Two-Pulse Photoluminescence Correlation Technique for Studying Ultrafast Carrier Dynamics in Deep-UV Few Monolayer Thick Nitride Quantum Wells

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Abstract: We a present a new two-pulse photoluminescence correlation technique and use it to study ultrafast dynamics of photoexcited carriers in few monolayer thick deep-UV GaN/AlN quantum wells with picosecond time resolution. Our results show that our technique can be used to study carrier dynamics in nanostructures using photoluminescence in a very simple way.

OCIS codes: (260.712) Ultrafast Phenomena; (320.7150) Ultrafast spectroscopy; (040.4200) Multiple quantum well

1. Introduction

The III-nitride semiconductor nanostructures have been shown to be the most suitable materials for optoelectronic devices in this wavelength range [1-3]. One of the lowest pump threshold Deep-UV lasers has been realized using few monolayer thick GaN quantum wells and AlN barriers [1]. Optoelectronic characterization of Deep-UV nitride materials is therefore important. Time-resolved photoluminescence techniques have proven to be the most useful and reliable methods in characterizing the ultrafast dynamics associated with the relaxation and recombination of photoexcited carriers in semiconductors. Time-correlated photon counting techniques have been used to characterize deep-UV nitride nanostructures [1,4]. Time resolution in this technique is typically limited to a few hundred picoseconds given the efficiency-bandwidth product limitations of most fast photon detectors. Although ultrafast nonlinear up-conversion of the photoluminescence via Ker gating can provide sub-picosecond time resolution, the use of this technique in the deep-UV range runs into difficulties because of the lack of commonly available suitable optical materials and devices in the up-converted sub-200 nm wavelength range [5].

Fig.1: A schematic of the two-pulse photoluminescence correlation technique.

2. Two-Pulse Photoluminescence Correlation (TPPC) Technique

In this paper, we present a new two-pulse photoluminescence correlation (TPPC) technique for studying ultrafast dynamics of photoexcited carriers in materials via photoluminescence measurement, and use this technique to study the carrier dynamics in few monolayer thick, deep-UV wavelength, GaN/AlN quantum wells. In contrast to other techniques (mentioned above), TPPC requires only a visible wavelength ultrafast pulse source and a slow photon counter and provides sub-picosecond time resolution for studying ultrafast photoluminescence. The technique is depicted in Fig.1. The pump pulses consist of two identical time-delayed pulses derived from a single femtosecond Ti:Sapphire system.

Fig.2: (LEFT) TEM of 2 monolayer GaN/AlN MQW structure. (RIGHT) PL depicting optical transition at ~250 nm wavelength.

These pulses excite photocarriers in a GaN/AlN MQW sample via two-photon absorption. The resulting photoluminescence (PL) and the DC photocurrent $I(\tau)$ are measured using a relatively slow photon detector as a function of the time delay τ between the pulses. The PL correlation signal, captured in the current $I(\tau)$, contains information on the carrier dynamics. The dynamics captured by DC photocurrent $I(\tau)$ can be understood as follows. If the carrier recombination dynamics were governed by processes completely linear in the photoexcited carrier density then $I(\tau)$ would be independent of the time delay τ. Because carrier dynamics are almost always described by processes nonlinear in the carrier density, $I(\tau)$ is a strong function of the
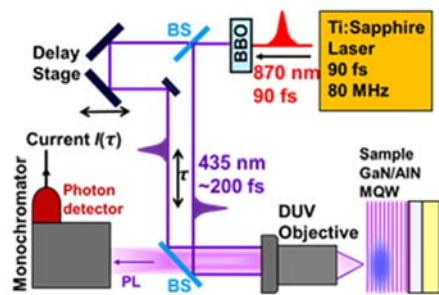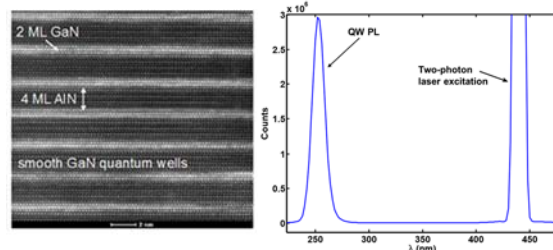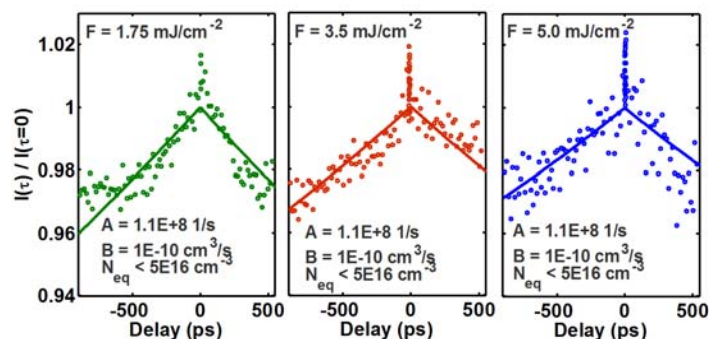
Fig.3: (Dots) The measured $I(\tau)$ (normalized) is plotted for different values of the pump pulse fluences (F) as a function of the delay τ. (Solid) Model fits using the indicated parameter values. Same parameter values fit all the data.

time delay τ. For example, if the time separation of the two pump pulses is much longer than the photoexcited carrier density decay times, the total time-integrated PL would be twice the PL generated by a single pulse. On the other hand if the two pump pulses arrive close to each other in time, the larger resulting carrier density would result in faster recombination times (due to nonlinear mechanisms) and the resulting integrated PL would be larger (if the dominant nonlinear mechanism is radiative recombination) compared to the case when the time separation between the pulses was very long. By comparing the measured values of $I(\tau)$ vs τ for different values of the pump pulse energies, all the rate constants for the carrier recombination processes can be reliably extracted.

3. Measurements and Results

The GaN/AlN quantum well samples used in this work were grown by MBE on AlN templates grown on Sapphire substrates. Samples consisted of 1-3 monolayer thick GaN quantum wells sandwiched by thicker AlN barriers (Fig.2). In Fig.3, the measured $I(\tau)$ is plotted for different values of the pump pulse fluence (F) as a function of the time delay τ between the pump pulses. Note that ideally $I(\tau) = I(-\tau)$. The slight asymmetry in the data is due to the non-ideal linear translation stage. The data can be modeled using the following carrier recombination model for the photoexcited carriers appropriate for the ultrathin GaN quantum wells,

$$dN/dt = -A(N - N_{eq}) - B(NP - N_{eq}P_{eq}) \qquad dP/dt = -A(P - P_{eq}) - B(NP - N_{eq}P_{eq})$$

Here, N_{eq} and P_{eq} are the equilibrium electron and hole densities in our mildly n-doped quantum well samples. A is the linear recombination rate constant and models defect-assisted exciton recombination via carrier capture. B is the non-linear recombination rate constant and equals the sum of B_r (spontaneous emission rate constant) and B_{nr}. B_{nr} is the bimolecular non-radiative recombination rate constant and models defect-assisted recombination involving carrier capture via exciton-exciton or exciton-carrier Auger-like collisions. The recombination processes going as the cube of the carrier density (e.g. direct or indirect interband Auger recombination) can be ignored [6]. It will be shown in the talk that the values of the normalized slope $dI(\tau)/d\tau\big|_{\tau=0}/I(\tau=0)$ and the scaling of $I(\tau=0)$ (normalized to its value at any one pump fluence value) with the pump fluence can be used to extract the values of the parameters A and B very reliably. The large measured values of B (compared to theory estimates of B_r) indicates that non-radiative recombination dominates over radiative
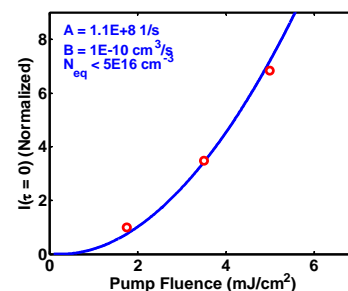

Fig.4: (Dots) Scaling of $I(\tau)$ (normalized to its value for the smallest pump fluence of 1.75 mJ/sq-cm) is plotted as a function of the pump fluence. (Solid) The fit from the model.

recombination in our samples. The talk will discuss in detail the physics and the mechanisms associated with non-radiative carrier recombination in ultrathin deep-UV Nitride nanostructures.

4. References

[1] J. Sellés et al., "Deep-UV nitride-on-silicon microdisk lasers," Scientific Reports, 6, 21650 (2016).
[2] Z. Bryan et al., "High internal quantum efficiency in AlGaN multiple quantum wells grown on bulk AlN substrates," Appl. Phys. Lett. 106, 142107 (2015).
[3] Z. Bryan, "Strain dependence on polarization properties of AlGaN and AlGaN-based ultraviolet lasers grown on AlN substrates," Appl. Phys. Lett. 106, 232101 (2015).
[4] Y.-H. Cho, "Comparison of carrier dynamics in GaN quantum dots and GaN quantum wells embedded in low-Al-content AlGaN waveguides," Appl. Phys. Lett. 89, 251914 (2006).
[5] J. C. Blake et al., "Ultraviolet femtosecond Kerr-gated wide-field fluorescence microscopy," Optics Letters, 41, Issue 11, 2462 (2016).
[6] E. Kioupakis, "First-principles calculations of indirect Auger recombination in nitride semiconductors," Phys. Rev. B, 92, 035207 (2015).