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2007 Semicond. Sci. Technol. 22 158

(http://iopscience.iop.org/0268-1242/22/2/027)

INSTITUTE OF PHYSICS PUBLISHING SEMICONDUCTOR SCIENCE AND TECHNOLOGY

Semicond. Sci. Technol. **22** (2007) 158–162 doi:10.1088/0268-1242/22/2/027

Temporal evolution of effects of ultrafast carrier dynamics in $In_{0.33}Ga_{0.67}N$: above and near the bandgap

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Received 21 June 2006, in final form 27 November 2006
Published 9 January 2007
Online at stacks.iop.org/SST/22/158

Abstract

Ultrafast carrier dynamics in the $In_{0.33}Ga_{0.67}N$ epilayer were investigated in detail, using femtosecond transient differential non-degenerate optical absorption measurements. Following an excitation at 400 nm with fluence ranging from 25 μJ cm^{-2} to 3000 μJ cm^{-2}, probing was carried out above and near the bandgap using different wavelengths generated from a super continuum source. We have found that bandgap renormalization plays a key role when probing at photon energies well above the bandgap and it is clearly distinct from other effects at the lowest fluence. The critical carrier density for the onset of noticeable bandgap renormalization effects in this material when probing well above the bandgap is approximately 5×10^{18} carriers cm^{-3}. We have observed a decrease in the energy loss rate of this material as a function of photogenerated carrier density which is attributed to phonon bottleneck effect. For the lowest carrier density, we have extracted an optical phonon lifetime to be approximately 45 ± 9 fs.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

The applications of semiconductors based on GaN in optoelectronic devices have attracted a great deal of attention. Such a semiconductor is the ternary alloy $In_xGa_{1-x}N$, where even small concentration of indium is believed to lead to the 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. There have been many reports on stimulated emission in InGaN epilayers [2–4] and multiple quantum well structures [5, 6].

Furthermore, it is well known that the optical properties of the material are dependent on their energy band structure. Most of the work carried out so far has been investigating the dynamic behaviour of the electron–hole system using time-resolved photoluminescence [7]. This technique allows only the radiative recombination dynamics to be directly characterized. In spite of the progress made using this technique in recent years, significant work is needed to further improve the fundamental understanding of carrier dynamics in these materials. Key quantities, influencing the dynamic behaviour in these systems can be directly investigated, using ultrafast pump–probe techniques [8–10]. The temporal evolution of state filling, bandgap renormalization and presence of free carriers are some of the effects that can be studied in detail, giving an important insight to the dynamic behaviour in these materials. In this work, we report on the dynamic behaviour of $In_{0.33}Ga_{0.67}N$ following ultrafast excitation with energy above and near the band gap. We use a super continuum technique to probe the dynamic behaviour

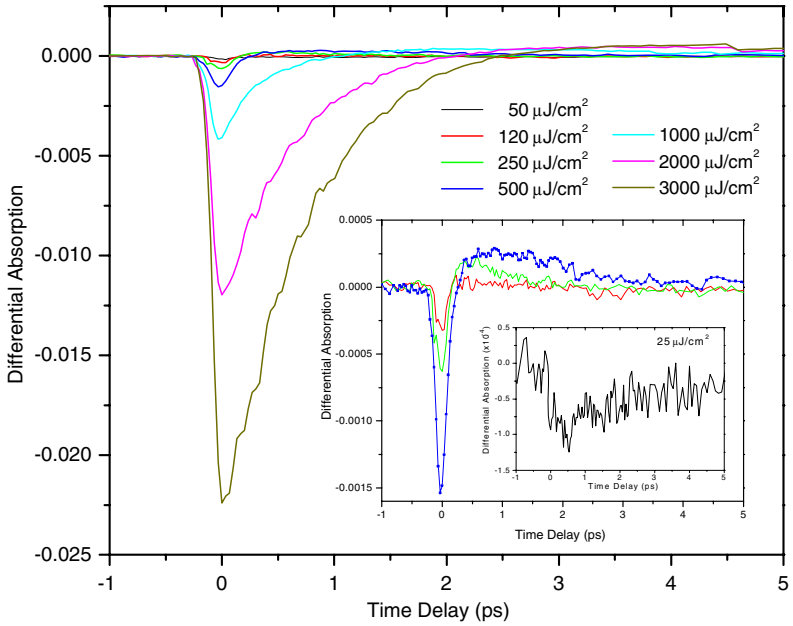

Figure 1. Transient differential absorption for the $In_{0.33}Ga_{0.67}N$ epilayer excited at 400 nm with white light probe wavelengths at 450 nm at fluences ranging from 25 to 3000 μJ cm^{-2}.

of the carriers over a broad range of bandgap energy states providing us with additional insight into the various time-dependent processes.

In view of this, we performed ultrafast transient differential non-degenerate optical absorption measurements in an $In_{0.33}Ga_{0.67}N$ epilayer under a broad range of excitation. These measurements give a complete picture of the dynamic behaviour of the carriers, as well as, their temporal evolution for this material.

2. Experimental details

The source of short pulses consisted of a self-mode-locked Ti:Sapphire oscillator generating 100 fs pulses at 800 nm. A regenerative amplifier system was used to amplify the pulses to approximately 1 mJ at a repetition rate of 1 kHz. The ultrashort pulses were used in a pump-probe setup where the pump beam was frequency doubled at 400 nm using a nonlinear crystal (BBO). A half wave plate and a polarizer in front of the BBO crystal were utilized to control the intensity of the pump beam, incident on the sample. A small part of the fundamental energy was used to generate a super continuum spectrum, by focusing the beam on a thin sapphire plate. The white light generated was used in a non-collinear geometry in a pump–probe configuration, to carry out transient reflectivity and transmission measurements. Narrow bandpass filters were utilized, in front of the detectors, to select the wavelength. The differential reflected and transmitted signals were measured using lock-in amplifiers, with reference to the optical chopper frequency of the pump beam. The temporal variation in the index of refraction was monitored as a change in the reflectivity and transmission, which is a direct measure of the photo-excited carrier dynamics within the probing region.

The sample used in these experiments consisted of a 500 nm $In_{0.33}Ga_{0.67}N$ epilayer, grown by nitrogen radio-frequency plasma source molecular-beam epitaxy (RFMBE) [11] on a Ga-face GaN/Al_2O_3 (0 0 0 1) substrate. The GaN layer on the sapphire substrate consisted of two sublayers: 30 nm grown by RFMBE and 2 μm by metal-organic chemical vapour deposition. Room temperature photoluminescence measurements performed on this sample revealed broad spectral profile (FWHM 100 nm) centred at 620 nm. The composition of the sample was confirmed by x-ray diffraction measurements. The broad photoluminescence and the asymmetry in the XRD measurements suggest strong composition fluctuations and cluster formations in this material.

3. Experimental results

In what follows, a description of transient differential absorption measurements obtained with a fluence ranging from 25 μJ cm^{-2} to 3000 μJ cm^{-2} at 400 nm over a broad range of probing wavelengths will be given. The absorption changes were obtained from transient reflection and transmission measurements taken on these samples. Results will be grouped into two distinct categories, depending on the range of probing energies; namely for probing well above the bandgap and near the bandgap.

3.1. Probing above the bandgap

Figure 1 shows the induced transient absorption changes measured with pump fluence from 25 μJ cm^{-2} to 3000 μJ cm^{-2}, at a probing wavelength of 450 nm. At low fluences (50 μJ cm^{-2} up to 250 μJ cm^{-2}) it appears that the induced absorption has a sharp drop followed by a fast recovery to positive values and then a recovery towards equilibrium (inset in figure 1). It is interesting to point out that measurements carried out at 25 μJ cm^{-2} (the lowest possible value with a distinguished signal) do not show the fast recovery

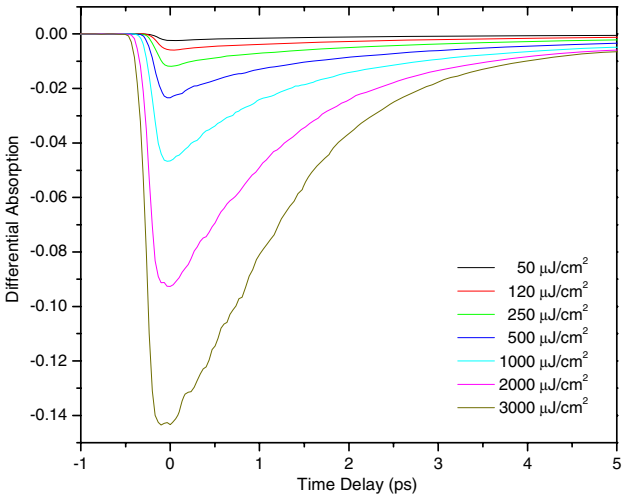

Figure 2. Transient differential absorption for the $In_{0.33}Ga_{0.67}N$ epilayer excited at 400 nm with above the bandgap white light probe wavelengths at 500 nm at fluences ranging from 50 to 3000 μJ cm^{-2}.

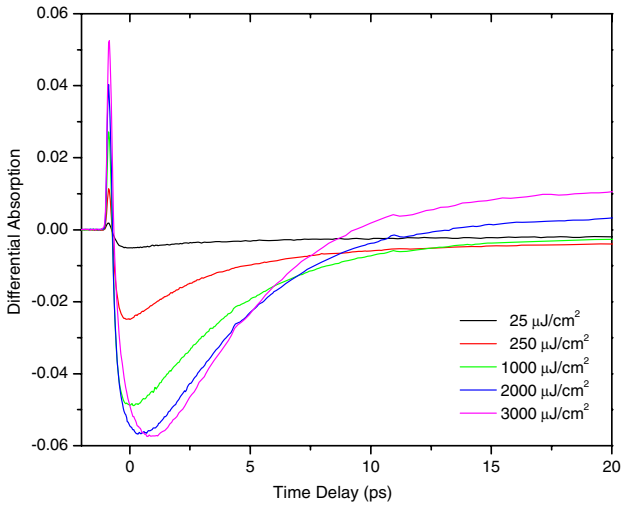

Figure 3. Transient differential absorption for the $In_{0.33}Ga_{0.67}N$ epilayer excited at 400 nm with near the bandgap white light probe wavelength at 630 nm at fluences ranging from 25 μJ cm^{-2} to 3000 μJ cm^{-2}.

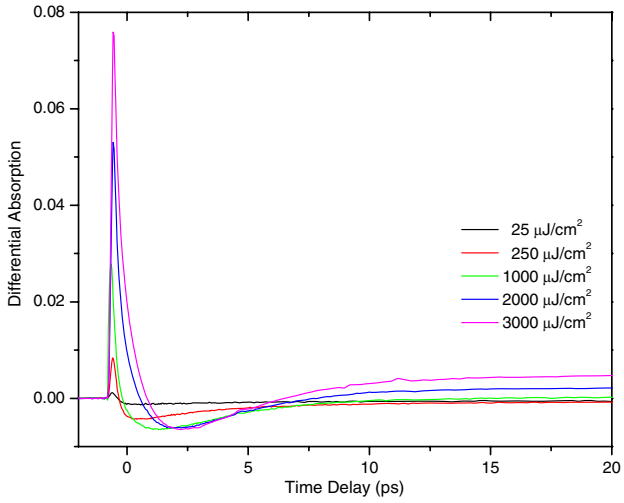

Figure 4. Transient differential absorption for the $In_{0.33}Ga_{0.67}N$ epilayer excited at 400 nm with near the bandgap white light probe wavelength at 650 nm at fluences ranging from 25 μJ cm^{-2} to 3000 μJ cm^{-2}.

to positive values, rather a slower recovery towards equilibrium (∼3 ps). This behaviour reveals a threshold of the observed fast recovery seen at higher fluences. Furthermore, at higher fluences (500 μJ cm^{-2} up to 3000 μJ cm^{-2}), we observe an enhancement of the negative differential absorption peak and a longer recovery towards equilibrium.

To further investigate the dynamic behaviour of the carriers at different energy states above the bandgap we have performed transient absorption measurements over a wavelength range between 470 and 600 nm. Typical behaviour of the transient absorption at the energy states corresponding at 500 nm probing wavelength is seen in figure 2. It is interesting to point out that with increasing probing wavelength the recovery time also increases, with the differential absorption always remaining negative for the observed time scales.

3.2. Probing near the bandgap

Figure 3 shows transient differential absorption measurements for fluences ranging from 25 μJ cm^{-2} to 3000 μJ cm^{-2} at a probing wavelength of 630 nm. This corresponds to the peak photoluminescence emission from the sample. The transient measurements for all the fluences under investigation exhibit a fast rise followed by a sharp drop below the zero level and a slow recovery towards equilibrium. Clearly evident with increasing fluences the positive peak change in absorption increases linearly whereas the negative peak reaches saturation at higher fluences. In addition, at the higher fluences there is a noticeable positive contribution to the differential absorption at longer delay times.

In order to investigate the carrier dynamics around the photoluminescence peak we have performed transient absorption measurements for longer probing wavelengths. Figure 4 shows differential absorption measurements at a probing wavelength of 650 nm. At this wavelength we observe an enhancement in the positive contribution of absorption with longer decays to negative values at which point we have a slow recovery towards equilibrium. With increasing fluence, the initial decay increases evident from the shift in the negative peak of the absorption.

4. Discussion and analysis

First we will analyse the results for the probing region above the energy bandgap corresponding to figures 1 and 2. The observed changes, following above energy bandgap excitation, can be attributed to changes in the dielectric function associated with the presence of free carriers and changes in the dielectric function associated with interband transitions. The interband contribution can arise from three separate effects, namely, state filling, lattice temperature changes and bandgap renormalization. Furthermore, effects such as carrier trapping and diffusion should be included in the analysis of the observed carrier dynamics. We should point out that for the material under investigation, the broad photoluminescence and the asymmetry in the

XRD measurements suggest strong composition fluctuations and cluster formations. This may be considered as a key contributing factor in the time evolution of the photogenerated carriers. The effect of carrier diffusion at different probing wavelengths in our time-resolved measurement was estimated using linear absorption measurements. Given that the carrier diffusion coefficient is 0.6 $cm^2\ s^{-1}$ [12] the estimated diffusion time over the penetration depth has values greater than 400 ps. As a consequence, diffusion effects are negligible in this material for the short time scales considered in this work.

Starting with the results shown in figure 1 at 450 nm with low fluence, the negative change in the differential absorption is due to the photogenerated carriers occupying the available energy states (state filling). The observed fast recovery is attributed to induced absorption due to bandgap renormalization. Furthermore, the recovery of the induced absorption is attributed to the capture of the carriers due to the presence of traps in the $In_{0.33}Ga_{0.67}N$ sample. At this point it is important to note that measurements performed at the lowest possible fluence (25 $\mu J\ cm^{-2}$) in this work have shown no bandgap renormalization contribution. Therefore we believe the critical carrier density for the onset of bandgap renormalization in this material is approximately 5×10^{18} carriers cm^{-3} for probing wavelengths well above the bandgap.

With increasing fluence, the larger number of photogenerated carriers result in occupying larger number of available energy states. As a consequence, bandfilling effect becomes more pronounced overcoming the contribution from bandgap renormalization. Furthermore, with increasing fluence we note an increase in the recovery time from 0.38 ps for 2×10^{20} carriers cm^{-3} to 0.78 ps for 6×10^{20} carriers cm^{-3}. Given this behaviour, we believe that Auger recombination has a negligible contribution to the recovery in this material. Measurements performed at probing wavelengths ranging from 470 to 600 nm reveal similar behaviour with longer recovery times with increasing wavelength. This is attributed to the fact that the lower energy states require longer time to recover due to large carrier density occupying states above the probing state.

In general, the observed recovery of the induced absorption in this material appears to have three components. The first fast component corresponds to redistribution and energy relaxation of carriers within the bands. The second component corresponds to the capture of the carrier by traps in the sample and stimulated emission (for fluence higher than 100 $\mu J\ cm^{-2}$ [4]). For the longer probing wavelengths, we observed a third much slower component which is attributed to radiative recombination.

Measurements near the peak photoluminescence emission depict a different initial behaviour from those at shorter wavelengths. The interestingly observed different behaviour at 630 nm and 650 nm we believe is mainly due to bandgap renormalization and band filling effect. With the generation of carriers and many body effects, bandgap renormalization occurs, resulting in an increase of the available density of states at the probing region. This results in an initial rise to the induced absorption which is followed by a sharp decrease associated with state filling. This sharp drop corresponds to the time required by the photogenerated carriers to reach the probing energy state (carrier energy relaxation). Following the minimum differential absorption, the temporal recovery of the signal is the removal of the carriers from the probing region through capturing of carriers by traps [13]. With increasing fluence, bandgap renormalization becomes more important, further increasing the available energy states making the induced absorption more positive.

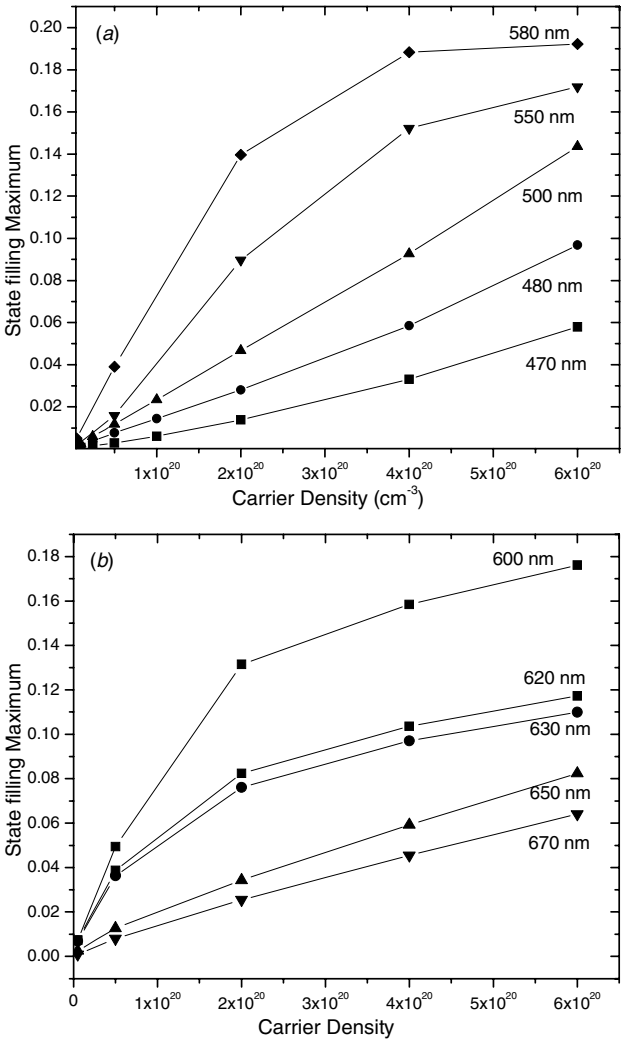

Figure 5. Maximum state filling as a function of the carrier density induced by the pump laser beam for different probing wavelengths.

State filling is a critical parameter that may be used to obtain a better understanding of density of states in semiconductors. In figures 5(*a*) and (*b*), we present the maximum change in absorption due to state filling as a function of the carrier density generated by the pump. These graphs provide interesting information about the density of states of the $In_{0.33}Ga_{0.67}N$ epilayer at the different probing energy states. For small probing wavelengths (450–500 nm), the occupation of the states increases almost linearly, where the slight deviation is believed to be due to bandgap renormalization. This shrinkage of the gap provides a slight increase in the available density of states resulting in a noticeable increase in the occupation of states at the particular probing wavelengths.

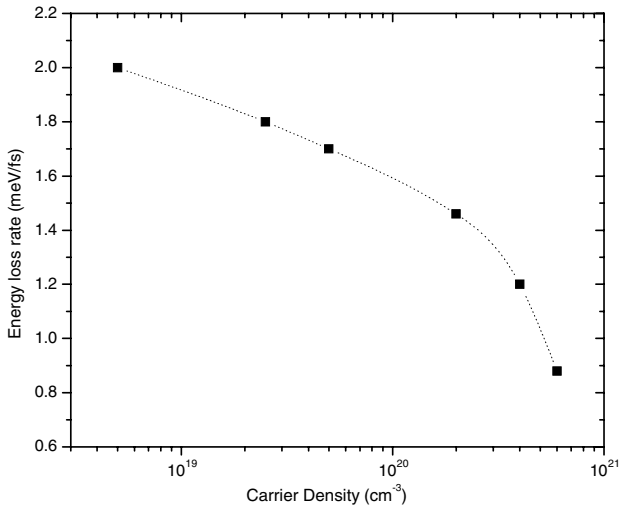

Figure 6. Energy loss rate as a function of photogenerated carrier density in the $In_{0.33}Ga_{0.67}N$ sample. The line serves as a guide to the eye.

This is a further evidence that bandgap renormalization plays a key role in this material. With increasing wavelengths (550–600 nm), hence probing at lower energy states closer to the bandgap, saturation is clearly evident for photogenerated carrier densities starting as low as 2×10^{20} cm^{-3}. Furthermore, it is interesting to point out that at probing wavelengths (620–670 nm) close to the peak of photoluminescence (figure 5(*b*)) the maximum occupied density of states follows the similar behaviour. However, the overall occupied density of states is smaller as expected closer to the band edge where the available density of states is smaller.

Finally, we should point out that from the time-resolved measurements we may resolve the rate of energy relaxation through phonon emission in this material. This parameter may be extracted from initial drop to maximum state-filling value in the differential absorption measurements for all probing wavelengths. We have performed linear fits of required times to reach maximum as a function of the energy difference between the pump and probe energy states. At the lowest fluence of 25 μJ cm^{-2}, we have estimated an energy loss rate of 2.0 ± 0.1 meV fs^{-1}. At the higher fluences of 250 μJ cm^{-2} and 2000 μJ cm^{-2}, we have 1.7 ± 0.1 meV fs^{-1} and 1.2 ± 0.1 meV fs^{-1} respectively. Figure 6 shows the energy loss rate as a function of the photogenerated carrier density in the $In_{0.33}Ga_{0.67}N$ sample. Clearly evident is the decrease of the energy loss rate with increasing carrier density. We believe this decrease may be attributed to phonon bottleneck effects which become more pronounced with increasing carrier density. It is interesting to note that at the lowest carrier density where saturation effects are minimum, the estimated value of the optical phonon (assuming 90 meV phonon energy [9]) lifetime is approximately 45 ± 9 fs for this material.

5. Conclusions

A comprehensive study of ultrafast carrier dynamics in the $In_{0.33}Ga_{0.67}N$ epilayer has been carried out using non-degenerate super continuum transient differential absorption measurements. Measurements have been performed with probing wavelengths above and near the band edge of the material with fluence ranging between 25 μJ cm^{-2} and 3000 μJ cm^{-2}. Bandgap renormalization plays a crucial role in the absorption changes for carrier densities used in this work. Measurements at low fluence reveal a critical carrier density of 5×10^{18} cm^{-3} for noticeable changes due to bandgap renormalization for probing wavelengths well above the bandgap. We have found that contributions from state filling and bandgap renormalization to differential absorption have a complex relation between them depending on the probing position on the bandgap and the carrier density generated in the material. Furthermore, we must note that the available energy states of this material increase linearly for probing energy states well above the bandgap (450–580 nm) whereas for energy states near the band edge (600–670 nm) saturation occurs. The extracted critical carrier density for the observed saturation is 2×10^{20} cm^{-3}. In addition, we have estimated the energy loss rate of this material as a function of photogenerated carrier density. The observed decrease in the energy loss rate is attributed to the phonon bottleneck effect. Finally, for the lowest carrier density we have extracted an optical phonon lifetime to be approximately 45 ± 9 fs.

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