Observation of huge nonlinear absorption enhancement near exciton resonance in GaN

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Abstract: Huge nonlinear-absorption enhancement was observed in GaN system at the excitonic transition wavelength. This excitonic enhancement is attributed to four-wave-mixing type nonlinear processes. The peak nonlinear coefficient is at least 3000 cm/GW corresponding to an enhancement factor of >200. The dephasing time of the exciton was also observed to be 60-160 fs in GaN thin film at room temperature.
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1. Introduction
In this paper, we report the first observation of huge resonance of TPA coefficient near exciton resonance in semiconductor ever. This excitonic enhancement is not just attributed to four-wave-mixing (FWM) type nonlinear processes, but also due to the large exciton binding energy in GaN.

2. Sample and Experimental setup

To study the TPA with the excitation near the bandgap, we performed UV pump-probe experiments on the 2.5 μm thick undped GaN and observed the significant enhancement of TPA.

3. Experimental results

Fig. 1 shows the typically measured probe transmission change $\Delta T/T$ of the nominally undoped GaN thin film as a function of probe delay at different central wavelengths near the exciton resonance photoexcitation. The gaussian-like component of the traces at zero time delay is contributed by the TPA process. The traces also reveal that the processes of excitons ionization and free carriers thermalization were 100 – 250 fs and 600 – 800 fs, respectively. The detail of the fast carriers dynamics were discussed in our previous studies [1]. The magnitude of the TPA component is obviously enhanced near the photoexcitation of 362 nm, which is very close to the exciton excitation energy in our sample.

4. Discussion

By assuming the pulse is gaussian-shaped in time and the beam is also gaussian-shaped in space, a TPA coefficient value β can be estimated as

$$\beta \approx -\frac{\Delta T}{T}\sqrt{\frac{\pi}{2\ln 2}}\frac{\alpha}{I_e}.$$

α is the linear absorption coefficient and I_e is the effective intensity of the pulse. The TPA coefficient values with different central wavelengths are showed in Fig. 2. For example, at a wavelength of ~362 nm, a TPA coefficient value of 2900 cm/GW can be obtained, which is much higher than the already high below-bandgap TPA coefficient value of 15 cm/GW at 400 nm [2].

To explain the enhancement of the TPA coefficient values near exciton resonance frequency, we considered a simplified three levels system. The system is initially in the ground state $|g\rangle$ with no excitons, and the final state $|f\rangle$ is the elctron-hole pair state. For the intermediate states, we assume that the contribution of the bandtail states is small and only take the exciton state $|n\rangle$ into consideration. The relation between the TPA coefficient and the photoexcitation frequency could be derived as

$$\beta \propto \frac{1}{\left(\omega - \omega_{ng}\right)^2 + \Gamma_{ng}^2} \, ,$$

where ω_{ng} is the exciton resonance frequency and Γ_{ng} is the dephasing term. The fitting results are also shown in fig. 2. An exciton dephasing time ~ 66 fs can be obtained. If we use a Lorentzian function convolved by the laser spectrum, the dephasing time is ~ 155 fs. This results show that the room temperature dephasing time of excitons is close to the exciton ionization time of ~ 200 fs.

5. Summary

We have observed the enhancement of nonlinear absorption near exciton resonance in GaN thin films using femtosecond pump-probe techniques. The TPA coefficient values of GaN near exciton resonance are at least 3000 cm/GW, and the dephasing time is 60-160 fs at room temperature. Detailed analysis and discussion will be presented in the talk.

6. References

[1] Y. –C. Huang, et al., "Femtosecond dynamics of exciton bleaching in bulk GaN at room temperature," Appl. Phys. Lett. **81**, 85 (2002).
[2] C. –K. Sun, et al., "Two-photon absorption study of GaN," Appl. Phys. Lett. **76**, 439 (2000).
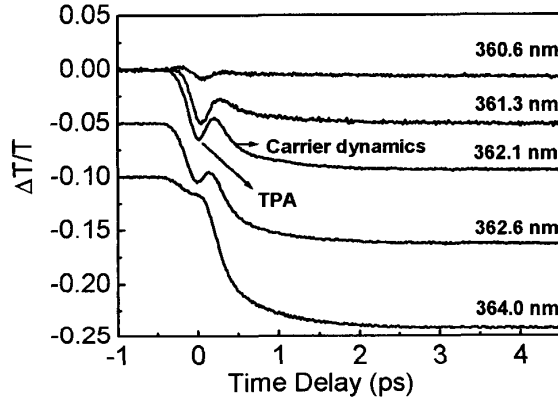

Fig. 1. Measured probe transmission transient response of the nominally undoped GaN thin film as a function of probe delay at different central wavelengths near the exciton photoexcitation.
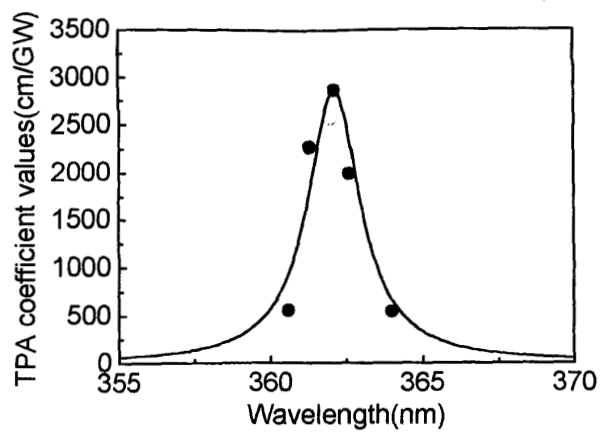
Fig. 2. The solid circles are the TPA coefficients calculated from the measured traces near the exciton resonance, and the solid line is the Lorentzian fitting curve.