9:45am - 10:00am
ThB5

Ultrafast Electron Dynamics and Intervalley Scattering in GaN

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Understanding ultrafast relaxation processes on the time scales of femtoseconds to picoseconds is important for applications of high speed electronics and optical devices. Our investigation on GaN-based semiconductors are motivated by GaN's applications as light emitters in the blue to UV wavelength range [1]. In our previous study [2], we have investigated ultrafast carrier dynamics in InGaN using femtosecond single-wavelength measurements, where electron dynamics was not able to be separated from that of hole. In order to investigate the electron dynamics in GaN, we use a multiple-wavelength pump-probe technique to study the intra-conduction-band dynamics in n-type GaN. Interesting information in electron-dynamics and intervalley scattering is obtained in the current study.

In our investigation of GaN, a short optical pulse is used to excite the conduction band electron distribution out of equilibrium. The internal thermalization of the electron gas and equilibration of the electronic and lattice temperature (external thermalization) are subsequently monitored in the time domain using a femtosecond probe pulse. The experiments were performed using a multiple-wavelength femtosecond pump-probe technique [3,4] where a below-bandgap infrared femtosecond pump pulse perturbed the electron gas and the changes in the distribution was probed by tuning the probe wavelengths in the vicinity of the absorption change peak, which is around the valence band to conduction band-edge transition. The use of the widely separated pump and probe wavelengths is essential to avoid the influence of the valence band carriers on the measured response. The GaN:Si film was grown by MOCVD on c-plane sapphire. After annealing the substrate and deposition of a nucleation layer, an unintentionally doped GaN layer of 1070 Å thickness and a Si doped GaN layer of 1000 nm thickness were grown. The resulted n-type carrier concentration was $2x10^{18}$ cm^{-3}. The sample was finished with a 6 nm thick undoped GaN cap layer. The crystal structure was wurzite.

Femtosecond pump-probe measurement was performed with a standard pump-probe geometry using a Ti:sapphire laser generating 120 fs pulses. One portion of the infrared beam passed through a variable delay stage and was used as the pump. The other half of the infrared beam was focused into a 500-μm-thick BBO crystal to produce frequency-doubled probe pulses tunable within 360 - 370 nm (3.44 - 3.35 eV), corresponding to the vicinity of valance-conduction band-edge transition (3.39 eV at room temperature). After the BBO crystal, the infrared pulses were removed using a color glass filter. The duration of the doubled UV pulses was 140 fs. The pump and probe were focused into the sample with a lens. The transmitted infrared were rejected using an iris and color glass filters so that only the UV probe signal was detected. The pump beam was chopped and the detected probe signal was measured as a function of the temporal delay between the pump and probe using a lock-in amplifier. Within the tuning range of the Ti:sapphire laser, free carrier absorption can be assumed to be frequency-independent and the difference in the initial electron distribution generated by different pump photon energy should be negligible. Therefore, the experimental results can thus be interpreted as probing the evolution of the same initial population by wavelength tunable pulses [3]. The time constant of external thermalization for all different probe wavelengths should thus be the same. However, if the pump IR photon energy is close to the energy difference between the U-point and Γ-point conduction band minimum, the external thermalization time constant will be strongly modified by the contribution from intervalley scattering and different time constants will appear for different pump wavelengths [4].

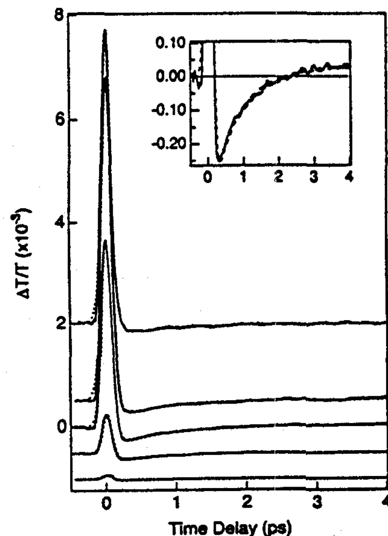

Figure 1. Measured transient transmission changes (solid lines) for probe wavelengths of 360, 362.5, 365, 367.5, and 370 nm (from top to bottom). (Inset) Enlargement of the negative component for 365 nm trace. Dotted lines are convolution fits.

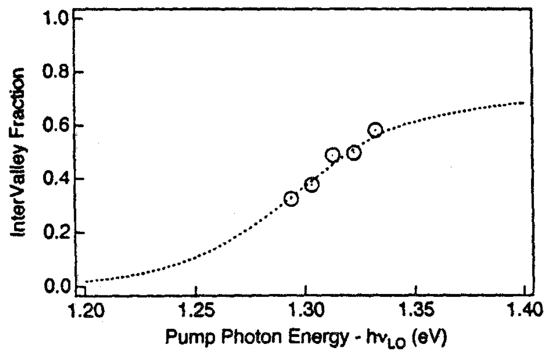

Figure 2. Intervalley scattering fraction vs. pump photon energy minus $h\nu_{LO}$. The dashed line is a computer generated fit.

Figure 1 shows the measured probe transmission changes $\Delta T/T$ for GaN:Si film against probe wavelength and probe delay for a normalized pump fluence of 70 $\mu J/cm^2$. The signal gradually vanished when probe wavelength was tuned below bandgap. At zero time delay, a positive transmission peak with a width of pump-probe cross-correlation was observed. We believe that the positive transmission peak in our experiments should have contributions from ionization of midgap states which were responsible for the observed yellow luminescence from our sample. Inset of Figure 1 shows an enlargement of the 365 nm trace. The ionization of midgap states was evidenced by the positive residue signal at long delays. After zero time delay, a negative transmission change was observed for all traces with a $\Delta T/T$ signal size on the order of 2×10^{-4}. This negative transmission change was attributed to carrier heating. However, this negative component can not be fitted using a fixed-time-constant single exponential decay for all wavelengths, indicating a contribution from intervalley scattering [4].

We have fitted our negative signals using a model developed by D. J. Dougherty *et al.* [4] with two time constants. The heating was described by a response function given by [4]

$$h(t) = \frac{a_1}{\tau_1} e^{\frac{-t}{\tau_1}} + \frac{a_2}{\tau_2 - \tau_1} e^{\frac{-t}{\tau_2}} \left\{ 1 - e^{-t\left(\frac{1}{\tau_1} - \frac{1}{\tau_2}\right)} \right\}$$

where τ_1 and τ_2 are the cooling time and intervalley scattering time. The first term describes the initial cooling response due to electron remained in the Γ valley and the second term describes the delayed cooling due to the intervalley returned electrons. The fraction of electrons scattered to the second valley is given by $a_2/(a_1+a_2)$ [4]. Excellent fits for all traces can be obtained with τ_1 = 500 fs and τ_2 = 1 ps. Delayed cooling time due to the intervalley returned electrons of 1 ps is similar to but faster than those of GaAs and ZnSe[4] material systems. This is probably due to higher density of states and larger LO phonon energy in the GaN material systems.

Figure 2 plots the intervalley scattering fraction against the pump photon energy minus one LO phonon energy of 90 meV. A drastic change in the intervalley scattering fraction was observed as we tuned the pump wavelengths. Tuning the pump across the intervalley energy threshold accounted for this behavior [4]. The fraction of the electron raised to the conduction band valley can be estimated with the overlap of density of states and free-carrier-absorption induced carrier distribution. The dotted line in Figure 2 is the result of a calculated overlap integral assuming an energy separation between valley minimums ΔE = 1.34 eV. According to A. Rubio *et al.* [5] and M. Palummo *et al.* [6], the second conduction band minimum should be located at U-point (U_1^c). The energy separation between the nearby conduction band L-point ($L_{1,3}^c$) and the valence band Γ point maximum (Γ_6^v) is 4.4, 4.54, or 5.66 eV according to the calculations of A. Rubio *et al.* [5], M. Palummo *et al.* [6] and Y. C. Yeo et al. [7], respectively. According to our experiments, the separation between the U_1^c and Γ_6^v is 4.73 eV, which is the summation of the bandgap energy (Γ_6^v- Γ_1^c, 3.39 eV) and the conduction valley energy difference (Γ_1^c- X_1^c, 1.34 eV). This work is supported by National Science Council of Taiwan, R.O.C. (Grant No. 87-2112-M-002-022).

Reference:

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