Resonantly enhanced femtosecond second-harmonic generation and nonlinear luminescence in GaN film grown on sapphire

Hui Yang, S. J. Xu, Q. Li, and Jie Zhang

Citation: [Applied Physics Letters](#) **88**, 161113 (2006); doi: 10.1063/1.2197310

View online: <http://dx.doi.org/10.1063/1.2197310>

View Table of Contents: <http://scitation.aip.org/content/aip/journal/apl/88/16?ver=pdfcov>

Published by the [AIP Publishing](#)

Articles you may be interested in

[Enhanced second- and third-harmonic generation and induced photoluminescence in a two-dimensional GaN photonic crystal](#)

Appl. Phys. Lett. **87**, 101106 (2005); 10.1063/1.2037849

[Enhancement of visible second-harmonic generation in epitaxial GaN-based two-dimensional photonic crystal structures](#)

Appl. Phys. Lett. **84**, 1245 (2004); 10.1063/1.1649800

[Second-harmonic generation in periodically poled GaN](#)

Appl. Phys. Lett. **83**, 1077 (2003); 10.1063/1.1599044

[Optical and structural studies of hydride vapor phase epitaxy grown GaN](#)

J. Vac. Sci. Technol. A **21**, 701 (2003); 10.1116/1.1568346

[Phase-matched optical second-harmonic generation in GaN and AlN slab waveguides](#)

J. Appl. Phys. **85**, 2497 (1999); 10.1063/1.369611

The image shows the cover of an Applied Physics Reviews journal. It features a blue and orange color scheme with a background of blue spheres and a bright light source. The text 'NEW Special Topic Sections' is prominently displayed in white. Below this, it says 'NOW ONLINE' in yellow, followed by 'Lithium Niobate Properties and Applications: Reviews of Emerging Trends' in white. The AIP Applied Physics Reviews logo is in the bottom right corner. On the left, there is a small image of the journal cover showing a diagram of a device structure.

NEW Special Topic Sections

NOW ONLINE
Lithium Niobate Properties and Applications:
Reviews of Emerging Trends

AIP | Applied Physics
Reviews

Resonantly enhanced femtosecond second-harmonic generation and nonlinear luminescence in GaN film grown on sapphire

Hui Yang, S. J. Xu,^{a)} and Q. Li

Department of Physics and HKU-CAS Joint Laboratory on New Materials, The University of Hong Kong, Pokfulam Road, Hong Kong, China

Jie Zhang

Laboratory of Optical Physics, Institute of Physics, Chinese Academy of Sciences, Beijing 100080, China

(Received 6 November 2005; accepted 23 March 2006; published online 19 April 2006)

At room temperature, by using a tunable broadband femtosecond laser as excitation source we observed second-harmonic generation (SHG) and nonlinear photoluminescence (NPL) in GaN film grown on sapphire simultaneously or individually. In addition to the observation of the resonance effect of the nonlinear response when the SHG is tuned to coincide with the near-band-edge emission, we carefully measured dependence of the SHG and NPL signals on polarization of the excitation light. The results reveal that the reabsorption of the SHG photons with energies higher than the fundamental gap of GaN significantly contributes to generation of the efficient NPL signal. © 2006 American Institute of Physics. [DOI: 10.1063/1.2197310]

Since the demonstration of high-bright short-wavelength light emitting diodes based on III-V nitrides in the beginning of 1990s,¹ III-V nitrides have attracted a great deal of interest.^{2–4} Without any doubt, binary GaN occupies a central position within the whole family of III-V nitrides. Usually, GaN epilayers are grown on foreign substrates such as sapphire because of lacking of large size native GaN substrate. In the past decade, the linear optical properties of GaN have been studied extensively, for example, single-ultraviolet (UV)-photon excited photoluminescence (PL) of GaN.⁵ It has been well established that under excitation of single UV photon, i.e., the excitation photon energy exceeds the fundamental band gap of GaN, the PL spectrum of GaN heteroepilayers usually contains two characteristic emission bands: one is sharp UV band at 3.4 eV due to annihilation of band-edge excitons,^{6,7} and the other is the broad yellow band centered at 2.2 eV which is believed to be due to defect complexes.^{8–10}

Very recently, nonlinear optical properties of GaN have attracted an increasing interest.^{11–19} For example, Kim *et al.* observed multiphoton excited PL in GaN with a tunable picosecond laser.¹² After the theoretical work done by Hughes *et al.*,¹³ Angerer *et al.* experimentally investigated the ultrafast second-harmonic generation (SHG) signal from GaN thin films grown on sapphire.¹⁴ Sun *et al.* reported scanning second-harmonic and third-harmonic generation microscopies of GaN.^{15,16} On the other hand, Schmidt *et al.* demonstrated the large interband second-order susceptibilities in InGaN/GaN quantum wells.²⁰ They not only determined the second-order susceptibility leading to the SHG in the InGaN/GaN quantum wells but also observed a resonance effect of the nonlinear response when the SHG peak is tuned to approach the fixed nonlinear photoluminescence (NPL) peak. A further investigation of such a resonance effect as well as the relationship between the SHG and the NPL signals in III-V nitrides, in particular, in GaN, is thus interesting and desirable. Here we attempt to do such a study

in a high-quality GaN film grown on sapphire. The experimental results reveal the interrelation between the SHG and NPL signals when the SHG photon energies are higher than the band gap of GaN.

The GaN film studied in the work was grown on a *c*-plane sapphire substrate with metalorganic vapor phase epitaxy method. The thickness of the film is about 2.88 μm . The optical quality of the film is very high, which was indicated by previous PL studies already reported elsewhere.²¹ The excitation laser is a tunable mode-locked Ti:sapphire femtosecond (fs) laser with pulse width of ~ 100 fs (Spectra-Physics Tsunami system). The laser wavelength can be tuned from 700 to 930 nm. The laser pulse was focused onto the surface of sample with a $f=10$ cm lens and the incident angle is about 60° . The luminescence and SHG signals from the sample were collected and collimated by a lens with a focus length of $f=3.5$ cm. After blocking the excitation laser with an optical filter, the PL and SHG were guided into an Acton spectrometer by a lens with $f=20$ cm. The PL and SHG signals dispersed by the spectrometer were finally detected by a Hamamatsu R928 photomultiplier tube and amplified by a Stanford Research digital lock-in amplifier.

Figure 1 shows the measured luminescence spectra of

FIG. 1. (Color online) Measured room-temperature NPL spectra of the sample under excitation of 712 nm fs laser with two polarization states. The inset shows the quadratic dependence of the NPL intensity on the excitation power.

^{a)} Author to whom correspondence should be addressed; electronic mail: sjxu@hkucc.hku.hk

FIG. 2. (Color online) Measured room-temperature NPL/SHG spectra of the sample. The vertical dashed line marks the location of the band-edge emission. The inset shows the peak intensity of the SHG signal vs the two-photon energy of the excitation light. A clear resonant enhancement of the SHG can be seen when the two-photon energy of the excitation light is approaching the band gap of GaN.

the sample at room temperature and under excitation of 712 nm fs laser with two polarization states. For 712 nm laser, the corresponding two-photon energy is 3.483 eV which is well above the band gap of GaN. Being similar to the single-ultraviolet-photon excited luminescence spectrum, the NPL spectrum consists of a near-band-edge emission peak at 3.379 eV (~ 367 nm) and a weak broad yellow luminescence peak centered at 2.3 eV (~ 539 nm). The band-edge emission peak exhibits a dependence on the polarization of the incident fs laser whereas the broad yellow peak does not depend on the polarization of the excitation light. As shown later, the polarization dependence of the band-edge emission is due to reabsorption of the SHG photons of the excitation laser. It is also due to the strong reabsorption of the SHG photons that the detected SHG signal is very weak at the excitation wavelength of 712 nm. In order to verify the nonlinearity of the observed NPL signal, excitation-power dependence of the band-edge emission peak was measured when the wavelength of excitation light is fixed at 712 nm. The inset of Fig. 1 plots the square root of the band-edge emission intensity against the excitation power. The solid squares represent the experimental data while the solid line is the least-square fitting result. Indeed, the band-edge emission intensity exhibits a quadratic dependence on the excitation power, which indicates the nonlinearity of the NPL process. It is important to address that both direct two-photon absorption and reabsorption of the SHG photons contribute to the observed luminescence signal (the band-edge transition) when the energies of incident photons exceed the half band gap of GaN. Since the SHG itself is a second-order nonlinear optical process, it is not surprising that the total intensity of the band-edge emission depends on square of the excitation intensity.

Keeping the power density of the excitation laser at 0.4 GW/cm^2 , we tuned the laser wavelength in a range of 710– ~ 750 nm so as to observe evolution of the band-edge emission and SHG signal from the sample. For this wavelength range, corresponding laser photon energy varies in the range $\frac{1}{3}E_g < \hbar\omega < E_g$. The measured NPL/SHG spectra were shown in Fig. 2. Looking at the figure, a resonance effect of the nonlinear optical response is immediately observed as shown in the inset of the figure. That is, when the two-photon energy of the excitation laser is swept over the band

FIG. 3. Measured dependence of the SHG and NPL signals on the polarization of the excitation lights with wavelengths of 753 and 712 nm, respectively. The solid line is the theoretical polarization dependence of the SHG signal.

gap of GaN the SHG signal rapidly increases and then decreases. Simultaneously, the intensity of the band-edge emission whose position is marked by vertical dashed line strongly decreases as the energies of the excitation photons decreases. The strong increase of the SHG signal and the correspondingly rapid decrease of the NPL signal indicate that a close relationship between the SHG and NPL signals likely exists. This makes us to believe that efficient reabsorption of the SHG photons inside the sample could occur when their energies are higher than the GaN band gap and then make important contribution to the generation of the band-edge luminescence. A more clear evidence (polarization dependence measurement) supporting this idea will be given later. We now turn to talk about another mechanism leading to the nonlinear luminescence. It is the direct two-photon absorption under the condition of the excitation photon energies higher than $\frac{1}{2}E_g \approx 1.7 \text{ eV}$ (corresponding ~ 729 nm). The two-photon absorption coefficients of GaN have been determined both experimentally¹⁵ and theoretically.¹⁷ For example, a two-photon absorption coefficient of $3 \pm 1.5 \text{ cm/GW}$ at 720 nm wavelength was determined by Sun *et al.* using transient transmission experiments.¹⁵ Recently, a huge excitonic enhancement of two-photon absorption near the exciton-transition energy was even observed by Lin *et al.* in the GaN system.²² From the results obtained in the present work, the direct two-photon absorption is still a dominant mechanism producing the NPL, in particular, when the excitation photon energies are much higher than the half band gap of GaN. However, when the excitation photon energies are tuned to be below $\frac{1}{2}E_g$ the nonlinear absorption mechanism will change from the two-photon absorption to the three-photon absorption. As one more photon is necessitated to excite electrons across the band gap the absorption coefficient will strongly decrease according to E_g^{4N-5} .²³ Here N is photon number involved in an optical transition. The strong decrease of the nonlinear absorption coefficient certainly results in a significant reduction of the NPL intensity.¹³

To further investigate the interrelation between the SHG and the NPL in GaN, we measured dependence of the luminescence signal centered at 367 nm and the SHG signal on the polarization of 712 and 753 nm excitation lights, respectively. The experimental results are depicted in Fig. 3 with symbols. As expected, the dependence of the SHG on the polarization of the excitation light clearly exhibits a rotational symmetry. For generation of second-harmonic light in

a thin film, its intensity is proportional to the square of the second-order polarization induced by electric field of the incident fundamental light,²⁴

$$I_{\text{SHG}} \propto [P^{(2)}(\omega = 2\omega_0)]^2, \quad (1)$$

where $P^{(2)}(\omega = 2\omega_0) = \chi_{ijk}^{(2)} E_j E_k$, $\chi_{ijk}^{(2)}$ is the second-order susceptibility of the film. Because wurtzite GaN possesses C_{6v} (or $6mm$) point group symmetry, the nonzero second-order susceptibility elements are given as follows:¹⁴ $\chi_{zzz}^{(2)} = -3.28 \times 10^{-8}$ esu, $\chi_{xxz}^{(2)} = \chi_{yzy}^{(2)} = \chi_{xzx}^{(2)} = \chi_{yyz}^{(2)} = 1.64 \times 10^{-8}$ esu, and $\chi_{zxx}^{(2)} = \chi_{zyy}^{(2)} = 2.4 \times 10^{-8}$ esu. Considering that the incident angle of the excitation light is about 60° in the present study, we can derive the second-order susceptibility as a function of the linear polarization angle ϕ of the incident light,

$$P^{(2)}(\omega = 2\omega_0) \propto (1.05 \cos^2 \phi + 2.4 \sin^2 \phi). \quad (2)$$

Using Eqs. (1) and (2) we calculated the SHG intensity versus the laser polarization angle as shown in Fig. 3. It is seen that satisfactorily good agreement between experiment and theory is achieved. Note that the NPL also exhibits a similar but weaker polarization dependence. This is a strong evidence indicating that the strong reabsorption of the SHG photons significantly contributes to the observed luminescence. Therefore, the observed nonlinear luminescence originates from the two kinds of optical transitions: the direct two-photon absorption and reabsorption of SHG photons whose energies are higher than the band gap of GaN.

In conclusion, we have studied the second-harmonic generation and nonlinear luminescence from the GaN epilayer at room temperature using an intense tunable femtosecond laser as the excitation source. The resonant enhancement of the ultrafast SHG was demonstrated when the two-photon energy of the excitation light was tuned to approach the band gap of GaN. We also show that both the direct two-photon absorption and the reabsorption of the SHG photons are responsible for the efficient luminescence signal when the SHG photon energies are higher than the GaN band gap.

The work was supported by Competitive Earmarked Research Grant, Research Grants Council of HKSAR under

Contract No. HKU-7036/03P and Research Grants of University of Hong Kong under Contract No. 10205780.

¹H. Nakamura, M. Senoh, and T. Mukai, Appl. Phys. Lett. **62**, 2390 (1993).

²H. Morkoc and S. N. Mohammad, Science **267**, 51 (1995).

³I. Akasaki, S. Sota, H. Sakai, T. Tanaka, M. Koike, and H. Amano, Electron. Lett. **32**, 1105 (1996).

⁴F. A. Ponce and D. P. Bour, Nature (London) **386**, 351 (1997).

⁵See, for example, P. P. Paskov, T. Paskova, P. O. Holtz, and B. Monemar, Phys. Rev. B **70**, 035210 (2004), and references therein.

⁶W. Shan, T. J. Schmidt, X. H. Yang, S. J. Hwang, J. J. Song, and B. Goldberg, Appl. Phys. Lett. **66**, 985 (1995).

⁷F. A. Ponce, D. P. Bour, W. Götz, and P. J. Wright, Appl. Phys. Lett. **68**, 57 (1996).

⁸T. Ogino and M. Aoki, Jpn. J. Appl. Phys. **19**, 2395 (1980).

⁹P. Perlín, T. Suski, H. Teisseyre, M. Leszczynski, I. Grzegory, J. Jun, S. Porowski, P. Boguslawski, J. Bernholc, J. C. Chervin, A. Polian, and T. D. Moustakas, Phys. Rev. Lett. **75**, 296 (1995).

¹⁰S. J. Xu, G. Li, S. J. Chua, X. C. Wang, and W. Wang, Appl. Phys. Lett. **72**, 2451 (1998).

¹¹J. Miragliotta and D. K. Wickenden, Phys. Rev. B **53**, 1388 (1996).

¹²D. Kim, I. H. Libon, C. Veolkmann, Y. R. Shen, and V. Petrova-Koch, Phys. Rev. B **55**, R4907 (1997).

¹³J. L. P. Hughes, Y. Wang, and J. E. Sipe, Phys. Rev. B **55**, 13630 (1997).

¹⁴W. E. Angerer, N. Yang, A. G. Yodh, M. A. Khan, and C. J. Sun, Phys. Rev. B **59**, 2932 (1999).

¹⁵C.-K. Sun, J.-C. Liang, J.-C. Wang, F.-J. Kao, S. Keller, M. P. Mack, U. Mishra, and S. P. DenBaars, Appl. Phys. Lett. **76**, 439 (2000).

¹⁶C.-K. Sun, S.-W. Chu, S.-P. Tai, S. Keller, U. K. Mishra, and S. P. DenBaars, Appl. Phys. Lett. **77**, 2331 (2000); C.-K. Sun, S.-W. Chu, S.-P. Tai, S. Keller, A. Abare, U. K. Mishra, and S. P. DenBaars, Scanning **23**, 182 (2001).

¹⁷S. Krishnamurthy, K. Nashold, and A. Sher, Appl. Phys. Lett. **77**, 355 (2000).

¹⁸V. I. Gavrilenko and R. Q. Wu, Phys. Rev. B **65**, 035405 (2001).

¹⁹Y. Toda, T. Matsubara, R. Morita, M. Yamashita, K. Hoshino, T. Someya, and Y. Arakawa, Appl. Phys. Lett. **82**, 4714 (2003).

²⁰H. Schmidt, A. C. Abare, J. E. Bowers, S. P. DenBaars, and A. Imamoğlu, Appl. Phys. Lett. **75**, 3611 (1999).

²¹S. J. Xu, W. Liu, and M. F. Li, Appl. Phys. Lett. **77**, 3376 (2000).

²²K.-H. Lin, G.-W. Chern, Y.-C. Huang, S. Keller, S. P. DenBaars, and C.-K. Sun, Appl. Phys. Lett. **83**, 3087 (2003).

²³B. S. Wherrett, J. Opt. Soc. Am. B **1**, 67 (1984).

²⁴Y. R. Shen, *The Principles of Nonlinear Optics* (Academic, New York, 1993).