Optical Transitions and Dynamic Processes in III-Nitride Epilayers and Multiple Quantum Wells

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Abstract. Fundamental optical transitions in GaN and InGaN epilayers, InGaN/GaN and GaN/AlGaN multiple quantum wells (MQWs) grown both by metal-organic chemical vapor deposition and reactive molecular beam epitaxy have been studied by picosecond time-resolved photoluminescence spectroscopy. The exciton binding energies and radiative recombination lifetimes of the free excitons and bound excitons have been obtained. Effects of well thickness on the optical properties of $In_xGa_{1-x}N$/GaN and GaN/$Al_xGa_{1-x}N$ MQWs have also been studied.

1. Introduction

GaN based devices offer great potential for applications such as UV-blue lasers, solar-blind UV detectors, and high-power electronics. Researchers in this field have made extremely rapid progress toward materials growth as well as device fabrication.[1] The commercial availability of super-bright blue light emitting diodes (LED) and the demonstration of the room temperature blue lasers based on the GaN system are clear indicative of the great potential of this material system.[2,3] Recently, there has been much work concerning the fundamental optical transitions in GaN.[4-7] It is expected that all optoelectronic devices based on GaN will take advantages of multiple quantum well (MQW) structures of GaN/AlGaN and InGaN/GaN. Thus a better understanding of the fundamental optical transitions in nitride epilayers and MQWs is needed.

2. Experimental

Samples used in this work include GaN and $In_xGa_{1-x}N$ epilayers and $In_xGa_{1-x}N$/GaN and GaN/$Al_xGa_{1-x}N$ MQWs grown both by metal-organic chemical vapor depositions (MOCVD) and by reactive molecular beam epitaxy (MBE). Their structures and parameters are summarized in Table I. All samples studied were of wurtzite structure grown on sapphire (Al_2O_3) substrates with AlN buffer layers.

Low temperature time-resolved PL spectra were measured by using a picosecond laser spectroscopy system with an average output power of about 20 mW, a tunable photon energy up to 4.5 eV, and a spectral resolution of about 0.2 meV.[4,5] A micro-channel-plate photomultiplier tube (MCP-PMT) together with a single photon counting system was used to collect time-resolved PL data and the overall time resolution of the detection system was about 20 ps.

3. Results and Discussions

3.1 GaN epilayers. Figure 1 shows four low-temperature (10 K) photoluminescence (PL) spectra obtained for samples A, B, C, (MOCVD) and sample G (MBE). For high purity epilayer grown by MOCVD (sample A), two emission lines with energy peak positions at about 3.485 eV and 3.491 eV are identified as due to the recombination of the ground state of free A and B excitons [A(n=1) and B(n=1)].[8] The narrow linewidths of these two emission bands are clear indication of its high crystalline quality of sample A. Our results show that the energy separation between the A and B valance bands is about 6 meV with the assumption that A and B exciton binding energies are comparable.[5] An emission line due to the recombination of the first excited state of the A-exciton A(n=2), which is about 14.3 meV above the A(n=1) emission line, is also observable in sample A at T>40 K. For an as-grown sample with a higher

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Growth Method	Sample	Materials	x	Structure	Type	Carrier Concentration (cm^{-3})
MOCVD	A	GaN		Epilayer	n	5×10^{16}
	B	GaN			n	2.4×10^{17}
	C	GaN		Epilayer	p	1×10^{17}
	D	$In_xGa_{1-x}N$	0.12	Epilayer	n	
	E	25 A $In_xGa_{1-x}N$/GaN	0.15	MQW		
	F	90 A $In_xGa_{1-x}N$/GaN	0.15	MQW		
MBE	G	GaN		Epilayer	Insulating	
	H	25 A GaN/$Al_xGa_{1-x}N$	0.07	MQW		
	I	50 A GaN/$Al_xGa_{1-x}N$	0.07	MQW		

Table I. Structures and parameters of GaN, InGaN epilayers, InGaN/GaN and GaN/AlGaN MQW samples used in this work. Well thicknesses of MQWs are also indicated.

native donor concentration (sample B), the dominant emission line occurs at 3.476 eV and is due to the recombination of the excitons bound to neutral donors, called I_2.[9] The shoulder at about 3.484 eV in sample B is due to the free A exciton (n=1) recombination. Thus the binding energy of the neutral-donor-bound exciton is about 8-9 meV. For Mg doped p-type epilayer (sample C), the dominant emission line at 3.459 eV is due to the recombination of neutral-acceptor-bound excitons, called I_1.[10] A value of about 26 meV is thus obtained for the binding energy of the neutral-acceptor-bound exciton in GaN. For a MBE grown high quality and purity epilayer (sample G), three emission lines at 3.483 eV, 3.489 eV, and 3.498 eV are observable, which correspond to the transitions of the ground state of A- and B-excitons and the first excited state of the A-exciton (n=2), respectively.[4] The energy difference between the first and the ground states of the excitons thus gives the binding energy of the A exciton, E_b=(4/3)x(3.498-3.483) eV = 20 (meV). The energy separation between the A and B valence bands obtained from sample G (6 meV) is consistent with that obtained from sample A. The slight energy difference in the peak positions of the A(n=1) and B(n=1) emission bands for sample A and G (3.485 eV vs. 3.483 eV and 3.489 eV vs. 3.491 eV) may be due to a slight difference in strain in these two different samples.

3.2 InGaN epilayers. Figure 2(a) shows a PL emission spectrum for a MOCVD grown InGaN epilayer (sample D) measured at 10 K. By comparing with the PL spectra of GaN epilayers (Fig. 1(a) and Fig. 1(d)), the emission peak position is clearly shifted toward lower energy due to In incorporation. Another effect is that the emission linewidth of the InGaN epilayer is more than one order of magnitude larger than those of GaN epilayers due to alloy disorder. The emission peak position in sample D is at about 3.193 eV with a full linewidth at half maximum (FWHM) of about 55 meV, which is due to the recombination of localized excitons. In an alloy, the exciton localization is caused by energy fluctuations in the band edge induced by alloy disorder and the linewidth is correlated with the exciton localization energy.

3.3 InGaN/GaN MQWs . The MQW sample with well thickness of 25 Å (sample E shown in Fig. 2(b)) emits an exciton line at 3.211 eV. The emission linewidth seen in MQWs is broader than that in the InGaN epilayer. Quantum confinement is also evident by comparing the PL spectrum of MQWs (sample E) shown in Fig. 2 (b) with that of InGaN epilayer (sample D) shown in Fig. 2(a). For MQWs with large well thicknesses, e.g. L_W= 90 Å (sample F shown in Fig. 2(c)), a dominant emission at 2.963 eV is observed. From the fact that the energy position of this emission line is below the exciton emission line in InGaN epilayers, we attribute it to an impurity related transition. Since the growth conditions for 25 and 90 Å MQWs are identical, our results indicate that the 90 Å well thickness is above the critical thickness of the InGaN/GaN MQW system. Above the critical thickness, strain is relieved by the creation of large density of misfit dislocations, which leads to the dominance of the impurity transition.

3.4. GaN/AlGaN MQWs. As shown in Fig. 3, in GaN/$Al_xGa_{1-x}N$ MQWs, the dominant emission lines always result from the well region at low temperatures. Comparing the PL spectra shown in Fig. 3(a) and

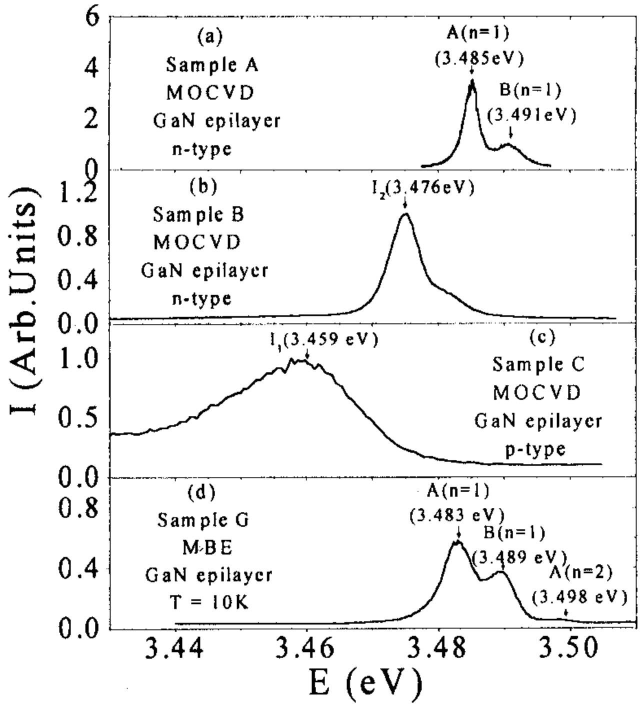

Fig.1. PL emission spectra of GaN epilayers measured at T=10 K.

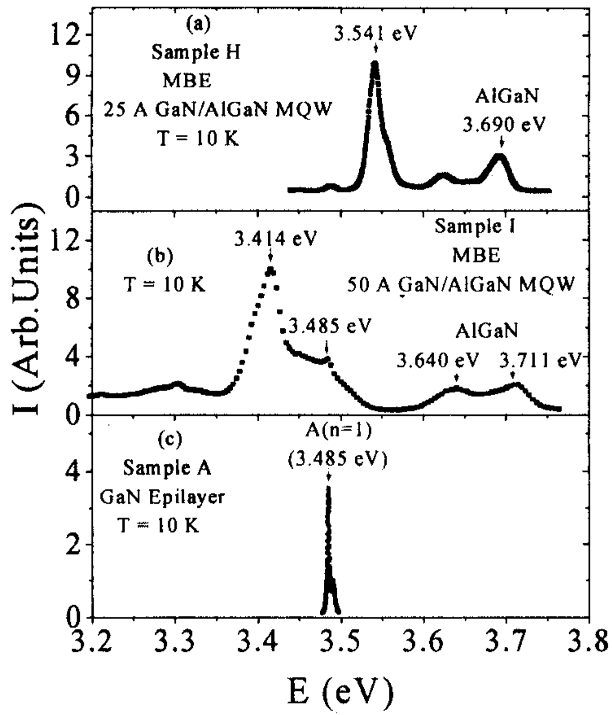

Fig.3. PL emission spectra of $GaN/Al_xGa_{1-x}N$ MQWs (a) and (b), and GaN epilayer (c) measured at T = 10 K.

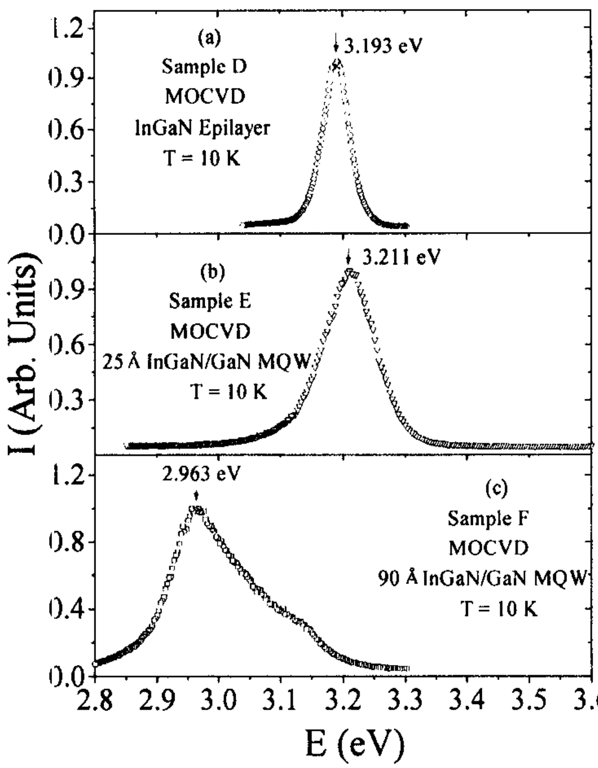

Fig. 2. PL emission spectra of $In_xGa_{1-x}N/GaN$ MQWs (b) and (c), and $In_xGa_{1-x}N$ epilayer (a) measured at T=10 K.

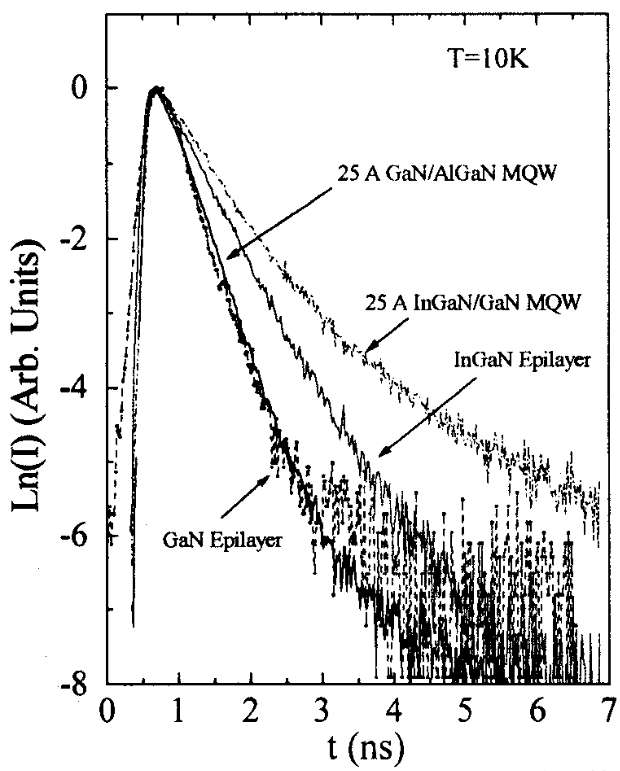

Fig. 4. PL temporal responses measured at the spectral peak positions for MBE and MOCVD samples: GaN and $In_xGa_{1-x}N$ epilayers, $GaN/Al_xGa_{1-x}N$ and $In_xGa_{1-x}N/GaN$ MQWs.

3(c), the exciton emission line resulting from the 25 Å MQWs (sample H) is blue shifted by 56 meV with respect to the GaN epilayer (sample A). Comparing the PL spectra shown in Fig 3(b) and 3(c), the dominant emission line in the 50 Å MQWs is 71 meV below the exciton emission line (sample I). Our results for GaN/AlGaN MQWs can be summarized as follows: (i) the optical transitions in MQWs with narrow well widths (L_W < 40 Å) are blue shifted with respect to the GaN epilayer, however, no such blue shift was evident for the MQW samples with well thickness greater than 40 Å and (ii) the band-to-impurity transitions are the dominant emission lines in MQWs of large well thicknesses (L_W > 40 Å).

3.5. Recombination lifetimes. Recombination lifetimes of various emission lines in GaN and InGaN epilayers and InGaN/GaN and GaN/AlGaN MQW samples used here have been measured. Fig. 4 shows PL temporal responses of exciton emission lines in GaN and $In_xGa_{1-x}N$ epilayers, $GaN/Al_xGa_{1-x}N$ and $In_xGa_{1-x}N$ MQWs measured at their respective spectral peak positions. In general, the exciton lifetimes in MQWs are enhanced compared with those in epilayers. The typical exciton lifetime is around 0.3 ns in GaN epilayers and 0.6 ns in InGaN epilayers. Moreover, the exciton recombination lifetime in GaN/AlGaN MQWs of narrow well thickness (< 40 Å) increases linearly from about 0.3 ns to about 0.45 ns as temperature increases from 10 to 60 K, which is a hallmark of radiative recombination in MQWs.[11]

4. Summary

Our results have revealed that the optical transitions at low temperatures are dominated by I_2 (I_1) in n-type (p-type) epilayers and free exciton transitions in high quality and purity GaN epilayers. The binding energies of free and bound excitons have been determined. The dominant optical transitions in $In_xGa_{1-x}N$/GaN and $GaN/Al_xGa_{1-x}N$ MQWs are localized excitons at low temperatures due to alloy disorder and/or well width fluctuation. Quantum confinement effect has been observed for MQWs of narrow well widths. Exciton lifetimes in different materials and structures have been measured and compared.

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