Femtosecond Nonlinear Coherence Spectroscopy of the Blue Band of GaN

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Abstract

The nonlinear optical properties of an undoped GaN epitaxial layer have been investigated in the spectral region 400 - 450 nm using two-colour multiple-pulse photon echo measurements. Variation of the laser wavelengths and pulse sequence and the use of spectrally resolved measurements allows us to study the dephasing of the donors and acceptors and the energy relaxation properties for the blue band of GaN.

Keywords - Photon echo, Laser spectroscopy, Optical properties of GaN.

I. INTRODUCTION

Due to the high chemical, thermal and mechanical stability GaN and its composite alloys have great potential for fabricating high power, high temperature optoelectronic devices over an extremely large wavelength range from the ultraviolet to the visible. Recently, different experimental methods have been used to study the linear and non-linear optical properties of GaN over a wide range of temperatures and time scales. However, the nonlinear properties of GaN are still not well understood, and, in particular, the influence of defect levels on the nitride device is still not well explained [1-5]. These defect levels give rise to radiative and nonradiative effects, most of which are related to the emission of broad luminescence bands in the regions near 2.3 eV and 2.8 eV, the so-called "yellow band" and "blue band".

In this paper we investigate the nonlinear optical properties of GaN using spectrally resolved two-colour three-pulse photon echo measurements with wavelengths in the spectral region of the blue band (400 - 450 nm). Variation of the wavelength and the sequence of the laser pulses allows us to study the optical nonlinearity of this band on a time scale of less than 1 ps. The time evolution of the photon echo spectra in different phase matching directions versus the delay time between the laser pulses yields detailed information about the optical properties of the material.

II. EXPERIMENTS

The nonlinear spectroscopy experiments [6,7] were carried out using three femtosecond laser pulses (duration ~ 100 fs) with wavelengths in the range of 400 - 450 nm and pulse energies of 1 - 10 μJ. Two of the laser pulses have wave vectors $\mathbf{k_1}$ and $\mathbf{k_2}$ and the same wavelength, and the third pulse has wave vector $\mathbf{k_3}$ and a wavelength which may differ from that of the other two pulses. The first pulse excites the GaN layer to create electrons and holes in the band or in trap levels. The second pulse then interacts with the freely evolving system, resulting in a phase and population distribution of electrons and holes that depends on the distribution of the carriers in the respective energy levels. The time evolution of this distribution is dependent on the time separation of the first two interacting pulses. The third pulse interacts with the electron and hole systems resulting in a nonlinear signal in the phase-matching directions $\mathbf{k_4} = \mathbf{k_3} + \mathbf{k_2} - \mathbf{k_1}$ and $\mathbf{k_6} = \mathbf{k_1} + \mathbf{k_2} - \mathbf{k_3}$. For two-colour experiments with $\omega_1 = \omega_2 \neq \omega_3$, where ω is the carrier frequency of the laser pulses, conservation of momentum and energy leads to the following phase-matching directions and signal frequencies: $\mathbf{k_4} = -\mathbf{k_1} + \mathbf{k_2} + \mathbf{k_3}$; $\mathbf{k_6} = -\mathbf{k_3} + \mathbf{k_1} + \mathbf{k_2}$; $\omega_4 = \omega_3 + \delta\omega_4$; $\omega_6 = -\omega_3 + 2\omega_1 + \delta\omega_6$, where $\delta\omega$ represents the frequency shift associated with the transfer of optical coherence to other transitions of different frequency. The time evolution of the signal spectra in the $\mathbf{k_4}$ and $\mathbf{k_6}$ directions provide detailed information about the dephasing and population relaxation of excited carriers and also about the relevant energy structures.

The ultrafast pulses are generated by two independently tunable optical parametric amplifiers (OPA, 100 fs, 400 - 450 nm, 1 - 10 μJ) pumped with 1 mJ amplified pulses (80 fs, 800 nm, 1 kHz). The output of one OPA is split into two beams, which act as the pump pulses ($\mathbf{k_1}$ and $\mathbf{k_2}$), and part of the output of the second OPA acts as the probe pulse ($\mathbf{k_3}$). The signals are measured in the phase-matching directions $\mathbf{k_4}$ and $\mathbf{k_6}$ and detected by two spectrometers equipped with Garry CCD-arrays with a spectral resolution < 1 nm. In the three-pulse experiment one of the pulses ($\mathbf{k_2}$) has a fixed time delay, and the time delay between the $\mathbf{k_2}$ pulse and one positive-sign wave vector represents the population time, e.g., t_{23} between pulses 2 and 3 for observation in the $\mathbf{k_4}$ direction and t_{12} between pulses 2 and 1 for observation in the $\mathbf{k_6}$ direction. The other delay time (t_{12} or t_{23} for $\mathbf{k_4}$ and $\mathbf{k_6}$, respectively) is the coherence time. The spectra are measured at different fixed coherence times or population times by scanning the other delay time [8]. The sample under investigation is a 2 μm undoped GaN epitaxial layer grown by metal-organic chemical vapour deposition (MOCVD) on a sapphire substrate.

III. RESULTS AND DISCUSSION

i) Population time scan: Figures 1 and 2 show the time evolution of the signal spectrum versus the population time (t_{23} for the signal in the $\mathbf{k_4}$ direction and t_{12} for the signal in the $\mathbf{k_6}$ direction) with the coherence time fixed at zero. When the coherence time is zero the experiment represents the special case of a population grating experiment in which the

population time is scanned. For time delays close to zero, where the dephasing relaxation time makes a large contribution to the carrier dynamics, the spectrum of the signal exhibits additional broadening. The relaxation in the time domain leads to spectral broadening of the signal in the frequency domain. When different optical transitions are excited coherently, which is possible with the broad bandwidth of the short laser pulses, the radiated signal can consist of a broad range of frequencies.

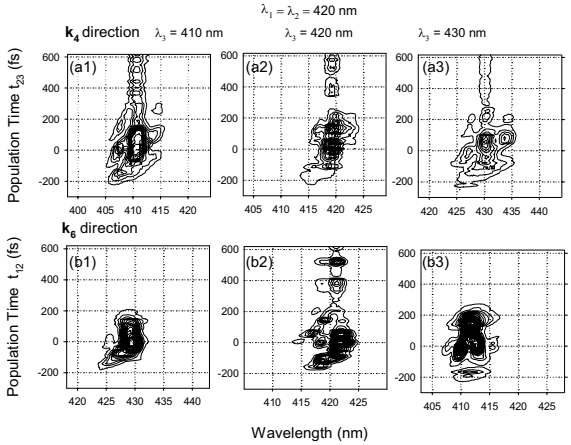
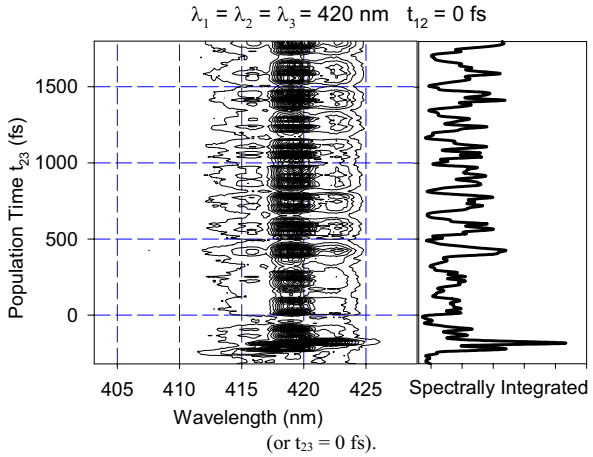


Figure 1. Time evolution of nonlinear signal spectra for 2 μm undoped GaN epitaxial layer detected in the $\mathbf{k_4}$ and $\mathbf{k_6}$ directions versus population time t_{23} (t_{12}) at various third-pulse wavelengths (λ_3) and fixed coherence time $t_{12} = 0$.



(or $t_{23} = 0$ fs).

Figure 2. Long population time scans for three pulses with the same wavelength at 420 nm and detected in the $\mathbf{k_4}$ direction.

A long time decay component, which can be considered as the diffraction of the probe beam from a grating induced by the two pump pulses, is observed in the $\mathbf{k_4}$ direction and only for the case when the three pulses have the same wavelength for observation in $\mathbf{k_6}$ direction. The diffraction signal-traces contain some beat frequencies (25 meV and 34 meV) originating from interference between the different optical transitions which are coherently excited by the broad band emission of the short laser pulses. The beating is present for the duration of the population time [6,7]. When the wavelengths of two pump pulses ($\mathbf{k_2}$ and $\mathbf{k_3}$ for the signal in $\mathbf{k_6}$ direction) are different the transient grating signal is not observed (see Fig.1b1 and Fig 1b3). The observation indicates that the different wavelength pulses interact with different donor-acceptor transitions and the coherence transfer between the transitions is rather small. This suggests that the blue band in GaN originates from many shallow donor - shallow acceptor transitions.

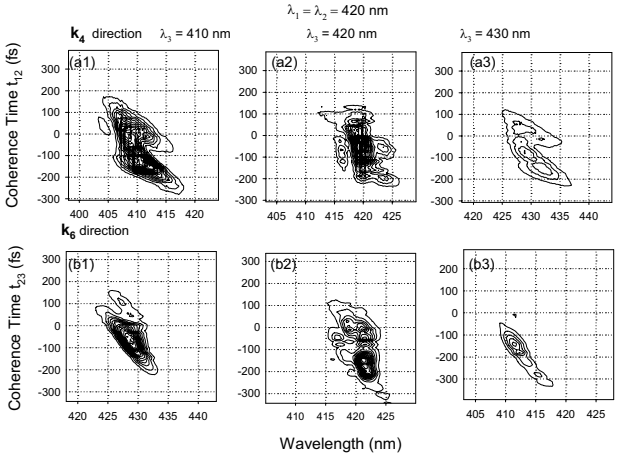


Figure 3. Time evolution of the nonlinear signal spectra from 2 μm undoped GaN epitaxial layer detected in the $\mathbf{k_4}$ (and $\mathbf{k_6}$) directions versus coherence time t_{12} (t_{23}) at various third-pulse wavelengths (λ_3) and fixed population time $t_{23} = -150$ fs (or $t_{12} = -150$ fs).

ii) Coherence time scan: In the $\mathbf{k_6}$ direction a 'pure' four-wave mixing signal with wavelength $\approx -\lambda_3 + 2\lambda_1$ is observed at times close to zero delay (Fig. 1b1 and 1b3 and Fig. 3b1-b3). The four-wave mixing signal is expected to be observed when the three pulses are present at the same time and to decay rapidly at the effective optical dephasing time T_2. When the time delay between the laser pulses is smaller than the dephasing time the time evolution of the spectral pattern reflects the dephasing time and the coupling between the transitions. Figure 3 shows the signal spectrum in the $\mathbf{k_4}$ and $\mathbf{k_6}$ direction versus coherence time for a fixed population time of -150 fs.

For a qualitative interpretation of the observations we consider a four-level system consisting of two donor levels $|D_1\rangle$ and $|D_2\rangle$ and two acceptor levels $|A_1\rangle$ and $|A_2\rangle$, as illustrated in the energy level diagrams in Fig. 5 (A and B). When $\lambda_3 > \lambda_1 = \lambda_2$ (Fig. 5A) , the first pulse $\mathbf{k_1}$ with wavelength λ_1 can generate optical coherence between levels $|A_1\rangle$ and $|D_1\rangle$. The scanning pulse $\mathbf{k_3}$ with wavelength $\lambda_3 > \lambda_1$ takes the system down to level $|A_2\rangle$ to store the coherence in the population of level $|A_2\rangle$. The third pulse $\mathbf{k_2}$ with wavelength $\lambda_2 = \lambda_1$ can then create optical coherence between levels $|A_2\rangle$ and $|D_2\rangle$, allowing the generation of a 'pure' four-wave mixing signal for the transition $|D_2\rangle \rightarrow |A_1\rangle$ at wavelength $\approx -\lambda_3 + 2\lambda_1$ in the phase-matching direction $\mathbf{k_6}$. The four-wave mixing signal is expected to decay at the effective optical dephasing time T_2. A similar set of transitions

can be considered for the case of $\lambda_3 < \lambda_1 = \lambda_2$, as illustrated in Fig. 5B, for storing the coherence in

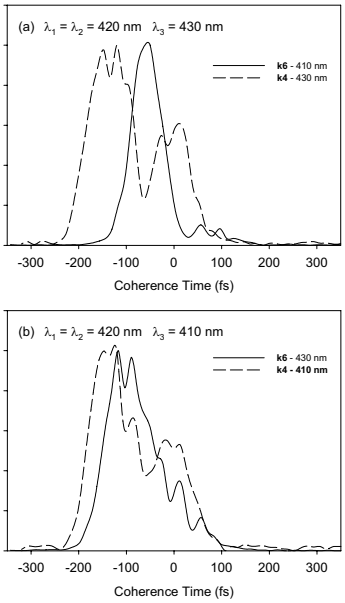


Figure 4. Time evolution of signal intensity taken from Fig. 3 at the four-wave mixing wavelength and the probe wavelength (in the $\mathbf{k_6}$ direction).

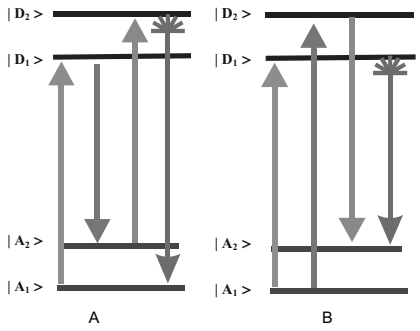


Figure 5. Pulse sequences for a four-level system.

the donor level $|D_2\rangle$. When the stored optical coherence can be transferred from level $|A_2\rangle$ to $|A_1\rangle$ or from level $|D_2\rangle$ to $|D_1\rangle$ during relaxation then the development of a photon echo signal at wavelength λ_6 reflects the time delay characteristics of the relaxation in the donor or acceptor level. The dephasing time of the donor can be deduced from the time delay of the signal for the case $\lambda_3 < \lambda_1 = \lambda_2$, which is ~ 200 fs (Fig. 4b), and the dephasing time of the acceptor is ~ 50 fs (Fig. 4a) for the case $\lambda_3 > \lambda_1 = \lambda_2$. The time evolution of the signal in the $\mathbf{k_4}$ direction is more complicated (Fig. 4) because the second and third pulses have the same wavelength and a strong phase relationship.

In conclusion, we have investigated the blue band of GaN using spectrally resolved two-colour three-pulse photon echoes and shown that the spectrally resolved measurement provides more detailed information about the relaxation and dephasing processes. The results of the population grating experiment suggest that the blue band of GaN originates from many shallow donor - hallow acceptor transitions.

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