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Strong Electron-LO Phonon Scattering and Hot Carrier Relaxation in GaN

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Wide-bandgap GaN is currently of great interest because of its applications in short-wavelength lasers. GaN is strongly excitonic, the binding energy E_X ~23meV [1], and the band-edge optical properties are dominated by pronounced excitonic resonances arising from the three closely-spaced valence bands in the Wurzite structure. However, femtosecond time-resolved reflection studies [2] reveal that the onset of stimulated emission coincides with the Mott transition, so that lasing in fact arises from an electron-hole plasma. It is also of great interest in the production of high-field electronic devices. Because it is strongly polar, electron-phonon scattering rates in GaN are expected to be comparable to or possibly faster than electron-electron scattering (τ_{LO} ~ 8 fs), even at high carrier density. This suggests that energy relaxation may occur before there is any significant thermalization of the photoexcited plasma. In order to resolve these issues, and to provide a detailed understanding of the electron-hole dynamics in GaN, we have made the first comprehensive study of exciton and hot carrier relaxation dynamics in this material.

The time-resolved measurements reported here were made using an optical parametric amplifier pumped by a frequency-doubled amplified Ti:sapphire laser. A 0.5μm epilayer of GaN grown on sapphire was measured using the pump-probe differential transmission technique. The pump energy could be tuned over a wide energy range extending from the exciton resonances to well into the electron-hole continuum. A white-light continuum probe with a resolution of 120fs was used.

Carrier relaxation has been investigated using non-resonant excitation. Electrons were photoexcited with an excess energy above the LO phonon energy (E_{LO} = 92meV) over a range of densities extending above the Mott limit (~1×10^{19} cm^{-3}). At high density, during the risetime of the pulse, we observe remarkably rapid electron relaxation into a highly non-thermal distribution characterized by measurable bleaching due to LO phonon satellites. Analysis of the normalized difference spectra (NDS) obtained shows a rapid decrease in the energy loss rate as a population of hot phonons is built up. Non-thermal high-energy carrier distributions are also observed, consistent with earlier experiments [3].

In a preliminary theoretical study we have performed a series of Monte Carlo simulations of hot carrier dynamics in a parabolic four-band model in which excitonic effects are neglected. We considered unscreened optical phonon scattering by a single LO mode together with optical deformation potential scattering, intravalley acoustic phonon scattering, and Coulombic scattering within the static screening approximation. We also included hot phonons and degeneracy effects, which proved to be critically important at the high carrier densities used in these experiments. The simulations reproduce the important experimental observations: in particular, they confirm the development of the carrier distribution at early times, where phonon satellites are seen, and a strong non-thermal electron distribution in the region near E_{LO} arising from a remarkably strong electron-LO phonon interaction.

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References

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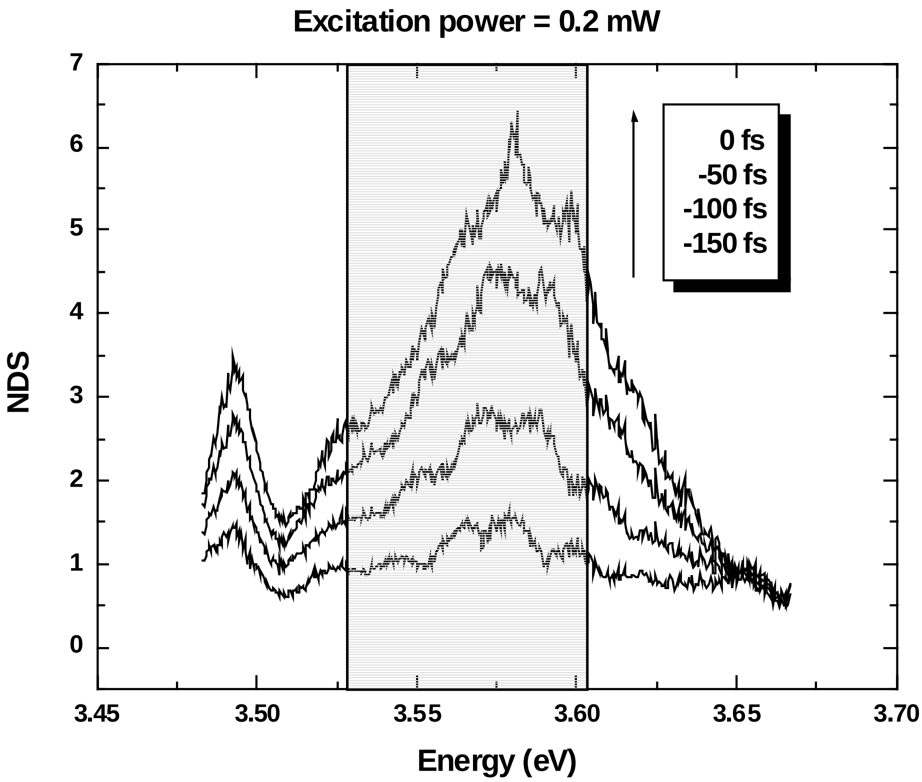
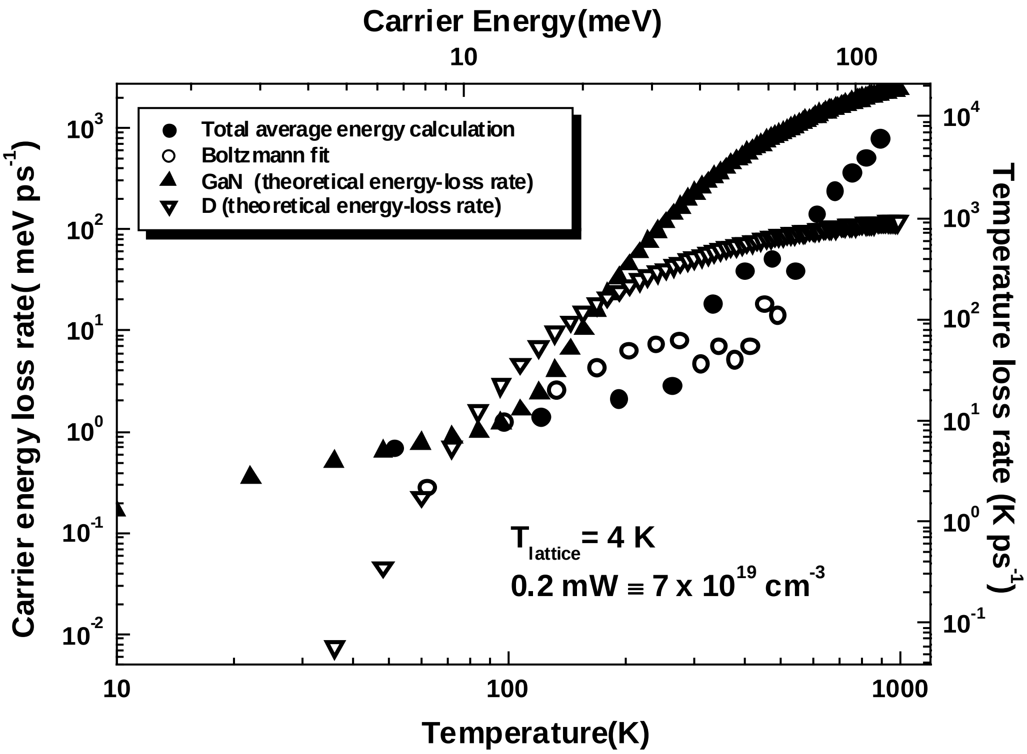


The figure above shows the bleaching induced by pulsed excitation at 3.667 eV. The shaded area shows the region where bleaching occurs due to LO phonon emission.



This figure compares the calculated energy loss rates for GaAs and GaN assuming a lattice temperature of 4 K, and equilibrium conditions. Also plotted are the measured energy loss rates from our experiment. The figure shows that at early times (high initial mean energies), the theory and experiment agree well. As the carriers cool, the loss rate drops – this is due to non-equilibrium LO phonons populating the relevant wavevectors where both LO phonon emission and re-absorption can occur.