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Plasma heating in highly excited GaN/AlGaN multiple quantum wells

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Time-resolved photoluminescence (PL) spectroscopy was used to investigate carrier distributions in a GaN/AlGaN multiple quantum well (MQW) sample under high excitation intensities necessary to achieve lasing threshold. Room temperature PL spectra showed optical transitions involving both confined and unconfined states in the quantum well structure. Analysis of the experimental results using a microscopic theory, indicates that at high excitation the carrier distributions are characterized by plasma temperatures which are significantly higher than the lattice temperature. The implications of our findings on GaN MQW laser design are also discussed. © *1998 American Institute of Physics.* [S0003-6951(98)03243-4]

Group III–V nitride semiconductors have been recognized as promising 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 must take advantage of multiple quantum well (MQW) structures such as GaN/AlGaN and InGaN/GaN MQWs. Recently, extensive efforts have been devoted towards the fabrication and understanding of MQW lasers.[2–5] For the design of these lasers, one important issue is to maximize optical emission from the lasing states. The population of these states depends strongly on the carrier dynamical processes under high excitation conditions. This letter describes experimental and theoretical studies aimed at understanding the physical processes governing the carrier distributions in MQW structures.

In this letter, the optical properties of GaN/AlGaN MQWs under high excitation intensities were probed by picosecond time-resolved photoluminescence (PL). Since most GaN based devices operate at or above room temperature (RT), we have focused on RT optical studies. Our results revealed that in the GaN/AlGaN MQWs, plasma heating strongly effects the carrier distribution between the confined and unconfined states of the MQW structure. The confined states are the bound states of the quantum well, while the unconfined states have energies above the quantum well confinement potential, and spatially extend into the barrier regions. The plasma temperature T_p may be a few hundred degrees higher than the lattice temperature T_ℓ and increases rapidly with the injected carrier density.

The MQW sample studied was grown by reactive molecular beam epitaxy on sapphire substrates with a 50 nm AlN buffer layer. The 10 period MQW was composed of alternating 25 Å GaN wells and 50 Å $Al_xGa_{1-x}N$ barriers with $x \approx 0.07$. The sample was nominally undoped and GaN epilayers grown under similar conditions were insulating. A description of the laser and detection system used for time-resolved PL spectroscopy can be found elsewhere.[6] For typical low excitation measurements, we used a lens with a focal length of 8 cm to focus the excitation laser beam to a spot of about 50 μm in diameter. For high excitation measurements, the laser beam was focused to a spot of 2 μm in diameter by an objective with a focal length of 2.7 mm. The excitation intensity was controlled by a set of neutral density filters. A single photon counting system with an overall time resolution of 20 ps was used to measure time-resolved PL spectra.

Room-temperature continuous wave (cw) PL spectra of the $GaN/Al_{0.07}Ga_{0.93}N$ MQW sample under high ($I_{exc}=I_0$) and low ($I_{exc} \approx 0.01\, I_0$) excitation intensities are presented in Fig. 1(a) and 1(b), respectively. For comparison, the PL spectrum of a GaN epilayer (grown under similar conditions) under low I_{exc} is also shown in Fig. 1(c). For the GaN epilayer, the main peak at 3.423 eV is dominated by the band-edge transitions, including the band-to-band and free excitonic transitions.[7] In the MQW sample under low excitation [Fig. 1(b)], the main peak at 3.500 eV is blue shifted by an amount of 77 meV with respect to the GaN epilayer. The blue shift is due to quantum confinement and biaxial strain effects. Under very high excitation, the PL spectrum is quite different. As shown in Fig. 1(a), it has a much broader emission band due to band filling and carrier-carrier collisions. A second PL line with peak position around 3.610 eV is attributed to the unconfined states as determined from the band structure.

In order to understand the optical transitions in the $GaN/Al_{0.07}Ga_{0.93}N$ MQW sample under high I_{exc}, we varied the excitation intensity by one order of magnitude from $0.1 I_0$ to I_0. The carrier density is estimated to be about $N = 10^{12}/\mathrm{cm}^2$ (at $I_{exc}=0.1\, I_0$) to $10^{13}/\mathrm{cm}^2$ (at $I_{exc}=I_0$). We plotted the PL spectra for four representative excitation in-

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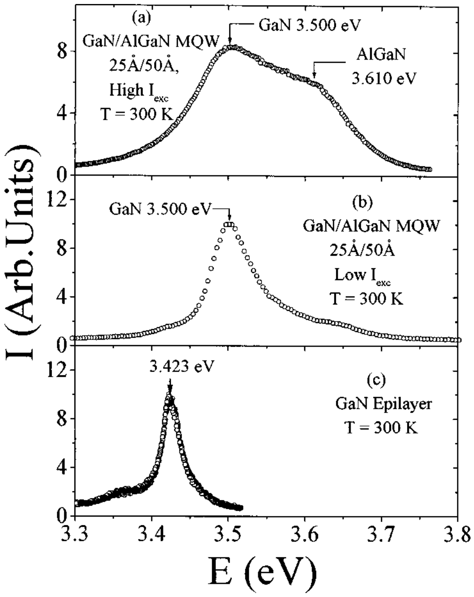

FIG. 1. Room-temperature cw PL spectra of the 25 Å well thickness GaN/$Al_{0.07}Ga_{0.93}N$ MQWs under (a) low and (b) high excitation intensity. For reference, the PL spectrum of a GaN epilayer grown under similar conditions as the MQWs is included in (c).

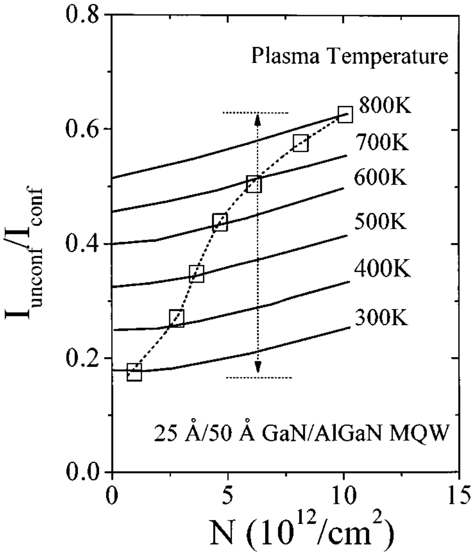

FIG. 3. The ratio of the PL intensities of the 25 Å GaN/$Al_{0.07}Ga_{0.93}N$ MQW sample from the unconfined states in the barriers and the confined states in the wells as a function of injected carrier density. The open squares are experimental data and the solid lines are calculations for different plasma temperatures T_p.

tensities in Fig. 2. Each emission spectrum can be fitted with a Lorentzian function (low energy peak) plus a Gaussian function (high energy peak) as shown by the dashed curves in Fig. 2. The low energy peak at 3.500 eV is due to transitions involving carriers in the confined states from the wells. The high energy peak around 3.610 eV is due to the transitions involving carriers in the unconfined states.

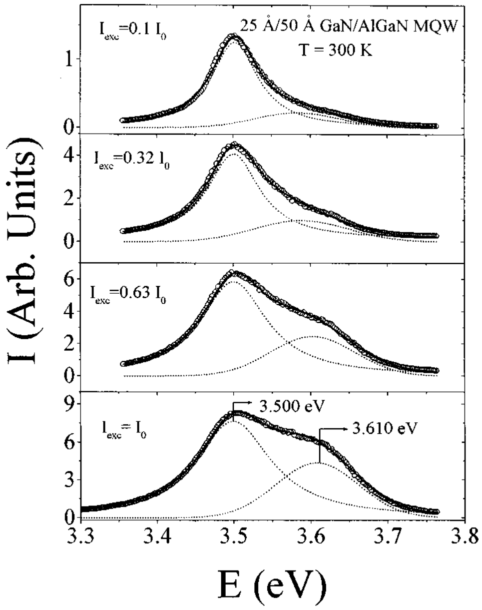

FIG. 2. Room-temperature PL spectra of the 25 Å GaN/$Al_{0.07}Ga_{0.93}N$ MQW sample measured at four representative high excitation intensities. The dotted lines are the least square fitting of the experimental data with a Lorentz function (lower energy peak) and a Gaussian function (higher energy peak), while the solid line is the sum of the Lorentz and Gaussian fitting curves.

The actual shape of the emission spectrum depends on the inhomogeneous broadening due to, e.g., quantum well thickness fluctuations in the sample. The integrated spectrum which is proportional to the spontaneous emission rate, however, is independent of inhomogeneous broadening. In Fig. 3, we plotted the ratio of the integrated PL spectra from the unconfined and the confined states, $I_{\mathrm{unconf}}/I_{\mathrm{conf}}$, as a function of injected carrier density N (open squares). The ratio starts at a value of about 18% for $N=10^{12}/\mathrm{cm}^2$ ($I_{\mathrm{exc}}=0.1I_0$), and reaches a value over 64% for $N=10^{13}/\mathrm{cm}^2$ ($I_{\mathrm{exc}}=I_0$). This shows considerable contributions from the barrier regions even at relatively low carrier density. For device applications, the optical transition from the barrier regions is a loss to optical gain. The carrier density is very high in our experiment and an electron-hole plasma (EHP) state is expected. Because the carrier transfer process from barrier to well (normally a few tens to hundreds femtoseconds) is much faster than the carrier recombination lifetime (a few hundred picoseconds), it's reasonable to assume the carrier populations are described by quasiequilibrium distribution at some plasma temperature. The laser pump energy is about 4.3 eV, which is far above the energy band gap of the sample studied here. This may result in a hot carrier population with a significantly high plasma temperature.[8]

The experimental results are analyzed using a microscopic theory, which is based on the semiconductor Bloch equations with Coulomb correlation effects treated at the level of quantum kinetic theory.[3] The model gives the gain spectra at different carrier densities and plasma temperatures. Using a phenomenological expression based on energy balance,[9] these gain spectra are converted to the corresponding spontaneous spectra. The areas under the spontaneous emission spectra are then compared to the experimental results.

The calculated ratio of carriers in the unconfined to the confined states ($I_{\mathrm{unconf}}/I_{\mathrm{conf}}$) as a function of carrier density at different temperatures are plotted in Fig. 3 (solid lines). The figure shows that the experiment results can only be explained by plasma heating of the injected carriers at high $I_{\mathrm{exc}}(T_p>T_\ell)$. The transparency carrier densities for GaN/$Al_xGa_{1-x}N$ MQW structures with well thickness from

2 to 4 nm were calculated to be around $1\times10^{12}/\mathrm{cm}^2$.[10] It is thus obvious from Fig. 3 that under high carrier injection density above the transparency density, the plasma temperature, T_p, is no longer a constant. It rapidly increases with injected carrier density. Our results indicate that above the transparency carrier density, the carrier temperature may be a few hundred degrees above T_ℓ, reaching a value of 800 K when the carrier density reaches a value of $10^{13}/\mathrm{cm}^2$. Thus a significant PL contribution from the barrier regions is actually due to the carrier plasma heating effect.

Plasma heating makes it more difficult to obtain high quantum efficiency in the well regions of MQW laser structures. While with current injection, the carriers are likely to be introduced at lower energies (4.1 eV for $\mathrm{Al}_{0.3}\mathrm{Ga}_{0.7}\mathrm{N}$ cladding layers instead of 4.3 eV in our optically pumped experiment), our results can still provide suggestions on improving the quantum efficiency of future $\mathrm{GaN/Al}_x\mathrm{Ga}_{1-x}\mathrm{N}$ MQW laser structures, or increasing the percentage of carriers recombining in the well regions. The Al mole concentration in the barriers should be optimized to reduce the carrier density in the barrier regions. Our calculations showed that the unconfined carrier density is considerably reduced if we increase the Al mole concentration in $\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{N}$ barrier from 10% to 20%.[10] However, increasing the Al mole concentration in $\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{N}$ barrier may cause difficulty to the sample growth. One possibility is to grow graded $\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{N}$ barriers to improve carrier capture efficiency. It has been demonstrated that graded barriers improve the efficiency of carrier capture by the wells in group II–VI materials.[11]

In summary, the RT optical properties of the $\mathrm{GaN/Al}_{0.07}\mathrm{Ga}_{0.93}\mathrm{N}$ MQWs under high excitation intensity were investigated by picosecond time-resolved PL spectroscopy. Our results show that under high excitation intensity, (a) injected carriers form an EHP and (b) plasma heating of the injected carriers strongly affects the carrier distributions in the well and barrier regions. Our results indicate that at carrier densities above the transparency density, the carrier plasma temperature may be a few hundred degrees higher than the lattice temperature and increases rapidly with the injected carrier density. The importance of plasma heating has both theoretical and experimental implications. It complicates the modeling of III–N lasers because plasma temperature has to be treated as a variable. From the experimental aspect, carrier leakage problems are accentuated by the higher plasma temperature.

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