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
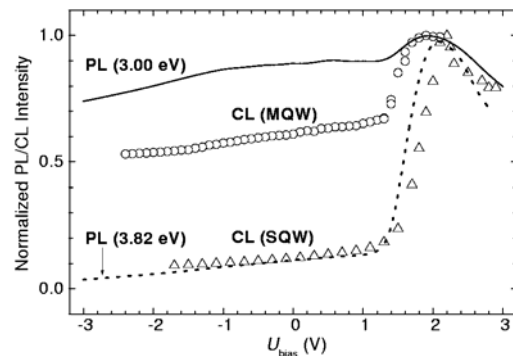
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1   detect the CL signal, respectively. The electron beam energy was chosen to be 15 keV. CL investigations
2   of the excitation density dependence were carried out by varying either the electron beam current be-
3   tween 1 and 200 pA or the excitation volume switching the electron beam from the focused to well de-
4   fined defocused states. [8] The variation of the beam current is limited by the signal-to-noise ratio on the
5   low-current side and by electron-beam-induced modifications of the optical properties of the
6   (In,Ga)N/GaN QWs on the high-current side. [9]
7
8   **3   Results and discussion**   Figure 1 displays the integrated PL (lines) and CL (triangles) intensities of
9   the SQW-LED and the CL intensity (I_{CL}) of the MQW-LED (circles) as a function of the applied bias
10 voltage (U_{bias}). Since the p-region of the (In,Ga)N/GaN LEDs is situated on the top of the structure (sur-
11 face side), the electric field of the p-n junction and the piezo-electric field of the QWs act in opposite
12 directions. Thus, an increasing p-n junction field (reverse bias) leads to a compensation of the polariza-
13 tion field in the QWs. Therefore considering the QCSE, an increase of the quantum efficiency (η) is
14 expected when an increasing reverse bias voltage is applied. Actually, the opposite is observed as shown
15 in Fig. 1. With decreasing forward bias, the PL and CL intensity increases only between 3 and 2 V, but
16 decreases abruptly for a further reduction of U_{bias} when the excitation energy (E_{exc}) exceeds the band gap
17 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
18 PL intensity (I_{PL}) decreases gradually for large reverse bias voltages. Horikoshi et al. [4] have shown that
19 PL quenching in reverse-biased GaAs/(Al,Ga)As QWs is due to carrier tunnelling and carrier drift in-
20 duced by the p-n junction field. The comparison of the U_{bias} dependencies obtained for high-energy (CL,
21 PL at 3.82 eV) and low-energy (PL at 3 eV) excitation reveals that for high-energy excitation and with
22 increasing p-n junction field, carrier drift contributes dominantly to the luminescence quenching. While
23 carrier tunnelling (excitation at 3 eV) affects η only for large reverse bias voltages, the carrier drift leads
24 to an abrupt decrease of η between 2 and 1 V.

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

Fig. 1   Normalized CL (triangles) and PL (lines) intensity of a (In,Ga)N/GaN-LED containing a SQW as a function of the bias voltage. Circles denote $I_{CL}(U_{bias})$ of an MQW-LED. The PL curves differ in the excitation energy.

40 As the drift of the carriers depends strongly on the
41 electric field of the p-n junction, one should expect a
42 significant impact of electric-field screening by ex-
43 cess carriers on the luminescence quenching in such structures. Therefore, we investigated the depend-
44 ence of the CL spectra of the SQW-LED on both the electric field and the excitation density. Figures 2
45 (a) and (b) display the U_{bias} dependence of the optical transition energy (E_t) derived from the CL spectra
46 and of I_{CL} for various values of the CL generation rate (G), respectively. G ranges from 4×10^{17} to
47 7.2×10^{20} cm^{-2} s^{-1} as indicated in Fig. 2 (a). For a decreasing forward and increasing reverse bias, we ob-
48 serve — besides the quenching of the CL intensity within the range of 1 to 2 V — a blue shift of E_t,
49 which is due to a compensation of the piezo-electric field of the QW by the increasing electric field in
50 the depletion region. This electric-field characteristics varies, however, significantly with the excitation
51 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 nonlinearities, 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
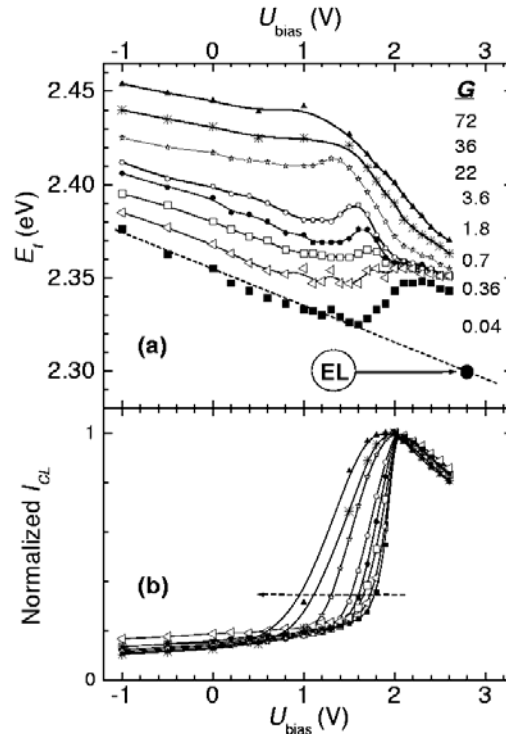
Fig. 2 (a) Optical transition energies and (b) normalized CL intensities as a function of the bias voltage for the (In,Ga)N/GaN LED containing a SQW. The various data sets differ in the CL generation rate, which is displayed in units of 10^{19} cm^{-2}s^{-1} in (a). EL represents the onset energy of the electroluminescence of the LED.

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_{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_{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(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_{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_{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_{bias}=2.1 V (closely below the onset of EL) should resemble the EL spectrum obtained for U_{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.
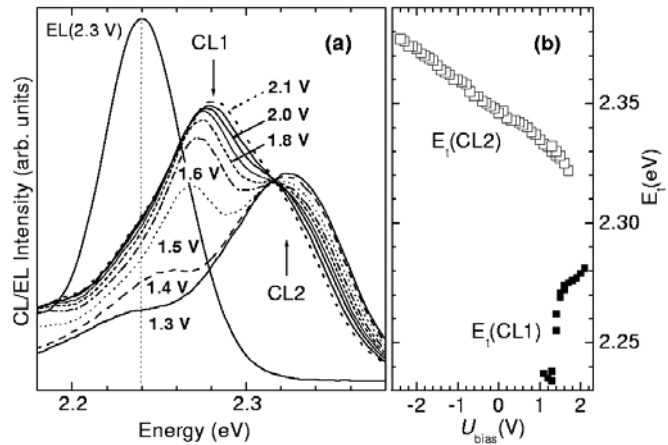
Fig. 3 (a) CL spectra of the MQW-LED for various values of the forward bias voltage. The CL generation rate amounted to about 10^{20} cm^{-2}s^{-1}. EL represents the electroluminescence spectrum at the onset voltage of 2.3 V. (b) Optical transition energy of the CL lines CL1 and CL" as a function of U_{bias}.

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