Hot Carrier Dynamics and Coherent Effects in GaN under Short Laser Pulse Excitation

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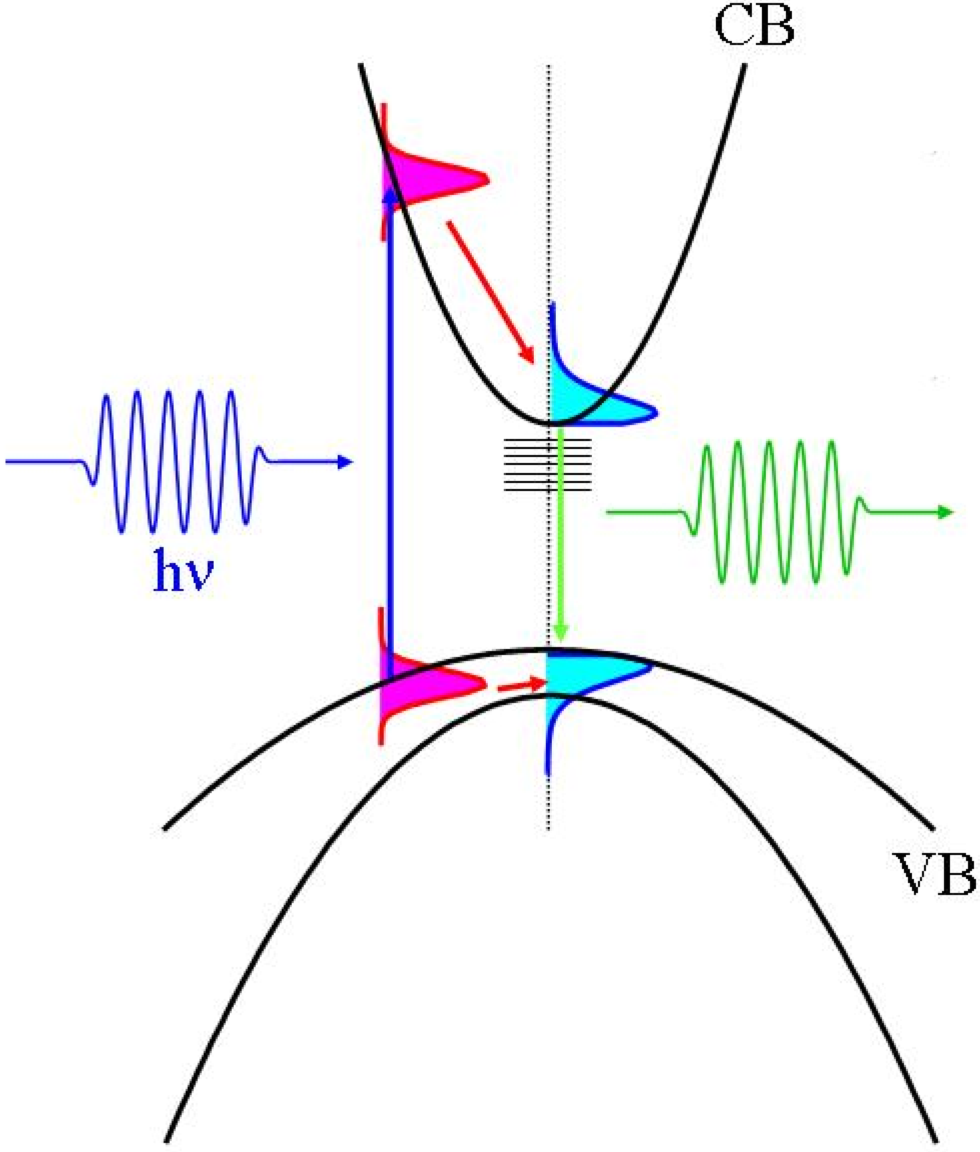
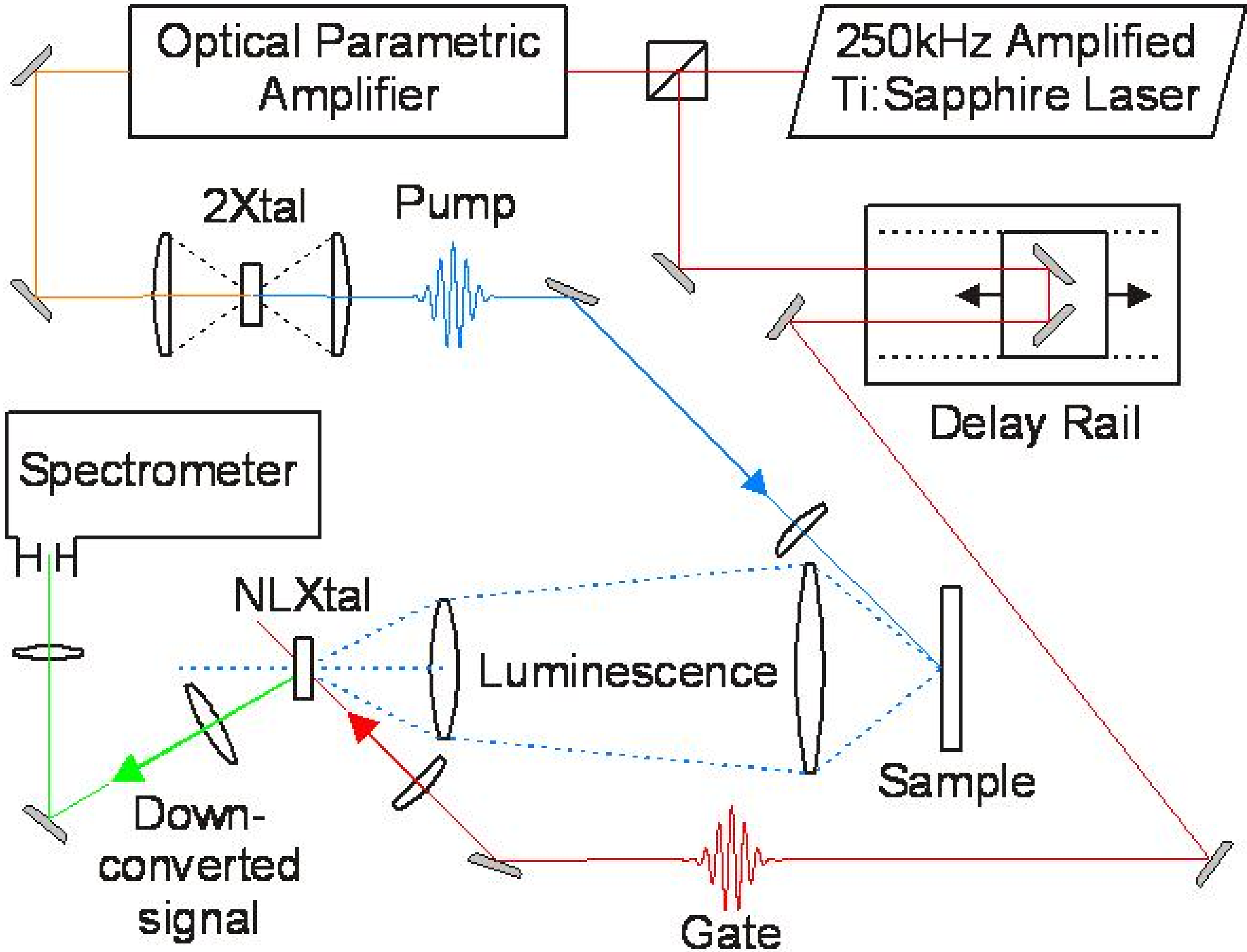
Carrier Dynamics

- Electrons and holes generated by ultrashort pulse with photon energy hν
- Intraband relaxation
- Interband recombination
- Localization and recombination through trap states

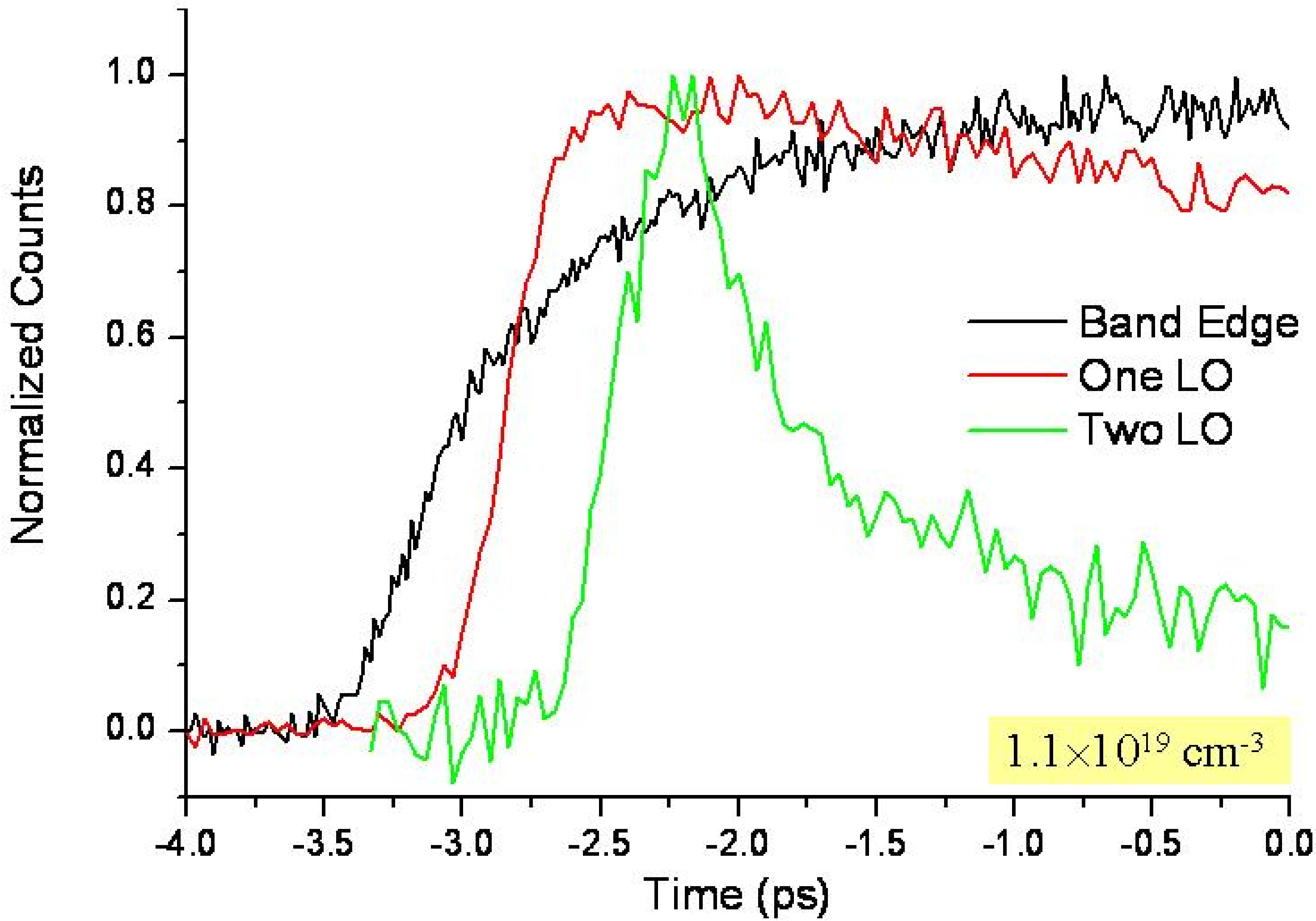


Experimental Setup

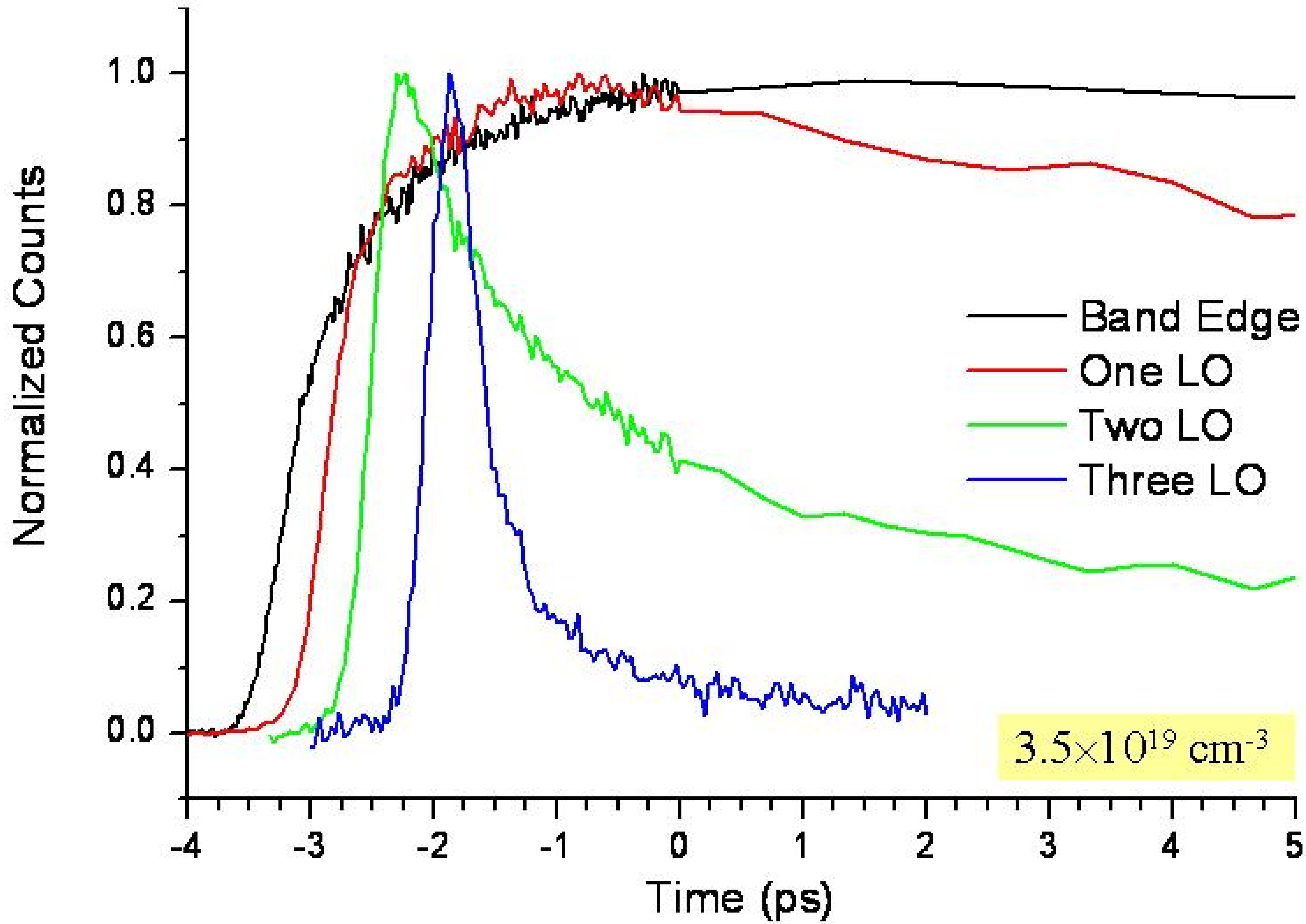
- Ti:Sapphire
  - 150 fs, 5 μJ, 800 nm pulses
- OPA
  - tunable from 720 to 450 nm, ~50 fs pulses
- PL excitation pulse
  - tunable from 360 to 225 nm, ~15 nJ pulses
  - pump energy from 3.44 to 5.5 eV, suitable for studying $Al_xGa_{1-x}N$ materials

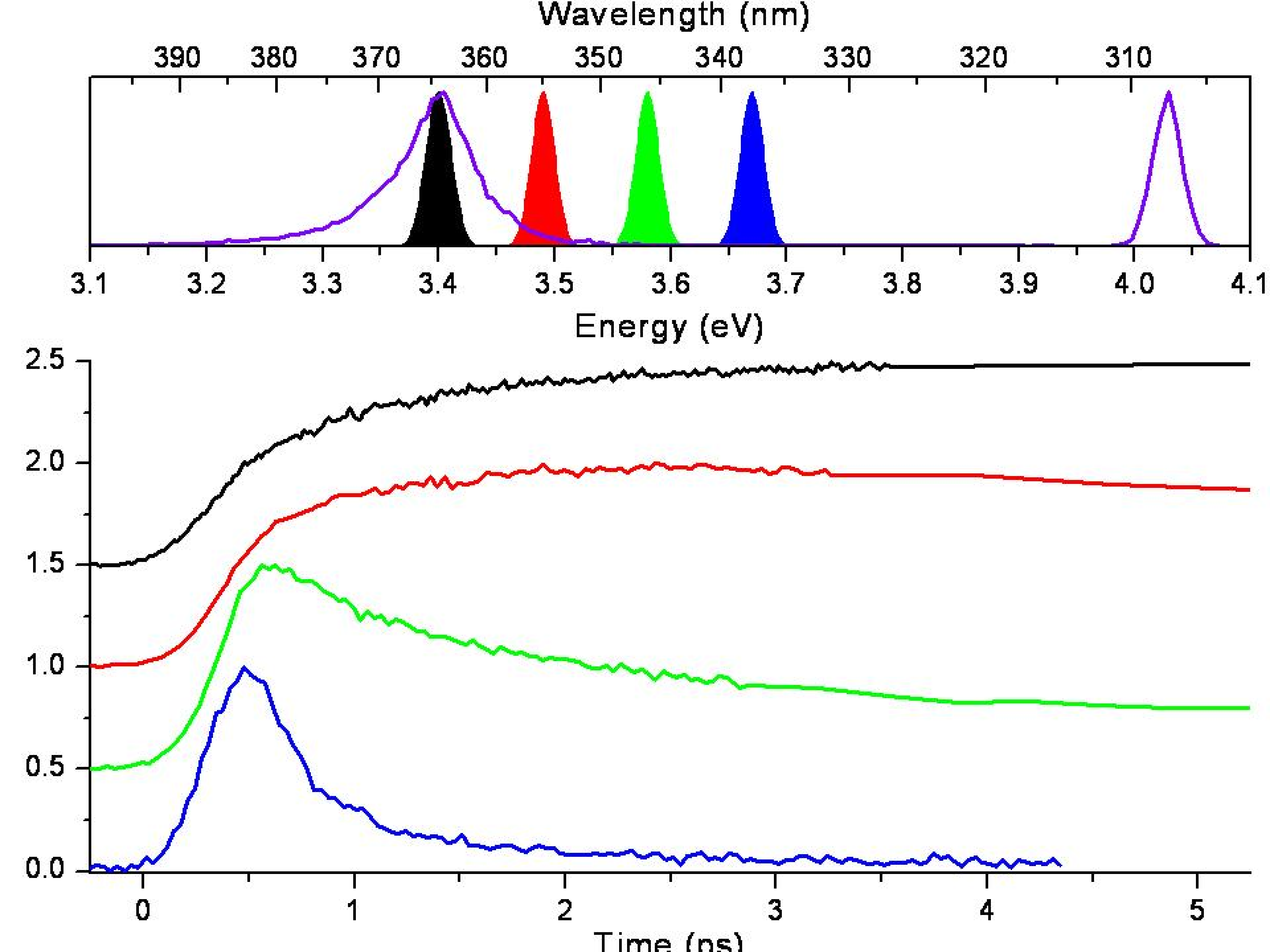


Pump 660 μW at 308 nm

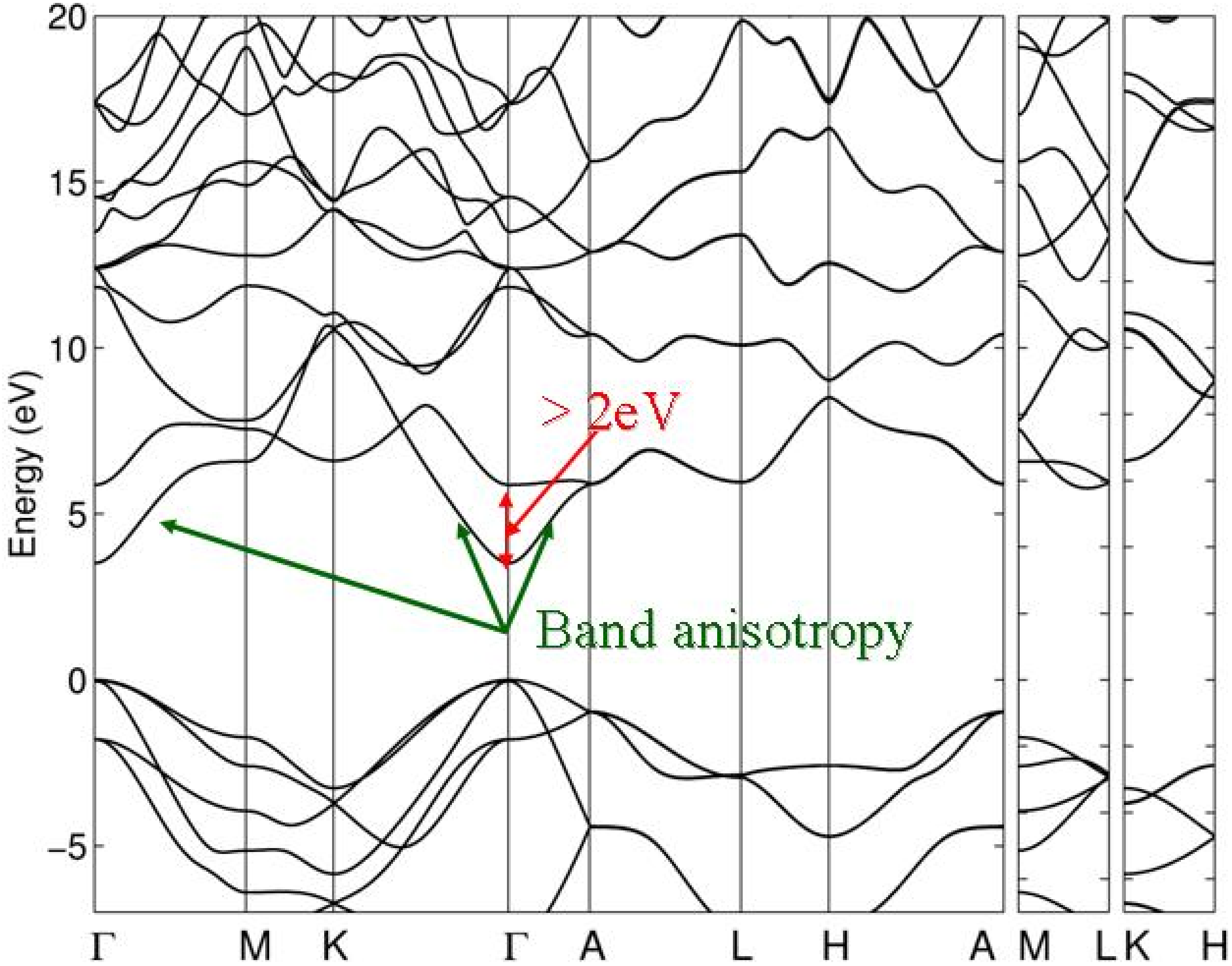


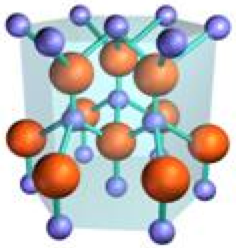
Pump 2.0 mW at 308 nm

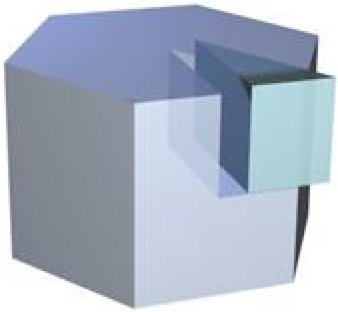



Wavelength (nm)
390
380
370
360
350
340
330
320
310
3.1
3.2
3.3
3.4
3.5
3.6
3.7
3.8
3.9
4.0
4.1
Energy (eV)
2.5
2.0
1.5
1.0
0.5
0.0
0
1
2
3
4
5
Time (ps)

GaN-Band Structure







M. Goano, E. Bellotti, E. Ghillino, G. Ghione, and K. F. Brennan, J. Appl. Phys. 88, 6467 (2000).

Model

- Two-band model for carriers
- Dipole and rotating wave approximations for carrier-light interaction
- Carrier-phonon scattering, only polar LO-phonons included. Adiabatic and Markov approximations, to the second order in interaction.
- Carrier-carrier scatterings, first order (Hartree-Fock terms) and second order contributions in Markov approximation.
- Static screening for carrier-carrier and carrier-phonon interactions.

$$H_0 = \sum_{\boldsymbol{k}} \varepsilon_e(k) c_{\boldsymbol{k}}^{+} c_{\boldsymbol{k}} + \sum_{\boldsymbol{k}} \varepsilon_h(k) d_{\boldsymbol{k}}^{+} d_{\boldsymbol{k}} + \sum_{\boldsymbol{k}} \hbar\omega(q) b_{\boldsymbol{q}}^{+} b_{\boldsymbol{q}}$$

$$+ \sum_{\boldsymbol{k}} \left[M_{\boldsymbol{k}} E_0(t) e^{-i\omega_p t} c_{\boldsymbol{k}}^{+} d_{-\boldsymbol{k}}^{+} + M_{\boldsymbol{k}}^{*} E_0(t) e^{i\omega_p t} d_{-\boldsymbol{k}} c_{\boldsymbol{k}} \right]$$

$$H_{c-ph} = \sum_{\boldsymbol{k},\boldsymbol{q}} \left[\gamma_{\boldsymbol{q}}^{e} c_{\boldsymbol{k}+\boldsymbol{q}}^{+} b_{\boldsymbol{q}} c_{\boldsymbol{k}} + \gamma_{\boldsymbol{q}}^{e*} c_{\boldsymbol{k}}^{+} b_{\boldsymbol{q}}^{+} c_{\boldsymbol{k}+\boldsymbol{q}} + \gamma_{\boldsymbol{q}}^{h} d_{\boldsymbol{k}+\boldsymbol{q}}^{+} b_{\boldsymbol{q}} d_{\boldsymbol{k}} + \gamma_{\boldsymbol{q}}^{h*} d_{\boldsymbol{k}}^{+} b_{\boldsymbol{q}}^{+} d_{\boldsymbol{k}+\boldsymbol{q}} \right]$$

$$H_{c-c} = \sum_{\boldsymbol{k},\boldsymbol{p},\boldsymbol{q}} V_q \left[\frac{1}{2} c_{\boldsymbol{k}}^{+} c_{\boldsymbol{p}}^{+} c_{\boldsymbol{p}+\boldsymbol{q}} c_{\boldsymbol{k}-\boldsymbol{q}} + \frac{1}{2} d_{\boldsymbol{k}}^{+} d_{\boldsymbol{p}}^{+} d_{\boldsymbol{p}+\boldsymbol{q}} d_{\boldsymbol{k}-\boldsymbol{q}} - c_{\boldsymbol{k}}^{+} d_{-\boldsymbol{p}}^{+} d_{-\boldsymbol{p}+\boldsymbol{q}} c_{\boldsymbol{k}-\boldsymbol{q}} \right]$$

$$V_q = \frac{4\pi e^2}{\varepsilon_0 \varepsilon_s \mathcal{V}} \frac{1}{q^2 + \kappa^2} \qquad \gamma_q = \frac{e^2 \hbar \omega_{LO}}{\varepsilon_0 \mathcal{V}} \left(\frac{1}{\varepsilon_\infty} - \frac{1}{\varepsilon_s} \right) \frac{q^2}{\left(q^2 + \kappa^2\right)^2}$$

$$\kappa^2 = -\frac{4\pi e^2}{\varepsilon_s \mathcal{V}} \sum_{\boldsymbol{k},\alpha} \left(\frac{\partial \varepsilon_\alpha(k)}{\partial k} \right)^{-1} \left(\frac{\partial f_{\boldsymbol{k}}^\alpha}{\partial k} \right)$$

$$f_{\boldsymbol{k}}^e(t) = \left\langle c_{\boldsymbol{k}}^+(t) c_{\boldsymbol{k}}(t) \right\rangle \quad f_{\boldsymbol{k}}^h(t) = \left\langle d_{\boldsymbol{k}}^+(t) d_{\boldsymbol{k}}(t) \right\rangle \quad p_{\boldsymbol{k}}(t) = \left\langle d_{-\boldsymbol{k}}(t) c_{\boldsymbol{k}}(t) \right\rangle$$

$$N_q = \left\langle b_q^+ b_q \right\rangle$$

$$i\hbar \frac{d}{dt} \hat{O}(t) = \left[\hat{O}, H \right]$$

$$\left.\frac{df_{\boldsymbol{k}}^{e}}{dt}\right|^{(0)} = \left.\frac{df_{-\boldsymbol{k}}^{h}}{dt}\right|^{(0)} = g_{\boldsymbol{k}}^{(0)}(t)$$

$$g_{\boldsymbol{k}}^{(0)}(t) = \frac{1}{i\hbar}\left[M_{\boldsymbol{k}} E_0(t) e^{-i\omega_p t} p_{\boldsymbol{k}}^{*} - M_{\boldsymbol{k}}^{*} E_0(t) e^{i\omega_p t} p_{\boldsymbol{k}}\right]$$

$$\left.\frac{dp_{\boldsymbol{k}}}{dt}\right|^{(0)} = \frac{1}{i\hbar}\left[\left(\varepsilon_e(\boldsymbol{k}) + \varepsilon_h(-\boldsymbol{k})\right)p_{\boldsymbol{k}} + M_{\boldsymbol{k}} E_0(t) e^{-i\omega_p t}\left(1 - f_{\boldsymbol{k}}^{e} - f_{-\boldsymbol{k}}^{h}\right)\right]$$

$$E_0(t) = E_0 e^{-t^2/\tau_P^2} \qquad M_k \approx d_{\mathrm{cv}}$$

$$\frac{d_{\mathrm{cv}}}{e} = \frac{\hbar p_{\mathrm{cv}}}{E_g m_0} \qquad \frac{m_0}{m_e} \approx 1 + \frac{2p_{\mathrm{cv}}^2}{m_0 E_g}$$

$$\frac{m_e}{m_0} = 0.2 \qquad E_g = 3.39\, eV \quad \Rightarrow \quad \frac{d_{\mathrm{cv}}}{e} \approx 2.12\, \mathring{A}$$

$$\Omega_R^{(0)} \equiv \frac{d_{\mathrm{cv}} E_0}{\hbar} \qquad \Omega_R^{(0)} \approx 0.5\ \mathrm{THz} \times \sqrt{\mathrm{W_{cw}}/1\mu\mathrm{W}}$$

Coherent optical Bloch equations with relaxation time approximation

$$U_{2k}(t) \equiv -i\left(e^{-i\omega_p t} p_k^*(t) - e^{i\omega_p t} p_k(t)\right) \qquad U_{1k}(t) \equiv e^{-i\omega_p t} p_k^*(t) + e^{i\omega_p t} p_k(t)$$

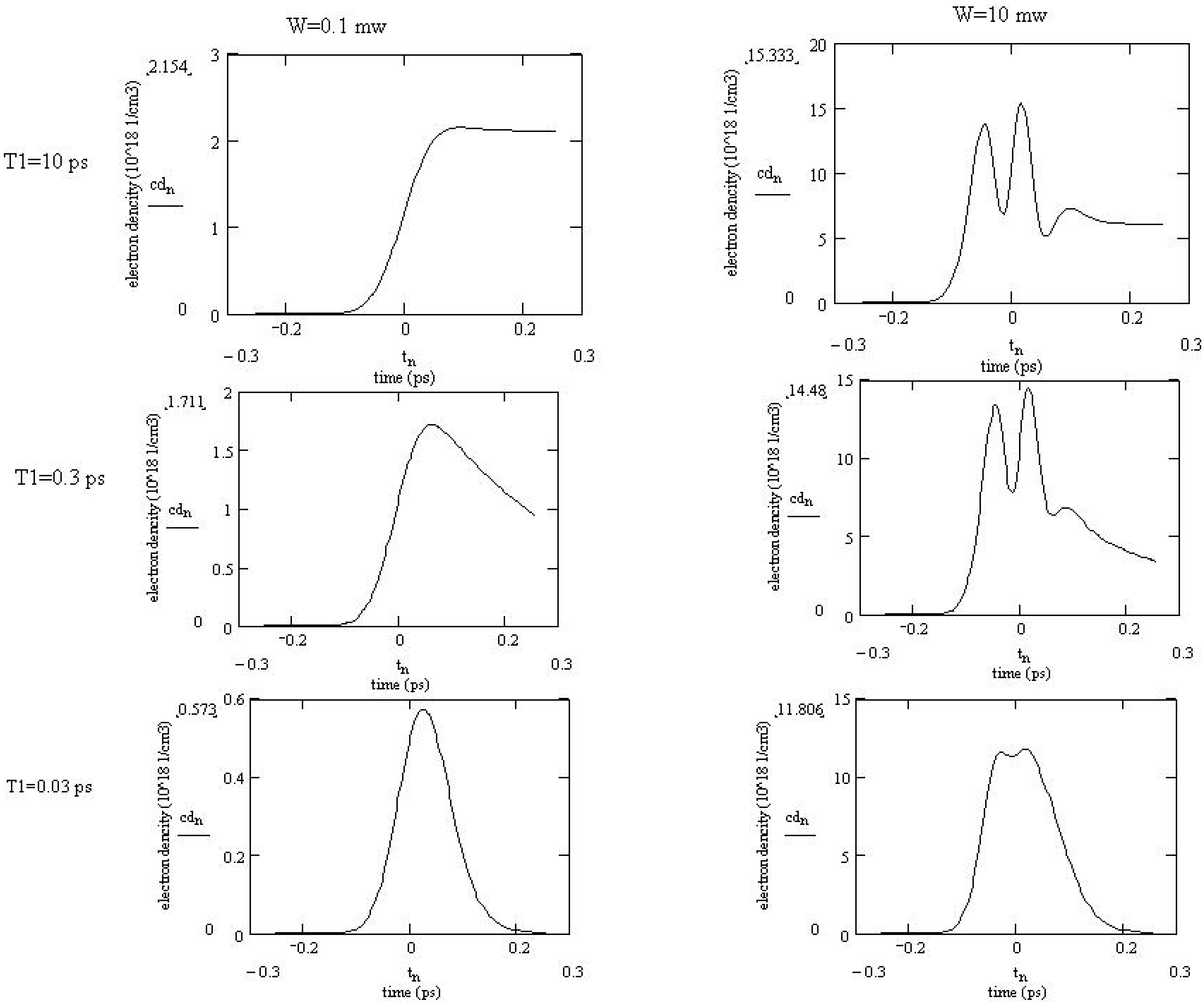
$$U_{3k} \equiv 1 - f_k^e(t) - f_k^h(t) \qquad \Delta_k \equiv \frac{\varepsilon_k^e + \varepsilon_k^h}{\hbar} - \omega_p$$

$$\dot{U}_{1k} = -\Delta_k U_{2k} - \frac{U_{1k}}{T_2}$$

$$\dot{U}_{2k} = \Delta_k U_{1k} + 2\Omega_R^{(0)} E_0(t) U_{3k} - \frac{U_{2k}}{T_2}$$

$$\dot{U}_{3k} = -2\Omega_R^{(0)} E_0(t) U_{2k} - \frac{1 - U_{3k}}{T_1}$$

$$U_{3k}(-\infty) = 1 \qquad U_{1k}(-\infty) = U_{2k}(-\infty) = 0 \qquad T_2 = 2T_1$$



W=0.1 mw
W=10 mw
T1=10 ps
T1=0.3 ps
T1=0.03 ps
electron dencity (10^18 1/cm3)
cd_n
t_n
time (ps)
2.154
15.333
1.711
14.48
0.573
11.806

Semiclassical limit :

polarization equation is treated with Markov and adiabatic approximations and Boltzmann equations are obtained

$$\frac{df_{\mathbf{k}}}{dt} = \sum_{\mathbf{k}'} \left[W_{\mathbf{k},\mathbf{k}'} f_{\mathbf{k}'} - W_{\mathbf{k}',\mathbf{k}} f_{\mathbf{k}'} \right]$$

Monte Carlo simulation

$$f_{\mathbf{k}}(t) = \sum_{\mathbf{k}'} G_{\mathbf{k},\mathbf{k}'}(t,t_0) f_{\mathbf{k}'}(t_0) \approx \sum_{\mathbf{k}'} \sum_{i=1}^{N_{\mathbf{k}'}} G_{\mathbf{k},\mathbf{k}'}(t,t_0) w_i = \sum_{i=1}^{N} G_{\mathbf{k},\mathbf{k}_i}(t,t_0) w_i, \quad N = \sum_{\mathbf{k}'} N_{\mathbf{k}'}$$

General structure of the kinetic equations

$$\frac{d}{dt}\Im_{\boldsymbol{k}}^{\alpha} = \frac{d}{dt}\Im_{\boldsymbol{k}}^{\alpha}\Big|_{coh} + \frac{d}{dt}\Im_{\boldsymbol{k}}^{\alpha}\Big|_{incoh}, \quad \alpha = e, h, p$$

$$\Im_{\boldsymbol{k}}^{e,h} \equiv f_{\boldsymbol{k}}^{e,h}, \quad \Im_{\boldsymbol{k}}^{p} \equiv p_{\boldsymbol{k}}$$

$$\frac{d}{dt}\Im_{\boldsymbol{k}}^{\alpha}\Big|_{coh} = \sum_{\alpha'} \mathscr{G}_{\boldsymbol{k}}^{\alpha',0}\left(\{\Im^{\alpha'}\}\right) + \sum_{j} \mathscr{G}_{\boldsymbol{k}}^{\alpha,j}\left(\{\Im^{\alpha}\}\right)$$

$$\frac{d}{dt}\Im_{\boldsymbol{k}}^{\alpha}\Big|_{incoh} = \sum_{j}\sum_{\boldsymbol{k}'}\left[W_{\boldsymbol{k},\boldsymbol{k}'}^{\alpha,j}\Im_{\boldsymbol{k}'}^{\alpha} - W_{\boldsymbol{k}',\boldsymbol{k}}^{\alpha,j}\Im_{\boldsymbol{k}}^{\alpha}\right]$$

Generalized Monte Carlo simulation:

Extension of Monte Carlo method to the analysis of coherent phenomena.

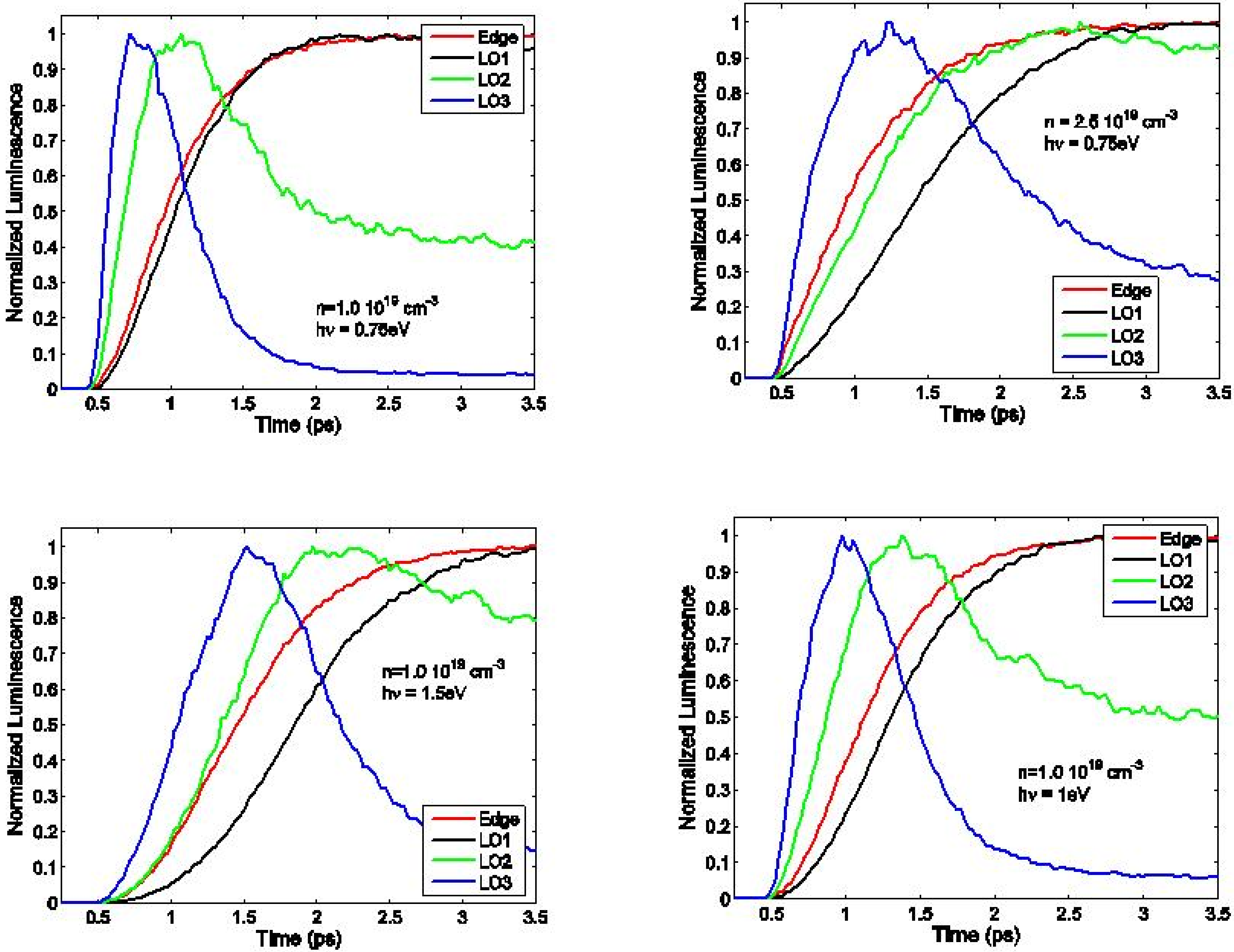
References:

S. Haas, F. Rossi, T. Kuhn, Phys. Rev. B 53, 12855 (1996)
F. Rossi and T. Kuhn, Rev. Mod. Phys. 74, 895 (2002)

Phase relations between different types of carriers (polarization phenomena)

Interaction of carriers with an external electromagnetic field

Correlation and renormalization effects associated with carrier-carrier interaction


Normalized Luminescence
Time (ps)
Edge
LO1
LO2
LO3
n=1.0 10^19 cm^-3
hν = 0.75eV
n = 2.5 10^19 cm^-3
hν = 0.75eV
n=1.0 10^19 cm^-3
hν = 1.5eV
n=1.0 10^19 cm^-3
hν = 1eV