# Observation of second harmonic emission and three-photon fluorescence from Gallium-Nitride

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## Abstract

In this study, optical emission from the undoped GaN epilayer sample was characterized using confocal micro-spectroscopy. A femtosecond laser was used to excite the sample. As the laser wavelength tuned from 762-796nm, the sample emitted a corresponding second harmonic signal from 381-398nm with a quadratic power dependence. A strong signal at 367nm resulted from a three-photon process with cubic power dependence was simultaneously observed for the first time.

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### Summary

GaN is a wide band-gap semiconductor (Eg=3.4eV at 300K) of important technological applications. Recently there has also been a growing interest in the studies of high-order optical nonlinearity in GaN[1,2]. Two- and three- photon conductivity and photocurrent using optical beam induced current were also reported[3]. On the other hand, it is well known that, the back ground noise for multi-photon excitation can be greatly reduced as a result of the limited excitation volume in confocal microscopy. In this work, we report for the first simultaneous observation of second harmonic (SH) emission and three-photon fluorescence from a GaN epilayer excited by a tunable Ti-sapphire femtosecond laser using a confocal micro-spectroscopic system.

The sample studied is an undoped GaN epilayer grown by hybrid vapor phase epitaxy on a c-plane sapphire substrate. The thickness of the layer on the sample is about 14.9 μm. The structure, surface morphology, and optical properties of the GaN sample studied have been reported previously[4]. These measurements indicate the GaN epilayer has good optical and structural quality. A typical linear photoluminescence (PL) spectrum of the sample excited by a 325nm He-Cd laser at room temperature is shown in Fig. 1(a). The sample emits a sharp peak centered at 367nm, corresponding to near band-edge emission with a weak broad band emission from 460nm to 700nm. As a reference, the PL spectrum of a blank sapphire substrate is also shown in the dash line of Fig. 1(a). This indicates that the luminescence shown in Fig. 1(a) is from the GaN epi-layer. A typical emission spectrum of the GaN sample excited by the femtosecond Ti-sapphire laser at main wavelength at 796nm is shown as the solid line of Fig. 1(b). Two main emission peaks at 367nm and 398nm and a little side peak around 390nm were observed in contrast to the single dominant PL emission peak at 367nm shown in Fig. 1(a). The small side peak near main peak of 398nm is attributed to an artifact due to a weak emission feature at 780nm in the main pumping wavelength of 796nm. The power dependence of these two emission peaks at pumping wavelength of 796nm is shown in Fig. 2. A crossover behavior of spectral luminescent intensity between 367nm line and 398nm line was observed. The short-dash line is a power-fitting curve for 398nm emission data (solid circles), while the dash line is that of 367nm luminescent data (open triangles). The 398nm peak shows quadratic power dependence, further indicating the second harmonic emission nature of this emission wavelength. Since the second order optical nonlinearity has reported for GaN wave-guide and thin film, the SH emission observed here is possible. As for the 367nm peak, a superlinear power dependence is observed. At low pumping power, the 367nm signal does not rise as fast as the 398nm peak. However the pumping intensity increases, the intensity of 367nm signal increases very rapidly, and exceeds SH signal at the same power with near cubic power dependence at high power level. Since at the pumping wavelength of 796nm, the two-photon process is not energetic enough ($E_{two\text{-}photon}$=3.12eV<$E_g$) to excite fluorescence emission of 367nm. This fluorescence emission peak at 367nm is thus as the result of a three-photon process. This is the first time, to our knowledge, simultaneous SH emission and three-photon fluorescence from GaN was observed.

In summary, optical emission from the undoped HVPE grown GaN epilayer sample was characterized using a confocal spectroscopy system. The sample was excited by a tunable femtosecond laser at 100fs. As he laser wavelength tuned from 762-796nm, the sample emits signals at second harmonic wavelength from 381-398nm with a quadratic power dependence. At the same time, a three-photon fluorescence signal with peak wavelength at

367nm was also observed with a cubic power dependence at high pimping power. The use of micro-spectroscopic configuration and tunable short pulse long wavelength laser excitation could be a powerful technique for observing multiple photon nonlinear optical phenomena in wide bandgap semiconductor materials.

## References


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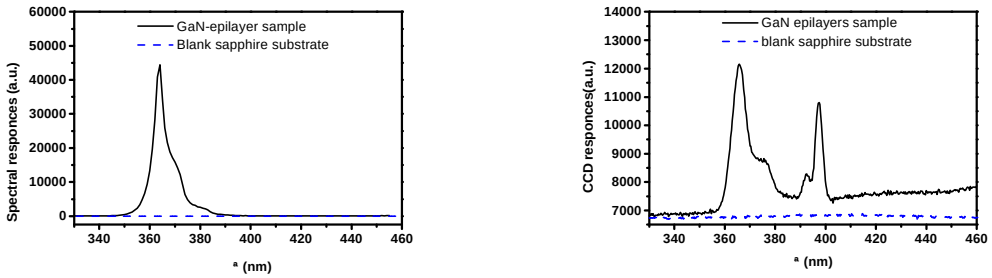


Fig. 1:GaN epilayer emission spectra: (a) single-photon excited the PL for the GaN epilayers sample(solid line) and the blank sapphire substrate reference line(dash line) and (b) The femtosecond laser excited emission spectrum(solid line) and the blank sapphire substrate reference line(dash line)

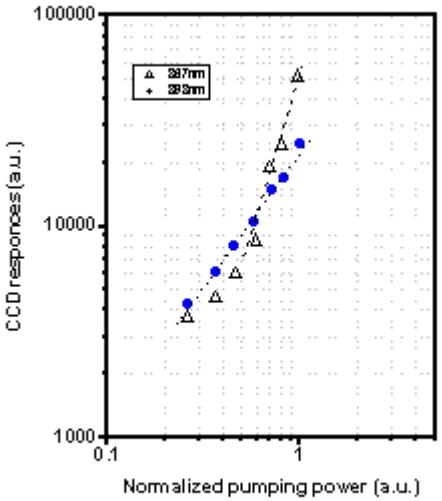


Fig. 2: Dependence of the two emission peaks intensity on pumping power at 796nm