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Two-photon excited photoluminescence in InGaN multi-quantum-wells structures

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***Abstract*— In this work, we report on the two-photon absorption induced luminescence of InGaN multiple quantum wells grown on sapphire. When the sample was excited by femtosecond near-infrared laser pulses at room temperature, an intense luminescence signal peaked at ∼415 nm from the sample was observed, which indicates strong nonlinear optical effect in InGaN quantum well structures. The interferometric autocorrelated luminescence traces were recorded to verify the second order nonlinearity of the luminescence. In addition, the strong second harmonic generation signal of the excitation laser was also observed. The mechanism of the two-photon excited photoluminescence in InGaN quantum wells was discussed.**

I. Introduction

As the active layer in GaN based light-emitting diodes [1], [2], linear optical properties of InGaN have been extensively investigated in recent years. However, very few studies of its nonlinear optical properties, especially two-photon absorption (TPA) induced luminescence (TPAL), have been reported so far. In the past several years, the TPA process in GaN was investigated [3]–[7] and the TPA coefficient of GaN was found to be as large as 1500 cm/GW [4]. For InGaN, a larger TPA effect is theoretically predicted [7]. In this work, we investigate the two-photon excited photoluminescence of InGaN multiple quantum wells (MQWs) grown on sapphire when the sample is excited by femtosecond (fs) near infrared laser. It is found that there exists strong nonlinear optical effect in InGaN MQWs. The interferometric autocorrelated luminescence trace measurements clearly demonstrate the nonlinearity of the TPAL process. We also observed strong second harmonic generation (SHG) signal from the sample excited by infrared fs laser. These results show that InGaN/GaN MQWs structures have potential applications in nonlinear optics.

II. Experiments and Discussions

The InGaN MQWs sample used in the present study was grown on sapphire by metalorganic vapor phase epitaxy. A 30 nm low-temperature GaN buffer was firstly grown, followed by 1 μm GaN epilayer. Ten periods of 3 nm $In_{0.13}Ga_{0.87}N$/5nm $In_{0.03}Ga_{0.97}N$ QW structures were then grown, which was capped by a 20 nm GaN layer. Near infrared fs laser pulses with 80-100 pulse width from a Ti:sapphire oscillator pumped by solid-state laser diode array were employed to excite the sample at room temperature. The wavelengths of the laser pulses can be tuned from 710 nm to 950 nm. The luminescence signal from the sample was dispersed and detected with conventional photoluminescence spectrometer. A standard phase lock-in amplification technique was used to improve the signal/noise ratio. The interferometric autocorrelated luminescence traces were recorded using an experimental arrangement shown in Fig. 1.

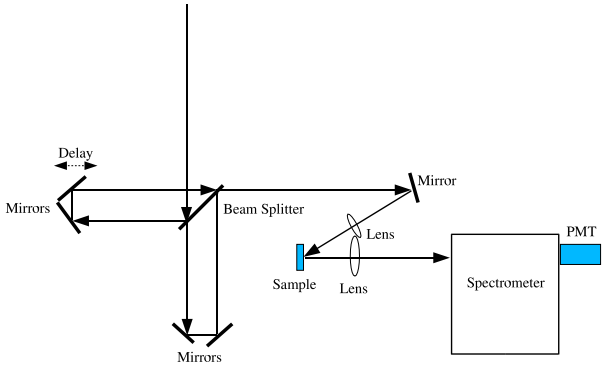

Fig. 1. Experimental arrangement for the interferometric autocorrelated luminescence trace.

Figure 2 shows the room-temperature TPAL spectra from the sample excited by 710 nm (1.75 eV) and 760 nm (1.63 eV), respectively. The excitation powers of the two excitation lines were the same. The spectra are dominated by the luminescence peak at $\sim$ 415 nm (2.99 eV), which is much higher than energies of the excitation photons. In the case of 760 nm excitation, a shoulder appears at the lower energy side of the main peak. From Fig. 2 it can be seen that the intensity of the TPAL signal increases when the wavelengths of the excitation lines become shorter. This indicates that the TPA coefficient increases when the excitation photon energy increases, which is in accordance with the relation between the TPA coefficient β of a direct band gap semiconductor and the photon energy $\hbar\omega$, proposed by Sheik-Bahae *et. al.* [8]:

$$\beta(\hbar\omega) \propto F(\hbar\omega/E_g),$$

where E_g is the band gap energy of the semiconductor and $F(x) = (2x-1)^{3/2}/(2x)^5$. According to this relation, when

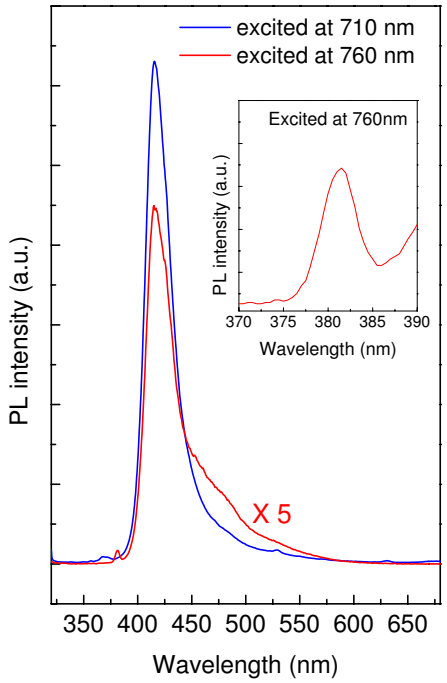

Fig. 2. Two-photon absorption induced luminescence spectra. The spectra excited at 760 nm was multiplied by a factor of 5. The inset shows the SHG signal of the 760 nm laser line.

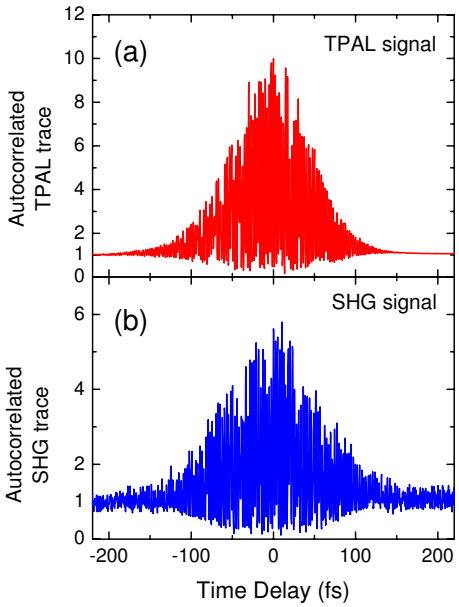

Fig. 3. Autocorrelated luminescence traces recorded by using a pump-probe technique.

$\hbar\omega/E_g \approx 0.71$, the TPA coefficient $\beta(\hbar\omega)$ reaches its maximum value. Taking E_g=2.99 eV, since (1.63 eV/E_g)<(1.75 eV/E_g)<0.71, one can conclude that the magnitude of β at 1.75 eV (710 nm) is larger than that at 1.63 eV (760 nm). Our experimental results are in agreement with such theoretical estimation.

Figure 3(a) shows the interferometric autocorrelation traces of the TPAL signal at 415 nm from the sample, when it was excited by the 760 nm laser line. The Gaussian-function fitting to the recorded trace in Fig. 3(a) shows that the ratio of luminescence signal at zero delay and that at delay time far from the zero point is ∼8.5. This ratio is slightly larger than the ideal value of 8 for the second-order nonlinear process [9].

Besides the TPAL signal, the SHG signal of the excitation laser was also observed in our sample. The inset in Fig. 2 shows the SHG signal of the 760 nm excitation line. The SHG signal intensity is found to be dependent on the wavelengths of the excitation lines. The recorded interferometric autocorrelation trace of the SHG of the 760 nm line is shown in Fig. 3(b). We note that the ratio of the SHG signal at zero time delay and that at the delay time far from the zero delay point is ∼6, which is less than 8. The reason causing the smaller the ratio is not clear at moment. Further investigations are needed to make it clear.

III. Conclusion

Nonlinear luminescence induced by two-photon absorption in InGaN/GaN MQWs structure was investigated. When the sample was illuminated by near-infrared pulsed laser with about 100 fs pulse width at room temperature, the intense luminescence signal was observed. The interferometric autocorrelation traces of the luminescence were recorded using a colinear autocorrelation technique and the ratio of the signal at the zero delay and that at the delay time far from the zero delay point is 8.5, which is almost identical with the ideal value 8 for second-order nonlinear process. These results show that the InGaN/GaN MQWS structure has strong optical nonlinearity.

Acknowledgment

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