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Time-resolved photoluminescence was employed to study optical transitions in GaN/Al$_x$Ga$_{1-x}$N multiple quantum wells (MQWs). The effects of quantum confinement on the optical transitions as well as on the exciton-phonon interactions in MQW were investigated. Recombination lifetimes of optical transitions were measured at different emission energies and temperatures from 10 to 300 K. It was found that the exciton recombination lifetime increases linearly with temperature up to 60 K, which is a hallmark of radiative exciton recombination in MQW. Observed optical transitions and their dynamics in GaN/Al$_x$Ga$_{1-x}$N MQW were also compared with those in GaN epilayers and GaAs/ Al$_x$Ga$_{1-x}$As MQW. © *1996 American Institute of Physics.* [S0003-6951(96)03043-4]

GaN based devices offer great potential for applications such as UV-blue lasers, solar-blind UV detectors, and high-power electronics.[1] The commercial availability of super-bright blue light emitting diodes (LEDs) and the demonstration of the room temperature pulsed 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] In particular, important fundamental parameters of GaN, including the detailed band structure near the Γ point, free and bound exciton binding energies, and exciton radiative recombination lifetimes, have been obtained. It is expected that all GaN based devices will take advantages of quantum well (QW) structures of GaN/AlGaN and InGaN/GaN. Thus a better understanding of the fundamental optical transitions in III–V nitride QW is urgently needed. In this letter, we report the results of optical transitions and their recombination dynamics in GaN/Al$_x$Ga$_{1-x}$N multiple quantum wells (MQWs) as obtained by time-resolved photoluminescence (PL) measurements. Exciton recombination lifetimes in GaN/Al$_x$Ga$_{1-x}$N MQW have been measured at different temperatures and emission energies and are compared with those of GaN epilayers and GaAs/Al$_x$Ga$_{1-x}$As MQW.

The wurtzite GaN epitaxial layer of about 4 μm thick and GaN/Al$_x$Ga$_{1-x}$N MQW used here were grown by reactive molecular beam epitaxy (MBE) on sapphire (Al_2O_3) substrates with 50 nm AlN buffer layers.[8] MQW are composed of 10 periods of alternating 25 Å GaN wells and 50 Å Al$_x$Ga$_{1-x}$N barriers with $x \approx 0.07$. The GaN layers were nominally undoped and insulating. Low temperature time-resolved PL spectra were measured by using a picosecond laser spectroscopy system.[4,5] The time resolution of our detection system has been improved compared with our previous setup.[4,5] A micro-channel-plate photomultiplier-tube (MCP-PMT) together with a single photon counting system were used to collect time-resolved PL data and the overall time resolution of the detection system is about 20 ps. However, the lifetimes obtained by our new detection system and the previous detection system (with the use of a deconvolution technique) are consistent.

In Fig. 1, we plotted the continuous-wave (cw) PL spectra of the GaN/Al$_x$Ga$_{1-x}$N MQW sample (open triangles) obtained at (a) $T=300$ K and (b) $T=10$ K. For comparison, PL spectra of a GaN epilayer are also shown. In the GaN epilayer, the dominating transition line at $T=10$ K is due to the recombination of the ground-state of A exciton.[4,7] In the MQW, the excitonic transition peak position is blue shifted due to the well known effect of quantum confinement of electrons and holes. The blue shift at room temperature (79 meV) is what we expected for our MQW structure with a 67% (33%) conduction (valence) band offset.[8] One of the interesting features shown in Fig. 1 is that the blue shift

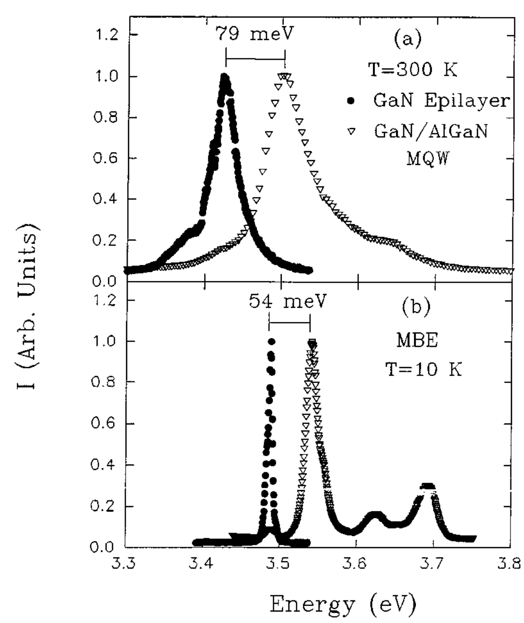

FIG. 1. cw PL spectra of a GaN epilayer (solid dots) and GaN/Al$_x$Ga$_{1-x}$N MQW (open triangles) measured at (a) $T=300$ K and (b) $T=10$ K.

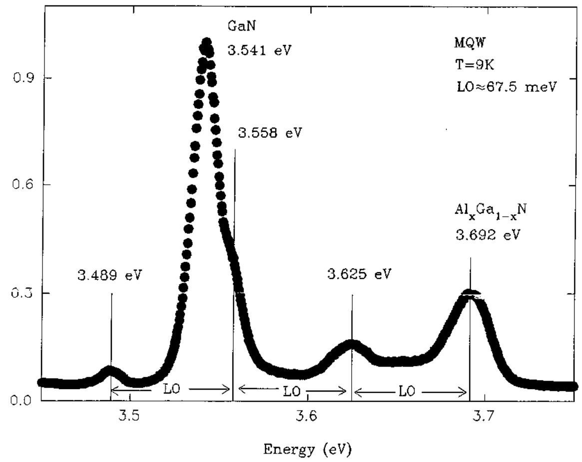

FIG. 2. cw PL spectra of $GaN/Al_xGa_{1-x}N$ MQW measured at $T=10$ K, showing several phonon replicas of the excitonic transition at 3.692 eV resulting from the $Al_xGa_{1-x}N$ barriers with a modified LO phonon energy of 67.5 meV. The excitonic transition resulting from the GaN wells at 3.541 eV is also shown.

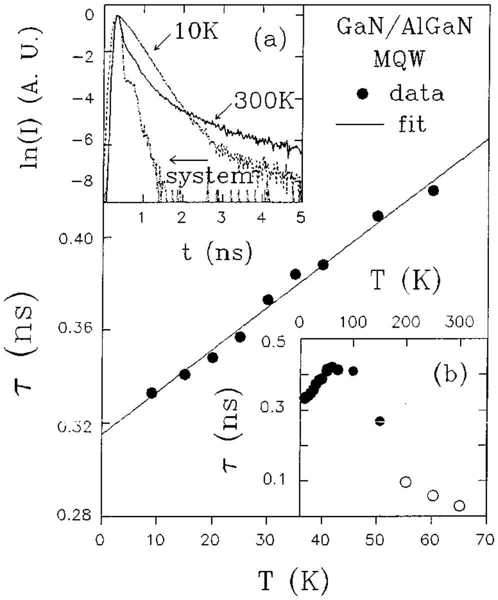

FIG. 3. Temperature dependence of the recombination lifetime of the excitonic transition resulting from the GaN wells up to 60 K. Inset (a) shows the temporal responses of the exciton recombination measured at $T=10$ and 300 K with ''system'' indicating the system response to the laser pulse which is about 20 ps. Inset (b) shows the temperature dependence of the recombination lifetime up to room temperature.

observed at 10 K is 54 meV, which is about 25 meV less than the shift of 79 meV seen at 300 K. We attribute this difference to the fact that the PL emissions in the MQW at low and room temperatures result from the recombination of localized excitons and free excitons, respectively. The exciton localization at low temperatures is caused by the interface roughness of the MQW.[9] As the temperature increases, the localization energy is no longer sufficient to localize excitons. Thus the difference in blue shift at 10 and 300 K of 25 meV measures the exciton localization energy, which gives a well thickness fluctuation of about ± 4 Å. On the other hand, one could also argue that the dominating transition peaks in MQW at low and room temperature correspond to the free exciton and band-to-band transitions, respectively. In such a context, the 25 meV difference would then correspond to the exciton binding energy in the MQW. However, the band-to-band transition was not observed in the GaN epilayer under the same experimental condition. Moreover, the enhancement of the exciton binding energy in MQW also makes the band-to-band recombination less likely.

In addition to the main exciton emission band resulting from the GaN wells, Fig. 1(b) also shows that there are more features in the higher emission energy side. For a more clear illustration, we have replotted the 10 K cw emission spectrum of the MQW sample in Fig. 2. Four transition peaks at 3.692, 3.625, 3.558, and 3.489 eV are clearly resolved. We assign these to 0–3 phonon replicas of the excitonic transition resulting from the $Al_xGa_{1-x}N$ barriers. This assignment is based on the fact that these transition lines are separated by an equal energy space (67.5 meV). Moreover, the relative emission intensities of these transitions also support our assignment. The excitonic transitions from the $Al_xGa_{1-x}N$ barriers are easily seen here because the AlN mole fraction in the barrier material is relatively low ($x \approx 0.07$), which makes the energy difference between excitons in the wells and the barriers relatively small. Another surprising feature is that the energy separation between these transition lines is 67.5 meV. In GaN epilayers, phonon replica emissions are usually associated with longitudinal optical (LO) phonons, which have an energy of 91 meV.[10] The measured phonon energy in the MQW of 67.5 meV corresponds to the transverse optical (TO) phonon energy in GaN epilayers.[10] Similar phonon replica emissions with TO phonon frequency in $GaAs/Al_xGa_{1-x}As$ MQW and with LO phonon frequency in GaAs epilayers have been observed previously.[11] It was shown theoretically that the LO phonons involved in the optical transitions in MQW vibrate at the bulk TO phonon frequency due to the symmetry properties of MQW.[11] Thus the phonon replica emissions observed in $GaN/Al_xGa_{1-x}N$ MQW shown in Fig. 2 are also associated with the LO phonons, except that they vibrate at the GaN bulk TO frequency.

We have also measured the recombination dynamics of the optical transitions in the MQW. Inset (a) of Fig. 3 shows the temporal response of the excitonic transition resulting from the wells (3.541 eV line at $T=10$ K) measured at $T=10$ and 300 K. The decay of the exciton recombination can be described very well by a single exponential at low temperatures (up to 150 K).

The exciton recombination lifetime, τ, resulting from the well regions was measured at the spectral peak positions. The main figure of Fig. 3 shows that τ increases linearly with temperature up to 60 K. To our knowledge, this is the first time that such a linear increase of the exciton recombination lifetime with temperature has been recorded for the nitride system. Such a linear behavior of τ vs T at low temperatures has been observed previously in GaAs/ $Al_xGa_{1-x}As$ MQW and is now regarded as a unique property of radiative recombination in MQW.[12] Thus our time-resolved data show that radiative recombination is the dominant process at low tem-

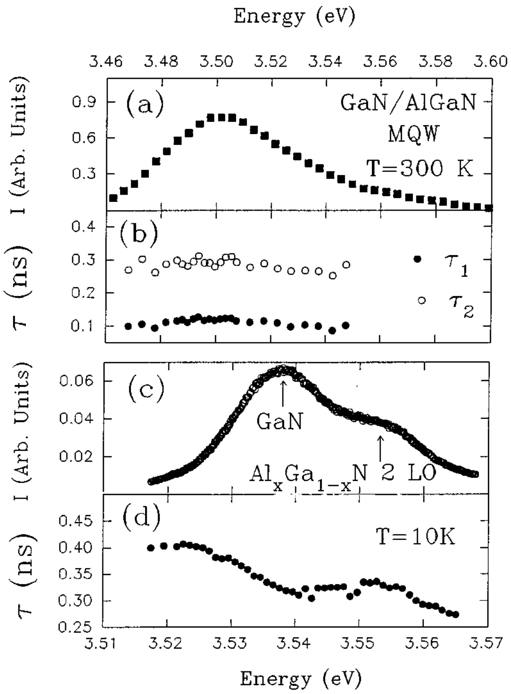

FIG. 4. cw PL spectra of the excitonic transitions in GaN/Al_xGa_{1-x}N MQW measured (at a higher excitation intensity) at (a) $T=300$ K and (c) $T=10$ K and the emission energy dependence of the exciton recombination lifetime measured at (b) $T=300$ K and (d) $T=10$ K. Arrows of GaN and Al_xGa_{1-x}N-2LO in (c) indicate the excitonic transitions resulting from the GaN wells and the 2 LO phonon replica of the excitonic transition resulting from the Al_xGa_{1-x}N barriers, respectively.

peratures, which demonstrates the high quality of the MQW sample used here. The longest radiative lifetime seen in the MQW sample here is 0.43 ns at 60 K, which is two times larger than a value of 0.22 ns seen in the GaN epilayer grown by the same method.[4] It is also of interest to compare the results of GaN/Al_xGa_{1-x}N MQW here to those of GaAs/Al_xGa_{1-x}As MQW. The low temperature radiative recombination lifetime is almost identical in both the GaN and GaAs systems and is about 0.3 ns. However, the slope of τ vs temperature T, $d\tau/dT$, is about 1.84×10^{-3} ns/K in GaN/Al_xGa_{1-x}N MQW as shown by the solid least-squares fit line in Fig. 3 and is about 1.6×10^{-2} ns/K in GaAs/Al_xGa_{1-x}As MQW of the same well width.[12] The inset (b) of Fig. 3 shows the temperature dependence of τ up to room temperature. The turnover behavior seen above 60 K is most likely due to an increased rate of dissociation of excitons and other nonradiative recombination processes at higher temperatures. Above 150 K, the decay of PL can no longer be described by a single exponential form and instead is of two exponential with the fast component contributing 90% of the PL signal. The recombination lifetime of the fast component obtained at $T>150$ K is plotted (open dots) together with the low temperature recombination lifetimes (solid dots).

In Fig. 4, we present cw PL spectra of the excitonic transition resulting from the GaN wells measured at (a) $T=300$ K and (c) $T=10$ K as well as the emission energy dependence of the exciton recombination lifetime measured at (b) $T=300$ K and (d) $T=10$ K. At $T=300$ K, the decay is two exponential with τ_1 and τ_2 representing the faster and slower decay components. The two arrows in Fig. 4(c) indicate the excitonic transition resulting from the GaN wells and the 2 LO phonon replica of the excitonic transition resulting from the Al_xGa_{1-x}N barriers. At 10 K, the exciton recombination lifetime decreases with an increase of emission energy and varies from 0.4 ns at the low energy shoulder (3.52 eV) to about 0.3 ns near the transition peaks (3.538 eV). This behavior is a well known characteristic of localized excitons due to interface roughness in QW.[9] To the contrary, the recombination lifetimes τ_1 and τ_2 are independent of emission energy at 300 K as shown in Fig. 4(b), which is consistent with our interpretation that the free exciton recombination is the dominant process at room temperature.

In summary, time-resolved PL has been employed to measure the optical transitions in GaN/Al_xGa_{1-x}N MQW. The results have been compared with those of GaN epilayers and GaAs/Al_xGa_{1-x}As MQW. Our results have revealed (i) strong quantum confinement of excitons in the GaN wells; (ii) enhanced exciton-LO phonon interactions in the Al_xGa_{1-x}N barriers; (iii) the vibration of LO phonons in MQW at the bulk TO phonon frequency due to the symmetry properties of MQW; (iv) the dominance of the localized (free) exciton recombination at low (high) temperatures; (v) a linear increase of the exciton recombination lifetime with temperature up to 60 K, which is a strong indication of radiative exciton recombination as well as the high quality of the MQW sample used here. These findings are expected to have important implications in the design of optoelectronic devices based on GaN/Al_xGa_{1-x}N MQW.

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