Electric-field dependence of luminescence spectra of (In,Ga)N/GaN LEDs containing quantum wells

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We have investigated the electric-field- and excitation-density-induced variation of the optical transition energy and cathodoluminescence (CL) as well as photoluminescence intensity of single and multiple (In,Ga)N/GaN quantum wells (QW) deposited in the depletion region of a p-n junction. The electric-field dependence of the CL intensity is governed by the competition between carrier drift within the depletion region and carrier capture by the QWs. A gradual screening of the p-n junction field with increasing excitation density causes a strongly nonlinear CL response. The electric-field dependence of the optical transition energy is governed by a gradual compensation of the polarization field of the QWs by the p-n junction field as well as by filling of band tail states of localized excitons. While the CL spectra of the QWs reflect radiative recombination of both free and localized excitons, EL spectra are dominated by the recombination of localized excitons.

1 Introduction
The optical response of quantum wells (QW) depends strongly on the electric field applied perpendicular to the layers offering attractive prospects for the development of electro-optical modulators. The quantum-confined Stark effect (QCSE) leads to both a quenching of the luminescence intensity and a red-shift of the optical transition energy ($E_t$) [1-3]. Moreover, it has been found that tunneling, thermally activated sweep-out, and carrier drift can also be responsible for an electric-field-induced luminescence quenching [4-6]. Recently, Jho et al. [5] have investigated the electric-field dependence of the recombination dynamics in a light-emitting diode (LED) consisting of (In,Ga)N/GaN QWs. They found that for high fields, tunneling of carriers through the tilted barriers contributes dominantly to the quenching of the luminescence. However, little is known about the dependence of those quenching effects on the excitation density in the GaN material system, which is interesting for optically controlled light modulators.

We have investigated the electric-field-induced variation of the Photoluminescence (PL) and cathodoluminescence (CL) spectra for single and multiple (In,Ga)N/GaN QWs deposited in the depletion region of a p-n junction. Our study focuses on the dependence of the electric-field characteristics of such diodes on the excitation density.

2 Experimental details
For the experiments, we used commercially available LEDs developed by Nakamura et al. [7], which contain (In,Ga)N/GaN single (SQW) and multiple (MQW) quantum wells. The latter consist of four wells. The thickness of the well layers amounts to 2.4–3 nm.

Photoluminescence investigations were performed at 10 K using the 325-nm-line of a HeCd laser for high-energy excitation and the 413-nm-line of a Kr+ ion laser for low-energy excitation. CL spectra were measured at 5 K in a scanning electron microscope equipped with a mono-CL2 and He-cooling stage system. A grating monochromator and a cooled charge-coupled-device array was used to disperse and...
detect the CL signal, respectively. The electron beam energy was chosen to be 15 keV. CL investigations of the excitation density dependence were carried out by varying either the electron beam current between 1 and 200 pA or the excitation volume switching the electron beam from the focused to well-defined defocused states. The variation of the beam current is limited by the signal-to-noise ratio on the low-current side and by electron-beam-induced modifications of the optical properties of the (In,Ga)N/GaN QWs on the high-current side.

3 Results and discussion

Figure 1 displays the integrated PL (lines) and CL (triangles) intensities of the SQW-LED and the CL intensity ($I_{CL}$) of the MQW-LED (circles) as a function of the applied bias voltage ($U_{bias}$). Since the p-region of the (In,Ga)N/GaN LEDs is situated on the top of the structure (surface side), the electric field of the p-n junction and the piezo-electric field of the QWs act in opposite directions. Thus, an increasing p-n junction field (reverse bias) leads to a compensation of the polarization field in the QWs. Therefore considering the QCSE, an increase of the quantum efficiency ($\eta$) is expected when an increasing reverse bias voltage is applied. Actually, the opposite is observed as shown in Fig. 1. With decreasing forward bias, the PL and CL intensity increases only between 3 and 2 V, but decreases abruptly for a further reduction of $U_{bias}$ when the excitation energy ($E_{exc}$) exceeds the band gap energy ($E_{GaN}$) of the GaN barriers (CL and PL at 3.82 eV). For $E_{exc} < E_{GaN}$ (PL at 3 eV), however, the PL intensity ($I_{PL}$) decreases gradually for large reverse bias voltages. Horikoshi et al. [4] have shown that PL quenching in reverse-biased GaAs/(Al,Ga)As QWs is due to carrier tunnelling and carrier drift induced by the p-n junction field. The comparison of the $U_{bias}$ dependencies obtained for high-energy (CL, PL at 3.82 eV) and low-energy (PL at 3 eV) excitation reveals that for high-energy excitation and with increasing p-n junction field, carrier drift contributes dominantly to the luminescence quenching. While carrier tunnelling (excitation at 3 eV) affects $\eta$ only for large reverse bias voltages, the carrier drift leads to an abrupt decrease of $\eta$ between 2 and 1 V.

Calculations by means of a rate equation model considering excess carriers in the barrier region and for free as well as localized excitons in the QW confirm the impact of the carrier drift to the CL quenching within the observed $U_{bias}$ range [10]. According to this model, $I_{CL}$ is determined by the competition between carrier drift and carrier capture, where the capture cross-section decreases exponentially with increasing p-n junction field. The degree of carrier capture can be increased by increasing the number of QWs situated within the depletion region of the p-n junction as can be seen in Fig. 1. While the CL intensity of the SQW decreases down to 15%, $I_{CL}$ of the MQW is only quenched to 65% of its maximum, when $U_{bias}$ is decreased from 2 to 1 V.

As the drift of the carriers depends strongly on the electric field of the p-n junction, one should expect a significant impact of electric-field screening by excess carriers on the luminescence quenching in such structures. Therefore, we investigated the dependence of the CL spectra of the SQW-LED on both the electric field and the excitation density. Figures 2 (a) and (b) display the $U_{bias}$ dependence of the optical transition energy ($E_t$) derived from the CL spectra and of $I_{CL}$ for various values of the CL generation rate ($G$), respectively. $G$ ranges from 4x10$^{17}$ to 7.2x10$^{20}$ cm$^{-2}$ s$^{-1}$ as indicated in Fig. 2 (a). For a decreasing forward and increasing reverse bias, we observe — besides the quenching of the CL intensity within the range of 1 to 2 V — a blue shift of $E_t$, which is due to a compensation of the piezo-electric field of the QW by the increasing electric field in the depletion region. This electric-field characteristics varies, however, significantly with the excitation density.
The first striking feature is a blue-shift of $E_t$ with increasing population in the QW by electron-beam excitation. The carrier concentration of the QW becomes larger by increasing both the generation rate and the capture probability. The latter is indicated by the steep increase of $I_{CL}$ within the bias range of 1 to 2 V. Therefore, we observe a blue-shift of $E_t$ (due to the increased population) not only for increasing $G$, but also for increasing forward bias keeping $G$ unchanged. For the generation rates used in our experiments, the carrier-density-related blue-shift of $E_t$ is rather caused by filling of band tail states than by screening of the piezo-electric field of the QW. [11]

Consequently, the field dependence of $E_t$ is governed by two competing processes: (i) the interaction of the piezo-electric field of the QW with the field of the $p$-$n$ junction and (ii) the filling of band tail states of localized excitons. The competition between the impact of the $p$-$n$ junction field via the QCSE and the band tail filling via field-induced occupation of the QW can be directly observed within the bias range of 1 to 2 V, particularly, for low generation rates.

The onset energy of the electroluminescence — marked by “EL” in Fig. 2(a) — agrees with the extrapolated $E_t(U_{bias})$ for the lowest generation rate used in our CL experiment (cf. dashed line). Since for EL the carrier injection rates are of a comparable low level as the lowest used $G$ values in Fig. 2, the comparison of CL and EL nicely confirms the picture of localized states being mainly involved in the electroluminescence of (In,Ga)N/GaN LEDs. We will come back to this point later.

The second striking feature is the shift of the onset of the steep increase of $I_{CL}$ towards lower forward bias voltages when $G$ is enhanced as indicated by the dashed arrow in Fig. 2 (b). This excitation-density-dependent shift of the onset voltage can be understood in terms of a partial screening of the built-in field by excited carriers as follows. For decreasing the drift field within the depletion region down to a certain value, which allows for a significant increase of the capture probability, the forward bias has to be increased. Under screening conditions, only a smaller value of the forward bias is necessary in order to obtain a comparably low drift field. Thus, for high concentrations of excited carriers, where screening becomes effective, the onset voltage for an increase of $I_{CL}$ shifts towards lower forward bias. Our corresponding model calculations [10] confirm the impact of excitation-induced screening of the $p$-$n$ junction field on $I_{CL}(U_{bias})$ of the SQW-LED.

As a consequence of the dependence of $I_{CL}(U_{bias})$ on the excitation density, the luminescence response of the QW shows significant nonlinearity, which in turn depend strongly on the applied bias voltage. While the CL quantum efficiency increases with increasing excitation density for a bias range between 1.4 and 2 V, it decreases for $U_{bias} > 2$ V. The latter is due to an increasing portion of free excitons compared with localized ones when $G$ is enhanced. [10,11]

Localized exciton states of (In,Ga)N/GaN QWs are usually discussed to be responsible for the high quantum efficiency of corresponding GaN-related LEDs or laser devices. [12,13] The impact of localized states became clearly visible in the $E_t(U_{bias})$ characteristics of Fig 2 (a), where within an $U_{bias}$ range of 1–2 V, the field-induced red-shift of $E_t$ is converted into a blue-shift by enhanced carrier capture of the QW.
and a respective filling of localized band tail states. In Fig. 3, we demonstrate the importance of localized excitons or rather their competition with the free ones by means of the electric-field dependence of the CL spectra of the MQW-LED. Figure 3 (a) depicts the corresponding CL spectra for various forward bias voltages covering a range between 1.3 and 2.1 V. Added is the EL spectrum for the onset voltage of 2.3 V. The CL spectra consist clearly of two distinct contributions. We tentatively assign the low- and high-energy part of the spectra — marked by CL1 and CL2 in Fig. 3 (a) — to localized and free excitons, respectively. Since for $U_{\text{bias}}<1.5$ V and for reverse bias voltages, the radiative recombination rate of free excitons increases due to a screening of the polarization field of the QW, the CL spectra are dominated by CL2 in the respective $U_{\text{bias}}$ range. As expected, the spectral peak position of this line exhibits a red-shift with decreasing reverse and increasing forward bias, which is clearly reflected by $E_t(\text{CL2})$ in Fig. 3 (b). When the forward bias exceeds 1 V, the low-energy line becomes visible. It exhibits, however, the opposite bias dependence of $E_t$, namely a blue-shift for $U_{\text{bias}}>1.3$ V caused by field-induced filling of the QWs. The field-induced occupation of the MQWs has been shown in Fig. 1, where $I_{\text{CL}}$ increase abruptly within a bias range of 1.3 and 1.6 V.

The EL spectrum of Fig. 3 (a) differs significantly from the CL spectra. Particularly, the CL spectrum obtained for $U_{\text{bias}}=2.1$ V (closely below the onset of EL) should resemble the EL spectrum obtained for $U_{\text{bias}}=2.3$ V (closely above the EL onset). The CL spectrum is, however, blue-shifted by about 40 meV with respect to the EL spectrum and it contains contributions of both localized and free excitons. The EL spectrum consists solely of a single line indicating — according to our assignment — the dominance of localized states in the recombination process of the EL. Moreover, the spectral peak position of the EL spectrum corresponds to the position of the low-energy shoulder of the CL spectrum obtained for 1.3 V (before field-induced filling of the QWs), which probably represents the tail states of the localized excitons in the QWs. Consequently, while EL is dominated by the recombination of localized excitons, CL under usual excitation conditions — in Fig. 3: 15 keV, 50 nA — is governed by the competition between free and localized excitons.

References
