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Time-resolved photoluminescence of quaternary AlInGaN-based multiple quantum wells

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Time-resolved photoluminescence (PL) dynamics has been studied in AlInGaN/AlInGaN multiple quantum wells (MQWs) grown by a pulsed metalorganic chemical vapor deposition (PMOCVD) procedure. The PL decay kinetics was found to be sensitive to the emission energy and temperature. The PL decay time increases with decreasing emission energy, which is a characteristic of localized carrier/exciton recombination due to alloy fluctuations. Its temperature dependence shows radiative recombination to be the dominant process at low temperatures, indicating a high quality of PMOCVD grown quaternary AlInGaN MQWs and establishing them as promising structures for the active region of deep ultraviolet light emitting diodes. © *2002 American Institute of Physics.*
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III-Nitride materials have attracted a great deal of interest due to their potential applications in UV/blue light emitters,[1–4] UV detectors,[5] and high power, high temperature electronic devices.[6,7] UV light emitting diodes (LEDs) with emission in the range of 300–350 nm are ideal for pumping the fluorescent film of full color display devices. The most promising materials for active layers of such UV emitters are either ternary $Al_xGa_{1-x}N$ or quaternary $Al_xIn_yGa_{1-x-y}N$ due to the desired wide direct band gap range. Several groups have recently reported on UV LEDs at 350 nm using AlGaN multiple quantum wells (MQWs) in the active region.[8,9] We have also reported on deep UV LEDs with emission from 305–340 nm using AlInGaN MQWs in the active region.[10,11] In our work the AlInGaN MQW layers for the active region were deposited using a pulsed atomic layer epitaxy (PALE) approach. More recently, we have also demonstrated a pulsed metalorganic chemical vapor deposition (PMOCVD) technique.[12] Although both techniques operate with group III metalorganic (MO) precursor gases (for Al, In and Ga) being supplied in a pulsed mode, the essential difference is in the group V (NH_3) precursor supply. In PMOCVD the group III precursors were pulsed and the NH_3 supply was kept on all the time, whereas in PALE both the MO gases and the NH_3 were pulsed and alternatively supplied in the growth chamber. Thus, PALE promotes essentially high quality two-dimensional (2D) growth of all GaN, AlN, and InN, whereas PMOCVD includes both 2D and 3D growth. In the latter case there is a possibility for compound segregation (especially In-related).[13] However, as yet, the photoluminescence (PL) emission dynamics of this PMOCVD quaternary AlInGaN material has not been systematically studied. In this letter, we report on the time-resolved PL studies of quaternary AlInGaN/AlInGaN MQWs grown by the PMOCVD procedure.

The AlInGaN/AlInGaN MQW samples for the study were grown on (0001)-oriented sapphire substrates. First, a 1-μm-thick intrinsic GaN epilayer was deposited using a 25-nm-thick low temperature AlN buffer layer and a conventional low pressure MOCVD process. The quaternary MQW structures were grown by a PMOCVD process at 750 °C.[12] In this procedure the group V precursor flow (NH_3) is kept constant throughout the growth. The group III precursors (Al, In, and Ga) are pulsed. The unit-growth cell consisting of the group III precursor pulses is repeated to build up the quaternary AlInGaN layer thickness. The number of Al, In, and Ga pulses in the unit-growth cell determines the alloy composition and the number of unit-growth cell repeats controls the layer thickness. The MQW structures consisted of three 5-nm-thick AlInGaN wells, and four 7-nm-thick AlInGaN barriers. Al molar fractions in the wells and barriers were close to 10% and 16%, respectively, and the In content in both wells and barriers was around 2%. The other details of the PMOCVD growth procedure can be found elsewhere.[12]

The quasi-stationary (with excitation pulses longer than radiative recombination decay times) PL measurements were performed at 10 K using a pulsed excimer laser ($\lambda = 193$ nm, $\tau = 8$ ns) as an excitation source. The luminescence was analyzed in backscattering geometry using a monochromator (SPEX550) with a UV enhanced charge coupled device array detector. In order to study the carrier dynamics in the AlInGaN MQWs, time-resolved PL was measured by using a time-correlated single photon counting system with time resolution better than 20 ps.[14] The excitation source was a picosecond mode-locked Ti:sapphire laser pumped by a frequency-doubled Nd:Vanadate ($Nd:YVO_4$) laser. The output laser pulses from the Ti:sapphire laser were frequency tripled ($\lambda = 273$ nm) by using a nonlinear crystal

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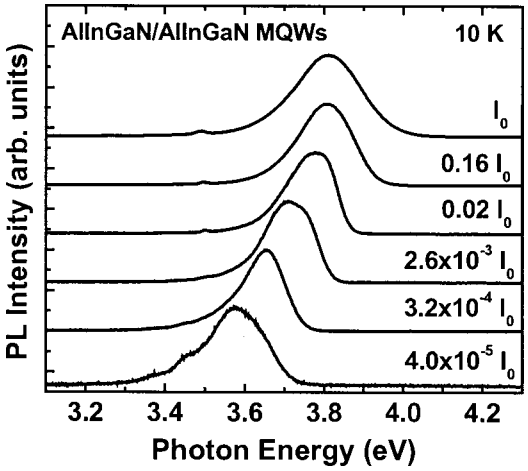

FIG. 1. PL spectra of $Al_{0.10}In_{0.02}Ga_{0.88}N/Al_{0.16}In_{0.02}Ga_{0.82}N$ MQWs for various excitation power densities at 10 K. The emission peak energy shows a clear blueshift with increasing excitation power density.

for sample excitation. They had a duration of less than 3 ps at 3.8 MHZ repetition rate and a average power of 100 μW.

Figure 1 shows the 10 K PL spectra of the $Al_{0.10}In_{0.02}Ga_{0.88}N/Al_{0.16}In_{0.02}Ga_{0.82}N$ MQWs with excitation power densities (maximum $I_0 \sim 2\ \mathrm{MW/cm^2}$) varying over five orders. As seen, under lowest excitation density ($I_{\mathrm{exc}} = 8.0\times10^{-5}\ \mathrm{MW/cm^2}$) the PL peak position is at 3.57 eV and has a full width at half maximum of about 170 meV. It shifts from 3.57 to 3.81 eV (blueshifts) with excitation power density increasing from 8.0×10^{-5} to $2\ \mathrm{MW/cm^2}$. This strong blueshift ($\sim$240 meV) is attributed to the combined effects of screening of quantum-confined Stark effect due to polarization fields and band filling of tail states caused by alloy compositional fluctuations.[13,15]

The emission energy dependence of the PL decay profiles in the quaternary AlInGaN/AlInGaN MQWs are shown in Fig. 2(a). The decay contains a fast and a slow component, and thus it cannot be described by a single exponential dependence. For analysis it is convenient to describe the decay process by a two-exponential function, $I(t) = A_1 \exp(-t/\tau_1) + A_2 \exp(-t/\tau_2)$, where τ_1 and τ_2 are the fast and slow decay times, respectively, and A_1 and A_2 are the contribution (e.g., in %) of the corresponding parts to the total PL intensity. The analysis of the data of Fig. 2(a) shows that in the energy

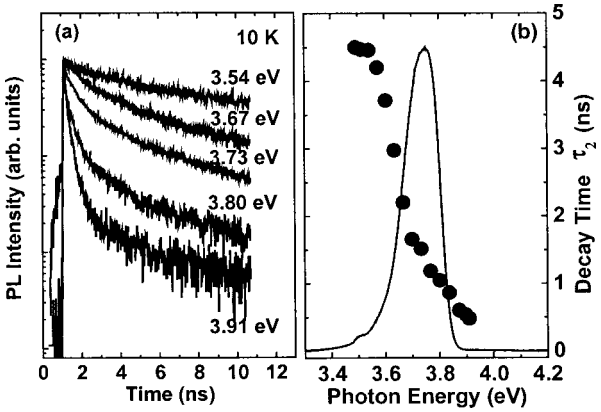

FIG. 2. (a) PL decay profiles and (b) decay time τ_2 and PL spectra ($I_{\mathrm{exc}} = 0.013\ \mathrm{MW/cm^2}$) of $Al_{0.10}In_{0.02}Ga_{0.88}N/Al_{0.16}In_{0.02}Ga_{0.82}N$ MQWs as a function of emission photon energy at 10 K.

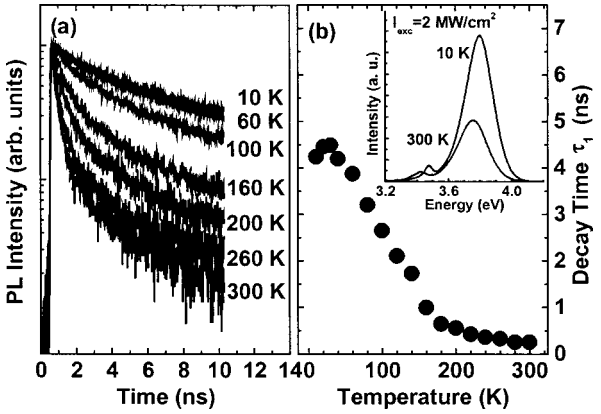

FIG. 3. Temperature dependence of (a) PL decay profiles and (b) PL decay time τ_1 of $Al_{0.10}In_{0.02}Ga_{0.88}N/Al_{0.16}In_{0.02}Ga_{0.82}N$ MQWs at the PL peak energy. The inset in (b) shows the PL spectra measured at 10 and 300 K.

region below 3.73 eV the slow component predominates, whereas in the higher energy region the role of fast component increases. With decreasing photon energy from 3.91 to 3.73 eV, τ_1 increases from 0.1 to 0.4 ns, and the corresponding fraction of PL amplitude decreases from 75 to 57 %. A similar behavior has been reported for InGaN/GaN MQWs,[14,16] in which the fast PL decay was assigned to nonradiative recombination and carrier capture at trap centers, whereas the slow component τ_2 was attributed to the radiative recombination of the localized states.

Figure 2(b) shows the slow-component decay time τ_2 as a function of energy in the range from 3.49 to 3.91 eV (points), as well as the corresponding PL spectra obtained under excitation power $I_{\mathrm{exc}} = 0.013\ \mathrm{MW/cm^2}$. As seen, the decay time is almost constant in the spectral range from 3.49 to 3.54 eV and then decreases from 4.5 to 0.5 ns as the photon energy increases from 3.54 to 3.91 eV. This variation of τ_2 with emission energy may be attributed to the recombination of excitons/carriers localized at potential wells due to compositional or other inhomogeneities in our AlInGaN/AlInGaN MQW structures. The decay (recombination) rate of the localized electron–hole pairs can be expressed by a radiative recombination rate plus a relaxation rate from higher energy sites to lower energy sites, e.g., tail states. The relaxation rate depends strongly on the density of the final lower-energy states, and thus should decrease with energy if the final states density is lowered. For the studied AlInGaN MQWs this explains data of Fig. 2(b). The emission energy dependence of PL decay time is ascribed to the recombination of localized excitons at band tail states. In previous studies a similar behavior of decay time was observed in InGaN[14,16–18] systems.

Figure 3(a) shows the time-resolved PL spectra for the quaternary AlInGaN MQWs as a function of temperature. The decay profiles were measured at the spectral peak position. As can be seen, at low temperatures the slow decay is predominant in the PL. However, at higher temperatures the fast component becomes more important. Assuming the radiative recombination as a two-exponential process, we analyzed the behavior of the fast component with temperature. The dots in Fig. 3(b) show temperature dependence of the decay time τ_1 in the range from 10 K to room temperature. The decay time increases with temperature up to 30 K. Such

an increase of the low temperature decay time τ_1 is generally accepted as a characteristic feature of radiative recombination in MQWs.[17] Our data therefore show that at low temperatures radiative recombination is the dominant process in PMOCVD grown AlInGaN MQWs. This also indicates the high quality of the quaternary AlInGaN MQWs used in this study. At high temperatures above 150 K, the PL decay becomes sufficiently faster. The fast decay component in this temperature region contributes more than 75% of the PL signal. The decrease of the PL decay time τ_1 with temperature might be related to the increased influence of nonradiative recombination process due to ionization of localized states and impurities.

The comparison of the present results with those of the PL for the AlInGaN layers grown by PALE process[10,19] revealed that the latter compounds had fewer band tail states and exhibited predominantly band-to-band UV emission. However, the higher number of band tail states in PMOCVD AlInGaN layers lead to an increase of spontaneous emission efficiency in these layers, which is similar to the results from localized band tail states in the InGaN material system. Our time-resolved PL results also show that the recombination kinetics in PMOCVD grown quaternary AlInGaN MQWs is dominated by electron–hole pairs (presumably excitons) localized at band tail states. These states can be attributed to alloy compositional fluctuations and/or interface disorders.

In summary, we have reported a study of time-resolved PL of quaternary AlInGaN/AlInGaN MQWs grown by PMOCVD. The strong blueshift of PL emission band with excitation power was ascribed to the combined results of the band filling of tail states and the screening of quantum-confined Stark effect. The emission energy and temperature dependence of the PL decay time indicates that the MQW recombination is dominated by localized carriers (presumably excitons) at band tail states due to alloy compositional fluctuations and/or interface disorders. The observed PL properties of PMOCVD grown AlInGaN MQWs agree well with those for the InGaN/GaN MQWs with a high density of localized states. This indicates that PMOCVD grown quaternary AlInGaN MQWs are promising materials for the active region of high power deep UV LEDs.

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