27</sup> this proposal agreeing with previous theoretical calculations.<sup>28</sup> However, acceptor levels in *n*-type GaN with energies lower than 25 meV have been neither experimentally observed nor theoretically predicted. Thus, if the peaks simultaneously observed in PC and CL spectra were related to transitions involving deep acceptors and very shallow levels, the latter should be donors. Dominant donors at room temperature in undoped GaN are Si and O. Oxygen binding energies between 32 and 34 meV are usually reported in the literature.<sup>29–31</sup> On the contrary, there is a higher dispersion for Si energies reported by different authors. Some investigations reveal a binding energy of (29–31) meV in undoped samples,<sup>29–31</sup> while according to other experiments the position of the Si donor level in GaN:Si band gap lays 22 meV below the conduction band.<sup>32,33</sup> The existence of donors with binding energies lower than 25 meV—probably linked to intrinsic defects—has been also reported in GaN PL investigations.<sup>32</sup> Hence, the possibility of these PC and CL bands being due to transitions involving deep levels and very

shallow donor levels cannot be ruled out only on the basis of the obtained PC and CL spectra. We will further address this point when analyzing the CL transient decays.

CL emissions have been observed in this work not only in the yellow range of the visible spectrum but also in the green and red ranges. Regarding the green range, the close peak energies of the bands found by CL (2.38 eV) and PC (2.43 eV), respectively, suggest that the same deep level could be involved in both transitions. Green luminescence centered at about 2.35 eV has been previously observed in Si-doped<sup>34</sup> and undoped<sup>35</sup> GaN films by time-resolved PL spectroscopy. Decay times of 3 ms at 50 K and about 600  $\mu$ s at 100 K were measured for this emission. Either in the Si-doped samples or in the undoped films, this characteristic time was found to be much longer than the decay time corresponding to the yellow luminescence ( $\sim 200$   $\mu$ s) observed centered near 2.25 eV. These results are in good agreement with our TRCL spectra showing a 2.38 eV band as the dominant emission for delay times above 400  $\mu$ s (Fig. 3). The nature of the CL emission detected in the red range of the visible spectrum at about 1.89 eV is difficult to ascertain. The intensity of this band is nearly independent of temperature up to 295 K. Such temperature behavior is similar to that shown by the red PL recently observed at about 1.92 eV in Si-doped GaN grown by hydride vapor phase epitaxy.<sup>36</sup> This emission was attributed to  $V_{\text{Ga}}$ -related defects bound to structural imperfections, which are also expected to occur in thin films grown over mismatched sapphire substrates.

Transient decay times measured for the yellow band by different research groups in time-resolved PL investigations are controversial. Hoffmann *et al.*<sup>5</sup> and Korotkov, Reshchikov, and Wessels<sup>8</sup> reported nonexponential decay of the yellow luminescence in the range  $10^{-1}$ – $10^3$   $\mu$ s and explained their results in the frame of the Thomas–Hopfield<sup>37</sup> model for distant DAP recombination. On the contrary, very fast decays below 1 ns were found by other authors<sup>9</sup> and explained by a strong contribution of free-to-bound transitions. In the present study, exponential decays with characteristic times of a few hundreds of  $\mu$ s were measured for all the deep level emissions appearing in the GaN:Si investigated (Table II), although it should be mentioned that observation of possible radiative transitions with decay times lower than 100 ns is hindered by experimental constraints. Nevertheless, even taking into account the time resolution of the experimental setup used, our CL observations are, in principle, not in agreement with the Thomas–Hopfield DAP recombination theory.<sup>37</sup> According to this model, the radiative recombination rate depends on the donor and acceptor separation. In particular, lifetimes of holes bounded to acceptor levels will be much longer for distant pairs than for close ones. This implies a wide distribution of instantaneous decay times, usually extending from the ns to the ms range.<sup>5,8,37</sup> Hence, if the transition considered is of the DAP type, the corresponding luminescence decay transient will not be fitted by a single exponential curve. The single exponential character of the CL decay transients measured in this work indicates then that the observed emissions do not correspond to DAP transitions but to transitions involving deep levels and the conduction band. In view of the present results, some

models previously proposed to explain the origin of the yellow band, based on different kinds of DAP transitions,<sup>2–6</sup> can then be ruled out.

The discrepancy between the decay times measured in this work and those reported for the yellow emission by different authors can be explained considering both the different nature of the excitation source and the different experimental conditions used in our CL measurements and their PL experiments. As is well known, as compared with PL, CL is a technique which can excite a higher amount of radiative recombination processes in semiconductors. Furthermore, the abovementioned PL investigations<sup>5,8</sup> were carried out at liquid helium temperature, while our CL transients were recorded at 87 K. A change in the recombination mechanism by increasing temperature has been very recently reported for the GaN deep level PL emission by Reshchikov *et al.*<sup>38</sup> While a DAP-type recombination involving shallow donors was observed at 15 K, the PL decay curve was found to approach an exponential one by increasing temperature. This behavior was attributed to an increase of the free electron concentration and a decrease of the number of neutral donors. A similar process could account for the present CL results. In addition, in comparison with typical conditions used for PL excitation much higher excitation conditions were used for CL measurements. In this context, our measurements reveal that the duration of the excitation pulse is an important parameter. In this study, electron pulses of 20  $\mu$ s were used to investigate the decay transients of Si-doped GaN and it was found that CL intensity saturation—corresponding to a steady state condition—was never reached for electron pulses shorter than a few hundreds of  $\mu$ s (Fig. 4). This situation strongly contrasts with PL experimental conditions, where the sample is commonly excited with ns light pulses.<sup>5,8,38</sup> Our observations suggest that certain recombination processes, as those observed in the present work using TRCL, may not be excited under typical PL experimental conditions due to the slow filling kinetics of the involved traps.

## V. CONCLUSIONS

Time-resolved cathodoluminescence and photocurrent spectroscopies have been used to investigate the yellow band of Si-doped GaN. Our measurements unambiguously reveal the complex nature of this broad emission, which is actually composed of several bands, and confirms that different deep defect levels are involved in the observed luminescence.

Five emission bands centered at 1.89, 2.03, 2.16, 2.29, and 2.38 eV were found in steady state and time-resolved CL measurements, while PC spectra showed transitions at about 2.01, 2.14, 2.28, and 2.43 eV. The behavior of the deep level emissions intensity as a function of the excitation pulse width indicates that saturation—corresponding to a steady state condition—is never reached for electron pulses shorter than a few hundreds of  $\mu$ s. These observations could be indicative of slow filling kinetics of the involved traps. TRCL measurements reveal different decay times for the observed CL emissions. The shorter decay time, 245  $\mu$ s, corresponds to the 2.29 eV emission band, while longer decay times of

315 and 340  $\mu$ s are respectively observed for the 2.16 and 2.38 eV bands, in respectively, agreement with the obtained TRCL spectra. Simultaneous appearance of the 2.03, 2.16 and 2.29 eV peaks in PC and CL spectra suggest that these bands could be related to deep acceptor to band transitions, as supported by the single exponential character of the corresponding decay transients.

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