Spectral and temporal resolution of recombination from multiple excitation states in modulation-doped AlGaN/GaN multiple quantum well heterostructures

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Time-resolved photoluminescence measurements of carrier lifetimes in modulation-doped (100 Å) $Al_xGa_{1-x}N$/(100 Å) GaN multiple quantum well heterostructures are reported. The photoluminescence (PL) spectrum exhibits several lines associated with recombination of carriers from multiple excited electron states to the hole ground state. The PL decay times associated with ground state recombination, e_0h_0, are found to be much longer than the inverse repetition rate of our system (20 μs) and estimated to be 9 ms. The experimental lifetimes associated with carrier recombination from excited states vary between 4 μs for the first excited state, e_1h_0, and 4.5 ns for the fourth excited state, e_4h_0. These lifetimes are in very good agreement with a self-consistent calculation of radiative recombination lifetimes which takes into account piezoelectric and spontaneous polarization. The significant differences in recombination lifetimes are the result of the large built-in electric field in the wells (0.5 MV/cm). © *2005 American Institute of Physics.* [DOI: 10.1063/1.1905785]

AlGaN/GaN heterostructures are of great interest for the development of optical devices operating in the UV range due to the large band gap of $Al_xGa_{1-x}N$ extending from 3.4 eV (for $x=0$) to 6.12 eV (for $x=1$). The photoluminescence (PL) properties of these quantum wells (QWs) structures are significantly influenced by the presence of large built-in spontaneous and piezoelectric fields[1–4] which result in the spatial separation of electron and holes and a large red shift of the photoluminescence spectrum (quantum confined Stark effect). It was recently demonstrated that *p*-type modulation doped AlGaN/GaN multiple quantum wells (MQWs) structures possess excellent electrical properties such as low resistivity, a temperature-independent free carrier concentration,[5] as well as very well resolved optical emission lines.[6] At low temperatures, hole mobilities as high as 36 cm²/V s and carrier concentrations of 2.5×10^{18} cm⁻³ were measured in such structures.[5] The percentage of activated acceptors is very large in these structures because the acceptor level in the barrier aligns with the hole energy levels in the wells.[7]

Continuous wave (CW) photoluminescence experiments on such a *p*-type modulation doped (100 Å)$Al_{0.2}Ga_{0.8}N$/(100 Å)GaN MQW structure performed by Waldron *et al.* revealed for the first time several peaks in the PL spectrum, associated with recombination of electrons from excited states in the GaN quantum well.[6] This assignment was based on a self-consistent Poisson–Schrödinger calculation of the transition energies associated with the recombination of electrons from five confined states in the conduction band well with ground-state holes in the valence band well.

The calculation also predicted the spontaneous recombination rates are dramatically influenced by the presence of the built-in electric fields since the spontaneous recombination rate is proportional to the momentum matrix element between the electron and holes states, in the dipole approximation. The momentum matrix element in turn is proportional to the electron and hole envelope function overlap.

In this work, we report on time-resolved photoluminescence studies of the *p*-type modulation doped $Al_{0.2}Ga_{0.8}N$/GaN MQWs. The time-integrated luminescence spectra reproduce the CW data obtained by Waldron *et al.*[6] confirming the presence of several distinct and well-resolved peaks in the emission spectrum indicating the high quality of the quantum wells. Dramatic changes in the relative intensities between the features in the spectrum are observed as a function of the electron concentration inside the wells. Most importantly, the radiative lifetimes vary significantly across the PL spectrum and are in good agreement with the calculations of Waldron *et al.*

The sample under study was the same Ga-faced $Al_{0.2}Ga_{0.8}N$/GaN MQW structure grown on a *c*-plane sapphire substrate by molecular beam epitaxy, used in Refs. 5 and 6. The structure is nominally modulation-doped (barriers only) with a Mg level of $N_{Mg}=1 \times 10^{19}$ cm⁻³ and consists of 20 periods with an equal barrier and well width of 100 Å. This width was chosen in order to minimize the effects of growth irregularities and alloy scattering. The epitaxial lay-

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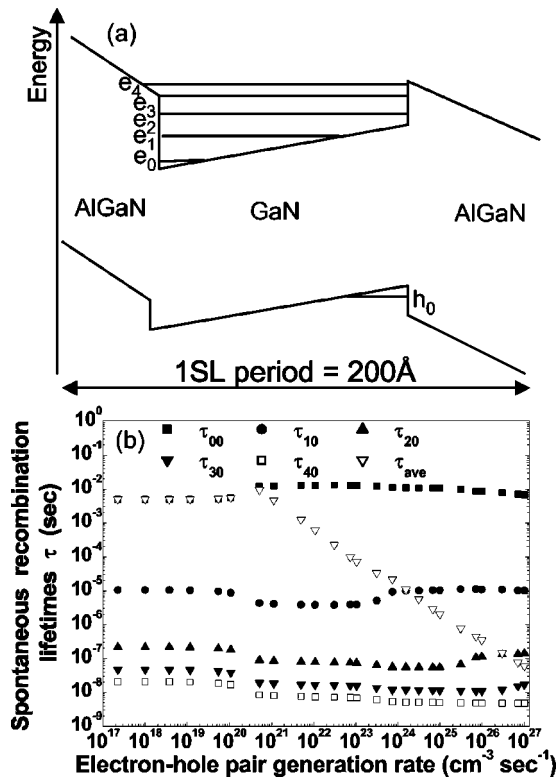[b)]Author to whom correspondence should be addressed; electronic mail: anc@buffalo.edu

FIG. 1. (a) Schematic band diagram for a 100 Å $Al_{0.2}Ga_{0.8}N/GaN$ QW structure. Calculations predict the existence of five electron bound states in the conduction band well. (b) Calculated spontaneous radiative recombination lifetimes associated with the five electronic bound states in an 100 Å $Al_{0.2}Ga_{0.8}N/GaN$ QW as a function of electron-hole pair generation rate.

FIG. 2. Time-integrated PL spectra from the sample under study for integration times ranging from 2 ns to 10 ms.

ers were grown pseudomorphically on a thick GaN buffer layer which, in turn, was grown on a nucleation layer and a sapphire substrate, resulting in the presence of a biaxial tensile strain in the barriers.

The frequency tripled 800 nm pulsed output of a Coherent REGA 9000 regenerative amplifier provided 266 nm excitation pulses at a repetition rate of 50 kHz. The excitation beam was focused onto the sample placed into a closed cycle refrigerator using conventional optics. The PL was collected in the backscattering geometry and spectrally and temporally resolved by a ChroMEX 250IS spectrometer equipped with a Hamamatsu Streak Camera (C3443) characterized by a 50 ps jitter. All measurements were performed at 10 K.

Figure 1(a) shows the schematic of the band diagram for the AlGaN/GaN quantum well. The energy bands are tilted as a result of the polarization effects. Under these conditions, electrons and holes are driven towards opposite sides of the quantum well and the overlap of the ground-state wave function is significantly reduced in comparison to the hypothetical flatband situation. Furthermore, the excited states are less localized and thus less affected by built-in fields. The wave function overlap between excited-state electrons and ground-state holes is considerably larger. The calculated spontaneous recombination lifetimes $\tau_{n,0}$, corresponding to recombination of electrons in the nth state (e_n, $n = 0$, 1, 2, 3, or 4) and ground state (h_0) holes, taken from Ref. 6, are summarized in Fig. 1(b). The recombination lifetime associated with the ground state is expected to be at least four orders of magnitude larger than the lifetime of the fourth excited state which remains largely unaffected by the built-in polarization. As a result, at high excitation intensities, when the number of pho-
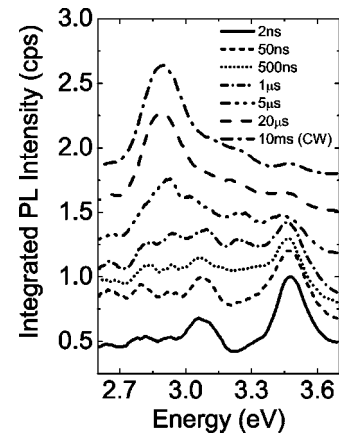
togenerated electrons is large enough to occupy the higher-energy states in the quantum well, transitions between excited electron states and ground-state holes become observable.

Figure 2 presents a series of time-integrated PL spectra from the sample under study for integration times ranging from 2 ns to 10 ms. The relative intensity between the features in the spectrum changes dramatically as a function of integration time, an indication that the radiative transitions contributing to the PL spectrum are characterized by different recombination lifetimes. Integrating over different periods of time after the laser pulse was incident on the sample is equivalent to performing a CW power study. The carrier concentration injected in each well is of the order of 10^{13} cm^{-2}/pulse, which is enough to fully screen the built-in electric field present in the well. Consequently, the PL spectrum sampled immediately after pulsed excitation is dominated by the features associated with recombination of the few short-lived electrons in the excited states. This is exemplified by the 2 ns spectrum shown in Fig. 2. This spectrum is very similar to the high power CW spectrum reported previously.[6] At longer delay times the spectrum becomes dominated by the recombination of the long-lived electrons
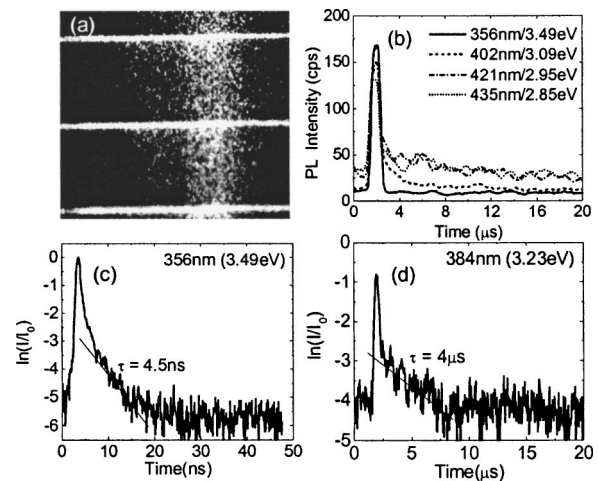

FIG. 3. (a) A streak camera image of the PL spectrum sampled over two consecutive 20 μs intervals between pulses. (b) Vertical cross section through the camera image in (a) at four different energies across the spectrum. (c) PL intensity decay at 3.49 and (d) 3.23 eV.

TABLE I. Calculated and measured lifetimes associated with the radiative recombination of an electron on one of five different bound states in the conduction band and a ground state hole from the valence band.

Transition	E(eV)	$\tau_{calculated}$(s)	$\tau_{measured}$(s)
$e_0 h_0$	3.084	9.3×10^{-3}	$\gg 20 \times 10^{-6}$
$e_1 h_0$	3.228	8.2×10^{-6}	4×10^{-6}
$e_2 h_0$	3.350	1.1×10^{-7}	3×10^{-7}
$e_3 h_0$	3.443	2.1×10^{-8}	1×10^{-7}
$e_4 h_0$	3.521	9.1×10^{-9}	4.5×10^{-9}

from the ground state and resembles the low power CW spectrum.

In order to obtain a quantitative measure of recombination lifetimes for each of the bound states in the conduction band, we analyzed the PL intensity decay at several energies across the spectrum, energies which correspond to the $e_n h_0$ ($n=0$, 1, 2, 3, or 4) recombination. Figure 3(a) represents a streak camera image of the PL spectrum sample over two consecutive 20 μs intervals between pulses. This image indicates that, on the low energy side of the spectrum, there are still electrons left in the conduction band when the next pulse is incident on the sample. For this low-energy state, the lifetimes are considerably longer than the time interval between two excitation pulses. A vertical cross section through the camera image at four different energies across the spectrum [Fig. 3(b)] indicates the decay is nonexponential with a component much longer than the repetition rate (which manifests itself through the constant background measured at low energies). On the high energy side of the spectrum, the lifetimes are shorter as illustrated in Figs. 3(c) and 3(d). A comparison between these lifetimes and the corresponding theoretical estimates at energies associated with the $e_n h_0$ transitions is presented in Table I. With the exception of the lifetime of the electrons in the ground state, which is too long in comparison with the inverse repetition rate of our laser system, all other lifetimes are in good agreement with the predicted values, confirming that the features observed in the PL spectrum are associated with recombination of electrons in excited states.

In conclusion, time-resolved photoluminescence measurements performed on a p-type modulation doped GaN/AlGaN superlattice structure identified several distinct emission lines observed in the PL spectrum. The emission lines are attributed to recombination of electrons occupying five confined states in the conduction band well with ground-state holes from the valence band well. The radiative recombination lifetimes associated with each of the five electronic states are dramatically different and range from 4.5 ns to $\gg$20 μs. This large range is attributed to the spatial separation of electrons and holes as a result of the built-in electric fields present in the GaN wells.

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