Femtosecond Exciton Dynamics and the Mott Transition in GaN Under Resonant Excitation

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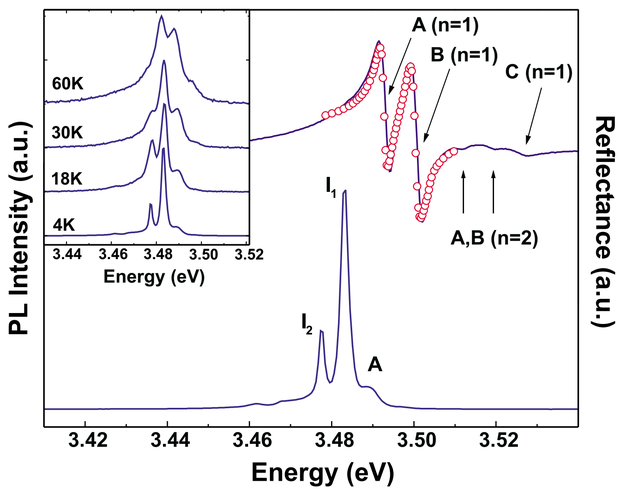

Figure 1. Reflectivity and luminescence spectra of an ELOG grown GaN epilayer at 4K. Bound exciton peaks in the luminescence are labelled I_1 and I_2. The open circles are a fit to the reflectance spectrum. E_A = 3.492 eV E_B = 3.499 eV. The effective linewidths for the A- and B-excitons are 1.5 meV.

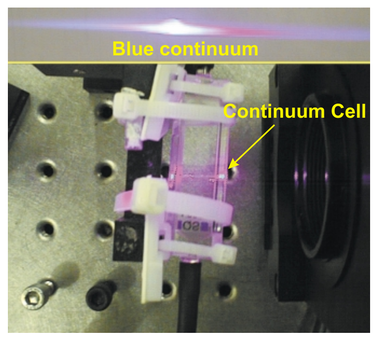

Plate 1. Flowing water cell showing conversion of 400nm probe light into a blue continuum. The upper picture shows the continuum dispersed by a grating onto a white card.

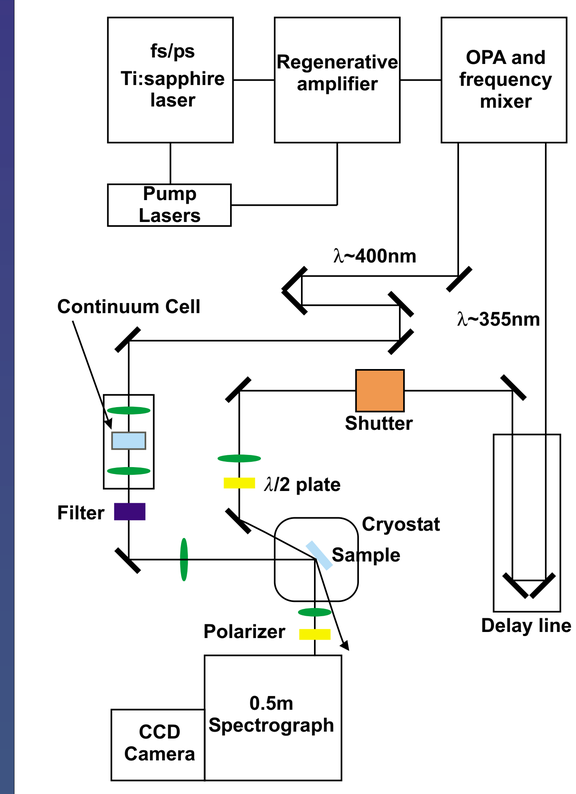

Figure 2. Schematic diagram of the time-resolved reflectance experiment. A femtosecond Ti:sapphire laser is amplified in a regernerative amplifier. The amplified output is frequency doubled to ~400 nm and some of this light is further shifted to ~355 nm by frequency mixing with red light generated in an optical parametric amplifier. This is used as the pump beam. The probe beam is generated by focussing the remaining doubled light into a cell of water to generate a continuum probe. The net time-resolution of the whole system was ~800fs.

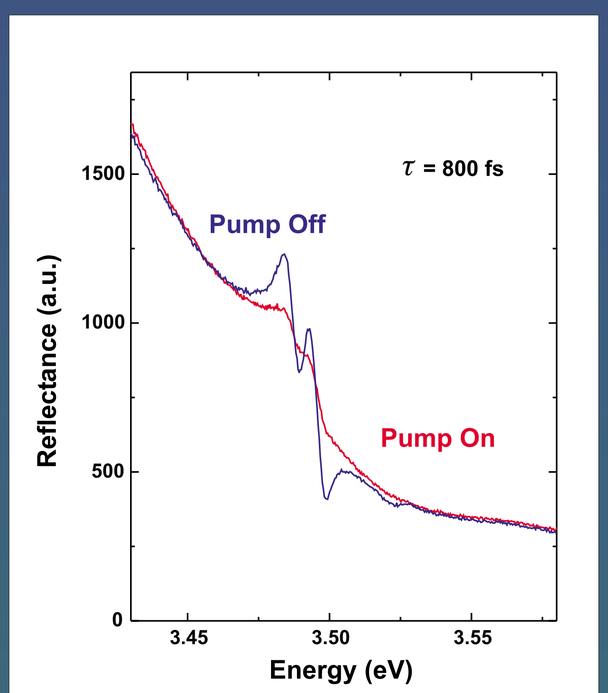

Figure 3. Time-resolved reflectance spectra from sample at 4 K at a time delay of 800 fs. The black curve shows the reflectance with the pump beam blocked, the red curve with the pump beam present. Note the bleaching of all three excitons.

Experiment

In this poster we present a time-resolved pump-probe reflectance study of high quality GaN epilayers in reflection geometry. The GaN epilayers are grown by lateral epitaxial overgrowth. In order to probe primarily the radiative exciton population, we resonantly excite excitons with a 250 fs laser pulse, resulting in an exciton population with wavevector q ~ 0. The sharp exciton resonances in the reflectance spectrum bleach as the density of photoexcited excitons increases, and the change in intensity of the reflected light as a function of time is a measure of the q ~ 0 exciton dynamics. At 4 K the dominant excitonic relaxation process is trapping at impurities and defects with a characteristic time t = 16 ps. At much higher temperatures (> 60 K) we observe the emergence of long-lived radiative recombination with t ~ 350-400 ps. The longer lifetime agrees well with the predicted value for the radiative decay of thermalised excitons at the polariton bottle-neck.

The time-resolved reflectance spectroscopy was performed on the A- and B-exciton transitions with an average pump power of 0.1 mW. This pump power corresponds to a maximum exciton density of 2.0 x 10^{19} cm^{-3}. The onset of bleaching occurs within the 800 fs time resolution of our system. The recovery of the excitonic reflectivity takes place on a slower timescale, and even after 35 ps there is a small residual bleaching. A noteworthy feature of the measurements is that the energies of the A- and B-excitons are apparently unshifted even in the presence of this relatively strong photoexcitation. This result is consistent with theoretical predictions of excitonic transitions in bulk semiconductors which include only exciton self-screening and no free carrier contribution, an approximation that is likely to apply in the present case of resonant excitation: in this model the bandgap renormalisation is exactly cancelled by the simultaneous reduction of the exciton binding energy. It is also noteworthy that the C-exciton, which arises from the spin-split-off valence band, features quite strongly in the differential reflectance at 3.52 eV, although it is not apparent in the CW spectrum.

An increase in temperature to 60 K and 100 K results in the appearance of a long-lived component in the differential reflectance data. The decay is now clearly bi-exponential, with decay times of 16 ps and 375 ps giving a good fit to the data. Comparison with the CW data presented, which show that a significant free exciton population exists at 60 K in thermal equilibrium with the neutral donor bound excitons, suggests that the longer decay component is due to intrinsic radiative recombination. Theoretical predictions based on a polariton model using our reflectance data suggest a radiative lifetime of 300 ps. This value is in remarkably good agreement with the experimentally observed long-lived component.

A measurement of the excitonic bleaching as a function of excitation density shows that the bleaching saturates at a density of 2.2 x 10^{19} cm^{-3}. This density can be identified with the Mott density in this sample.

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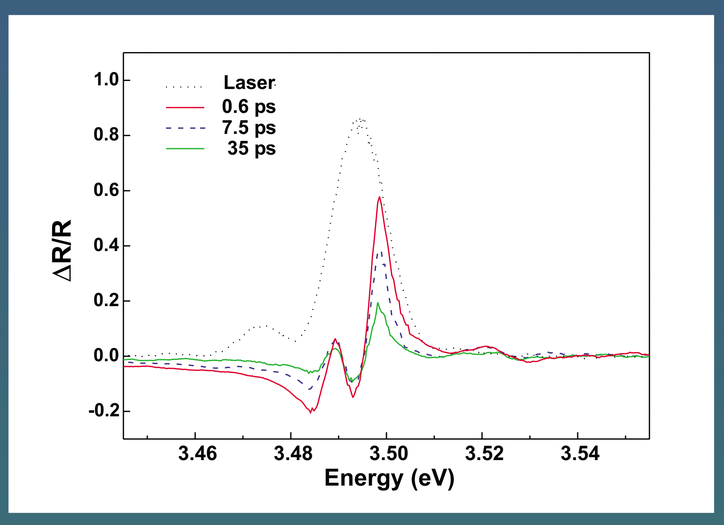

Figure 4. Differential reflectance spectra measured at various time delays at 4K. The laser excitation spectrum is shown as a dotted curve.

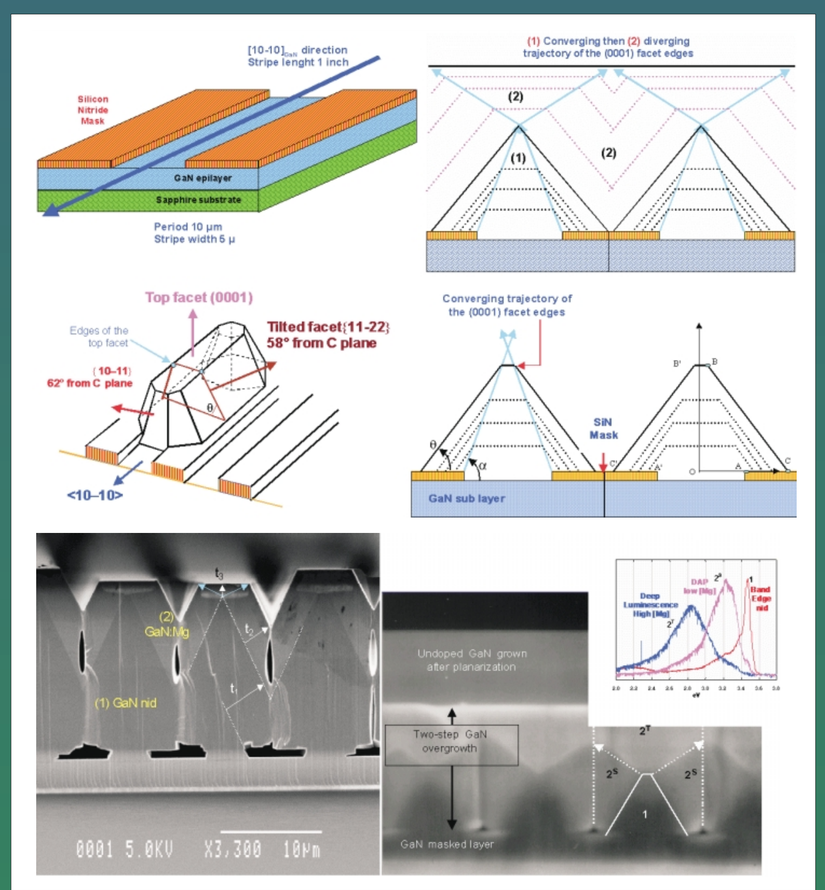

Figure 5. The drawings show the various stages involved in the growth of GaN on sapphire using the epitaxial layered overgrowth technique (ELOG). After initial GaN growth a silicon nitride film is deposited and etched into strips. GaN is then overgrown, and the pattern of growth seen is a v-pattern as shown. When Mg doping is introduced the v-pattern fills in to form a smooth surface. The plates show the large reduction in the density of threading dislocations possible using this technique.

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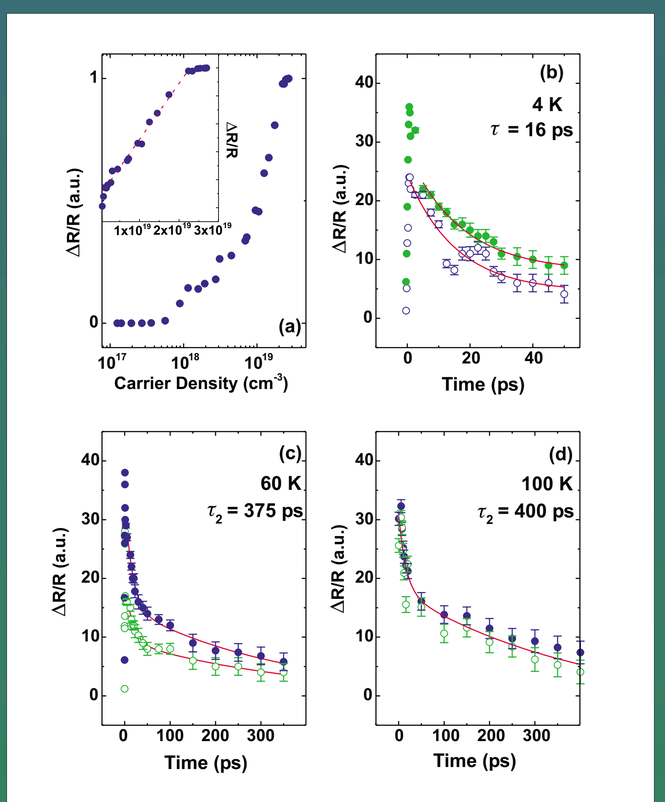

Figure 6. (a) Carrier density dependence of the differential reflectance spectra under resonant excitation. The inset shows the same data, but with both axes on a linear scale; (b) - (d) Time-dependence of the differential reflectance signal at different temperatures from A-excitons (hollow circles) and B-excitons (filled circles). The solid lines show the results of a single (b) or double (c) and (d) exponential fit to the data (τ = 16 $\pm$ 5 ps).