

# Ultrafast differential transmission spectroscopy of excitonic transitions in InGaN/GaN multiple quantum wells

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Room-temperature carrier dynamics in InGaN/GaN multiple quantum wells are studied by employing ultrafast pump-probe spectroscopy. Specifically, the observed differential spectral signatures are characteristic of changes in the absorption coefficient through both a reduction of the quantum-confined Stark shift due to the photoinduced in-well field screening (low carrier densities) and excitonic absorption quenching (high carrier densities). The comparison of the differential absorption spectra at different injected carrier densities allows us to separate field screening from excitonic bleaching. The estimated in-well field at the transition point between field screening and excitonic bleaching is consistent with the theoretical value of the piezoelectric field in the strained InGaN well. © 2003 American Institute of Physics. [DOI: 10.1063/1.1559432]

Recent developments of wide band gap materials using the InGaN system have led to the commercialization of blue, green, and amber light-emitting diodes and violet laser diodes.<sup>1,2</sup> The emission mechanisms of these materials are not fully understood due to the complex material physics. Several groups have discussed the blueshift of the photoluminescence (PL) peak energy with increasing excitation intensity and the long recombination lifetimes at lower photon energies in InGaN active layers. The observed behavior has been attributed to either the reduction of the quantum-confined Stark effect (QCSE) due to in-well field screening<sup>3-5</sup> or through bandfilling of the energy band tail states<sup>6</sup> or both.<sup>7,8</sup>

Traditional time integrated PL and time resolved PL measurements allow only the radiative recombination dynamics to be directly characterized. Ultrafast differential transmission spectroscopy, on the other hand, can provide additional insight on carrier transport and recombination dynamics by separating the field screening behavior from the bandfilling effects. Moreover, pump/probe techniques, when combined with time resolved PL, generally allow for the determination of carrier transport, recombination, and trapping. A few such measurements have been reported that included differential transmission studies on stimulated emission processes and carrier capture times in InGaN/GaN quantum wells (QWs).<sup>9-11</sup> However, of those experiments, none has spectrally and temporally resolved the resulting changes in absorption coefficient due to in-well field screening. In this article, the transition of the differential absorption spectral signature in InGaN/GaN multiple quantum wells (MQWs) from the signature for excitonic bleaching to that for an excitonic blueshift, as the injected carrier density is reduced, is reported.

The sample for the study reported here was an undoped In<sub>0.1</sub>Ga<sub>0.9</sub>N/GaN MQW structure deposited by metalorganic

vapor phase epitaxy on a hydride vapor phase epitaxy GaN/Sapphire substrate. The MQW structure consists of six layers of 100 Å In<sub>0.1</sub>Ga<sub>0.9</sub>N wells alternating with seven layers of 150 Å GaN barriers, as determined by x-ray diffraction. The strain in each of the wells is 1.09% (compressive) and the barriers are considered to be lattice matched to the GaN buffer. Using a method developed by Fiorentini *et al.*,<sup>12</sup> the estimated built-in electric field (parallel to the *c* axis) in the well is -0.96 MV/cm and in the barrier is 0.62 MV/cm.

Ultrafast single color pump-white-light continuum probe spectroscopy was used to time resolve the differential transmission signatures at room temperature. For these measurements, a portion of the 760 nm, 150 fs laser pulses from a 250 kHz regenerative amplifier (REGA) was frequency doubled to 380 nm to serve as the pump source for carrier excitation above the In<sub>0.1</sub>Ga<sub>0.9</sub>N band gap, but below the GaN band gap. The remaining output from the REGA was used to create a broadband white-light continuum, with spectral components from 385 nm to 1000 nm (1.24 eV to 3.22 eV), which served as the probe beam.<sup>10</sup> The time resolution of this system is limited to ~300 fs due to the broadened pulse width of the frequency-doubled pulse and the white-light continuum. The probe beam was focused to an 80 μm diameter spot on the sample and the transmitted light was spectrally resolved using a spectrometer. The pump spot size was chosen to be 160 μm to ensure that the probe beam was monitoring a relatively constant injected carrier density. Standard lock-in techniques were used to measure the difference in the probe transmission ( $\Delta T$ ) with and without the pump present as a function of probe wavelength, pump fluence, and delay time between pump and probe pulse. The differential absorption spectrum was then extracted from differential transmission using the expression:  $\Delta\alpha = -1/L[\ln(1+\Delta T/T)]$ , where *L* is the total thickness of the QWs.<sup>10</sup>

Two fundamentally different types of absorption nonlinearities have been clarified in the studies on GaAs-based self-electro-optic-effect devices and [111] oriented piezoelec-

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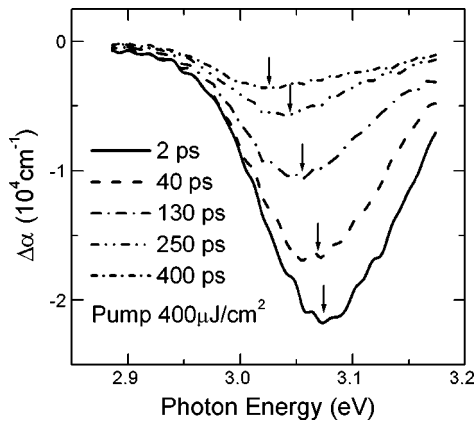


FIG. 1. (a) Differential absorption spectra at different delay times under high pump fluence ( $400 \mu\text{J}/\text{cm}^2$ ).

tric InGaAs devices: Excitonic bleaching and excitonic blueshifting.<sup>13–15</sup> Excitonic bleaching induces a decrease and broadening of the excitonic absorption and the corresponding change in absorption is dominated by a large negative peak centered at the excitonic peak energy. In excitonic blueshifting, the blueshift of the excitonic absorption due to field screening (and reduction of the QCSE) accompanied by a narrowing of the exciton resonance produces a change in absorption coefficient which will consist of positive and negative peaks, with a zero crossing that blueshifts as the injected carrier density is increased.<sup>15</sup>

Figure 1 shows the differential absorption spectra at different delay times under the pump fluence of  $400 \mu\text{J}/\text{cm}^2$ , corresponding to a maximum two-dimensional photoexcited carrier density for each well of  $7.2 \times 10^{13} \text{cm}^{-2}$  (much larger than the sheet charge carrier density,  $4.8 \times 10^{12} \text{cm}^{-2}$ , needed to screen the estimated in-well field). Therefore, at this pump fluence, the in-well field is totally screened and a single negative peak due to excitonic bleaching dominates the differential absorption spectra. It should be noted that with increasing delay time, the energy of the peak signal in the differential absorption spectra exhibits a redshift but the spectra is still dominated by a single negative peak. This large negative peak is a clear indication that we still have sufficient carriers to completely screen the in-well field. Moreover, the observed redshift with increasing time is consistent with carrier relaxation to lower-energy states.

By contrast, Fig. 2(a) shows that at low pump fluence, the observed spectral signature is consistent with the presence of excitonic bleaching and excitonic blueshifting. This signal is similar to that observed by differential transmission of AlGaIn/GaN QW with a negative–positive–negative swing due to blueshifting and narrowing of the excitonic absorption.<sup>16</sup> Figure 2(b) shows the differential absorption spectra for a fixed pump fluence of  $20 \mu\text{J}/\text{cm}^2$  versus time delay from 40 ps to 4  $\mu\text{s}$ . (The measurement at 4  $\mu\text{s}$  was made by setting the delay so that the probe pulse arrived at the sample 40 ps before the next pump pulse.) Clearly, although the net change in absorption decreases with time delay as the carrier density decreases, the change in absorption due to screening of the in-well field remains relatively unchanged for short times. Moreover, the positive and negative

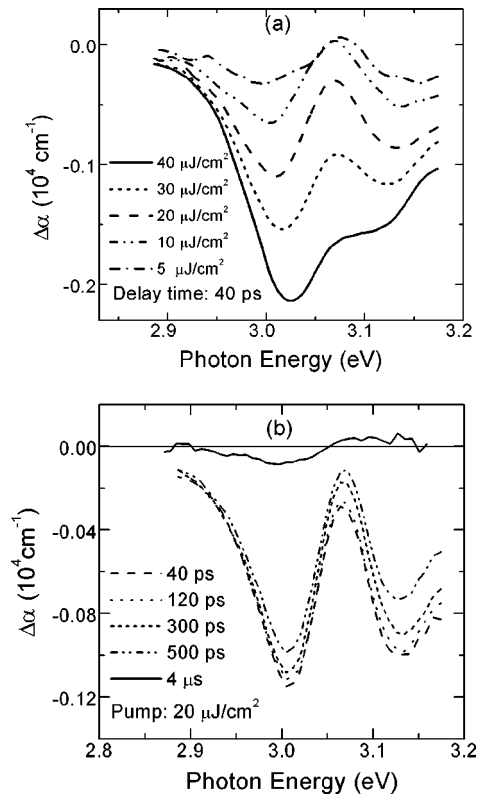


FIG. 2. (a) Pump fluence dependent differential absorption spectra at a delay time of 40 ps, and (b) differential absorption spectra at different delay times under a pump fluence of  $20 \mu\text{J}/\text{cm}^2$ .

peaks, indicative of a field induced excitonic blueshifting and narrowing, are still evident after 4  $\mu\text{s}$ . This extreme prolongation of recombination lifetime in the well is induced by the spatial separation between electrons and holes under the influence of the in-well field.<sup>15</sup>

The original differential absorption signal shown in Fig. 3 (solid line) is separated into two components by fitting the bleaching component with a spectral curve consistent with the bleaching shown in Fig. 1. The resulting field screening component is extracted and shown by the dashed–dotted line

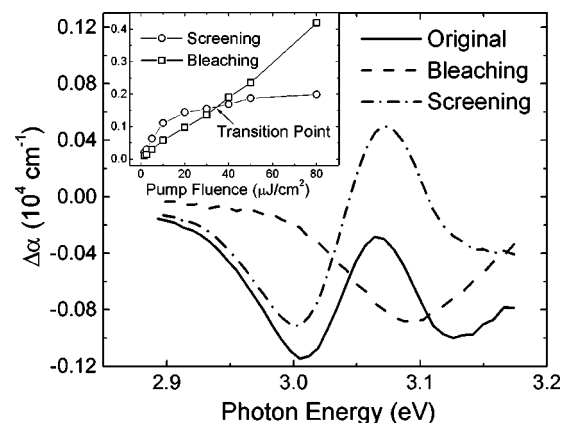


FIG. 3. The differential absorption spectrum (solid line) at a delay time of 40 ps under a pump fluence of  $20 \mu\text{J}/\text{cm}^2$  is separated into in-well screening (dashed–dotted line) and bleaching (dashed line) components. The inset shows the extracted peak change in absorption due to field screening and bleaching vs pump fluence.

in Fig. 3. The pump fluence dependent peak absorption changes caused by these two components, as estimated by the fitting, are also illustrated in the inset of Fig. 3. As should be expected, when increasing the pump fluence, the screening component approaches a constant value while the bleaching component increases linearly. The transition point,  $35 \mu\text{J}/\text{cm}^2$ , at which the bleaching and screening signatures are comparable, is a reasonable choice for estimating the in-well field (i.e., the in-well field is being sufficiently screened that the overlap of the carriers increases and the bleaching signature becomes significant). At this fluence, the sheet charge density in each well is estimated as  $5.6 \times 10^{12} \text{ cm}^{-2}$  and the corresponding in-well field, 1.12 MV/cm, is close to the estimated theoretical value of 0.96 MV/cm provided earlier. It is important to realize that this is not an extremely accurate estimate but does provide ballpark estimates of the in-well field.

The experimental results presented here, illustrating excitonic bleaching and in-well field screening in these undoped InGaN/GaN MQWs, may be explained as follows. Initially, the femtosecond pulse generates excitons that are rapidly dissociated by the in-well field in the 10 nm InGaN well.<sup>16</sup> At low carrier densities, the electrons and holes drift to the opposite sides of the well under the influence of the in-well field on a very fast time scale ( $<1$  ps). Thus, the intrinsic in-well field is reduced and band bending within the well is reduced. As a result of the reduction of the magnitude of the in-well field, the differential transmission signature for excitonic blueshifting and narrowing is observed (in conjunction with excitonic bleaching). At high carrier densities, the in-well field is completely screened and the remaining carriers fill any localized states. Moreover, at sufficiently high carrier densities, the excitonic absorption bleaching obscures the signature caused by the excitonic blueshift.

In conclusion, in-well excitonic bleaching and screening in an InGaN/GaN MQW structure has been investigated by performing ultrafast differential transmission spectroscopy. This technique provides a method to separate the field screening behavior from the band-filling effects by observing the transition from the signature of excitonic absorption

bleaching to excitonic blueshifting. The estimated in-well field at the transition point between the two signatures is consistent with the calculated value resulting from piezoelectric polarization in strained InGaN well.

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