Low-energy electron-beam irradiation of GaN-based quantum well structures

U. Jahn[*1], **S. Dhar**[1], **H. Kostial**[1], **P. Waltereit**[1], **F. Scholz**[2], **J. Off**[3], **I. M. Watson**[4], and **K. Fujiwara**[5]

[1] Paul-Drude-Institut für Festkörperelektronik, Hausvogteiplatz 5–7, 10117 Berlin, Germany
[2] Abteilung Optoelektronik, Universität Ulm, Albert-Einstein-Allee 45, 89081 Ulm, Germany
[3] Universität Stuttgart, Pfaffenwaldring 57, 70569 Stuttgart, Germany
[4] University of Strathclyde, Glasgow G4 0NW, Scotland, UK
[5] Kyushu Institute of Technology, Tobata, Kitakyushu 804-8550, Japan

Received zzz, revised zzz, accepted zzz
Published online zzz

PACS 78.20.-e, 78.60.Hk, 78.66.-w, 78.66.Fd, 78.67.De

The electronic properties of (In,Ga)N/GaN quantum wells fabricated by MOCVD depend significantly on low-energy electron-beam irradiation (LEEBI), e. g., during cathodoluminescence (CL) investigations, when a certain exposure dose is exceeded. For unintentionally doped structures, we observe a LEEBI-induced activation of donors and acceptors. Thus, the resistivity of the layers is not varied, while the quantum efficiency and optical transition energy increases significantly by LEEBI. The electric field distribution in a *p*-*n* structure is changed towards the flat band condition during LEEBI indicating an electron-beam-induced passivation of acceptors in the *p*-type layer.

1 Introduction

Low-energy electron-beam irradiation (LEEBI) of Mg-doped GaN layers grown by metalorganic chemical vapor deposition (MOCVD) causes a substantial decrease of the resistivity and increase of the luminescence efficiency (η) of those layers [1]. There are also indications for a LEEBI-induced variation of η in undoped MOCVD-grown GaN [2, 3, 4]. While the activation mechanism of acceptors in *p*-type GaN has been comprehensively investigated and found to be a minority carrier-induced dissociation of a Mg-H-N complex [5, 6, 7], a consistent picture about LEEBI effects in undoped and *n*-type GaN does not exist yet. The optical transition energy (E_t) and η of quantum wells (QW) are expected to vary significantly by LEEBI due to screening of the internal electric field by activated free carriers and/or by the interaction of the QW field with the electric field arising from surface polarization. The latter has been investigated by Mayrock et al. [8] and Gfrörer et al. [9].

We have studied LEEBI effects in undoped (In,Ga)N/GaN QWs fabricated by MOCVD as well as in a commercial light-emitting diode (LED: Nichia Chemical Industry Ltd.) containing a single QW in the active region. For reference, an (In,Ga)N/GaN and a GaAs/(Al,Ga)As QW structure both fabricated by molecular-beam epitaxy (MBE) were exposed to an electron-beam under the same conditions.

* Corresponding author: e-mail: ujahn@pdi-berlin.de, Phone: +49 30 203 77 523, Fax: +49 30 203 77 515

Table 1 Growth method, substrate, well material, well width (d_w), and doping state of the samples 1–5 used in this study.

No.	growth method	substrate	well	d_w (nm)	doping state
1	MOCVD	sapphire	$In_{0.10}Ga_{0.90}N$	2.6	MQW, undoped
2	MOCVD	6H-SiC(0001)	$In_{0.10}Ga_{0.90}N$	2.5	SQW, undoped
3	MBE	6H-SiC(0001)	$In_{0.16}Ga_{0.84}N$	3.1	MQW, undoped
4	MBE	GaAs(100)	GaAs	4.8	SQW, undoped
5	MOCVD	sapphire	$In_{0.45}Ga_{0.55}N$	3	SQW, *p*-*n* structure

2 Experimental

Table 1 contains details about the samples used in this work. The growth conditions of the undoped (In,Ga)N/GaN QW structures 1 and 2 fabricated by MOCVD are described by Pecharroman-Gallego et al. [10] and Off et al. [11], respectively. The MBE-grown samples 3 and 4 serve as reference structures in order to study distinctions between MBE and MOCVD-grown QWs as well as between QWs with (GaN-based) and without (GaAs-based) internal electric fields. Sample 5 is a light-emitting diode developed by Nakamura et al. [12].

The samples were exposed to the electron-beam of a scanning electron microscope (SEM) at 5 and 300 K. The beam energy and current amounted to 5 to 15 keV and 0.05 to 8 nA, respectively. The exposed area amounted to 29 μm^2. After distinct charge doses (D_c), CL spectra were acquired by a charged-coupled device detector. For the LED, the electron-beam-induced current (EBIC) was measured simultaneously with the CL spectra. Furthermore, Hall measurements were performed at 300 K using van der Pauw structures of the MOCVD-grown MQW.

3 Results and discussion

3.1 Unintentionally doped QWs

Characterization of QWs by CL is usually done at low temperatures. Therefore, we firstly exposed the samples and acquired CL spectra at 5 K. Figures 1(a) and 1(b) depict the integrated CL intensity (I_{CL}) and the variation of the transition energy (ΔE_t) of the QWs as a function of D_c for the samples 1–4,

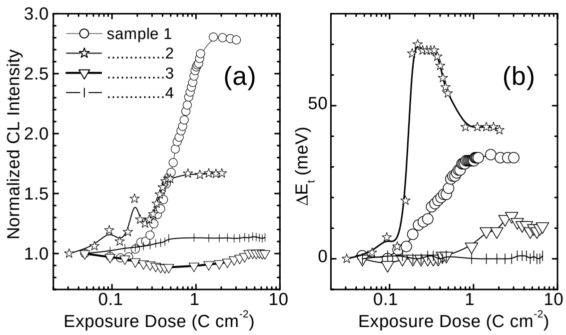

Fig. 1 (a) Integrated CL intensity and (b) variation of the optical transition energy of (In,Ga)N/GaN QWs fabricated by MOCVD (samples 1 and 2) and MBE (sample 3) as well as of a GaAs/(Al,Ga)As single QW (sample 4) as a function of the electron exposure dose at 5 K.

respectively. Both I_{CL} and ΔE_t of the MOCVD-grown QWs (samples 1 and 2) are significantly increased by LEEBI, while for the GaAs-based QW, η and ΔE_t remain almost the same during the electron exposure. For the MBE-grown GaN QW, only ΔE_t increases by about 10 meV after an exposure dose of 2 C cm^{-2}.

The observed increase of η accompanied by a blue-shift of E_t can be interpreted as a result of electric-field screening in the QWs. Similar results have been obtained at 300 K, where η is even more enhanced by LEEBI compared with the low temperature exposure. As has been shown by Gfrörer et al. [9], the electric field of QWs situated closely to the surface can be effectively screened by the electric field arising from spontaneous surface polarization. An electron-beam-induced variation of the electric field of the surface depletion region and of its width can occur by modification of the charge state of the surface and by activation of donors within the near-surface region of the structure. While the screening of the QW field reported in [9] is dominated by surface charge states, two arguments provided by the current study suggest that LEEBI-induced dopant activation can be additionally responsible for the modification of the electronic properties of GaN-related QW structures.

Firstly, the comparison of MOCVD- with MBE-grown QWs shows that LEEBI effects are much stronger in QWs fabricated by MOCVD. Since it is known that hydrogen – involved in MOCVD – effectively passivates dopants, an electron-beam-induced activation process can be expected, in particular, in QW grown by MOCVD. Secondly, besides a blue-shift and an increase of the QW CL of the MOCVD-grown samples, we observe also an increase of η in the GaN buffer layer as a function of D_c (not shown). For sample 1, the GaN CL (at 5 K) is dominated by free and bound exciton transitions before and after LEEBI, respectively. Moreover, at least two distinct bound exciton states evolve as a result of electron exposure a donor and an acceptor bound one. For sample 2, we observe an increase and red-shift of the bound exciton and an increase of the donor-acceptor-pair as well as of the yellow luminescence. Consequently, it appears that an activation of both hydrogen-passivated acceptors and donors occurs during LEEBI of unintentionally doped GaN QWs. In contrast to *p*-type GaN, the increase of η of the QWs and GaN buffer layer is not accompanied by a decrease of the resistivity after electron exposure. Hall measurements did not show any variation of the sheet resistivity before and after LEEBI at 300 K. Consequently, LEEBI results not in a significantly increased net free-carrier concentration. This result is consistent with the observation of a simultaneous LEEBI-induced activation of acceptors and donors preserving the compensating state of the material, while the probability for optical transitions and thus η increases significantly. An increased concentration of charged donors within the surface depletion region leads to an enhancement of the surface polarization field (narrowing of the depletion region) and therefore, to a blue-shift of the QW CL.

3.2 QW within a *p-n* junction

Figure 2 shows the EBIC signal as a function of the bias voltage (U_{bias}) of an unexposed (squares) and exposed (dashed line) region of the LED. Clearly, LEEBI leads to a significant reduction of the EBIC signal. Moreover, spatially resolved electroluminescence (EL) (not shown) revealed that the EL intensity is reduced by a factor of two within the exposed regions. This LEEBI-induced "damage" is almost completely annealed after 4 hours operation of the diode at 10 mA (self-annealing). Even a thermal annealing for an hour in N_2 atmosphere at a temperature as low as 250 °C recovers the EBIC signal almost completely (cf. solid line in Fig. 2). The exposed regions become visible as dark rectangles in the EBIC images shown at the right hand side of Fig. 2. For zero bias, this contrast disappears after thermal annealing, but appears again if the bias voltage is increased in the annealed sample. These results suggest that the *p-n* junction has been disturbed by LEEBI, which appears as the system is turned towards the forward bias condition.

This statement is confirmed by the comparison of the bias and exposure dose dependencies of I_{CL} and E_t of the QW situated within the *p-n* junction (not shown). For zero-bias, an electron exposure up to 0.7 C cm^{-2} leads to a red-shift of E_t by 40 meV, to an increase of I_{CL} by an order of magnitude and to a decrease of the EBIC signal (as shown in Fig. 2). The same behavior is observed when U_{bias} is increased from 0 to 2 V in forward direction, while the electron dose is kept low enough ($<$5 mC cm^{-2}) in order to prevent exposure effects. This bias dependence can be understood in terms of the interaction of the internal

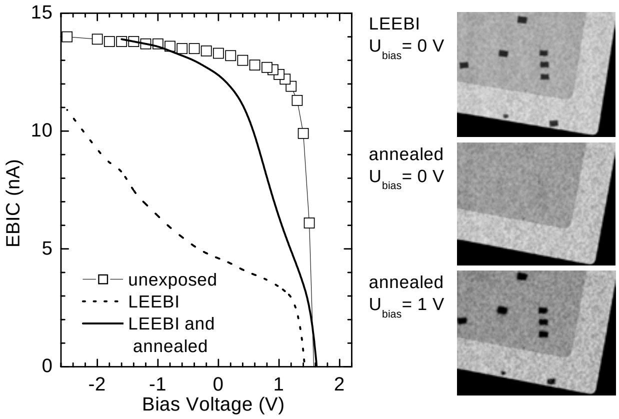

Fig. 2 EBIC of an (In,Ga)N/GaN LED as a function of the bias voltage at 300 K. Squares and the dashed line represent unexposed and exposed (1 C cm^{-2}) regions, respectively. The solid line has been obtained from the exposed region after thermal annealing for 1 hour in N_2 atmosphere. Right hand side: EBIC images of the LED before and after thermal annealing.

electric field of the QW with the external electric field of the *p-n* depletion region as has been described, e. g., by Jho et al. [13]. Briefly, since the electric field of the *p-n* junction and the one of the QW act in opposite directions, an increasing forward bias (decreasing external field) leads to an enhancement of the electric field in the QW and to a decreased escape probability of carriers out of the QW. Thus, E_t is red-shifted and I_{CL} is increased. Since the same is observed for zero bias but increasing values of D_c, we conclude that LEEBI turns the system towards the flat band condition.

A possible explanation for the variation of the field distribution during LEEBI is passivation of acceptors in the *p*-type layer by electron- beam-induced diffusion of hydrogen into the region of the *p-n* junction. The observed self-annealing of the LEEBI-related ”damage” is consistent with the activation of hydrogen-passivated acceptors by electrical injection of minority carriers as reported by Miyachi et al. [14] and with the low temperatures required for thermal annealing.

References

[1] H. Amano, H. Kito, K. Hiramatsu, and I. Akasaki, Jpn. J. Appl. Phys. Part 2 **28**, L2112 (1989).
[2] X. Li, S. Q. Gu, E. E. Reuter, J. T. Verdeyen, S. G. Bishop, and J. J. Coleman J. Appl. Phys. **80**, 2687 (1996).
[3] M. Toth, K. Fleischer, and M. R. Phillips, Phys. Rev. B **59**, 1575 (1999).
[4] S. Dassonneville, A. Amokrane, B. Sieber, J.- L. Farvacque, B. Beaumont, P. Gibart, J.-D. Ganiere, and K. Leifer, J. Appl. Phys. **89**, 7966 (2001).
[5] S. Nakamura, N Iwasa, M. Senoh and T. Mukai, Jpn. J. Appl. Phys. Part 1 **31**, 1258 (1992).
[6] S. J. Pearton, J. W. Lee, and C. Yuan, Appl. Phys. Lett. **68**, 2690 (1996).
[7] C. H. Seager, S. M. Myers, B. Vaandrager, and J. S. Nelson, Appl. Phys. Lett. **80**, 2693 (2002).
[8] O. Mayrock, H.- J. Wünsche, and F. Henneberger, Phys. Rev. B **62**, 16870 (2000).
[9] O. Gfrörer, C. Gemmer, J. Off, J. S. Im, F. Scholz, and A. Hangleiter, Phys. Stat. Sol. (b) **216**, 405 (1999).
[10] R. Pecharroman-Gallego, P. R. Edwards, R. W. Martin, and I. M. Watson, Material Science and Engineering B **93**, 94 (2002).
[11] J. Off, A. Kniest, C. Vorbeck, F. Scholz, and O. Ambacher, J. Crystal Growth **195**, 286 (1998).
[12] S. Nakamura, M. Senoh, N. Iwasa, S. Nagahama, T. Yamada, and T. Mukai, Jpn. J. Appl. Phys. Part 2 **34**, L1332 (1995).
[13] Y. D. Jho, J. S. Yahng, E. Oh, and D. S. Kim, Appl. Phys. Lett. **79**, 1130 (2001).
[14] M. Miyachi, T. Tanaka, Y. Kimura, and H. Ota, Appl. Phys. Lett. **72**, 1101 (1998).