APPLIED PHYSICS LETTERS **89**, 023103 (2006)

# Enhancement of free-carrier screening due to tunneling in coupled asymmetric GaN/AlGaN quantum discs

Kwan H. Lee, Jong H. Na, Robert A. Taylor,[a] and Sam N. Yi
*Department of Physics, University of Oxford, Parks Road, Oxford OX1 3PU, United Kingdom*

Stefan Birner
*Walter Schottky Institute and Physics Department, Technical University of Munich, D-85748 Garching, Germany*

Young S. Park, Chang M. Park, and Tae W. Kang
*Quantum-functional Semiconductor Research Center, Dongguk University, Seoul 100-715, South Korea*

(Received 6 February 2006; accepted 5 June 2006; published online 11 July 2006)

We present an investigation of free-carrier screening in coupled asymmetric GaN quantum discs with embedded AlGaN barriers using time-integrated and time-resolved microphotoluminescence measurements, supported by three-dimensional multiband **k·p** computational modeling. We observe that with increasing optical excitation the carrier lifetime decreases and emission energy blueshifts. This originates from the screening of built-in piezo- and pyroelectric fields in the quantum discs by photogenerated free carriers. Due to nonresonant tunneling of carriers from the smaller disk to the larger disk, free-carrier screening is enhanced in the larger disk. Computational modeling was in good agreement with the experimental results. © *2006 American Institute of Physics*.
[DOI: 10.1063/1.2220482]

Low dimensional nitride-based semiconductors incorporating quantum wells (QWs) and quantum dots (QDs) have attracted much attention due to their application in blue-ultraviolet wavelength optoelectronic devices.[1] In particular, self-organized GaN nanocolumns have gained interest as they possess desirable properties such as low defect density, good reproducibility, and quantum confinement effects,[2] and can be produced using conventional growth techniques without additional processing. These properties make nanocolumns an attractive basis for future quantum optoelectronic devices (such as embedded QDs in nanopillar cavities for quantum information processing[2]). Significant advancements have been made towards practical devices, with GaN nanocolumn experiments employing Bragg reflectors,[3] electrical excitation[4] and stimulated emission[5] having been undertaken. QW-like structures grown at the tip of GaN nanocolumns are known as quantum discs (Q-discs) and demonstrate lateral confinement. We have investigated coupled asymmetric GaN Q-discs separated by thin AlGaN barriers, as carrier tunneling in such structures has been suggested previously[6] and could play a role in future devices.

In this letter we present measurements and analysis of free-carrier screening and optical nonlinearity in a III-nitride Q-disc structure, with an enhancement in the emission from the larger Q-disc due to carrier tunneling. We have examined these effects using a combination of time-resolved (TR-) and time-integrated (TI-) photoluminescence (PL) spectroscopy and multiband **k·p** computational modeling with NEXTNANO[3].[7] From the TI- and TR-PL measurements we observe that increased optical excitation leads to a reduction in the lifetime and blueshift in the emission, with the effect being more pronounced in the larger Q-disc due to carrier tunneling.

The GaN/AlGaN Q-discs were grown at the tip of a 2.7-$\mu$m-high GaN nanocolumn using plasma assisted molecular beam epitaxy on a Si (111) substrate without any buffer layer.[8] The GaN nanocolumns exhibited an average diameter of 100 nm and a density of $1\times10^{9}$ cm$^{-2}$. The Q-disc sample consisted of ten periods of two alternating GaN Q-disc thicknesses of 4 and 3 nm, separated by an $Al_{0.5}Ga_{0.5}N$ barrier with an effective thickness of 1 nm. The larger Q-disc was referred to as DA, while the smaller Q-disc was referred to as DB. This structure was investigated by NEXTNANO[3], a nanostructure simulator capable of solving the self-consistent three-dimensional (3D) nonlinear Poisson-Schrödinger equation for wurtzite materials including strain, deformation potentials, and piezo- and pyroelectric charges. For the calculations, we employed the material parameters taken from Ref. 9. The fully strained Q-discs were simulated by minimization of the elastic energy within a continuum model approach that takes into account the symmetry of the hexagonal crystal structure. For the air-semiconductor interface we assumed that the atoms at the GaN boundary of the Q-disc were not allowed to relax into the surrounding air material. In order to calculate the wave functions a single-band model for the electrons and a six-band **k·p** Hamiltonian for the holes were considered. This could be justified as GaN and AlN have large band gaps and therefore the coupling between the conduction and valence bands can be neglected for our purpose.[10]

Figure 1(a) shows a visualization of the conduction and heavy hole valence band along the growth direction (i.e., *c* axis [0001]). Wurzite III-nitride heterostructures have a built-in piezo- and pyroelectric field (~ several MV/cm) which tilts the bands, giving rise to a quantum confined Stark effect (QCSE) for the carriers. This results in a redshift and the spatial separation of the confined electrons and holes. The calculated energy levels for the electrons and holes and the DA and DB emission energies are also presented in Fig. 1(a). The emission energies from DA and DB were calculated to be 3.47 and 3.58 eV, respectively. The calculations show that the tunneling of electrons from DB to DA is pos-

[a]Electronic mail: r.taylor1@physics.ox.ac.uk

 

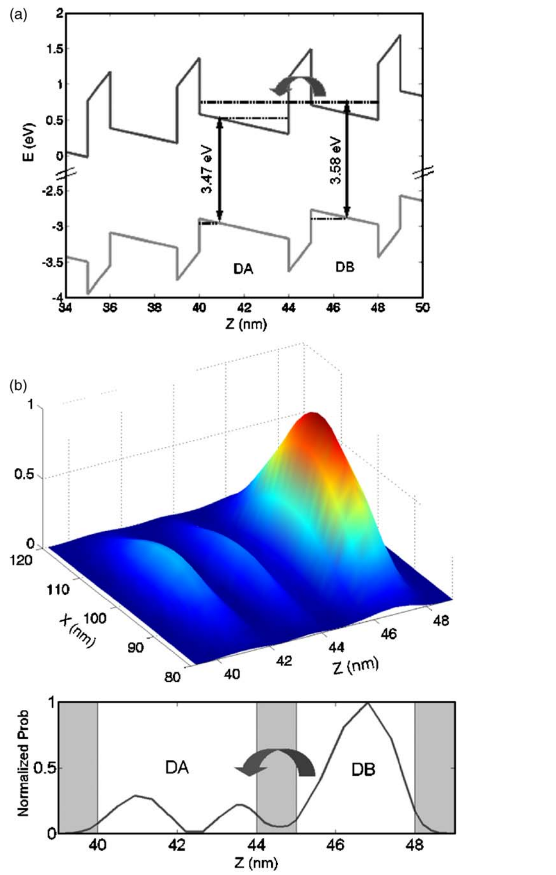


FIG. 1. (Color online) (a) Band diagram for electrons and holes. (b) Electron wave functions calculated using NEXTNANO[3] showing that the DB ground state extends into DA, giving rise to the possibility of tunneling from DB to DA.

sible, as the electron energy of DB lies above the triangular potential of the DA energy state. However, the converse is not true as the electron energy in DA was lower (by ~0.1 eV) and the energy of the electron lies within the triangular region of potential, implying stronger localization. The effect of tunneling was investigated by considering the squared wave functions that were obtained as the solutions of the Schrödinger equation, which was solved for the three-dimensional region that includes both Q-discs. Figure 1(b) shows that the probability density for the ground state of the electron in DB tunnels through the AlGaN barrier and extends into DA. Hence, even under nonresonant condition, there is evidence of weak coupling between the Q-discs.

TI-PL and TR-PL measurements were undertaken in order to look for the effects of the predicted tunneling. For these measurements, frequency-tripled femtosecond Ti:sapphire laser pulses at 266 nm were used for excitation. These pulses had duration of 150 fs and a repetition frequency of 76 MHz (Coherent MIRA). The sample was mounted in a continuous-flow helium cryostat (Janis ST-500) and the temperature was maintained at 4 K. A 36× reflecting objective (Ealing) was held above the cryostat to both focus the incident laser beam to a spot size of ~2 $\mu$m and to collect the resulting luminescence. The PL was dispersed by a 0.3 m spectrograph, which was equipped with a 1200 grooves/mm grating giving a spectral resolution of ~0.7 meV. For TI measurements the PL was detected by a Peltier-cooled charge-coupled device (CCD), and for TR measurements the PL was directed to a commercial time-correlated single pho-

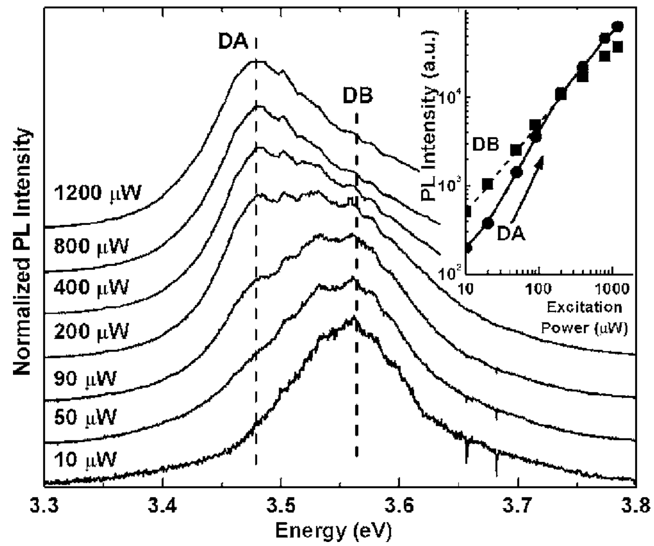


FIG. 2. Normalized TI-PL spectra of the GaN Q-discs. The inset shows a plot of peak intensity vs excitation power.

ton counting system with a rise time of 50 ps.

Optical nonlinearities in the Q-disc were investigated using TI-PL. Spectra as a function of excitation power ranging from 10 $\mu$W to 1.2 mW (at the focus) are presented in Fig. 2. Two peaks were observed at 3.48 and 3.56 eV, labeled DA and DB in the spectra. These were in good agreement with the calculated values (3.47 and 3.58 eV). As the excitation power increased the DA peak grew more rapidly, relative to DB peak, and it eventually dominated the spectra. This is reflected in the peak intensity versus excitation power plot (Fig. 2 inset), which showed that the DB peak intensity was proportional to the excitation power, while the DA peak intensity displayed a nonlinear dependence with excitation power. This observation is consistent with earlier studies involving GaAs/AlGaAs asymmetric QWs, which demonstrated nonlinear PL dependence with excitation power, due to carrier tunneling.[11]

Through TR-PL measurements, the effects of free-carrier screening were studied. Figure 3 shows the TR-PL trace collected from the DA and DB peaks, at high (600 $\mu$W) and low (40 $\mu$W) excitation power. The TR-PL traces show a single exponential decay at low excitation power, with fitted lifetimes of $\tau_{DA}=460$ ps for DA and $\tau_{DB}=360$ ps for DB. At high excitation power the TR-PL traces show a biexponential decay, with a fast component of $\tau_1=100$ ps, which was common to both DA and DB, and $\tau_{DA}=300$ ps and $\tau_{DB}=300$ ps. The fast component was identified with free carrier decay and screening, which has a more significant contribution with increasing optical excitation. The decrease in $\tau_{DA}$ and $\tau_{DB}$ with increasing optical excitation could be explained by free-carrier screening,[12] which compensates the built-in piezo- and pyroelectric fields (responsible for the QCSE). The photogenerated free carriers screen the built-in fields, which then leads to a reduction in the band structure tilt and consequently the overlap between electron and hole increases, which corresponds to an increased oscillator strength (stronger peak intensity and decreased carrier lifetime).

Enhancement of the free-carrier screening due to tunneling was investigated by reconstructing the spectrum from a series of TR-PL measurements, which were performed at 0.02 eV intervals (from 3.44 to 3.64 eV) for an incident

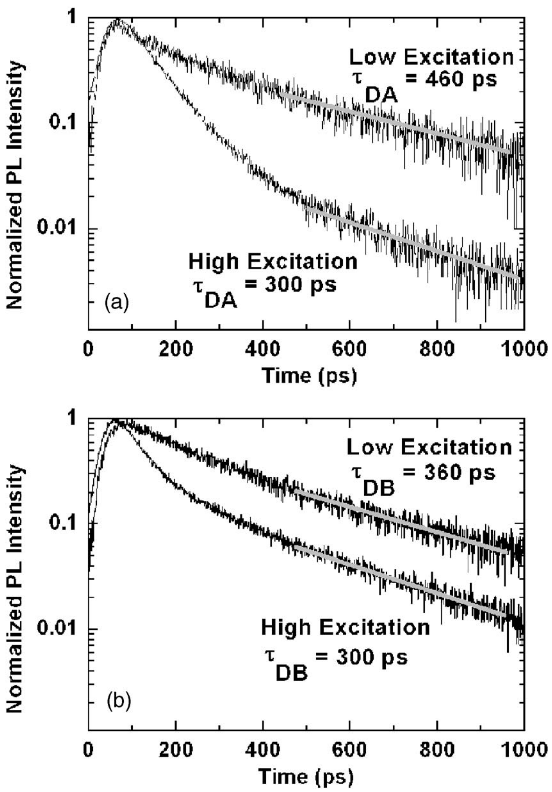


FIG. 3. TR-PL measurement from DA (a) and DB (b) at low and high excitation powers. The effect of the free-carrier screening can be seen, with the carrier lifetime decreasing with increasing optical power.

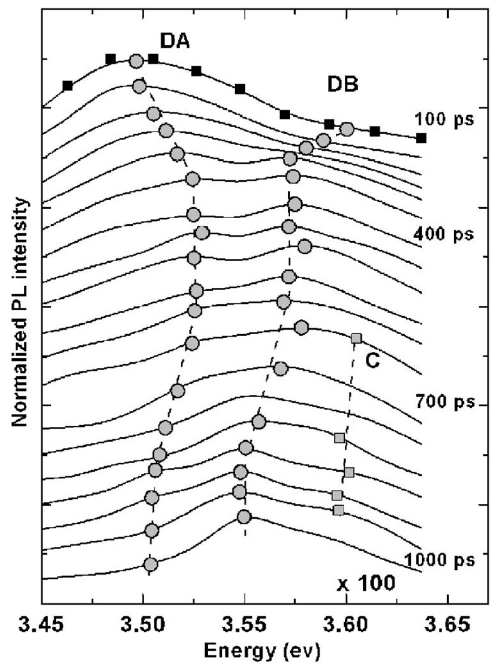


FIG. 4. Reconstruction of the spectrum from TR-PL measurements at an excitation power of 600 $\mu$W. The time increment between each line is 50 ps. Time origin as in Fig. 3.

power of 600 $\mu$W, as shown in Fig. 4. The measured response displays several interesting features. Initially the DA and DB peaks are blueshifted (3.50 and 3.60 eV) compared to TI-PL peaks. During the initial decay period (0–400 ps) DA peak blueshifts linearly at 0.15 meV/ps while DB peak redshifts and sharpens. This behavior arises from the transfer of electrons from DB to DA (i.e., tunneling) and hence the DA electron population increases, leading to a blueshift and decrease in the QCSE due to free-carrier screening. This phenomenon also corresponds to the transition region on the TR-PL curve for DA, at which the trace moves from the rapid to the slower decay characteristics. Between 400 and 700 ps the rate of tunneling decreases, with a reduced electron population in DB, and the carriers in DB and DA recombine (with a lifetime of ~300 ps). After 700 ps DA and DB experience a redshift as the carriers further recombine, and hence the effect of the free-carrier screening is reduced. Here, the peak energies for DA and DB return to the values seen at low excitation. The peak observed at ~3.60 eV (labeled C) corresponds to the GaN nanocolumn.[6] Overall, this response is in good agreement with behavior observed in symmetric III-nitride QWs (e.g., InGaN/GaN).[13] These measurements demonstrate that the free-carrier screening is enhanced in DA and agrees with predictions from TI-PL and computational modeling results. The dynamic response of the system explains why the DA blueshift could not be observed in the TI-PL spectrum, which is a summation of the PL slices in Fig. 4. Further studies reveal that this enhancement decreases as the barrier thickness increases (from 2 to 8 nm).

In conclusion free-carrier screening in a coupled asymmetric Q-disc system has been investigated using a combination of TI-PL and TR-PL techniques, and NEXTNANO³ multiband **k·p** computational modeling. We observe that with increasing optical excitation the carrier lifetime reduces and the emission energy is blueshifted. This is attributed to free-carrier screening of the built-in piezo- and pyroelectric fields. Furthermore, these results provide consistent evidence of electron tunneling from DB to DA and enhanced the free-carrier effect in DA. Changes in the carrier lifetime and blueshift were more pronounced in DA.

This research is part of the QIP IRC supported by EPSRC (GR/S82176/01). One of the authors (K.H.L.) acknowledges the support of the University College old members fund scholarship, Clarendon Fund bursary, Overseas Research Students award, and M. A. G. Jones for computational support. Three of the authors (Y.S.P., C.M.P., and T.W.K.) thank QSRC and AOARD at Dongguk University.

[1] S. Nakamura, M. Senoh, S. I. Nagahama, N. Iwasa, T. Yamada, T. Matsushita, H. Kiyokyu, and Y. Sugimoto, Jpn. J. Appl. Phys., Part 2 **35**, L74 (1996).
[2] M. Zamfirescu, M. Abbarchi, M. Gurioli, A. Vinattieri, J. Ristić, and E. Calleja, Phys. Status Solidi C **2**, 822 (2005).
[3] J. Ristić, E. Calleja, A. Trampert, S. Fernández-Garrido, C. Rivera, U. Jahn, and K. H. Ploog, Phys. Rev. Lett. **94**, 146102 (2005).
[4] A. Kikuchi, M. Kawai, M. Tada, and K. Kishino, Jpn. J. Appl. Phys., Part 2 **43**, L1524 (2004).
[5] A. Kikuchi, K. Yamano, M. Tada, and K. Kishino, Phys. Status Solidi B **241**, 2754 (2004).
[6] J. H. Na, R. A. Taylor, J. H. Rice, J. W. Robinson, K. H. Lee, Y. S. Park, C. M. Park, and T. W. Kang, Appl. Phys. Lett. **86**, 083109 (2005).
[7] NEXTNANO³ device simulator, the program is available at www.wsi.tum.de/nextnano3 and www.nextnano.de
[8] C. M. Park, Y. S. Park, Hyunsik Im, and T. W. Kang, Nanotechnology **17**, 952 (2006).
[9] I. Vurgaftman and J. R. Meyer, J. Appl. Phys. **94**, 3675 (2003).
[10] V. A. Fonoberov and A. A. Balandin, J. Appl. Phys. **94**, 7178 (2003).
[11] D. J. Leopold and M. M. Leopold, Phys. Rev. B **42**, 11147 (1990).
[12] F. D. Sala, A. Di Carlo, P. Lugli, F. Bernardini, V. Fiorentini, R. Scholz, and J.-M. Jancu, Appl. Phys. Lett. **74**, 2002 (1999).
[13] T. Kuroda, A. Tackeuchi, and T. Sota, Appl. Phys. Lett. **76**, 3753 (2000).