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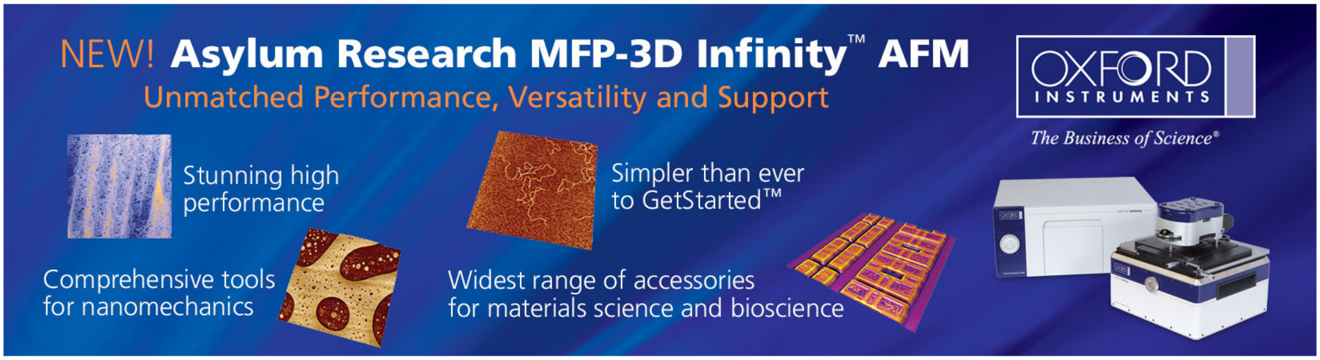

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Optimizing growth conditions for GaN/Al$_x$Ga$_{1-x}$N multiple quantum well structures

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We have investigated the optimal growth conditions for GaN/Al$_x$Ga$_{1-x}$N multiple quantum well (MQW) structures by metal organic chemical vapor deposition. Optical properties of a set of GaN/Al$_x$Ga$_{1-x}$N MQW samples grown under systematically varied growth conditions have been studied by employing picosecond time-resolved photoluminescence (PL) spectroscopy. The PL emission efficiency, the linewidth of the PL emission spectra, the ratio of the barrier emission intensity to the well emission intensity, and the temperature dependence of the PL decay lifetime of these GaN/Al$_x$Ga$_{1-x}$N MQW structures have been measured and compared with each other carefully. Based on our studies, we concluded that the optimal growth conditions for GaN/Al$_x$Ga$_{1-x}$N MQW structures are GaN-like rather than Al$_x$Ga$_{1-x}$N-like or other conditions. The GaN/Al$_x$Ga$_{1-x}$N MQW structures grown under the GaN-like growth conditions exhibited higher quantum efficiencies and narrower PL emission linewidths than those grown under other conditions. PL emission from barrier regions was not observed in the MQW structures grown under the GaN-like growth conditions, which is highly preferred for ultraviolet light emitter applications. © *2000 American Institute of Physics.* [S0003-6951(00)04107-3]

Group III–V nitride semiconductors have been recognized as important materials for many novel optoelectronic devices, such as blue ultraviolet (UV) light emitting diodes (LEDs), laser diodes (LDs), and high-temperature/high-power electronic devices.[1] As demonstrated by LDs, LEDs, and electronic devices, many III–V nitride based devices take advantage of multiple quantum well (MQW) structures such as GaN/Al$_x$Ga$_{1-x}$N and In$_x$Ga$_{1-x}$N/GaN MQWs for optimized performance. Recently, many efforts have been devoted towards the understanding and design of GaN/Al$_x$Ga$_{1-x}$N MQW structures for UV light emitter applications.[2–8] For the design and fabrication of these MQW structures, one important issue is to maximize the quantum efficiency, i.e., to maximize the optical emission from the confined states in the well regions and to minimize the optical losses in the barrier regions.[8] It is well known that structural parameters of MQWs, including both barrier and well thicknesses, have strong effects on the quantum efficiency of the MQWs. GaN/Al$_x$Ga$_{1-x}$N MQW structures with high optical qualities can be achieved when the well thickness is in between 25 to 40 Å[3,9,10] and the barrier thickness is around 80 Å.[11] However, effects of growth conditions on the quantum efficiency of these MQW structures have not been studied systematically. A detailed study on the growth conditions for obtaining GaN/Al$_x$Ga$_{1-x}$N MQWs with high quantum efficiency is needed in order to achieve high performance UV light emitters based on GaN/Al$_x$Ga$_{1-x}$N MQW structures.

The growth conditions for GaN and Al$_x$Ga$_{1-x}$N epilayers are quite different and have been well studied. For example, the growth temperature (pressure) for Al$_x$Ga$_{1-x}$N epilayers is higher (lower) than that for GaN epilayers. However, it is very difficult to change the growth conditions between the GaN well and the Al$_x$Ga$_{1-x}$N barrier layers during the metal organic chemical vapor deposition (MOCVD) growth of GaN/Al$_x$Ga$_{1-x}$N MQWs because the growth time for each well or barrier layer (around 10 s) is much shorter than the time needed for all the growth parameters to become stable (more than 60 s). One can thus choose the growth conditions for GaN/Al$_x$Ga$_{1-x}$N MQWs to be the optimal growth conditions of either GaN epilayers (GaN-like) or Al$_x$Ga$_{1-x}$N epilayers (Al$_x$Ga$_{1-x}$N-like), or choose conditions in between the optimal GaN- and Al$_x$Ga$_{1-x}$N-like. Al$_x$Ga$_{1-x}$N epilayers are more difficult to grow and are also more sensitive to the growth parameters than GaN epilayers. Thus one might expect that GaN/Al$_x$Ga$_{1-x}$N MQWs grown under Al$_x$Ga$_{1-x}$N-like conditions would have a better overall quality than those grown under GaN-like conditions. However, the argument which favors the GaN-like growth conditions for GaN/Al$_x$Ga$_{1-x}$N MQW structures is that the qualities of the GaN wells are more important for the well transitions and should be optimized.

In this work, the growth conditions for GaN/Al$_x$Ga$_{1-x}$N MQW structures have been studied by systematically varying the growth parameters between the GaN- and Al$_x$Ga$_{1-x}$N-like conditions. Picosecond time-resolved photoluminescence (PL) spectroscopy[12] has been employed to study the optical properties of these MQWs. We found that the optimal growth conditions for GaN/Al$_x$Ga$_{1-x}$N MQW structures are GaN-like rather than Al$_x$Ga$_{1-x}$N-like or other conditions.

A set of GaN/Al$_x$Ga$_{1-x}$N MQW samples has been grown by our MOCVD system with varying growth parameters. Before the growth of the MQWs, a 300 Å GaN nucleation layer and a 1 μm undoped GaN buffer layer were grown on the sapphire substrate. The MQW structures are composed of thirty periods of alternating GaN-well and Al$_x$Ga$_{1-x}$N-barrier ($x \sim 0.2$) layers. Detailed optical studies on four GaN/Al$_x$Ga$_{1-x}$N MQWs samples denoted as

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TABLE I. Summary of growth conditions and PL emission properties of samples A, B, C, and D.

Samples		A	B	C	D
Growth conditions		GaN-like	In between GaN- and AlGaN-like	In between GaN- and AlGaN-like	AlGaN-like
Growth parameters	Pressure (Torr)	300	150	300	150
	Temperature (°C)	1050	1050	1060	1060
	NH_3 (SL/min)	4.0	4.0	4.0	2.4
Well peak position (eV) @10 K		3.550	3.645	3.643	3.678
FWHM (meV) @10 K		32	59	58	65
Integrated well intensity @10 K		25	8.5	8	6
I_{emi}(barrier)/I_{emi}(well)($\times 10^{-4}$) @10 K		0	5	2.5	1.3

samples A, B, C, and D, representing four types of growth conditions have been carried out. Sample A represents those grown under the optimal GaN growth conditions (or GaN-like) with growth temperature and pressure being 1050 °C and 300 Torr, respectively. The well width of sample A is 30 Å based on the growth rate of GaN epilayers under the same growth condition. Sample D represents those grown under the optimal $Al_xGa_{1-x}N$ growth conditions (or $Al_xGa_{1-x}N$-like) with growth temperature and pressure being 1060 °C and 150 Torr, respectively. The barrier width is targeted at 50 Å for sample D based on the $Al_xGa_{1-x}N$ growth rate. Samples B and C represent those grown under growth conditions in between the optimal GaN and $Al_xGa_{1-x}N$ growth conditions with either the growth pressure or temperature being varied systematically. The detailed growth parameters for these MQWs are summarized in Table I.

The continuous-wave (cw) PL spectra measured at 10 K for samples A, B, C, and D are shown in Figs. 1(a), 1(b), 1(c), and 1(d), respectively. The main emission peaks in Fig. 1 are due to the excitonic recombinations in the GaN-well regions, which are all blue shifted with respect to an emission line at about 3.49 eV from the underneath GaN epilayer. This blue shift is due to the quantum confinement effects in the MQWs.[3,10] The position of the main emission peak of sample A grown under the GaN-like growth conditions is at 3.550 eV, which is consistent with the expected value from $GaN/Al_xGa_{1-x}N$ MQWs with a well thickness of 30 Å and Al content of 20% in the barrier regions. However the main emission peaks of samples B, C, and D grown under conditions other than the optimal GaN-like growth conditions are much higher than the expected value from $GaN/Al_xGa_{1-x}N$ MQWs with a well thickness of 30 Å. Time-resolved PL spectra revealed no obvious piezoelectric effects in these MQW samples (not shown), which is probably due to small well widths. Thus the most likely explanation for the much higher emission energy positions in samples B, C, and D is that the well thicknesses of these MQWs are less than 30 Å, due to a lower growth rate for the GaN-well layers under growth conditions other than the GaN-like growth conditions. In order to clarify this, we have also grown a MQW sample E with a well width of 18 Å under the optimal GaN growth conditions (all other structural parameters of sample E are the same as sample A). The PL spectrum of sample E is shown in Fig. 1(e). We see that the spectral peak positions of samples B, C, D nearly coincide with that of sample E. Thus the actual well thickness of sample B, C, and D is estimated to be around 18 Å. The most important feature shown in Fig. 1 is that PL efficiencies of samples A and E (grown under the GaN-like conditions) are about 2 to 4 times higher than those of other samples. The linewidths of the well transitions of samples A and E are about 32 and 28 meV, respectively. However, the linewidths of samples B, C, and D are between 54 and 65 meV. The broadening of the linewidth and the lower quantum efficiency are attributed to the relatively poor qualities of samples B, C, and D (grown under other than the GaN-like conditions). Thus, $GaN/Al_xGa_{1-x}N$ MQWs grown under the GaN-like conditions have the highest quantum efficiency and narrowest linewidth, i.e., the highest optical quality.

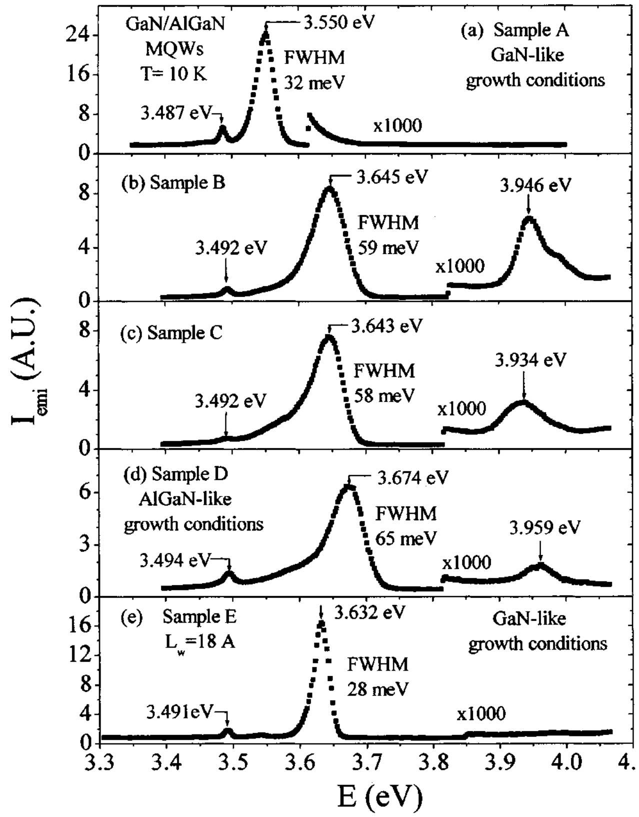

FIG. 1. Low-temperature (10 K) cw PL spectra of $GaN/Al_xGa_{1-x}N$ MQWs (a) sample A grown under the optimal GaN growth conditions (GaN-like) with well thickness $L_w = 30$ Å, (b) and (c) samples B and C grown under conditions in between the optimal GaN and $Al_xGa_{1-x}N$ growth conditions, (d) sample D grown under the optimal $Al_xGa_{1-x}N$ growth conditions ($Al_xGa_{1-x}N$-like) with barrier thickness of 50 Å, and (e) sample E grown under the GaN-like growth conditions with well thickness $L_w = 18$ Å. The growth time for the well and barrier layers are fixed for samples A, B, C, and D. Note that samples A and E exhibit the highest PL intensities and narrowest linewidths from the well regions as well as no PL emission from the barrier regions.

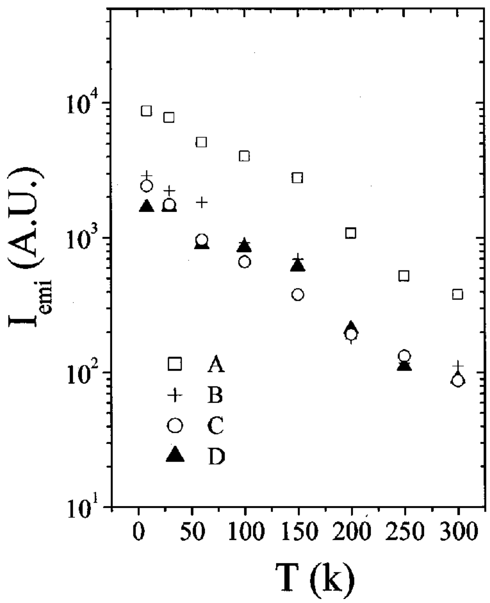

FIG. 2. Temperature dependence of the integrated PL emission intensity for samples A, B, C, and D.

For the design of MQW laser structures, an important issue is to minimize the optical transitions from the barrier regions, since emissions from the barrier regions represent losses in quantum efficiency or optical gain.[8] For samples A and E grown under the GaN-like growth conditions, there is hardly any observable emission intensities from the barrier regions. Samples B, C, and D, grown under other conditions, exhibit clear barrier emission peaks around 3.95 eV. The observed peak positions of the barrier transitions are consistent with the targeted Al concentration (about 20%). Our results show that for GaN/$Al_xGa_{1-x}N$ MQW structures carrier leakage from well to barrier regions can be minimized by employing the GaN-like growth conditions. Figure 2 shows the temperature dependence of the PL emission intensity of the four MQW samples A, B, C, and D. Sample A exhibits the highest emission efficiency at all temperatures.

Time-resolved PL measurements also provide additional information regarding the quality and quantum efficiency of these MQWs. Figure 3 shows the temperature dependence of the PL decay lifetime, τ, measured at the peak positions of the well transitions in these MQWs. For sample A grown under the GaN-like growth conditions [Fig. 3(a)], the PL decay lifetime increases with temperature up to 70 K. Such a linear increase of τ with T is a well-known characteristic of the exciton radiative recombinations in MQWs[13] and is observable only in samples of high optical qualities. As illustrated in Figs. 3(b)–3(d), a linear increase of τ with T is absent in samples grown under conditions other than the GaN-like conditions. Our time-resolved PL results suggest that the radiative recombination is dominant at low temperatures probably only in MQW samples grown under the GaN-like conditions. Thus time-resolved PL data further corroborate that GaN/$Al_xGa_{1-x}N$ MQWs grown under the GaN-like growth conditions have the highest optical qualities as well as the highest quantum efficiencies.

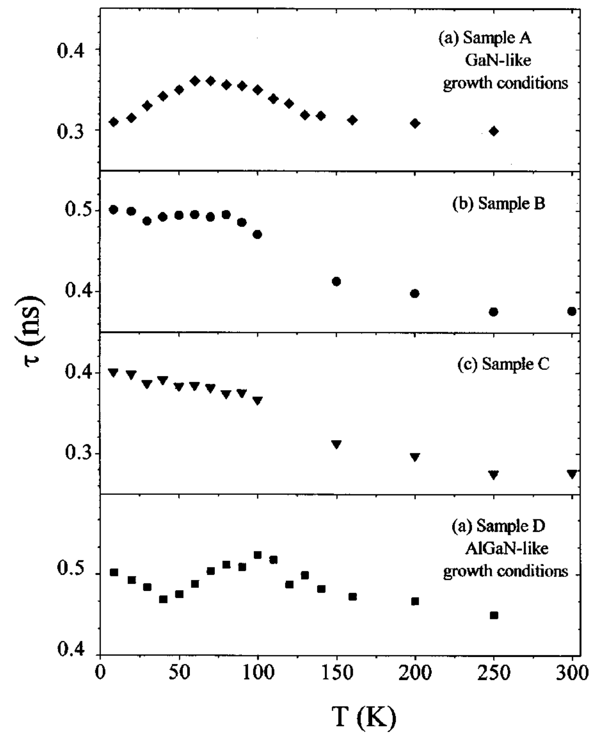

FIG. 3. Temperature dependence of the well transition PL decay lifetime, τ for samples A, B, C, and D. A linear increase of τ with temperature was observed for sample A up to 70 K (a).

In summary, GaN/$Al_xGa_{1-x}N$ MQW structures have been grown by MOCVD under systematically varied growth conditions. Picosecond time-resolved PL spectroscopy has been employed to monitor the optical properties of these structures. The optimal growth conditions for GaN/$Al_xGa_{1-x}N$ MQW structures are found to be GaN-like rather than $Al_xGa_{1-x}N$-like or conditions in between the two. GaN/$Al_xGa_{1-x}N$ MQW structures produced under the GaN-like conditions exhibited higher quantum efficiencies, narrower PL emission linewidths, a linear dependence of the PL decay lifetime on temperatures up to 70 K, and no observable barrier emissions, all of which are highly preferred for UV light emitter applications.

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