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Ultrafast carrier dynamics in $In_xGa_{1-x}N$ (0001) epilayers: Effects of high fluence excitation

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Ultrafast carrier dynamics in $In_xGa_{1-x}N$ (0001) epilayers were investigated, using femtosecond transient differential transmission and reflection measurements for $x=0.07$, 0.15, and 0.33, over a fluence range of $1-12\ \mathrm{mJ/cm^2}$. Stimulated emission as well as band gap renormalization play a crucial role in the dynamics of the photogenerated carriers. Threshold fluence leading to saturation of the differential reflectivity and transmission signals related to the In mole fraction has been observed, which is attributed to band gap renormalization, Auger process, and carrier recombination through In-rich nanoclusters. Furthermore, coherent acoustic phonon oscillations have also been observed in the $In_{0.15}Ga_{0.85}N$ at high fluence excitation. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2190456]

Semiconductors based on GaN have recently attracted a great deal of attention due to their potential applications as blue light optoelectronic devices. Of particular interest is the ternary alloy $In_xGa_{1-x}N$, where even moderate concentration of indium is believed to lead to formation of In-rich nanoclusters, which strongly influence the optical properties of the material.[1] The localized energy states caused by In composition fluctuation in the InGaN active layer are related to the high efficiency of the InGaN-based emitting devices. In spite of the progress made in recent years, significant work needs to be done to further improve the fundamental understanding of these materials. Key quantities influencing the dynamic behavior in these systems, such as carrier thermalization, are not very well understood, especially when these systems have undergone strong excitation. State filling, band gap renormalization, free carrier absorption, enhanced carrier recombination through stimulated emission,[2,3] and carrier trapping by recombination centers are some of the mechanisms evoked in the dynamics of these systems which need to be investigated. The available technology of ultrafast laser pulse excitation may help toward understanding the basic relaxation mechanisms of carriers in these materials.[4]

There have been many reports on stimulated emission in InGaN epilayers[2,3] and multiple quantum well structures.[5] However, very little work has been done in investigating ultrafast dynamics in such systems following strong fluence excitations which are well above stimulated emission threshold. In view of this, we report on the ultrafast transient differential degenerate reflectivity ($\Delta R/R$) and transmission ($\Delta T/T$) measurements in a set of $In_xGa_{1-x}N$ epilayers with x equal to 0.07, 0.15, and 0.33 under strong excitation. The source of short pulses consists of a self-mode-locked Ti:sapphire oscillator generating 100 fs pulses at 800 nm. A regenerative amplifier system is used to amplify the pulses to approximately 1 mJ at a repetition rate of 1 kHz. The ultrashort pulses are used in a noncollinear pump probe geometry, where the beams are frequency doubled at 400 nm using a nonlinear crystal [beta-barium borate (BBO)].

The samples used in these experiments consisted of 500 nm $In_xGa_{1-x}N$ epilayers, grown by nitrogen radio-frequency plasma source molecular-beam epitaxy[6] (RFMBE) on Ga-face GaN/Al_2O_3 (0001) substrates. The GaN layer on the sapphire substrate consisted of two sublayers: 30 nm grown by RFMBE and 2 μm by metal-organic chemical vapor deposition. Room temperature photoluminescence measurements performed on these samples revealed band gap energies at 3.04, 2.66, and 2.0 eV, corresponding to $In_xGa_{1-x}N$ samples with In mole fraction (x) equal to 0.07, 0.15, and 0.33, respectively. The compositions were confirmed by x-ray diffraction measurements. The asymmetric line shape of the InGaN x-ray diffraction peaks suggests the presence of compositional fluctuations, probably in the form of In-rich nanoclusters, in the films.

In the following, we will describe transient transmission and reflectivity measurements obtained with a range of fluence from 1 to 12 $\mathrm{mJ/cm^2}$ at 400 nm. The high fluences used in these experiments are well above the stimulated emission threshold for all indium concentration samples. Figure 1 shows the induced transient transmission and reflection changes measured with a pump fluence of 1 $\mathrm{mJ/cm^2}$ for all three samples. The upper curves correspond to the differential reflection $\Delta R/R$ with the scale shown on the left axis, the lower curves correspond to the differential transmission $\Delta T/T$ with the associated scale being on the right axis.

The $\Delta T/T$ measurements, for all the samples, appear to have a fast positive rise, followed by recovery toward equilibrium. Following the initial rise in the signal, the lowest x sample has a relatively long single exponential decay (~45.6 ps). On the other hand, the $x=0.15$ sample has a sharp drop reaching a minimum value below the zero level at 5 ps followed by a steady increase toward equilibrium with a time constant of 40 ps. Similarly, the highest indium concentration sample has depicted a fast drop to a minimum value

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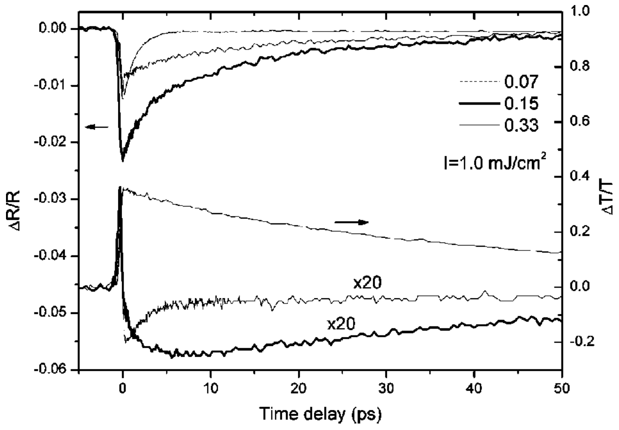

FIG. 1. Measured transient differential reflectivity and transmission responses of $In_xGa_{1-x}N$ samples, with $x=0.07$, 0.15, and 0.33, at fluence of 1.0 mJ/cm^2 (dashed line: $x=0.07$; bold solid line: $x=0.15$; normal solid line: $x=0.33$).

below the zero level at 0.5 ps and recovers toward equilibrium with a fast component of 3 ps and a much slower component.

The $\Delta R/R$ measurements exhibited a pulse-width limited drop, followed by a much longer decay toward equilibrium. The $x=0.07$ sample presents an initial fast recovery of 250 fs, which is followed by a double exponential recovery toward equilibrium with time constants of 3.8 and 34 ps. The $x=0.15$ sample appears to have a double exponential recovery of 2.7 and 20 ps toward equilibrium, whereas the highest x sample has a fast single exponential decay of 1.4 ps.

Measurements taken at the highest fluence of 12 mJ/cm^2 in these experiments are shown in Fig. 2. The behavior of the $\Delta T/T$ and $\Delta R/R$ data is distinctly different from the one observed at lower intensities as seen in Fig. 1. Clearly evident in all $\Delta T/T$ measurements is a fast rise, followed by a recovery toward equilibrium related to various relaxation mechanisms. The $\Delta T/T$ data for the lowest x sample have a fast 1.9 ps and a slow 47 ps exponential decay. The $In_{0.15}Ga_{0.85}N$ sample has depicted a faster single exponential decay (480 fs) toward equilibrium, whereas the highest x sample has a single exponential decay drop (560 fs) below the zero level, followed by a steady increase toward equilibrium with a much slower component.

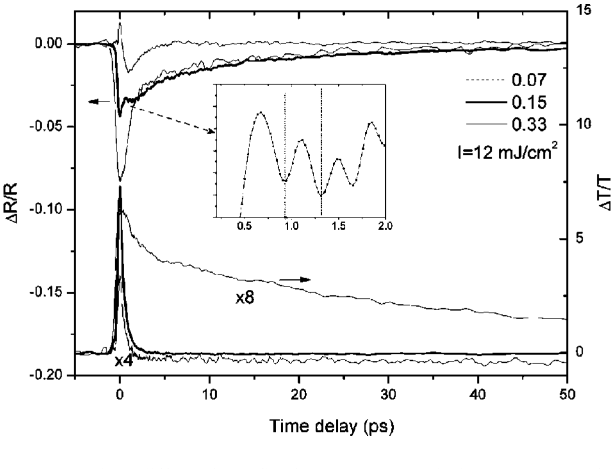

FIG. 2. Transient differential reflectivity and transmission responses of the $In_xGa_{1-x}N$ samples, with $x=0.07$, 0.15, and 0.33, at fluence of 12 mJ/cm^2 (dashed line: $x=0.07$; bold solid line: $x=0.15$; normal solid line: $x=0.33$).

The $\Delta R/R$ measurements of the two lower x samples appear to have a pulse-width limited drop and a recovery toward equilibrium. The $x=0.07$ sample has a fast recovery of 1.4 ps which is followed by a slow exponential decay toward equilibrium with a time constant of 33 ps. The $x=0.15$ sample presents a very fast recovery (360 fs) with a plateau of 1.5 ps together with small amplitude oscillations (see inset in Fig. 2) and a double exponential decay of 2.6 and 17 ps. We should point out that these amplitude oscillations appeared at 5 mJ/cm^2 and higher fluence. Finally, the $In_{0.33}Ga_{0.67}N$ sample exhibits a distinct behavior at high fluence from all the other samples. We notice a fast positive rise followed by a sharp drop to a value below the zero level (1 ps) and a fast increase toward equilibrium (2 ps) followed by a much slower component.

Following the above band gap excitation, the generated carriers induce an increase in the transmission due to state/band filling. The recovery to the equilibrium transmission is due to various effects, which include stimulated emission, carrier capture by traps, and diffusion of carriers from the probing region. In the $x=0.07$ sample, the carriers are generated with very little kinetic energy since the pump energy (3.1 eV) is very close to the band gap energy of 3.04 eV. The relatively slow recovery of the $\Delta T/T$ signal for this sample may be attributed to the normal radiative recombination of the carriers along with diffusion of the carriers out of the probing region. The observed biexponential recovery of the $\Delta R/R$ data for this sample further supported this assumption. We should point out that the sharp drop of $\Delta R/R$ after the state/band filling is due to the energy band gap renormalization effect[7] (accumulation of carriers in the band edges), which appears at this fluence. This drop is very fast given that the total kinetic energy acquired by the photogenerated carrier is only ~60 meV, thus requiring very little time for their energy relaxation to the band edges.

The $In_{0.15}Ga_{0.85}N$ sample has a very fast decay following the peak in the differential transmission which we believe is due to thermalization of electrons (holes) to the bottom (top) of the conduction (valence) band via the emission of optical phonons. Following thermalization and accumulation of the carriers at the edges of the bands, we have band gap renormalization bringing the overall $\Delta T/T$ to a value below the zero level. This signal recovers back to equilibrium at a slower rate due to stimulated emission, radiative recombination, and diffusion. At the same time, the $\Delta R/R$ data reveal again a biexponetial recovery of the signal, with the fast component attributed to band gap renormalization and the slow component due to stimulated emission, radiatative recombination, and diffusion effects. Here, we should point out that diffusion effects become more important as the optical penetration depth is smaller than the $x=0.07$ sample.

The $In_{0.33}Ga_{0.67}N$ sample has a similar behavior with the $x=0.15$ sample. Here, however, although the carriers are generated higher in the bands they appear to have a shorter thermalization time. This may be due to a stronger band gap renormalization effect. Although the fluence is the same, the penetration depth is smaller than the $x=0.15$ sample, thus generating a larger carrier density. With the accumulation of carriers at the band edges there is a strong contribution to stimulated emission seen as a recovery of the $\Delta T/T$ signal followed by a much slower recovery toward equilibrium due to radiative recombination and diffusion. The contributing factors used to explain the transient transmission data also explain the differential reflectivity measurements in this sample.

For the high fluence pumping, the differential transmission data of the $In_{0.07}Ga_{0.93}N$ sample have a faster drop toward equilibrium from the one seen at lower fluence due to enhanced stimulated emission from the high accumulation of photogenerated carriers at the band edges. Following the fast component, there is a slower component due to radiative recombination and diffusion. The $x=0.15$ sample appears to have a single exponential decay due to stimulated emission followed by a much slower recovery toward equilibrium due to the spontaneous emission and diffusion effects. The high concentration sample $x=0.33$ has enhanced stimulated emission followed by a more pronounced band gap renormalization and recovery toward equilibrium.

We should point out that for the high intensity pumping used in these experiments, the generated carriers will enhance stimulated emission and possibly other many body recombination mechanisms such as Auger. This is clearly seen in the high intensity time resolved measurements for the $x=0.15$ and 0.33 samples, where the $\Delta T/T$ signal recovery to the equilibrium appears to have small negative contribution. Similarly, the $\Delta R/R$ data appear to have a fast recovery toward equilibrium due to the above mentioned mechanisms. Especially in the $In_{0.07}Ga_{0.93}N$ sample, the $\Delta R/R$ measurements clearly show a fast stimulated emission effect and slow spontaneous emission and diffusion effects. Furthermore, in the $x=0.15$ sample the band filling effect together with the band gap renormalization bring the signal to a minimum value. The thermalization of the photogenerated carriers results in the generation of large number of optical phonons, which is evident from the small recovery of the signal reaching a plateau value. The observed oscillations with a period of ~0.42 ps (see inset in Fig. 2) in this plateau may be attributed to coherent acoustic phonon emission.[8] The fluence threshold for these observed oscillations was determined to be 5 mJ/cm^2. Furthermore, due to the large number of carriers present, Auger recombination and enhanced stimulated emission remove the carriers from the band edges. Simultaneously, spontaneous emission via photons, after the carrier density decreases below the stimulated emission threshold and diffusion effects, remove the carriers out of the probing region recovering the system in the steady state.

The $x=0.33$ sample appears to have a different behavior from the other samples. Due to the high accumulation of carriers and high kinetic energy of ~1.1 eV, the $\Delta R/R$ data appear to have an initial positive rise. This positive signal is attributed to a large number of phonon emissions increasing the lattice temperature and bringing the carriers at the band edges. However, a strong band gap renormalization effect and Auger recombination drop the reflectivity signal below the zero level. Stimulated emission (fast component) and spontaneous emission along with diffusion effects (much slower component) bring the system toward equilibrium.

Figure 3 shows the $\Delta R/R$ peak dependence on fluence for the samples under investigation in this work. All the samples exhibited a nearly linear dependence at low fluences and a saturation behavior above a threshold. We believe that the photogenerated carriers fill the bands providing a negative contribution to the reflectivity until a threshold fluence is reached, at which point nonlinear effects such as Auger recombination and band gap renormalization (positive contribution) become equally important resulting in the saturation of the peak $\Delta R/R$. Furthermore, the In traps in the samples[9] provide an additional recombination mechanism for the carriers contributing to saturation of the peak signal. Similar analysis may be used to explain the peak $\Delta T/T$ dependence on fluence.

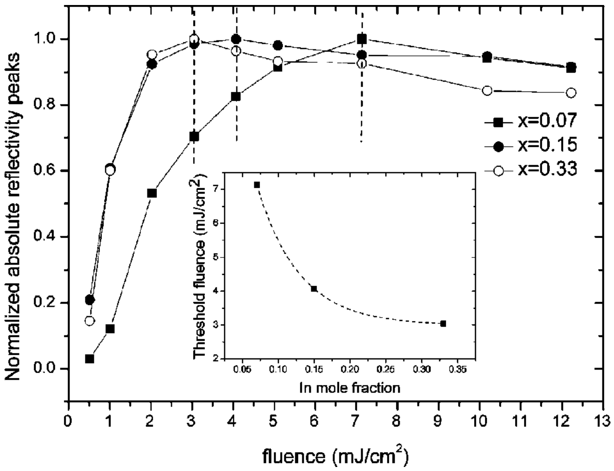

FIG. 3. Normalized absolute reflectivity peaks as a function of fluence under investigation in this work. The inset shows the threshold fluence for each In mole fraction.

We note that this threshold in the $\Delta R/R$ and $\Delta T/T$ peaks is dependent of the In mole fraction of the epilayers. It appears that with increasing x value the threshold moves to lower fluences (see inset of Fig. 3). We believe that this threshold dependence on fluence may be attributed to band gap renormalization, Auger process, and carrier recombination through In-rich nanoclusters.

In conclusion, we have investigated carrier dynamics in $In_xGa_{1-x}N$ ternary alloys, with x between 0.07 and 0.33, using ultrafast laser pulses at very high fluences. In these high fluences, the large number of photogenerated carriers causes band structure shrinkages (band gap renormalization). Simultaneously, the stimulated emission acts as an accelerating recombination factor for carriers above the threshold, where spontaneous emission and diffusion effects remove the carriers out of the probing region, recovering the system in the steady state. The $\Delta R/R$ and $\Delta T/T$ peak dependences on fluence appear to have a threshold related to the In mole fraction of the samples. Furthermore, at the highest fluence level we have observed pronounced oscillations which we believe to be due to the coherent acoustic phonons.

[1] S. Nakamura, Science **281**, 956 (1998).
[2] A. Satake, Y. Masumoto, T. Miyajima, T. Asatsuma, F. Nakamura, and M. Ikeda, Phys. Rev. B **57**, R2041 (1998).
[3] J. K. Shmagin, J. F. Muth, R. M. Kolbas, S. Krishnankutty, S. Keller, U. K. Mishra, and S. P. DenBaars, J. Appl. Phys. **81**, 2021 (1997).
[4] A. Othonos, J. Appl. Phys. **83**, 1789 (1998).
[5] T. J. Schmidt, S. Bidnyk, Y. H. Cho, A. J. Fisher, J. J. Song, S. Keller, U. K. Mishra, and S. P. DenBaars, Appl. Phys. Lett. **73**, 3689 (1998).
[6] E. Dimakis, A. Georgakilas, M. Androulidaki, K. Tsagaraki, G. Kittler, D. Cengher, E. Bellet-Amalric, D. Jalabert, and N. T. Pelekanos, J. Cryst. Growth **251**, 476 (2003).
[7] C.-K. Sun, F. Vallee, S. Keller, J. E. Bowers, and S. P. DenBaars, Appl. Phys. Lett. **70**, 2004 (1997).
[8] C. Sun, J. C. Liang, C. J. Stanton, A. Abare, L. Coldren, and S. P. DenBaars, Appl. Phys. Lett. **75**, 30 (1999).
[9] F. Chen, A. N. Cartwright, H. Lu, and W. J. Schaff, J. Cryst. Growth **269**, 10 (2004).