Yellow luminescence and related deep states in undoped GaN

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Photocapacitance spectra in undoped, metal-organic vapor-phase-epitaxy-grown GaN layers, in a range of photon energies from 0.6 to 3.5 eV, reveal two main persistent features: a broad increase of the capacitance from 2.0 to 2.5 eV, and a steep *decrease* at 1 eV, only observed after a previous light exposure to photon energies above 2.5 eV. A deep trap (E_v+1 eV) that captures photoelectrons from the valence band, after being emptied with photons above 2.5 eV, is proposed as the origin of these features. Optical-current deep-level transient spectroscopy results also show the presence of a trap at 0.94 eV *above* the valence band, *only* detected after light excitation with photon energies above 2.5 eV. A correlation is found between the ''yellow band'' luminescence intensity at 2.2 eV and the amplitude of the photocapacitance decrease at 1 eV, pointing to a deep trap at 1 eV *above* the valence band as the recombination path for the yellow band. The detection of the yellow band with below-the-gap photoluminescence excitation supports the proposed model.
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I. INTRODUCTION

In spite of the achievement of efficient devices (LEDs) out of GaN layers with a high dislocation density (10^8–10^{10} cm^{-2}), the presence of extended and point defects can be detrimental to the device performance. Point defects like V_N and Ga_I have been proposed as responsible for the high residual (10^{17}–10^{20} cm^{-3}) *n*-type character of undoped GaN.[1,2] However, recent calculations by Neugebauer and Van de Walle[3] indicate that individual V_N cannot account for the residual *n*-type conductivity of GaN. Impurities have been proposed as another possibility to explain this *n*-type residual character, and, indeed, oxygen contamination, either from the substrate (Al_2O_3) or from water vapor, can also account for lower residual *n*-type levels, since O generates shallow donor states.[4,5] On the other hand, Ga_I defects are thought to strongly modify the acceptor doping efficiency,[1] and similar effects are found in Zn compensation behavior due to O contamination.[4] The ubiquitous yellow band (YB) observed from photoluminescence (PL) at around 2.2 eV, is detected independently of the substrate and the epitaxial technique used, with its intensity being more pronounced at room temperature. Ga_I defects acting as double donors have been proposed to play a substantial role in the recombination path for this transition in undoped GaN,[6–8] that would proceed from a deep (A_1) donor state ($E_c-0.8$ eV) to an effective mass (EM) like acceptor.[6,7] An opposite model, by Ogino and Aoki[9] and Hofmann *et al.*,[10] relates the YB to a recombination path from a *shallow* donor to a deep donor (acceptor). A similar conclusion was reached by Suski *et al.*[11] from PL measurements under hydrostatic pressure, suggesting the final state to be a deep state of either donor or acceptor character.

Deep-level transient spectroscopy (DLTS) measurements by Götz *et al.*[12] in Si-doped GaN, by Hacke and co-workers[13,14] in undoped and Mg-doped GaN, and by Lee *et al.*[15] in undoped and Si-doped GaN up to 530 K, show no evidence of the origin of the found traps. Photoemission capacitance spectroscopy and optical transmission results by Götz *et al.*,[16] Balagurov and Chong[17] and Yi and Wessels,[18] show a variety of optical thresholds ranging from 0.87 to 3.1 eV that might support almost any model for the YB. However, most of these optical transitions are quite small signals not well resolved, in some cases just a guess,[18] making it rather difficult to reach clear conclusions.

Related to the YB recombination path, there are several contradictory results in the literature concerning the effects of *n*- and *p*-type doping on the intensity of this optical transition. Ogino and Aoki[9] showed that Si and O doping (shallow, *n*-type) did not affect the YB relative intensity, whereas *C* doping seemed to enhance this transition. In Nakamura's *et al.*[19] work, an increase of the doping level with Si and Ge produced a moderate decrease of the YB amplitude to band-edge luminescence ratio. Assuming the shallow donor to deep-trap model,[9] no significant changes in the YB signal should be expected with *n*-type doping, since undoped samples are already *n* type, with the deep-trap concentration being the limiting factor. This argument only applies assuming that the deep-trap concentration is not modified by the doping itself. However, this seems not to be the case reported by Zhang *et al.*,[20] where Ge doping, or growth under Ga-rich conditions, seems to wipe out the YB. When GaN is *p* type doped with Mg, most published works show that the YB intensity decreases, or even disappears.[20] In some cases, a luminescence signal at energies higher than 2.2 eV shows up, suggesting a different origin than that for the YB in undoped GaN.

Since the YB appears in GaN grown by various techniques, under dissimilar growth conditions and on different

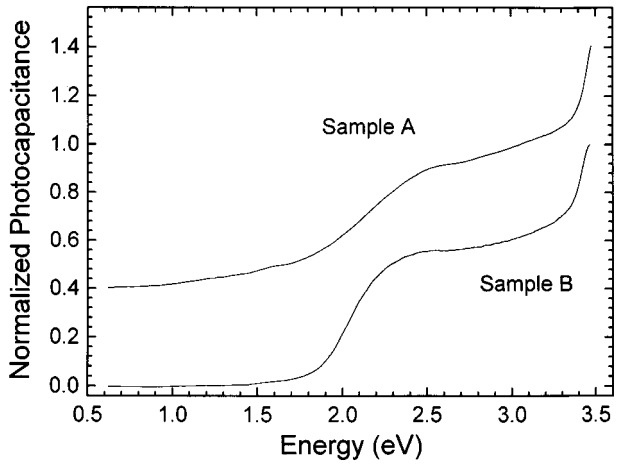

FIG. 1. Photocapacitance spectra of two typical undoped (*n* type) GaN samples.

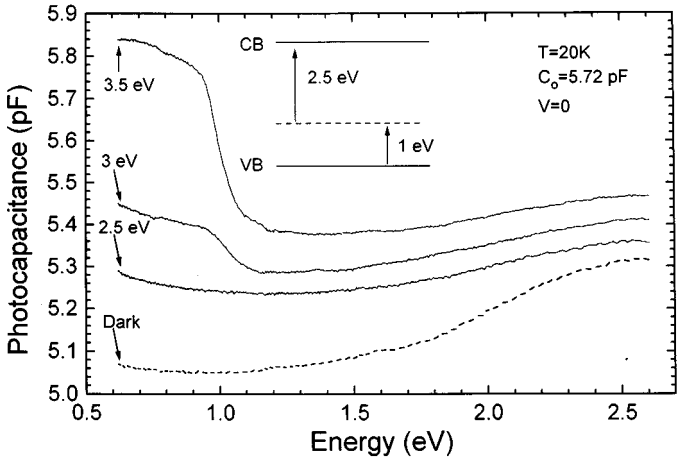

FIG. 2. Photocapacitance spectra taken after sample illumination at 20 K with different photon energies.

substrates (SiC, GaAs, Al_2O_3, lithium gallate, etc.), it is sensible to conclude that this luminescence is not related to impurities and/or contamination, but rather to point defects. Cathodoluminescence (CL) measurements show that the YB intensity is enhanced when increasing the probing depth toward the substrate.[21] Similar results are observed when the GaN layer is excited from the substrate side (sapphire).[20] From these results it seems that the origin of the YB, and its intensity changes from sample to sample, might be related to dislocations and point defects decorating them, and to crystal morphology differences, respectively. Very recent results from PL and CL, by Christiansen *et al.*,[22] relate the yellow luminescence to screw dislocations, and an early work by Pankove and Hutchby[23] suggested the damage after ion implantation as the origin of a PL band centered at 2.15 eV.

The aim of this work is to ascertain experimentally the model that describes the recombination path of the yellow emission. We present data from photocapacitance spectra showing two clear optical thresholds at 1 and 2.2 eV. A correlation between photocapacitance step amplitudes at 1 eV and relative YB intensities measured by PL is established. Capacitance and optical-current DLTS measurements have been performed in the range of 20–540 K to determine the thermal activation of deep traps.

II. EXPERIMENT

Undoped wurtzite GaN layers were grown by metal-organic vapor-phase epitaxy on *c*-oriented sapphire substrates using TMG and ammonia.[24] The overall sample structure was an AlN buffer layer (100 Å) grown at 1050 °C (some samples also incorporated a GaN buffer grown at 600 °C), followed by the active GaN layer grown at 1050 °C. Layer thicknesses were between 1 and 3 μm and the residual (*n*-type) carrier concentration (Hall) ranged from 10^{17} to 10^{18} cm^{-3}. Schottky barriers were formed with Au and Pt, whereas Ohmic contacts were formed with Ti/Al. A Boonton 7200 capacitance meter and a monochromator Jobin-Yvon H25 with a 600-W globar (quartz-tungsten) lamp were used for photoemission capacitance measurements. Band-pass interferential filters were used. Samples were cooled down in a He-closed-cycle cryostat. Photoluminescence was excited with the 334-nm (3.71 eV) line of an Ar laser, and detected with an UV-enhanced GaAs photomultiplier. DLTS measurements at high temperatures were performed in a specially designed cryostat working from 77 to 550 K. Pulsed blue and yellow LED's were used as excitation sources for optical-current DLTS.

III. RESULTS AND DISCUSSION

A. Photocapacitance

Figure 1 shows typical photocapacitance spectra, taken from 0.6 to 3.5 eV, where small deviations from a pure flat response will not be taken as meaningful if they are not reproducible and cannot be clearly resolved. There is a first capacitance increase between 2.0 and 2.5 eV, and a signature at 3.3 eV, probably due to transitions from EM residual acceptors located at some 200 meV from the valence band (VB). The capacitance increase from 2.0 to 2.5 eV (a threshold around 2.2 eV) was always observed, although its sharpness was sample dependent (samples *A* and *B*). The spectra were taken after cooling the sample down to 20 K in the dark. Once a thermal equilibrium was reached (steady capacitance after 1-h delay), a very slow photon energy scan (4 h) was performed, with the sample always kept at zero bias. Correction of the spectra by the system response slightly changed the relative amplitudes but not the threshold positions. It is worth mentioning that the capacitance, once light excitation (at any photon energy) was turned off, was partially recovered by thermal capture reaching a final *persistent* value. This thermal recovery was always quite nonexponential and very slow.

Figure 2 shows several photocapacitance spectra taken at 20 K after sample illumination with different photon energies. The measuring process, always at zero bias, was the following: (a) cooling down from room temperature (RT) to 20 K in the dark; (b) sample illumination at a given photon energy for 30 min, (c) the sample remains in the dark until a complete capacitance stabilization is reached (60 min), and

(d) a scan like the ones in Fig. 1 is performed. This procedure was repeated for each scan in Fig. 2. Step (c) was required because of the strong nonexponential, partial electron thermal recapture (capacitance decrease) taking place once the light was turned off, so that the final *persistent* capacitance value was lower. On the other hand, a nonexponential capacitance increase was always found when illuminating with photon energies indicated in Fig. 2 with arrows, that took more than 30 min to saturate. This behavior, that might indicate the contribution of more than one trap to the photoionization process, is identical to that reported by Johnson *et al.*[25]

When excitation photon energies below 2.5 eV are used, data in Fig. 2 show just a persistent increase of the photocapacitance, with corresponding amplitudes similar to the ones in Fig. 1. However, for photon energies of 2.8 eV and above, an abrupt decrease of the capacitance is observed at 1 eV. This is a much sharper feature than the positive step at 2.2 eV in Fig. 1, and its amplitude increases with increasing photon energies. This behavior has been observed in all samples.

In an *n*-type sample, an increase of the capacitance is associated with an increase of the net positive charge. The positive step (increase) observed at 2.2 eV in Fig. 1 is interpreted in terms of an electron emission to the conduction band (CB). However, the photocapacitance *decrease* at 1 eV corresponds to electron emission *from* the VB to a trap located at 1 eV above it. The fact that this step at 1 eV is not detectable unless the sample is excited with photons above 2.5 eV (i.e., a trap at $E_c-2.5$ eV is *emptied*), points to a trap located at 1 eV *above* the VB as responsible for both optical transitions (inset in Fig. 2).

The thermal capture processes taking place after turning the light off, present at any temperature (even at RT), cannot be accounted for by residual shallow donors (samples are *n* type). The Hall carrier concentration is barely modified from RT down to 20 K, and the most probable candidates for residual *n*-type conductivity, V_N, Si, or O, have ionization energies of few tens of meV. There have to be, in addition to these shallow donors, other electron states, possibly spread over a rather wide range of energies, that might explain the long times required for capacitance stabilization when shining light as well as once light is turned off. There is actually a natural explanation for the origin of these distributed states: the grain boundaries of polycrystalline, ''columnarlike'' GaN layers, and/or extended defects like dislocations. The presence of grain boundaries with distributed states at the interface generating a CB potential bending, might explain the persistent character of the photocapacitance signatures, without the need to involve capture barriers of metastable origin. Qiu *et al.*[26] suggested the presence of a distribution of states in the gap to account for the photoconductive response of undoped GaN to photon energies from 1.5 to 3.0 eV. Indeed, the monotonically increasing baseline of the photocapacitance in sample *A* (Fig. 1) from 1 to 3.3 eV might be related to such sample dependent distributed states.

B. Photoluminescence

Figure 3(a) represents a typical PL spectrum of our samples, where the donor-bound exciton at 3.47 eV together with the LO-phonon replica can be seen. The YB emission

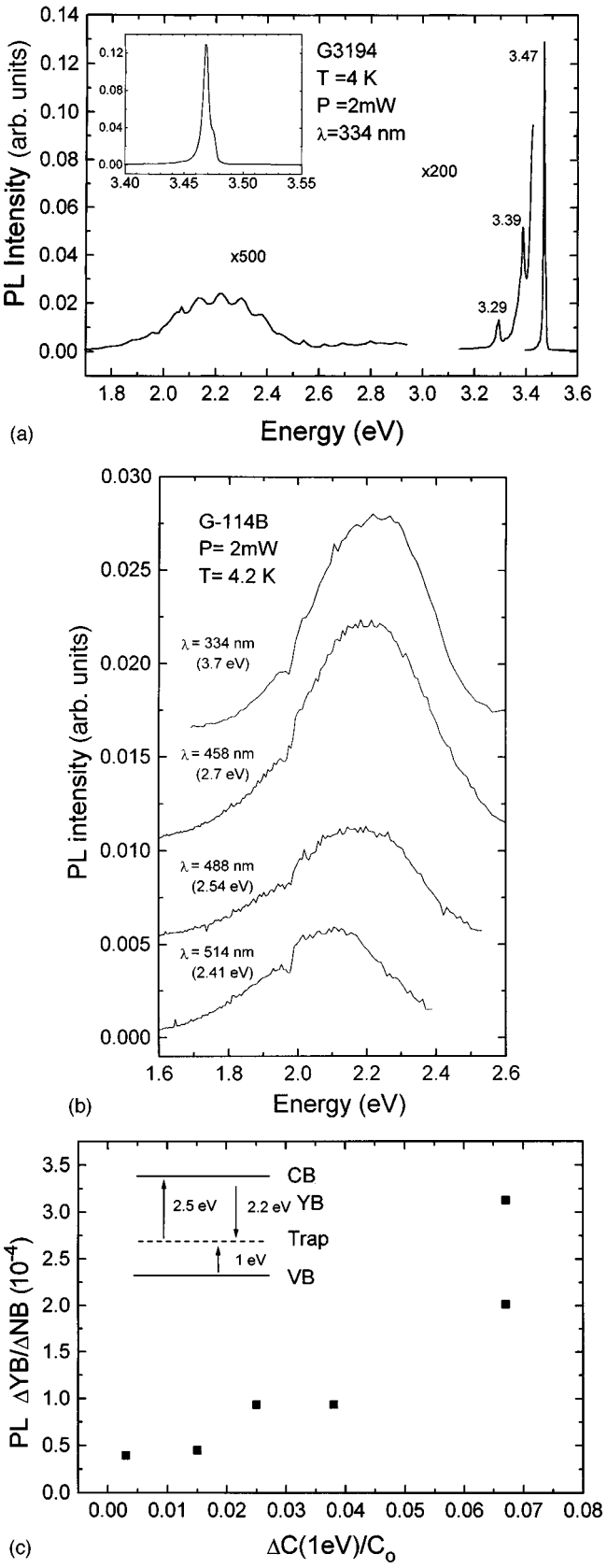

FIG. 3. (a) Typical PL spectrum of the undoped GaN samples. (b) PL spectra taken under below-the-gap excitation energies. (c) Correlation between the yellow band (YB) intensity normalized to the near-band (NB) PL signal, and the capacitance amplitude at 1 eV (Fig. 2) normalized to the total capacitance value (C_o).

centered at 2.2 eV is also shown. Figure 3(b) shows the evolution of the YB for excitation energies below the gap. Besides a moderate intensity decrease, probably due to a lower excitation efficiency, there is a clear redshift. Since no filters have been used that could distort the spectra, this shift might indicate that the broadening of the YB signal is due to emission from several closely spaced traps.

Figure 3(c) shows the experimental correlation found between the YB intensity (relative to the band-edge PL at 3.47 eV), and the capacitance step amplitude at 1 eV (relative to the total capacitance value) on different GaN samples. This capacitance amplitude was measured in all cases after light excitation at 3.5 eV. It seems that the trap located at 1 eV above the VB is also involved in the YB luminescence at 2.2 eV, and the inset in Fig. 3(c) sketches the proposed model to explain the recombination path, in which the initial stage can be either the CB or a residual shallow donor. It is also worth saying that the measured trap at 1 eV from the VB fits well with the deep acceptor found by Ogino and Aoki[9] (860–910 meV). This model is also consistent with the fact that we observe YB emission by PL with excitation below the band edge [Fig. 3(b)].

The broadening of the YB observed in PL spectra and that found in photocapacitance spectra (Fig. 1), from 2.0 to 2.5 eV, are analogous. They can be explained either as phonon-assisted transitions through a lattice-coupled trap with a zero-phonon line at about 2.5 eV,[9] or, most probably, as a multiple emission from several closely spaced traps [redshift in Fig. 3(b)]. However, the capacitance decrease observed at 1 eV is a rather sharp feature. This would mean that this transition involves a single trap with no phonon interaction. Both views can be conciliated, within the proposed model, if we assume that the capture of electrons from the VB at 1 eV takes place for the trap which exhibits the largest optical capture cross section among the closely spaced traps. This also might explain why the photocapacitance does not decrease further at 1 eV (Fig. 2). A recent work by Neugebauer and Van de Walle[27] suggested the V_{Ga} and related complexes as the deep acceptors responsible for the YB. They also pointed out that complexes like V_{Ga}-O and V_{Ga}-donor generate close deep acceptor states in the range of 0.9–1.1 eV. These results fit very well with the proposed picture of the YB as originated by several closely spaced deep acceptor states.

The work by Kennedy *et al.*[6] and Glaser *et al.*[7] suggested an opposite model where the initial state for the yellow emission is a deep donor, and the final state is an effective-mass-like acceptor, in undoped and lightly *p*-type doped samples. When *p*-type doping with Mg is increased, some reports indicate that the low-temperature donor-to-acceptor pair (DAP) signal, formerly at some 250 meV below the band-edge luminescence, shifts to lower energies, being replaced by a broadband peaking at some 2.8 eV.[28,29] However, other works show the DAP signal still present at around 3.2 eV (Refs. 6, 7, and 30–32) in samples with high Mg levels. In parallel, the yellow band is either non detectable[20] or replaced by a nonsymmetric and weak signal that is no longer centered at 2.2 eV.[29] In this last case, the above model for the yellow band[6,7] implies the shift of the deep-donor state, as well as the deepening of the acceptor state, or the generation of acceptor-related deep traps.[31] Leroux *et al.*[30] proposed that increasing the Mg-doping level generates compensating deep donors, instead of shifting the Mg acceptor binding energy to higher values, and indeed interstitial Mg and Mg in N sites are predicted to behave as donors.[33] These results agree with the fact that the Mg acceptor binding energy, derived from Hall measurements[29] and admittance spectroscopy[34] keeps constant ($\simeq$160 meV) regardless of the doping level. If the YB recombination path involves point defects, a change in stoichiometry due to heavy Mg doping can avoid the formation of these point defects,[1] as well as promote the formation of new ones,[33] thus explaining the strong differences observed in the YB ''signature'' of samples with high Mg doping levels. Given the rich phenomenology observed in heavy Mg-doped samples, a cautious approach to the YB problem advises not to mix it up with the case of undoped or lightly doped *p*-type samples.

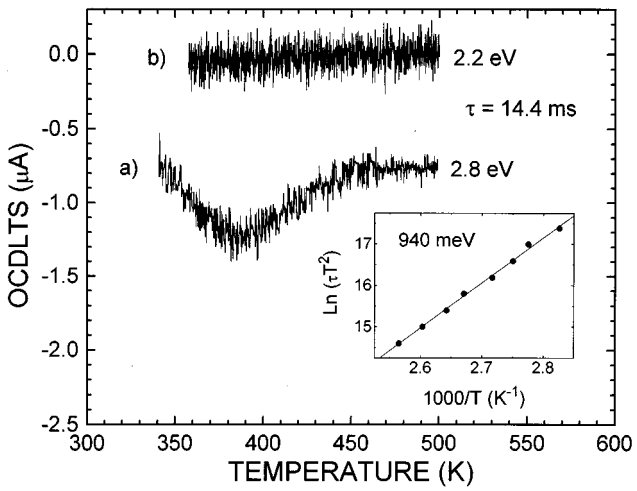

FIG. 4. Optical-current DLTS spectra with light excitations at 2.2 and 2.8 eV, showing the activation energy of a thermal electron capture process from the valence band.

Going back to the undoped case, the model above described[6,7] hardly explains that (a) the YB is observed from PL excited with energies below the gap; (b) no DLTS electron thermal emission, with activation energies around 1 eV, is observed in samples showing YB emission (see Sec. III C); and (c) the YB intensity seems not to increase with moderate *p*-type doping (we remark that the work of Ogino and Aoki[9] is the only one, to our knowledge, that shows a dependence of the YB intensity on the C doping level). However, if the YB recombination proceeds from a shallow donor state to a deep trap ($E_c-2.5$ eV) *generated by point defects*, then PL spectra excited at energies equal or larger than the trap depth (2.5 eV) should show the YB emission as a consequence of the trap photoionization through an efficient single-step process. In addition, these deep traps being generated by point defects, little effect might be expected from moderate *p*-type doping.

C. DLTS measurements

The essence of the optical-current DLTS technique is to perform a DLTS processing of the current signal of a GaN

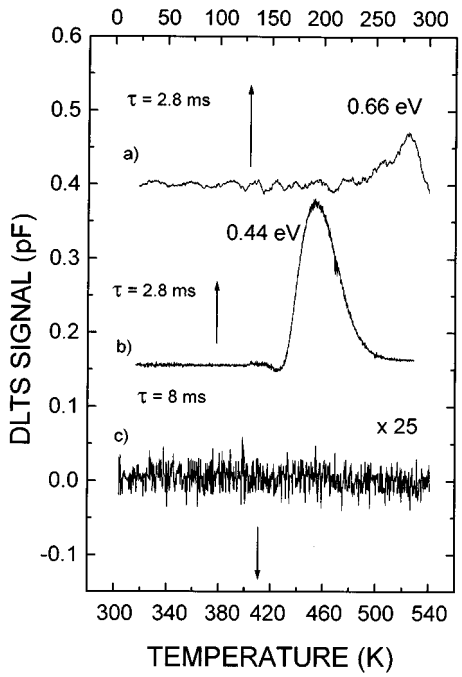

FIG. 5. DLTS spectra showing several electron emission processes, sample dependent. A flat response from 300 to 540 K is also observed.

sample between two Ohmic contacts. A pulsed light source (LED) illuminates the sample, that is kept under 5-V bias. When the light pulse is off, the current decay is analyzed by the DLTS system. If no thermal process is involved, the current decay will not be temperature dependent, and a flat DLTS signal will be recorded. However, if the light pulse photoionizes a trap which is able to thermally capture electrons at a given temperature, the current decay will be temperature dependent, and a DLTS signal will show up.

Optical-current DLTS spectra in Fig. 4 show a flat response when light excitation is below 2.5 eV. However, a clear peak is observed for light pulses of 2.8 eV (blue Nichia LED), that corresponds to a thermal process with an activation energy of 940 meV measured at around 350 °C (inset in Fig. 4). The strong similarity with data in Fig. 2, in terms of photon energy dependence, suggests that this thermal process involves the trap located at 1 eV above the VB (inset in Fig. 2). Once the photon energy is high enough to photoionize this trap (energy above 2.5 eV), a thermal electron capture takes place via the VB (hole emission) if the temperature is adequate. This electron capture will modify the current transient as a function of the temperature. For pulses with photon energies below 2.5 eV, there is no photoionization of this trap, and no thermal capture takes place. The activation energy of 940 meV is consistent with the values obtained from our photocapacitance experiments, and it really fits to the values given by Ogino and Aoki[9] for the thermal quenching of the yellow band (860–910 meV).

Capacitance DLTS measurements, from 20 to 540 K, have also been performed in Au and Pt Schottky diodes. Depending on the sample, electron traps with activation energies of 0.66 eV [Fig. 5(a)] and 0.44 eV [Fig. 5(b)] have been found. Similar traps, among others, have already been reported in the literature,[12–14] but, in our case, their small concentration and sample dependence rule out any relationship with the YB emission. We remark that the photocapacitance decrease at 1 eV (Fig. 2) has been observed in all our samples. DLTS performed in Pt-Schottky diodes at high temperatures shows no detectable signatures [Fig. 5(c)] up to 540 K. Lee *et al.*[15] recently published DLTS data up to 530 K where a peak corresponding to an activation energy of 1.6 ± 0.3 eV was shown. As the authors commented, this value, derived from an Arrhenius plot of just two points, has to be taken cautiously. We have also found in one sample (Au-Schottky) a DLTS signal that increases steadily from 300 to 540 K. This single result might be either a real transition or an artifact due to a progressive decrease of the diode turn-on voltage with temperature, allowing the current flow through the capacitance meter even for forward pulse voltages as low as 200 mV.

A simple estimation of emission rates for the temperature range covered in our experiments (300–540 K), assuming a capture cross section of 5×10^{-15} cm^2 (a mean value for most traps reported[13,14]), predicts that electron traps with activation energies between 0.7 and 1.2 eV should be observable by DLTS *if present*. These estimations, consistent with the fact that no optical transitions to the CB have been detected in this range (Fig. 1), cannot support the recombination model for the YB emission that assumes the presence of a deep donor between 0.7 and 1 eV below the CB.[6,7]

III. SUMMARY

Photocapacitance spectroscopy measurements reveal a trap located at 1 eV above the VB that accounts for the optical transitions observed. The correlation found between the photocapacitance step amplitude at 1 eV, and the yellow band intensity, points to this trap as responsible for the YB recombination path in undoped GaN. Optical-current DLTS measurements confirm the presence of this trap, the energies involved being in very good agreement with a previous work.[9] DLTS spectra show no traces of electron emission from traps to the CB in the range of 0.7–1.2 eV. These results back the model for the YB emission that involves a shallow donor (or CB) and a deep state (either donorlike or acceptorlike). The detection of the YB emission from PL excited with below-the-gap energies gives further support to this model, and the observed redshift suggests the existence of several closely spaced states. This picture is in very good agreement with recent calculations by Neugebauer and Van de Walle[27] that point to V_{Ga} and related complexes as deep acceptors responsible for the YB.

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