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Effects of well thickness and Si doping on the optical properties of GaN/AlGa_N multiple quantum wells

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Effects of well thickness and Si doping on the optical properties of GaN/AlGa_N (MQWs) have been investigated by picosecond time-resolved photoluminescence (PL) measurements. Our results have yielded that (i) the optical transitions in nominally undoped MQWs with narrow well thicknesses ($L_w < 40 \text{ \AA}$) were blue shifted with respect to the GaN epilayer due to quantum confinement, however, no such blue shift was evident for the MQWs with well thicknesses larger than 40 \AA , (ii) the band-to-impurity transitions were the dominant emission lines in nominally undoped MQWs of large well thicknesses ($L_w > 40 \text{ \AA}$) at low temperatures, and (iii) Si doping improved significantly the crystalline quality of MQWs of large well thicknesses ($L_w > 40 \text{ \AA}$). The implications of these results on the device applications based on III-nitride MQWs have been discussed. © 1997 American Institute of Physics. [S0003-6951(97)00836-X]

Group III nitride wide band-gap semiconductors have been recognized as technologically very important materials.^{1,2} They have recently attracted considerable interest due to their applications for optical devices which are active in the blue and ultraviolet (UV) wavelength regions and electronic devices capable of operation at high power levels, high temperatures, and harsh environments. As demonstrated by GaN laser diodes, light emitting diodes (LED), and GaN based electronic devices, all GaN based devices must take advantage of quantum well (QW) structures such as GaN/AlGa_N and InGa_N/GaN. In order to optimize the device design, it is necessary to study and understand the physical properties of nitride QWs as well as the QW structural effects on the device performance. In this letter, effects of well thickness and Si doping on the optical properties of GaN/AlGa_N MQWs have been investigated by picosecond time-resolved PL measurements. Our results have revealed that MQWs with high optical qualities can be achieved when the well thickness is below the critical thickness value. When the well layer thicknesses exceed the critical thickness value, large densities of misfit dislocations are generated and these defects degrade the electronic and optical properties of QWs. The critical thickness of the GaN/Al_{0.07}Ga_{0.93}N system is found to be around 40 \AA . Our results also indicate that Si doping improves the interface qualities of the GaN QW systems. These results are expected to have significant implications on device applications based on III nitride MQWs.

Time-resolved PL studies have been carried out for a set of GaN/AlGa_N MQW samples grown under identical conditions with well thickness varying from 20 to 60 \AA . These MQW samples were grown by reactive molecular beam epitaxy (MBE) on sapphire (Al₂O₃) substrates with 50 nm AlN buffer layers. The MQWs composed of ten periods of alternating GaN wells and 50 \AA Al_{*x*}Ga_{1-*x*}N ($x \approx 0.07$) barriers. All samples were nominally undoped and the GaN epilayers grown under similar conditions were insulating. For comparison, we also present results for a QW of well thickness 60 \AA doped with Si to a level of $5 \times 10^{17} \text{ cm}^{-3}$ in the well regions. Low-temperature time-resolved PL spectra were

measured by using a picosecond laser spectroscopy system with an average output power of about 20 mW and a spectral resolution of about 0.2 meV . The time resolution of our detection system was about 20 ps .³

Low-temperature (10 K) cw PL spectra for two representative GaN/Al_{*x*}Ga_{1-*x*}N MQW samples with well thicknesses $L_w = 25 \text{ \AA}$ and $L_w = 50 \text{ \AA}$ are presented in Figs. 1(a) and 1(b), respectively. For comparison, the PL spectrum of a GaN epilayer deposited under the similar conditions is also shown in Fig. 1(c). In the GaN epilayer, the dominant transition at 3.485 eV at 10 K is due to the recombination of the ground state of *A* exciton.⁴⁻⁶ In the 25 \AA well MQW sample, the excitonic transition peak position at 10 K is blue shifted with respect to the epilayer by an amount of 56 meV — 3.541 eV . This 56 meV blue shift is due to the well-known effect of quantum confinement of electrons and holes. If this blue shift is related to strain, then only a 25 meV shift would be seen for the GaN/Al_{0.07}Ga_{0.93}N system.⁷ The transition peaks at higher emission energies in MQWs are due to an exciton transition and its LO phonon replicas in the AlGa_N barrier regions. The observed LO phonon energy has been modified due to the symmetry properties of the MQWs. In a sharp contrast, the blue shift associated with the exciton transition at 3.485 eV is not observed in the 50 \AA well MQW sample at 10 K [Fig. 1(b)], indicating that there is no quantum confinement in this QW sample. Moreover, the dominant PL emission line at low temperatures in the 50 \AA MQW sample appears at 3.414 eV , which is 71 meV below the exciton transition line and is presumably due to an impurity related transition. We have also obtained data for MQW samples with 40 and 60 \AA well thicknesses, which behave similar to the 50 \AA well MQW sample. On the other hand, a 20 \AA well MQW behaves similar to the 25 \AA well MQW sample, however, with a larger spectral blue shift and a broader exciton emission linewidth due to the enhanced quantum confinement and interface roughness effects.

In order to further identify the origin of the optical transitions, the dynamical behavior of the optical transitions in these MQWs have also been studied and compared. The de-

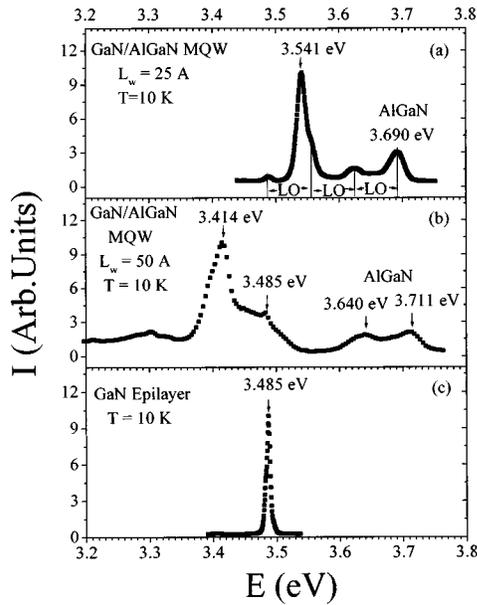


FIG. 1. Low-temperature (10 K) PL spectra of a nominally undoped (a) GaN/AlGaIn MQW sample with well thickness $L_w = 25 \text{ \AA}$; (b) GaN/AlGaIn MQW sample with well thickness $L_w = 50 \text{ \AA}$; and (c) GaN epilayer grown under identical conditions as the MQW samples.

decay of the exciton recombination resulting from the well regions of the 20 and 25 Å MQWs is exponential at temperatures below 150 K. An example of the PL temporal response is shown in the inset of Fig. 2(a). The main figure of Fig. 2(a) shows that the exciton recombination lifetime in both 20 and 25 Å MQWs increases linearly with temperature up to 60 K, similar to the behavior seen in the GaAs/AlGaAs QWs. This is a well-known property of the exciton radiative recombination in QWs, which is observable only in high quality samples. On the contrary, because of the coupling between the impurity related transition and the intrinsic transition, the decay of the dominant transition in the 50 Å MQW (at 3.414 eV at 10 K) follows two exponential functions, $I(t) = A_1 \exp(-t/\tau_1) + A_2 \exp(-t/\tau_2)$. The decay time constants τ_1 and τ_2 as functions of temperature ($T < 140 \text{ K}$) are shown in Fig. 2(b). The faster decay time constant τ_1 is almost independent of temperatures, whereas the slower decay time constant τ_2 decreases monotonically with temperature. We think that the slower decay component is predominantly due to the impurity related transition, while the faster decay component is related to the intrinsic transition (at 3.485 eV at 10 K). The impurity related transition in the 50 Å well MQW is of a band-to-impurity nature. This assignment is based on its nanosecond recombination lifetime and its spectral peak position as shown in Fig. 1(b). The emission intensity of the band-to-impurity transition is largely quenched at temperatures above 200 K. To be discussed later, the intrinsic transition in the 50 Å well MQW, which evolves from the excitonic to the band-to-band character with increasing temperature, becomes the dominant emission line at 300 K.

Figures 3(a) and 3(b) present the room-temperature PL spectra of these MQWs. The interesting feature shown in Fig. 3 is that at 300 K the band-to-impurity transition in the 50 Å well MQW is thermally quenched and the dominant

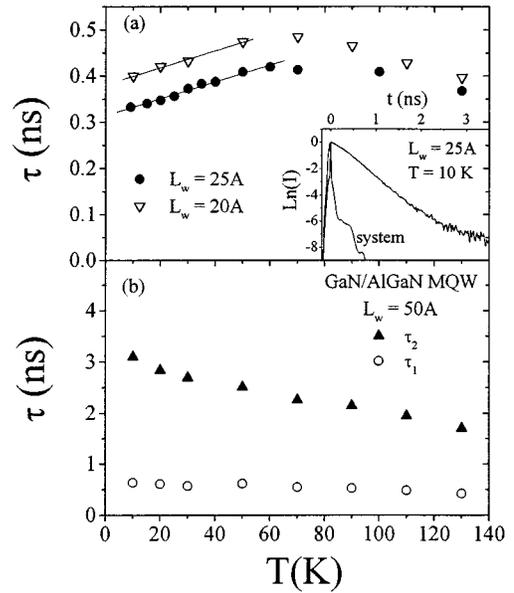


FIG. 2. (a) The recombination lifetime of the dominant PL emission line resulting from the GaN wells as a function of temperature for GaN/AlGaIn MQW samples with well thicknesses $L_w = 20 \text{ \AA}$ and $L_w = 25 \text{ \AA}$. The inset shows an example of the PL temporal response at 10 K, which illustrates that the low temperature ($T < 150 \text{ K}$) PL decay in these MQWs is exponential, $I(t) = A \exp(-t/\tau)$. (b) The decay time constants of the dominant PL emission line as functions of temperature for a GaN/AlGaIn MQW sample with well thickness $L_w = 50 \text{ \AA}$, where τ_1 and τ_2 are obtained by fitting the PL decay to a two exponential function, $I(t) = A_1 \exp(-t/\tau_1) + A_2 \exp(-t/\tau_2)$.

emission peak at 3.441 eV is blue shifted by 20 meV with respect to that in the GaN epilayer shown in Fig. 3(c). By considering the fact that no blue shift was seen at 10 K for this QW, the cause of this 20 meV shift at room temperature by quantum confinement can be precluded. We assign the transition peak at 3.441 eV in the 50 Å well MQW [Fig. 3(b)] and that at 3.421 eV in the epilayer [Fig. 3(c)] at room temperature to the band-to-band (or free electron to free hole) and exciton recombination, respectively. This is supported by the fact that the amount of blue shift (20 meV) observed is exactly equal to the binding energy of the exciton in GaN epilayers. The observation of the band-to-band transition in the 50 Å well MQW at room temperature is partly due to the presence of larger densities of dislocations which tend to break up the exciton into free carriers in this MQW.

These comparison results clearly indicate that when the well thickness is below a certain value, which we refer to as the critical thickness value, high quality QW samples can be obtained. Above the critical thickness, strain is relieved by the creation of large densities of misfit dislocations, which leads to the smearing of QW interfaces and the loss of quantum confinement as well as the dominance of the impurity transition. Our results indicate that the critical thickness of our GaN/AlGaIn QW system is less than 40 Å, which is consistent with a value of about 30 Å for the GaN films grown with an AlN buffer layer on sapphire substrate as determined by x-ray diffraction.⁸ It has also been shown that there is no quantum confinement in InGaIn/GaN MQWs with well thicknesses larger than $\sim 30 \text{ \AA}$.^{9,10}

An important result is shown in Fig. 3(d), where the

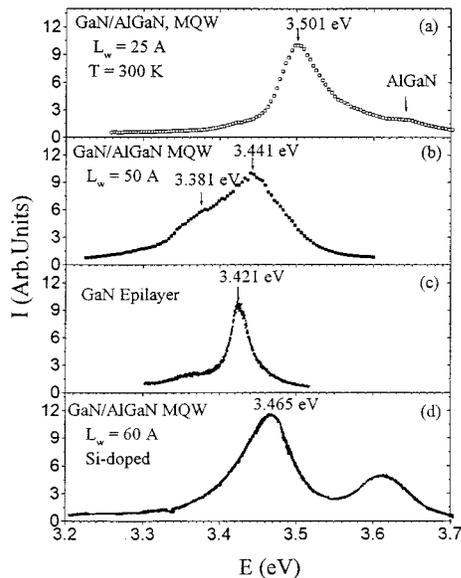


FIG. 3. Room-temperature PL spectra of the (a) undoped GaN/AlGaIn MQW sample with well thickness $L_w=25$ Å; (b) undoped GaN/AlGaIn MQW sample with well thickness $L_w=50$ Å; (c) undoped GaN epilayer grown under identical conditions; and (d) GaN/AlGaIn MQW sample with well thickness $L_w=60$ Å doped with Si in the well regions.

room-temperature PL spectrum of a 60 Å well QW doped with Si is presented. Due to the impurity screening effects, the transition in this QW sample is also of band-to-band nature.⁷ In a remarkable contrast to the nominally undoped QWs of similar well thickness, the band-to-band emission peak at 3.465 eV in Si doped QW is blue shifted by 44 meV with respect to that of the GaN epilayer. This large amount of blue shift obviously cannot be accounted for by the binding energy of the excitons in GaN (~ 20 meV). The quantum confinement effect must be taken into account in order to explain the spectral blue shift. The observation in Fig. 3(d) clearly indicates that the interface qualities and hence the optical properties of the Si doped QWs are improved over the nominally undoped QWs. This seems to suggest that Si doping suppresses the degradation effects due to misfit dislocations to a certain degree. Most recently, Nakamura *et al.*¹¹ have reported room-temperature continuous-wave operation of InGaIn MQW structure laser diodes and observed that Si doping reduces the threshold current density (from 10 to 3.6 kA/cm²) and operating voltage (from 20 to 5.5 V). It has been shown that Si dopants decrease dislocation density and hence improve the crystalline quality of GaN layers.¹² The observation here is consistent with this previous result and also with that in InGaIn MQW structure laser diodes.

Here, the observation of the band-to-band transition in MQWs is also interesting, as this transition is rarely observable in high quality and purity II–VI and other III–V semiconductors. A particular example is the GaAs/AlGaAs system in which the binding energy of the excitons is only a few meV, but the exciton transition is still the dominant transition even at room temperature. This is due to the much smaller radiative recombination rate of the band-to-band transition than that of the exciton transition in the GaAs system whereas in the GaN materials, our results presented here indicate that the recombination rates of these two intrinsic

transitions are of the same order, which is further supported by a recent observation of the band-to-band transition in a MOCVD grown epilayer at room temperature.¹³

In summary, effects of well thickness and Si doping on the optical properties of GaN/AlGaIn MQWs have been investigated. Our results have yielded that (i) the optical transitions in nominally undoped MQWs with narrow well thicknesses ($L_w < 40$ Å) are blue shifted with respect to the GaN epilayer due to quantum confinement, however, no such blue shift is evident for the MQWs with well thicknesses larger than 40 Å, (ii) the band-to-impurity transitions are the dominant emission lines in nominally undoped MQWs of large well thicknesses ($L_w > 40$ Å) at low temperatures, (iii) the exciton recombination lifetimes in MQWs with well thicknesses below 40 Å increases linearly with temperature up to 60 K, which is a hallmark of radiative recombination in QWs, and (iv) Si doping improves significantly the crystalline quality of MQWs of large well thicknesses ($L_w > 40$ Å). We have explained our results by the concept of critical thickness. MQWs with high optical qualities can be achieved when the well thicknesses are below the critical thickness value. When the well layer thicknesses exceed the critical thickness value, large densities of misfit dislocations are generated and these defects degrade the electronic properties of QWs. The critical thickness of the GaN/Al_{0.07}Ga_{0.93}N system is found to be less than 40 Å.

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