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Pulsed atomic layer epitaxy of quaternary AlInGaN layers

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In this letter, we report on a material deposition scheme for quaternary $\text{Al}_x\text{In}_y\text{Ga}_{1-x-y}\text{N}$ layers using a pulsed atomic layer epitaxy (PALE) technique. The PALE approach allows accurate control of the quaternary layer composition and thickness by simply changing the number of aluminum, indium, and gallium pulses in a unit cell and the number of unit cell repeats. Using PALE, AlInGaN layers with Al mole fractions in excess of 40% and strong room-temperature photoluminescence peaks at 280 nm can easily be grown even at temperatures lower than 800 °C. © 2001 American Institute of Physics. [DOI: 10.1063/1.1392301]

Solid-state white lighting is emerging as one of the most promising areas of application for III-nitride light emitting devices.¹ Phosphors pumped with high-power blue, GaN–InGaN multiple quantum well (MQW) light emitting diodes (LEDs) have in general been used to produce white light. However the use of blue LEDs, with an emission wavelength around 450 nm, severely limits the available phosphor choices and suffers from fundamental color rendering problems, especially when only one type (layer) of phosphor is used. These problems can be avoided by using LEDs with emission wavelengths in the 250–350 nm ultraviolet (UV) range combined with electrofluorescent-lighting conventional phosphors. III-N UV LEDs require the use of $\text{Al}_x\text{Ga}_{1-x}\text{N}$ or quaternary $\text{Al}_x\text{In}_y\text{Ga}_{1-x-y}\text{N}$ layers in the device's active region. The use of AlGaN, however, has been shown to significantly degrade the quantum well emission properties.² We are therefore exploring quaternary $\text{Al}_x\text{In}_y\text{Ga}_{1-x-y}\text{N}$ layers for the active region of MQW LEDs.

In the past, we have reported on the deposition of quaternary $\text{Al}_x\text{In}_y\text{Ga}_{1-x-y}\text{N}$ layers, GaN– $\text{Al}_x\text{In}_y\text{Ga}_{1-x-y}\text{N}$ heterojunctions,^{3,4} and InGaN– $\text{Al}_x\text{In}_y\text{Ga}_{1-x-y}\text{N}$ multiple quantum wells,⁵ using conventional low-pressure metalorganic chemical vapor deposition (MOCVD). Control of the quaternary layer composition was achieved by varying the precursor fluxes. The use of quaternary AlInGaN wells with InGaN quantum wells significantly increased the barriers' optical emission.⁵ The incorporation of In into ternary AlGaN barrier layers improves their structural properties, reduces the number of band-tail states, and thus yields smoother quantum well interfaces.⁶ In this previously reported work, the maximum Al-mole fraction had to be kept below 12% (a cutoff wavelength=332 nm) because a growth temperature of 950 °C was required to obtain good room-temperature photoluminescence (PL) from the quaternary AlInGaN layers. This made In incorporation and hence the deposition of quaternary AlInGaN material with a high indium/aluminum composition ratio very difficult.

In the past, we have also reported on the use of pulsed atomic layer epitaxy (PALE) to deposit high quality GaN,⁷

AlN,⁸ and AlGaN and GaN–AlN short period superlattices⁹ at temperatures 200–300 °C below those required for conventional low-pressure MOCVD. Nakamura *et al.* have also used this GaN–AlN short period superlattice approach for $\text{Al}_x\text{Ga}_{1-x}\text{N}$ barrier regions in their purple GaN–InGaN MQW lasers.¹⁰ We now report on using this PALE approach to deposit GaN/AlN/InN short period superlattices for high-optical/electrical quality quaternary $\text{Al}_x\text{In}_y\text{Ga}_{1-x-y}\text{N}$ layers on sapphire substrates. The use of PALE allowed us to reduce the growth temperatures well below 800 °C thereby significantly increasing the In incorporation and growing high Al fraction quaternary AlInGaN layers with transmission cut-offs in the 250–350 nm range. These layers exhibited strong room-temperature photoluminescence, thus establishing their suitability for use in UV LEDs for solid-state white lighting and solar-blind ultraviolet detectors.

For all the $\text{Al}_x\text{In}_y\text{Ga}_{1-x-y}\text{N}$ depositions, we used trimethyl aluminum (TMA), trimethylgallium (TMG), trimethyl indium (TMI), and NH_3 as the precursors and basal plane sapphire substrates. Prior to the quaternary layer, a 250 Å thick AlN buffer layer and a 1.5 μm thick intrinsic *i*-GaN layer were grown using conventional low-pressure MOCVD. The growth pressures for the two layers were kept at 40 Torr whereas their growth temperatures were 450 and 1000 °C, respectively. The quaternary $\text{Al}_x\text{In}_y\text{Ga}_{1-x-y}\text{N}$ layers were then grown at 760 °C by 150 repeats of a unit cell. The unit cell had the growth sequence outlined in Fig. 1. As can be seen, 6 s long pulses of the TMA, TMI, TMG, and NH_3 precursors were introduced alternately into the low pressure MOCVD reactor. An ammonia pulse always followed the metalorganic pulses. As an example, the unit cell in Fig. 1 consisted of three repeats of Al and N pulses, followed by one In and N and one Ga and N pulse. We refer to the resulting $\text{Al}_x\text{In}_y\text{Ga}_{1-x-y}\text{N}$ layer as the $(3\text{Al}, 1\text{In}, 1\text{Ga})_{150}$ or, simply, as a $(3,1,1)_{150}$ layer. The subscript 150 denotes the number of unit cell repeats. The precursor fluxes in individual pulses were adjusted in such a way that the deposited thickness in each unit cell, determined from the total layer thickness divided by the number of unit cell repeats, was around 6 Å.

In order to ascertain that the PALE approach yields high

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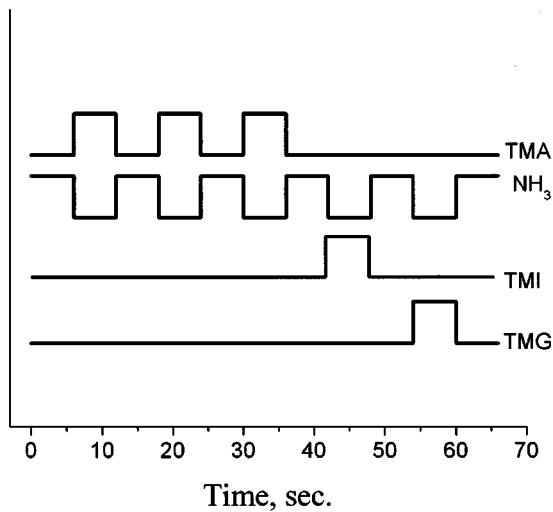


FIG. 1. Representative growth sequence of a unit cell using PALE. We denote this cell as $(3_{\text{Al}}, 1_{\text{In}}, 1_{\text{Ga}})$ or, simply, $(3,1,1)$.

quality quaternary AlInGaN layers and also allows simple composition control, a $(3,3,1)_{150}$ layer was grown first. We then used Rutherford backscattering spectroscopy (RBS) spectra to determine the composition of this $(3,3,1)_{150}$ layer to be about 2% In, 36% Al, and 62% Ga. Using atomic force microscopy (AFM) we measured the root mean square (rms) roughness for this layer to be only 9 Å. This compares favorably with the rms roughness value for a typical MOCVD grown AlInGaN layer with similar Al-mole fraction. Quaternary layers with unit cell configurations of $(3,0,1)$, $(3,1,1)$, $(3,3,1)$, and $(3,3^+,1)$, $(3,6,1)$ were then deposited. (Here the superscript “+” denotes pulses with the In-metalorganic flux doubled). In Fig. 2, we plot the relative indium composition in these layers as determined from energy dispersion x-ray (EDAX) analysis measurements. As is seen, the indium signal for the quaternary layers scales linearly with the number of In pulses in the unit cell and the indium flux (within the pulse). The data included in Fig. 2 clearly establish the viability of the PALE process in accurately controlling the quaternary AlInGaN material composition over a wide range. Using the RBS profiles for the $(3,3,1)_{150}$ sample, we also confirmed the thickness of this quaternary layer to be approximately 1000 Å. The RBS data clearly showed the si-

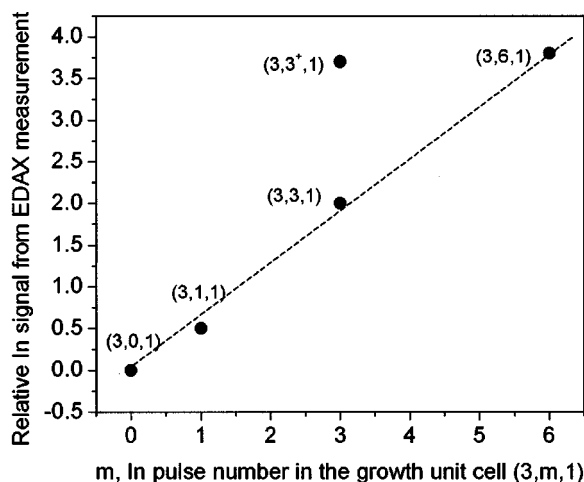


FIG. 2. Relative EDAX signals vs the number of In pulses in the unit cell.

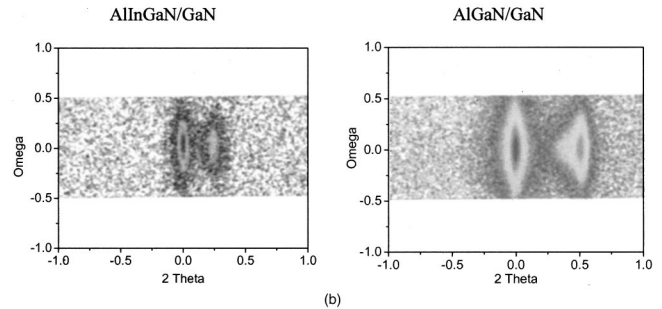
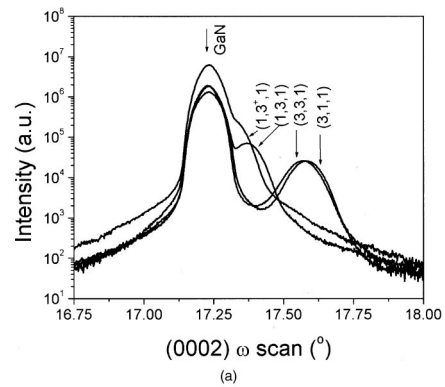


FIG. 3. (a) (0002) XRD ω - 2θ rocking curves of a set of quaternary samples. (b) (0004) diffraction reciprocal lattice map for two samples with $(1,3,1)$ and $(1,0,1)$ unit cells.

multaneous presence of Al, In, and Ga in the quaternary layer. Similar observations were also made from secondary ion mass spectroscopy profiling data.

In Fig. 3(a), we include ω - 2θ x-ray diffraction (XRD) rocking curves for a set of four quaternary $\text{Al}_x\text{In}_y\text{Ga}_{1-x-y}\text{N}$ samples grown under identical PALE conditions but with different unit cells: $(3_{\text{Al}}, 1_{\text{In}}, 1_{\text{Ga}})$, $(3_{\text{Al}}, 3_{\text{In}}, 1_{\text{Ga}})$, $(1_{\text{Al}}, 3_{\text{In}}, 1_{\text{Ga}})$, and $(1_{\text{Al}}, 3_{\text{In}}^+, 1_{\text{Ga}})$. As seen from the x-ray diffraction spectra, when Al repeats in the unit cell are kept constant, the lattice mismatch between the quaternary $\text{Al}_x\text{In}_y\text{Ga}_{1-x-y}\text{N}$ and the underlying GaN layers can be decreased by either increasing the number of In pulses in the unit cell or by increasing the precursor flux in the In pulse. The data thus clearly show the PALE approach to be capable of depositing lattice matched quaternary $\text{Al}_x\text{In}_y\text{Ga}_{1-x-y}\text{N}$ -GaN heterojunctions. Our data on diffraction reciprocal lattice mapping for two samples with a unit cell composition of $(1_{\text{Al}}, 3_{\text{In}}, 1_{\text{Ga}})$ and $(1_{\text{Al}}, 0_{\text{In}}, 1_{\text{Ga}})$ grown under identical deposition conditions show that the structural quality for the quaternary AlInGaN samples is significantly better than that of the ternary AlGaIn. Moreover, the mapping data show that a part of the AlGaIn layer (most probably that at the AlGaIn/GaN interface) is already partially relaxed due to the considerable lattice mismatch.

Figure 4 shows the room-temperature PL spectra for the quaternary $\text{Al}_x\text{In}_y\text{Ga}_{1-x-y}\text{N}$ films with unit cell configurations of $(3,0,1)$, $(3,1,1)$, $(3,3,1)$, $(3,3^+,1)$, $(2,2,1)$, $(1,3,1)$, and $(1,3^+,1)$. The growth temperature for all samples was 760 °C. We have also included the data for a 1000 Å thick quaternary layer deposited at the same temperature (760 °C) but using a conventional low pressure MOCVD process and precursor flows similar to that of the $(1,3,1)$ sample of Fig. 4. In addition, the data show the PL signal for PALE quaternary layers to be much stronger than that of the conventional MOCVD grown sample. Also, the PALE samples have no

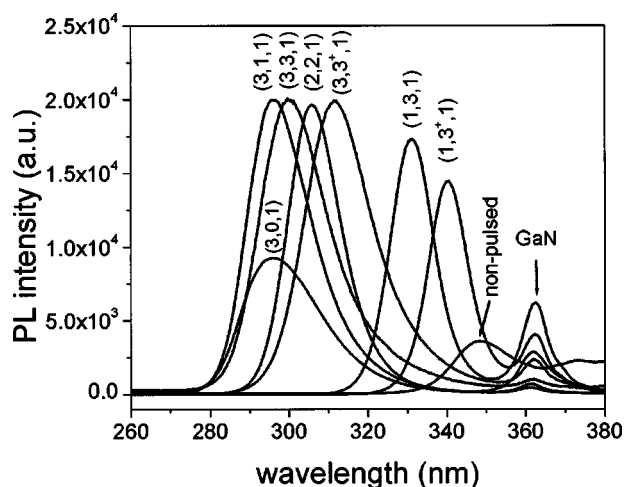


FIG. 4. Room-temperature excimer laser excited PL spectra of the quaternary samples.

deep level emissions. Figure 4 data further establish that addition of Al in the unit cell blueshifts the PL peak positions. The addition of In, on the contrary, results in a redshift. This is to be expected from the known band gaps of AlN, InN, and GaN. In comparing the PL signal strength from the (3,0,1) and the (3,1,1) samples we conclude that the addition of indium also improves the optical quality of the films. This observation confirms the results reported earlier for quaternary AlInGaIn layers grown by a conventional MOCVD process.⁶ Furthermore, in contrast to conventional $\text{Al}_x\text{Ga}_{1-x}\text{N}$ films, the PL peak intensity for the quaternary layers does not degrade with increasing the Al-mole fraction. This is true even for the material with a cutoff wavelength of 280 nm.

The PL peak positions for the PALE samples also coincide with the measured band-edge cutoff wavelengths. This confirms that the PL of the PALE deposited quaternary AlInGaIn layers at 760 °C is from band-to-band emissions. In contrast, Aumer *et al.* could only get AlInGaIn band-edge emission from conventional MOCVD grown AlInGaIn films when the growth temperature was greater than 850 °C.¹¹ Therefore, the data in Fig. 4 clearly establish the superiority of PALE to the conventional MOCVD process for producing high optical quality AlInGaIn films.

Using standard van der Pauw geometry, we measured the carrier concentration and mobility for the quaternary $\text{Al}_x\text{In}_y\text{Ga}_{1-x-y}\text{N}$ layers with the PL spectra shown in Fig. 4. The films were all *n* type and the room-temperature carrier concentration ranged from 1×10^{18} to $3 \times 10^{18} \text{ cm}^{-3}$. The room-temperature electron mobility ranged between 200 and 350 $\text{cm}^2/\text{V s}$. These values are very high given the high Al-mole fraction of the measured samples. In addition to better material quality, these measured values of conductivity and mobility may also be affected by contributions from the two-dimensional (2D) electron gas at the *i*-GaIn- $\text{Al}_x\text{In}_y\text{Ga}_{1-x-y}\text{N}$ interface. More experiments are currently underway to isolate these contributions and to accurately determine the bulk carrier density and mobility values of PALE deposited quaternary AlInGaIn material.

The structural, optical, and electrical characterization data presented here clearly show improved material quality

as a result of the PALE process. We believe there are several reasons for these improvements. First, in PALE material deposition the group III (Al, Ga, In) atoms and the group V species (NH_3) are supplied to the growth chamber at different times. This increases the surface mobility of the adatoms and enables them to find energetically favorable sites. Enhanced migration also allows the lowering of single crystal epitaxy temperatures, and increases indium incorporation into the quaternary films. This in turn reduces the band-tail states and the disorder in the AlGaIn films.⁶ Lowering of growth temperatures also helps in avoiding In segregation, which improves the material composition uniformity.

The introduction of material organics in separate pulses also helps in avoiding adduct formation. It is well known that Al/In/Ga adducts can be incorporated into the films and degrade their structural and optical properties. In addition, for PALE the growth thickness per pulse is less sensitive to variations in the metalorganic fluxes. Thus the thickness and composition control are only related to the number of adatom pulses in the unit cell and the total number of unit cell repeats. This therefore makes the uniformity of the PALE deposited layers superior to those from conventional MOCVD.

In summary, we have reported the use of a unique short period superlattice approach to grow quaternary $\text{Al}_x\text{In}_y\text{Ga}_{1-x-y}\text{N}$ layers. A pulsed atomic layer epitaxy procedure was developed to deposit $\text{Al}_x\text{In}_y\text{Ga}_{1-x-y}\text{N}$ films of high structural, electrical, and optical quality even at growth temperatures well below 800 °C. These low-temperature deposited $\text{Al}_x\text{In}_y\text{Ga}_{1-x-y}\text{N}$ layers with high aluminum content and indium/aluminum composition ratio are ideally suited for the active region of UV LEDs for solid-state white lighting and solar-blind ultraviolet photodetectors.

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