Microscopic Modelling of the Optical Properties of Quantum-Well Semiconductor Lasers

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- **OVERVIEW**
  - Outline of Theory
  - Gain/Absorption
  - Luminescence
  - Radiative and Auger Losses
  - Nonequilibrium Effects

- **COLLABORATORS**
  - A. Thränhardt, I. Kutzentsova, S.Becker, Marburg
  - J. Hader, J. V. Moloney et al., Tucson
  - W. Chow, Sandia

- **SUPPORT**
  - AFOSR F49620-02-1-0380, DFG

Overview

simple models: easy to use

- often poorly controlled, ad hoc assumptions
- rely on major experimental input
- (semi-)empirical fits of experimental data
- many and adjustable parameters
- limited potential for predictions
- ….

microscopic theory: numerically demanding

- systematic approach based on fundamental physics
- controlled approximations
- predicts experiments
- no need for detailed experimental input
- wide range of applicability
- can be used as basis for controllably simplified models
- ….

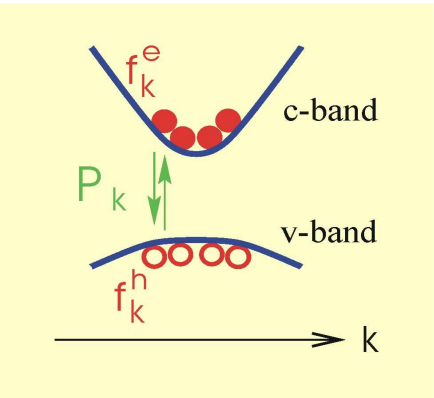
Semiconductor Optics: Semiclassical Theory

MAXWELL'S WAVE EQUATION

$$\left[\frac{\partial^2}{\partial z^2} - \frac{n^2(z)}{c^2}\frac{\partial^2}{\partial t^2}\right] E = \mu_0 \frac{\partial^2}{\partial t^2} P$$

macroscopic optical polarization

semiconductors: Bloch basis $P = \sum_k d^*_{cv} P_k + c.c.$

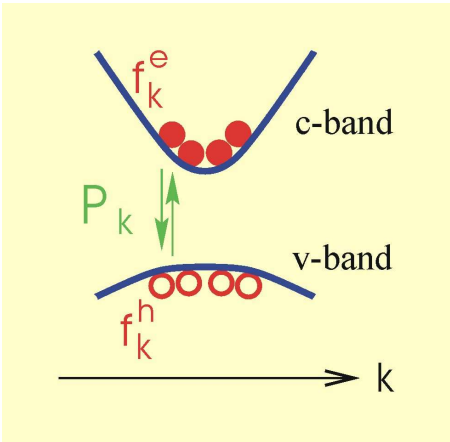


general theory: Haug/Koch, Quantum Theory of the Optical and Electronic Properties of Semiconductors, 4th ed., World Scientific (2004)

Semiconductor Density Matrix

two-band model, one QW subband

$$\begin{pmatrix} \langle a_k^\dagger \, a_k \rangle & \langle b_{-k} \, a_k \rangle \\ \langle a_k^\dagger \, b_{-k}^\dagger \rangle & \langle b_k^\dagger \, b_k \rangle \end{pmatrix} = \begin{pmatrix} f_k^c & P_k \\ P_k^* & f_k^v \end{pmatrix}$$



P_k … e-h-pair transition amplitude

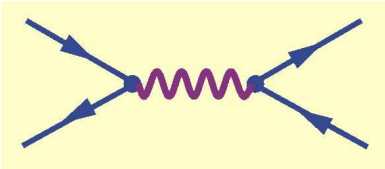
$f_k^{c,v}$ … occupation probabilities

Hamiltonian

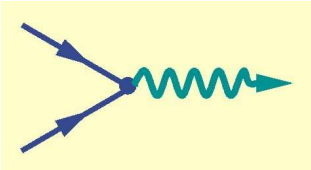
$$H = H_0 + H_{Coul} + H_{dip}$$

H_0 single particle (band structure)

H_{Coul} Coulomb interaction between carriers



H_{dip} dipole interaction with optical field



Calculation of Optical Response

Heisenberg equation of motion: $i\hbar \frac{\partial}{\partial t} \mathcal{O} = [\, \mathcal{O}, H \,]$

structure of resulting equations:

$$i\hbar \frac{\partial}{\partial t} \langle b_{-k}\, a_k \rangle = \varepsilon_k \langle b_{-k}\, a_k \rangle + \sum_{k',q\neq 0} V_q \left\langle a^\dagger_{k'+q}\, b_{q-k}\, a_{k'}\, a_k \right\rangle + \ldots$$

infinite hierarchy of many-body correlations

→ **systematic CLUSTER expansion**

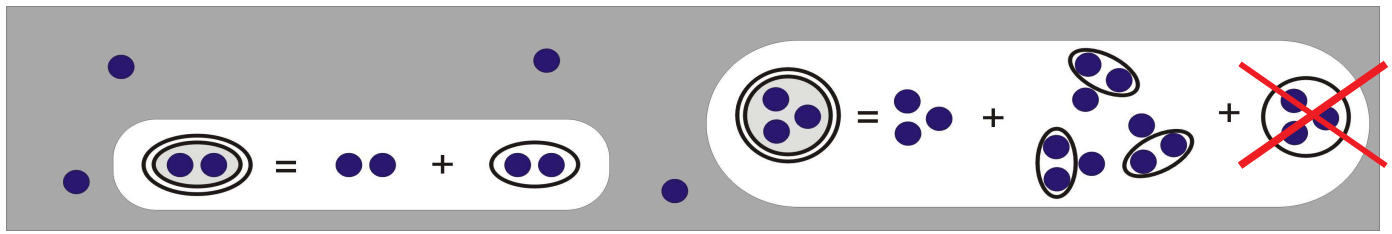
Schematics of Cluster Expansion

- two-point level (uncorrelated single particles)

$$
\begin{aligned}
i\hbar\frac{\partial}{\partial t}\langle\{1\}^\dagger\{1\}\rangle &= T\left[\langle\{1\}^\dagger\{1\}\rangle\right] + V\left[\langle\{2\}^\dagger\{2\}\rangle\right] \\
&= T\left[\langle\{1\}^\dagger\{1\}\rangle\right] + V\left[\langle\{2\}^\dagger\{2\}\rangle_{\mathrm{S}}\right] \\
&\qquad + V\left[\Delta\langle\{2\}^\dagger\{2\}\rangle\right]
\end{aligned}
$$

- four-point level (two particle correlations)

$$
\begin{aligned}
i\hbar\frac{\partial}{\partial t}\Delta\langle\{2\}^\dagger\{2\}\rangle &= T\left[\Delta\langle\{2\}^\dagger\{2\}\rangle\right] + V\left[\langle\{3\}^\dagger\{3\}\rangle\right] \\
&= T\left[\Delta\langle\{2\}^\dagger\{2\}\rangle\right] + V\left[\langle\{3\}^\dagger\{3\}\rangle_{\mathrm{S}}\right] \\
&\qquad + V\left[\langle\{3\}^\dagger\{3\}\rangle_{\mathrm{D}}\right] \\
&\qquad + V\left[\cancel{\Delta\langle\{3\}^\dagger\{3\}\rangle}\right]
\end{aligned}
$$

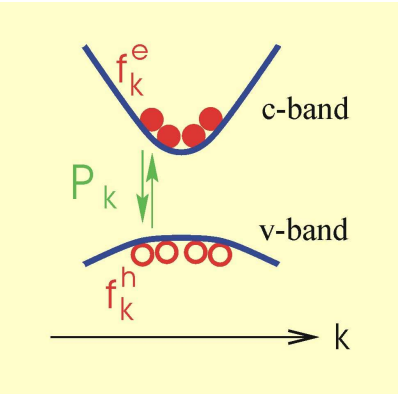


Semiconductor Bloch Equations

all contributions beyond singlets

$$\left[i\hbar\frac{\partial}{\partial t} - \epsilon_k^e - \epsilon_k^h\right] P_k = \left[1 - f_k^e - f_k^h\right]\Omega_k + \frac{\partial}{\partial t}P_k|_{corr}$$

$$i\hbar\frac{\partial}{\partial t}\, f_k^a = -\Omega_k(t)P_k^* + \Omega_k^* P_k + \frac{\partial}{\partial t}f_k^a|_{corr}$$



HF field renormalization $\quad \Omega_k(t) = d_{cv}\; E^{QW}(t) + \sum_{k'} V_{k-k'}\; P_{k'}(t)$

HF energy renormalization $\quad \epsilon_k^a(t) \;=\; \varepsilon_k^a - \sum_{k'} V_{k-k'}\; f_{k'}^a(t)$

- nonlinearities: phase-space filling, gap reduction, Coulomb enhancement
- correlation contributions: scattering, dephasing, screening

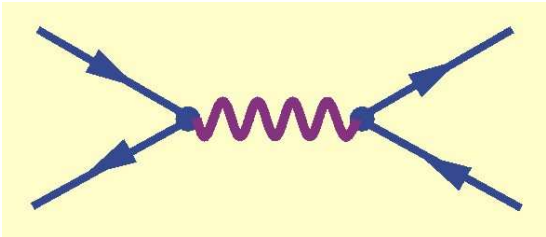
Second Born-Markov Approximation

- intraband Coulomb scattering
- phonon scattering

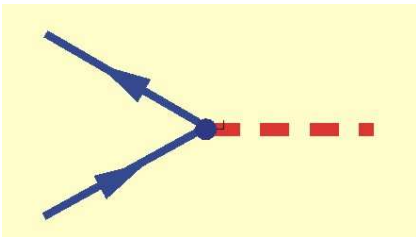
Born-Markov limit: Boltzmann equation

$$\frac{\partial}{\partial t} f_k^a(t)|_{corr} = \Sigma_k^{in,a}(t)[1 - f_k^a(t)] - \Sigma_k^{out,a}(t) f_k^a(t)$$

Coulomb scattering



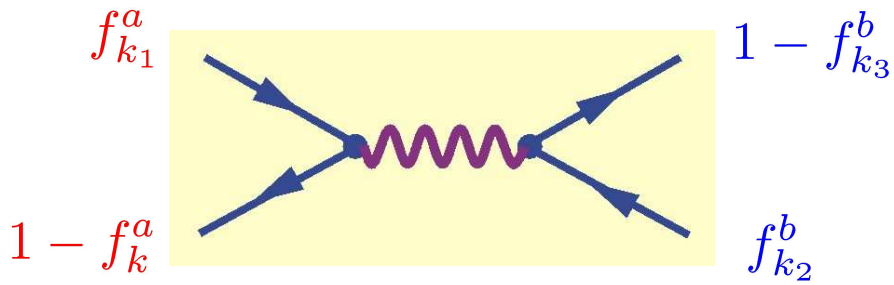
phonon scattering



Coulomb Scattering I

$$\frac{\partial}{\partial t} f_k^a(t)|_{corr} = \Sigma_k^{in,a}(t)[1 - f_k^a(t)] - \Sigma_k^{out,a}(t) f_k^a(t)$$

scattering rates:



$$\frac{\partial}{\partial t} f_k^a|_{corr} \approx \sum_{k_1,k_2,k_3,b=e,h} |W|^2 \, f_{k_1}^a f_{k_2}^b [1 - f_{k_3}^b][1 - f_k^a] - \ldots$$

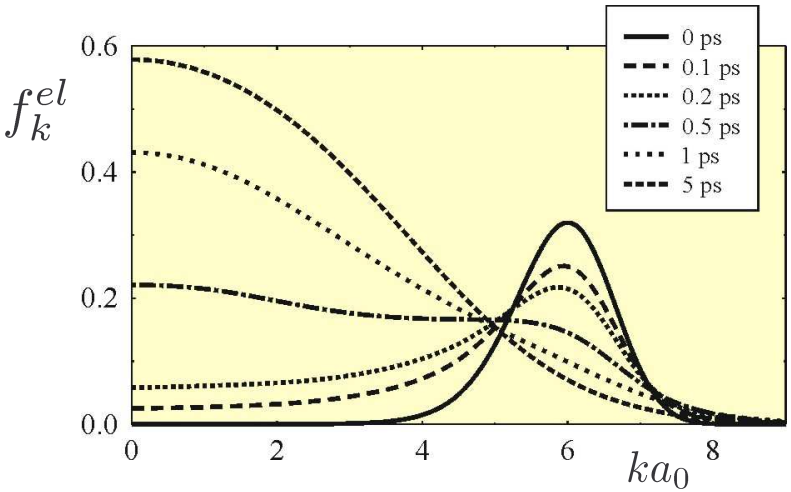
Coulomb Scattering II

$$\frac{\partial}{\partial t} f_k^a(t)|_{corr} = \Sigma_k^{in,a}(t)[1 - f_k^a(t)] - \Sigma_k^{out,a}(t) f_k^a(t)$$

quasi-equilibrium: $$\frac{\partial}{\partial t} f_k^a|_{corr} = 0$$

detailed balance

Fermi-Dirac distribution: $f_k^a \quad => \quad F_k^a = \frac{1}{e^{(E_k - \mu_a)/k_B T} + 1}$



Excitation Induced Dephasing

$$\left[i\hbar\frac{\partial}{\partial t} - \varepsilon^e_{\mathbf{k}}(t) - \varepsilon^h_{\mathbf{k}}(t)\right] P_{\mathbf{k}}(t) - \left[1 - f^e_{\mathbf{k}}(t) - f^h_{\mathbf{k}}(t)\right] \Omega_{\mathbf{k}}(t)$$

$$= i\left[\Gamma_{\mathbf{k}}(t) P_{\mathbf{k}}(t) + \sum_{k'} \Gamma_{\mathbf{k},\mathbf{k}'}(t) P_{\mathbf{k}'}(t)\right]$$

Re $\Gamma_k(t)$ … diagonal dephasing
$\Rightarrow$ generalized T_2 time

Im $\Gamma_k(t)$ … diagonal energy shift
$\Rightarrow$ generalized band-gap shift

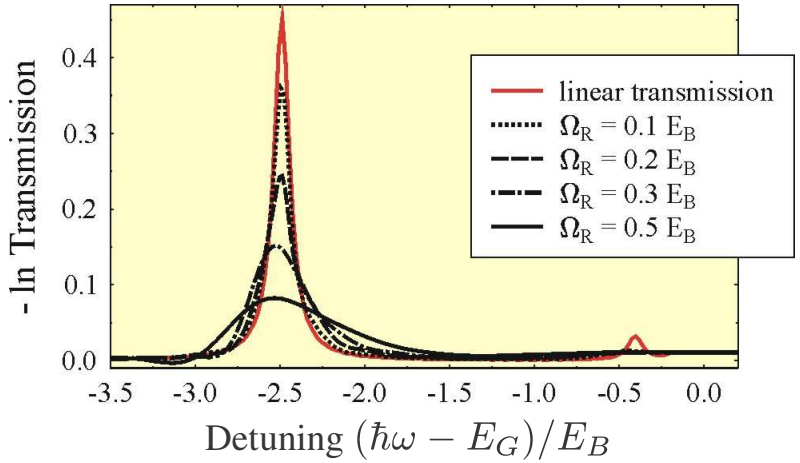
Re $\Gamma_{k,k'}(t)$ … off-diagonal dephasing

Im $\Gamma_{k,k'}(t)$ … off-diagonal energy shift

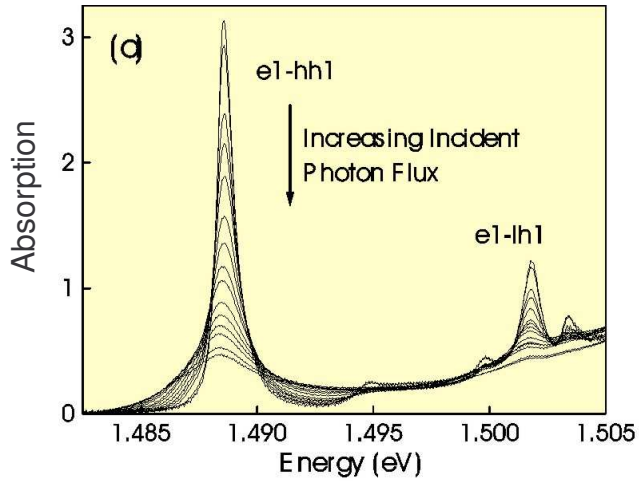
- $\Gamma_k(t)$ and $\Gamma_{k,k'}(t)$ are calculated from all terms quadratic in the screened Coulomb interaction
- strong compensation between diagonal and off-diagonal terms

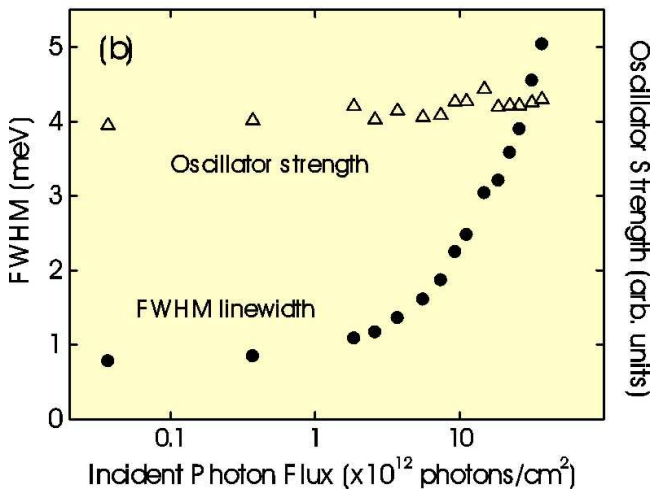
$\Rightarrow$ important for excitation saturation: Jahnke *et al.*, PRL 77, 5257 (1996).
and **gain calculations: Chow/Koch, Semiconductor-Laser Fundamentals, Springer Verlag (1999).**

Density Dependent Absorption



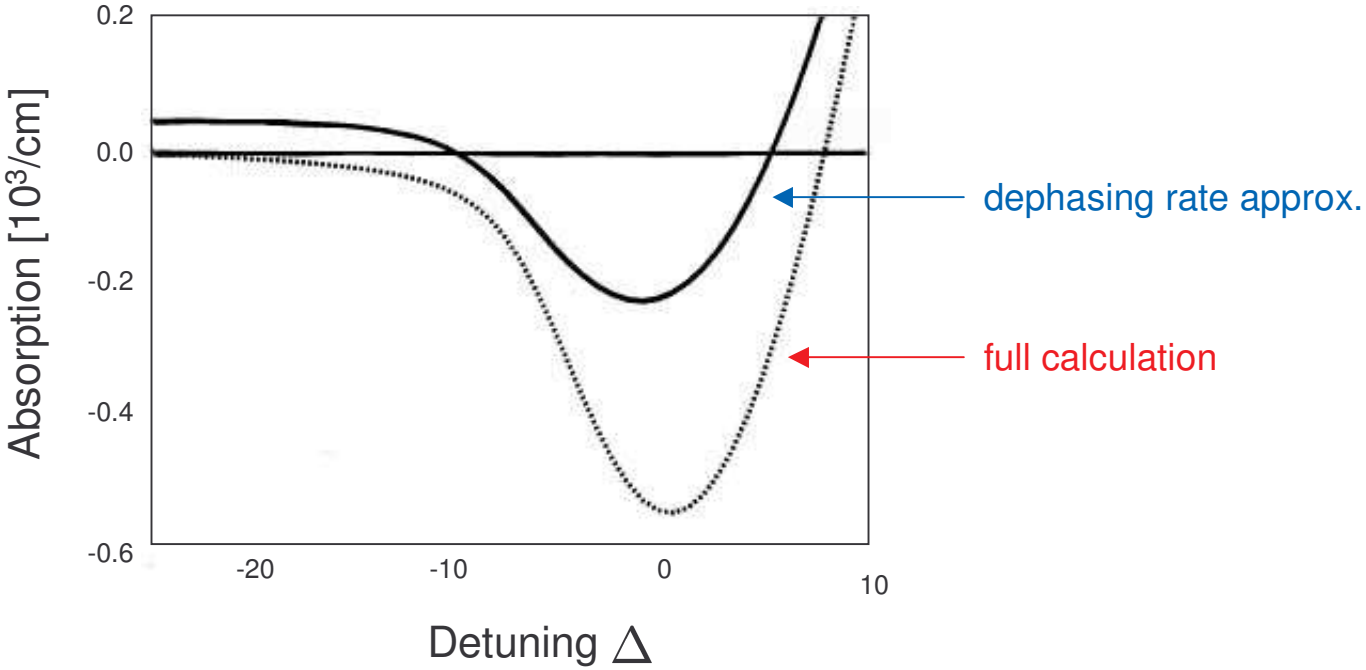
F. Jahnke, M. Kira, and S.W. Koch, Z. Physik B **104**, 559 (1997)





- experiment: InGaAs/GaAs QW
- Khitrova, Gibbs, Jahnke, Kira, Koch, Rev. Mod. Phys. **71**, 1591 (1999)
- EID first observed in 4-wave mixing, Wang *et al.* PRL **71**, 1261 (1993)

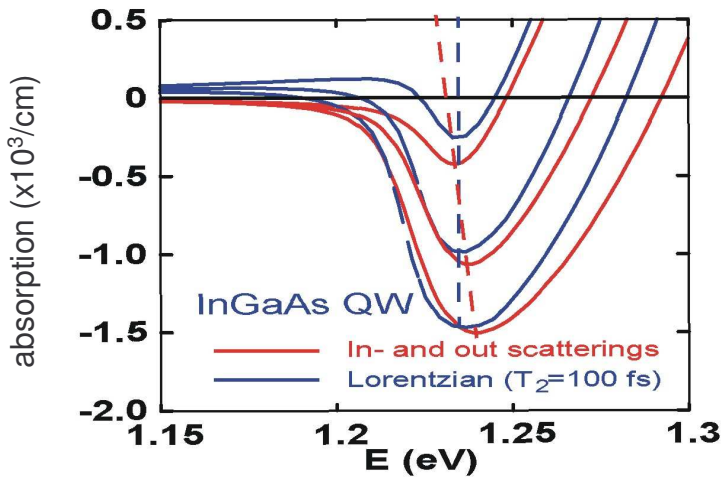
Lineshape Problem

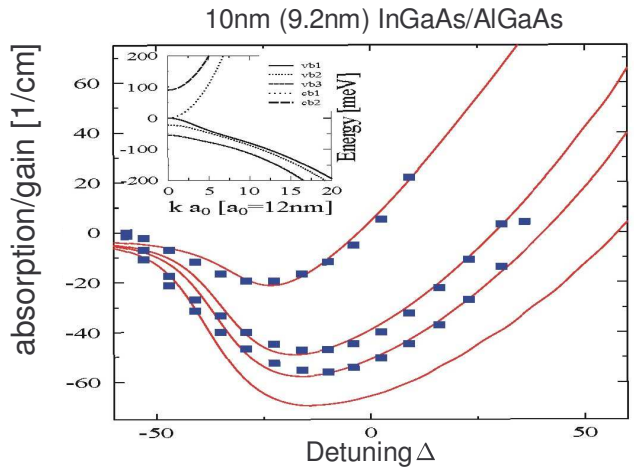


- $\Delta = (\hbar\omega - E_G)/E_B$
- gain of two-band bulk material
- nondiagonal scattering contributions → lineshape modification, no absorption below the gap

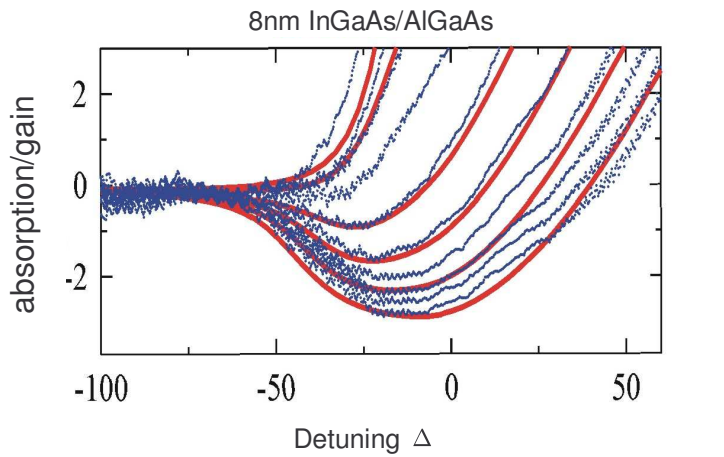
Hughes, Knorr, Koch, Binder, Indik, and Moloney
Solid State Comm. **100**, 555 (1996)

Optical Gain: Theory and Experiment

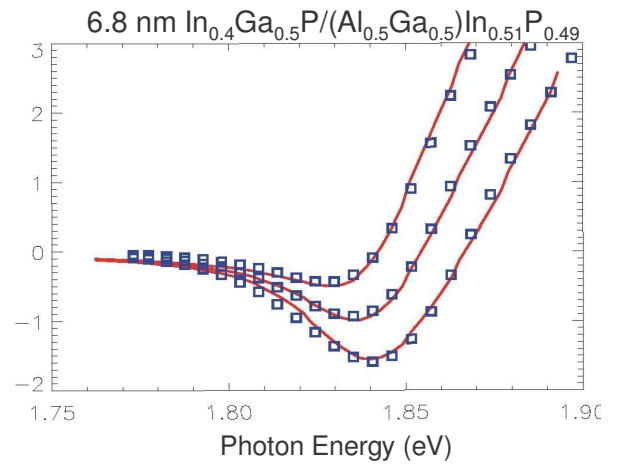




N=1.6, 2.2, 2.5, 3.0*10¹²/cm²
exp: D. Bossert et al., theory: A. Girndt et al., Marburg

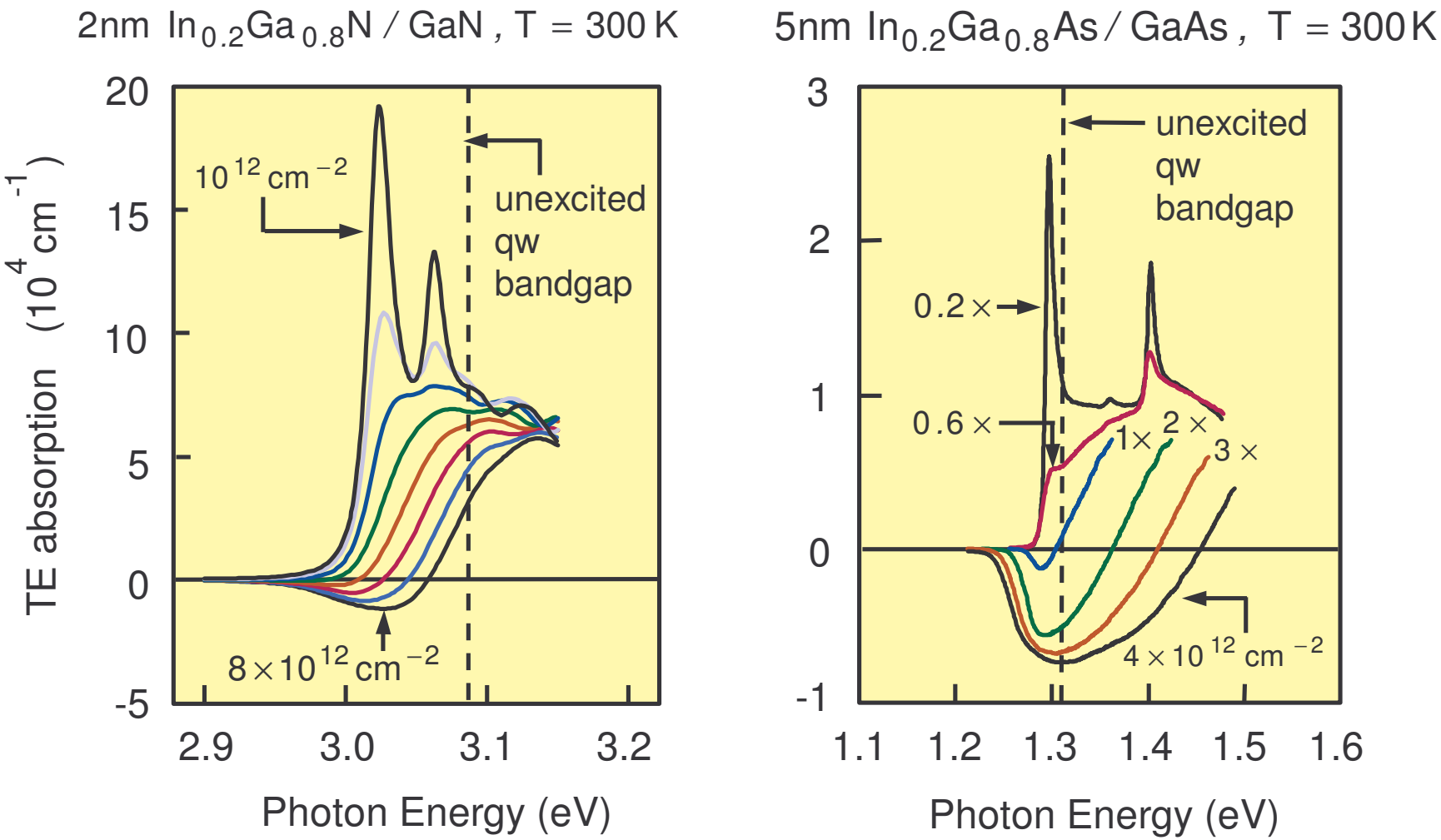


Current 0-20mA… Density 0.6-3.0x10^{12}cm^{-2}
exp: C. Ellmers et al., theory: A. Girndt et al. Marburg



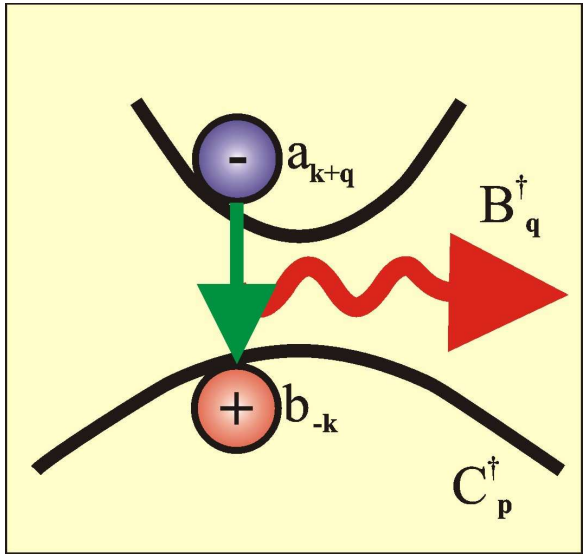
Courtesy of W.W. Chow, P.M. Smowton, P. Blood, A. Grindt, F. Jahnke, and S.W. Koch

Wide Bandgap Materials: InGaN/GaN



- strong Coulomb effects: gapshift and broadening
- excitonic effects even at elevated densities and temperatures

Semiconductor Luminescence



$$H_{cf}^{qm} = \sum A(k,q) a_{k+q} b_{-k} B_q^{\dagger} + \text{h.c.}$$

where $A(k,q)$ is proportional to dipole matrix element and mode strength at the QW position

Semiconductor Luminescence Equations I

- photon operator dynamics

$$i\hbar\frac{\partial}{\partial t}B_{q_z q_\parallel} = \hbar\omega_q B_{q_z q_\parallel} + F_q \sum_k b_{-k} a_{k+q_\parallel} - F_q^* \sum_k a^\dagger_{k-q_\parallel} b^\dagger_{-k}$$

- polarization operator dynamics

$$i\hbar\frac{\partial}{\partial t}b_{-k}a_{k+q_\parallel} = \left(\tilde{E}^e_{k+q_\parallel} + \tilde{E}^h_k\right) b_{-k}a_{k+q_\parallel}$$

$$+ \sum_{k'\neq k,k''} \left[V_{k'-k} b_{-k} \left(a^\dagger_{k'+k''-k} a_{k''} + b^\dagger_{k'+k''-k} b_{k''}\right) a_{k'+q_\parallel} + \ldots\right]$$

$$- \sum_{q_\parallel{}'} a^\dagger_{k+q_\parallel{}'} \sum_{q_z} F_q^* B_{q_z q_\parallel{}'} a_{k+q_\parallel} + \ldots$$

- Coulomb hierarchy problem (as in semiclassical theory)
- photon-carrier coupling hierarchy
- Cluster expansion to treat hierarchies on equal footing

Semiconductor Luminescence Equations II

$$i\hbar\frac{\partial}{\partial t}\Delta\left\langle B_q^\dagger B_{q'}\right\rangle = \hbar\left(\omega_{q'}-\omega_q\right)\Delta\left\langle B_q^\dagger B_{q'}\right\rangle + \sum_k\left(F_q\Pi_{k,q}^* - \text{c.c.}\right)$$

photon assisted interband polarization $\Pi_{k,q} = \Delta\left\langle B_q^\dagger b_{-k} a_{k+q_{||}}\right\rangle$

$$\left(i\hbar\frac{\partial}{\partial t} - \tilde{E}_{k,q_\parallel} + \hbar\omega_q\right)\Pi_{k,q} = \left(1 - f^e_{k+q_\parallel} - f^h_k\right)\sum_{k'} V_{k-k'}\Pi_{k',q}$$

$$+ \left.\frac{\partial\Pi_{k,q}}{\partial t}\right|_{col} + \Omega^{coh}_{k,q} + \Omega^{stim}_{k,q} + F_q S_{k,q}$$

incoherent source $S = \left\langle a^\dagger b^\dagger a b\right\rangle = f^e f^h + \Delta\left\langle a^\dagger b^\dagger a b\right\rangle$

coherent source $\Omega^{coh} \propto P, E$

feedback (cavity) Ω^{stim}

excitonic signatures $\left(1 - f^e - f^h\right)\sum V\Pi$

Quasi Stationary: Luminescence Elliott Equation

$$I_{\mathrm{PL}}(\omega) \propto \mathrm{Im}\Big[\sum_{\nu} \frac{\phi_\nu^r(r=0)}{E_\nu - \hbar\omega - i\gamma_\nu} \sum_{k,k'} \Big(\phi_\nu^l(k)\Big)^\star \langle a_{k'}^\dagger a_k b_{k'}^\dagger b_k \rangle\Big]$$

- $\langle N_\nu \rangle_{corr} \propto \sum_{k,k'} \langle a_{k'}^\dagger a_k b_{k'}^\dagger b_k \rangle = \langle N_\nu \rangle_{\mathrm{plasma}} + \Delta \langle N_\nu \rangle$

with
- $\langle N_\nu \rangle_{\mathrm{plasma}} \propto \sum_k f_k^e f_k^h$
- $\Delta \langle N_\nu \rangle$ „true“ excitons

- excitonic resonances independent of source term
- relative peak height depends on populations

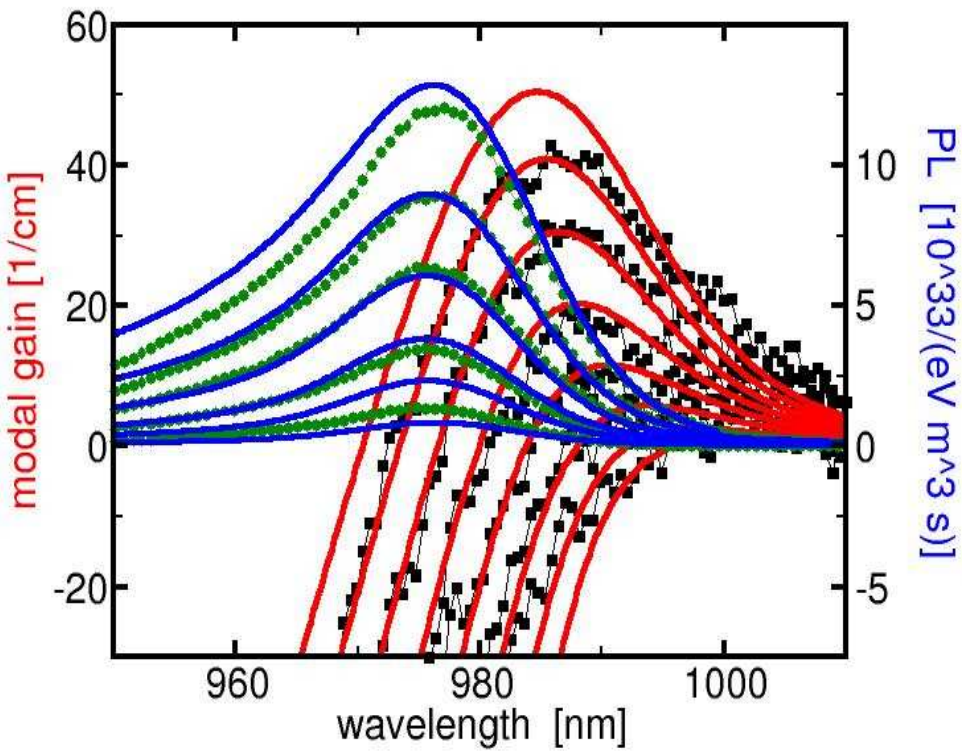
PRL **92**, 067402 (2004)

Gain/Absorption and Luminescence

5nm $Ga_{0.8}In_{0.2}As/GaAs$ pin-MQW

gain: theory: 1.0, 1.125, 1.25, 1.375, 1.5, 1.625, 1.75, 1.875
experiment: 6.0, 6.5, 7.0, 7.5, 8.3, 9.0 [mA]

PL: theory: 0.17, 0.30, 0.40, 0.53, 0.68, 0.86 [10^{12} / cm^2]
experiment: 12, 16, 18, 21, 24 [mW]



Luminescence Analysis

Input:

- nominal structural parameters
- experimental luminescence spectra at two moderate intensities

Analysis:

calculate ideal, homogeneously broadened luminescence spectra for given structure

↓

compare to experimental luminescence

↓

determine from deviations the inhomogeneous broadening, actual structural parameters, quality of device

↓

use optimized parameters to calculate gain/absorption spectra, refractive index, differential gain, linewidth enhancement factor, …

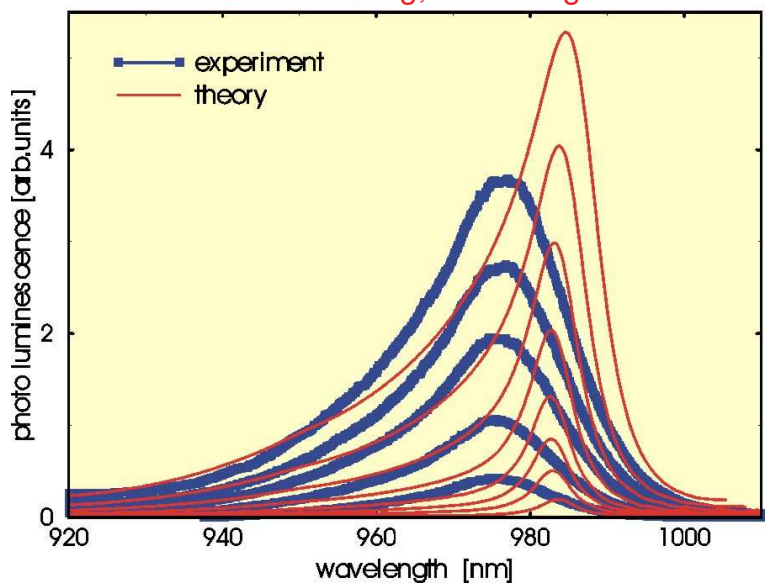
Ref.: Hader et al., IEEE Phot. Techn. Lett. **14**, 762 (2002)

Luminescence Analysis II

- sample with three quantum wells
- nominal composition: 5nm Ga(.8)In(.2)As/GaAs
- sandwiched between n and p doped buffer layers

experiment: 12, 16, 18, 21, 24 mW
theory: 0.1, 0.17, 0.23, 0.3, 0.4, 0.53, 0.68, 0.86 $10^{12}cm^2$

no broadening, no shifting



- needed: slight adjustment of composition
- inhomogeneous broadening

Luminescence Analysis

Input:

- nominal structural parameters
- experimental luminescence spectra at two moderate intensities

Analysis:

calculate ideal, homogeneously broadened luminescence spectra for given structure

↓

compare to experimental luminescence

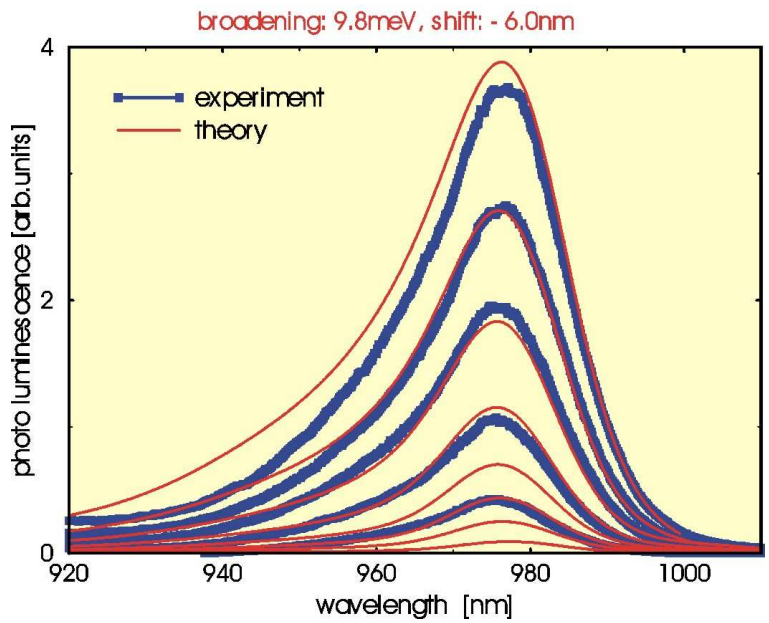
↓

determine from deviations the inhomogeneous broadening, actual structural parameters, quality of device

Luminescence Analysis III

- sample with three quantum wells
- nominal composition: 5nm Ga(.8)In(.2)As/GaAs
- sandwiched between n and p doped buffer layers

experiment: 12, 16, 18, 21, 24 mW
theory: 0.1, 0.17, 0.23, 0.3, 0.4, 0.53, 0.68, 0.86 $10^{12}cm^2$



- good fit to experiment yields actual composition
- less In (0.19) or thinner (- one monolayer)

Luminescence Analysis

Input:

- nominal structural parameters
- experimental luminescence spectra at two moderate intensities

Analysis:

calculate ideal, homogeneously broadened luminescence spectra for given structure

↓

compare to experimental luminescence

↓

determine from deviations the inhomogeneous broadening, actual structural parameters, quality of device

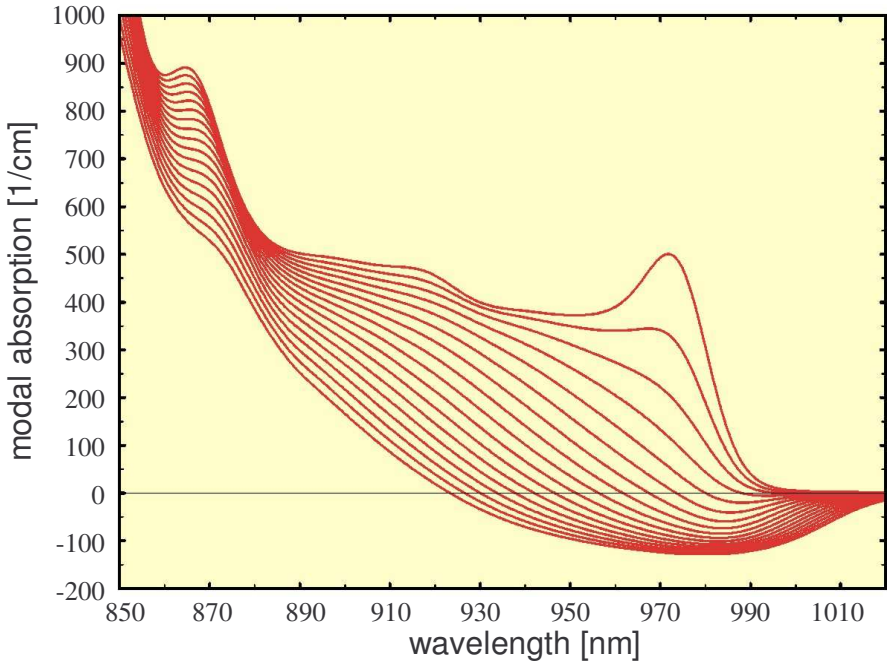
↓

use optimized parameters to calculate gain/absorption spectra

Absorption/Gain

- using optimized material parameters
- experimental inhomogeneous broadening

theory: 0.5, 0.75, 1.0, 1.25, …, 4.25, 4.5 [10^{12}/cm^2]

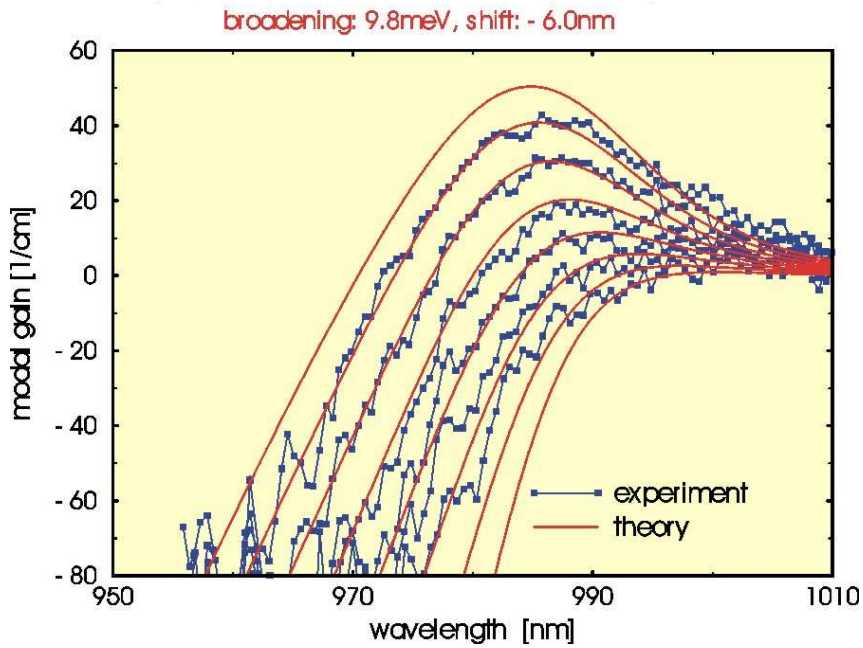


- other results:
- refractive index, alfa parameter, etc.

Test of Predicted Gain Spectra

- independently measured gain spectra
- NO additional fitting!

experiment: 5.0, 6.0, 6.5, 7.0, 7.5, 8.0, 8.6, 10.0 [mV]
theory: 1.0, 1.125, 1.25, 1.375, 1.5, 1.625, 1.75, 1.875 [10^{12}cm^2]

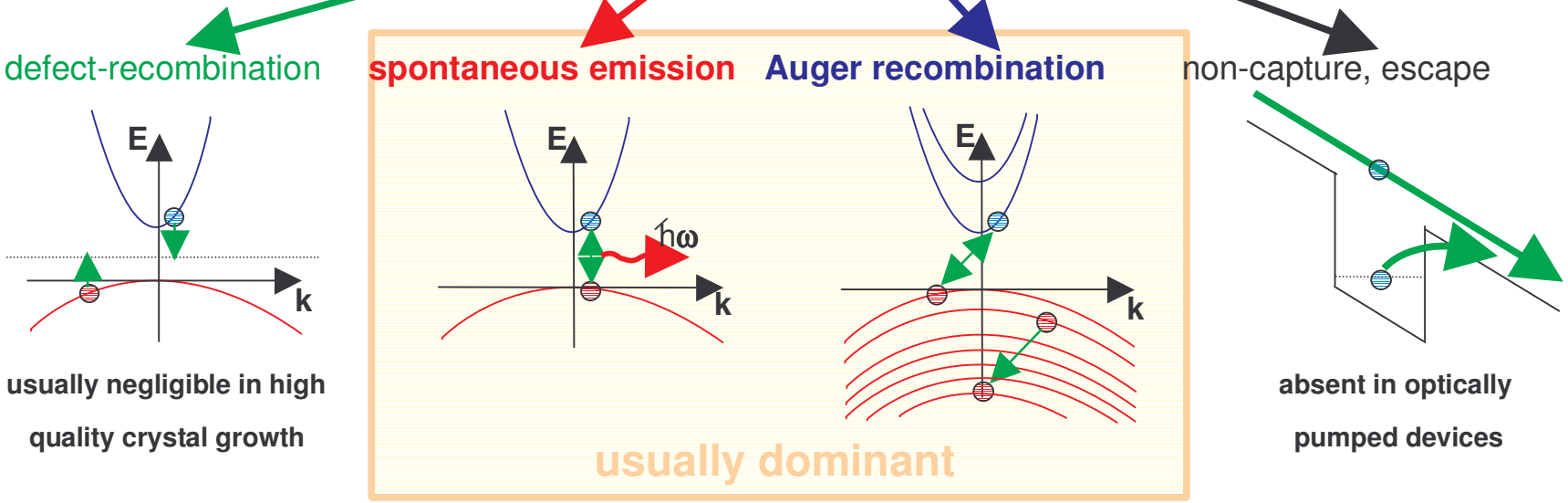


- very good theory/experiment agreement
- verification of microscopic theory
- predictive capabilities

Losses in Semiconductor Lasers

Classical Parametrization of Loss Current J_{loss}:

$$J_{loss} = \mathbf{AN} + \mathbf{BN^2} + \mathbf{CN^3} + J_{rest}$$



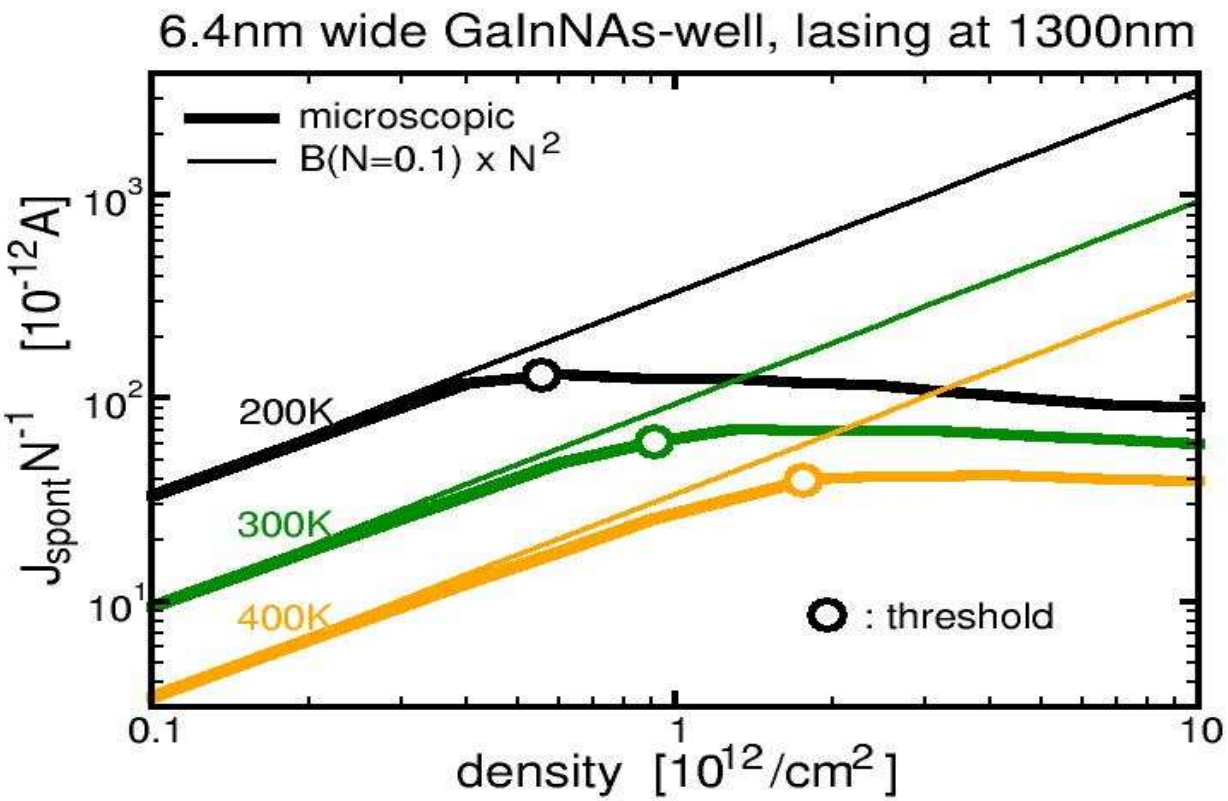
Problems with A, B, C - Parametrization:

- **parameters only very roughly known and only for special cases; depend on well- and barrier-materials, layer widths, temperatures, densities...**

- **simple density-dependence far from reality**

Spontaneous Recombination Current: Examples

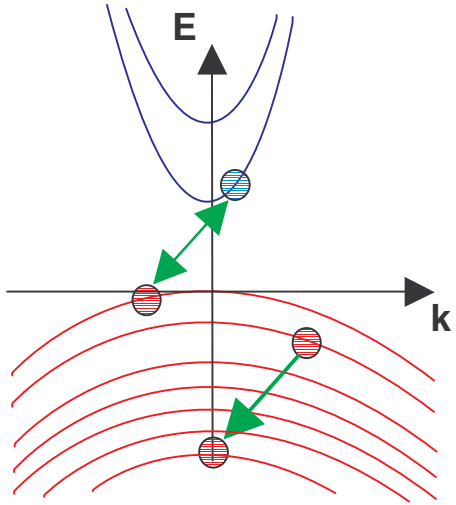
$$J_{spont} = eR_{spont} = e \int d\omega \; I_{PL}(\omega)$$



J_{spont} increases only linear with N at high densities

Auger Recombination I

$$J^e_{aug} = eR^e_{aug} = e\sum_{\mathbf{k},\mathbf{i}} \frac{\partial}{\partial t} f^i_k|_{aug}$$



Inclusion of particle number non-conserving terms:

Impact Ionization

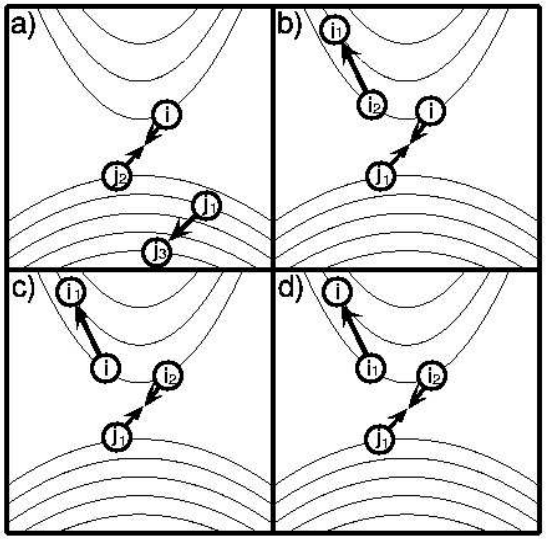
Auger Recombination

$$H_V = 2\sum \Big[a^\dagger_{i_1,s,k+q} b^\dagger_{j_1,s',k'-q} b^\dagger_{j_2,-s,-k} b_{j_3,s',k'} \, V^{i_1,j_3,j_1,j_2}_{s,s',s',-s}(q)$$

$$+ \, b^\dagger_{j_3,s,k+q} b_{j_2,-s',-k'+q} b_{j_1,s,k} a_{i_1,s',k'} \, V^{j_2,j_1,j_3,i_1}_{-s',s,s,s'}(q) \Big] - a \leftrightarrow b$$

Auger Recombination II

Quantum-Boltzmann scattering
in 2. Born-Markov approximation:



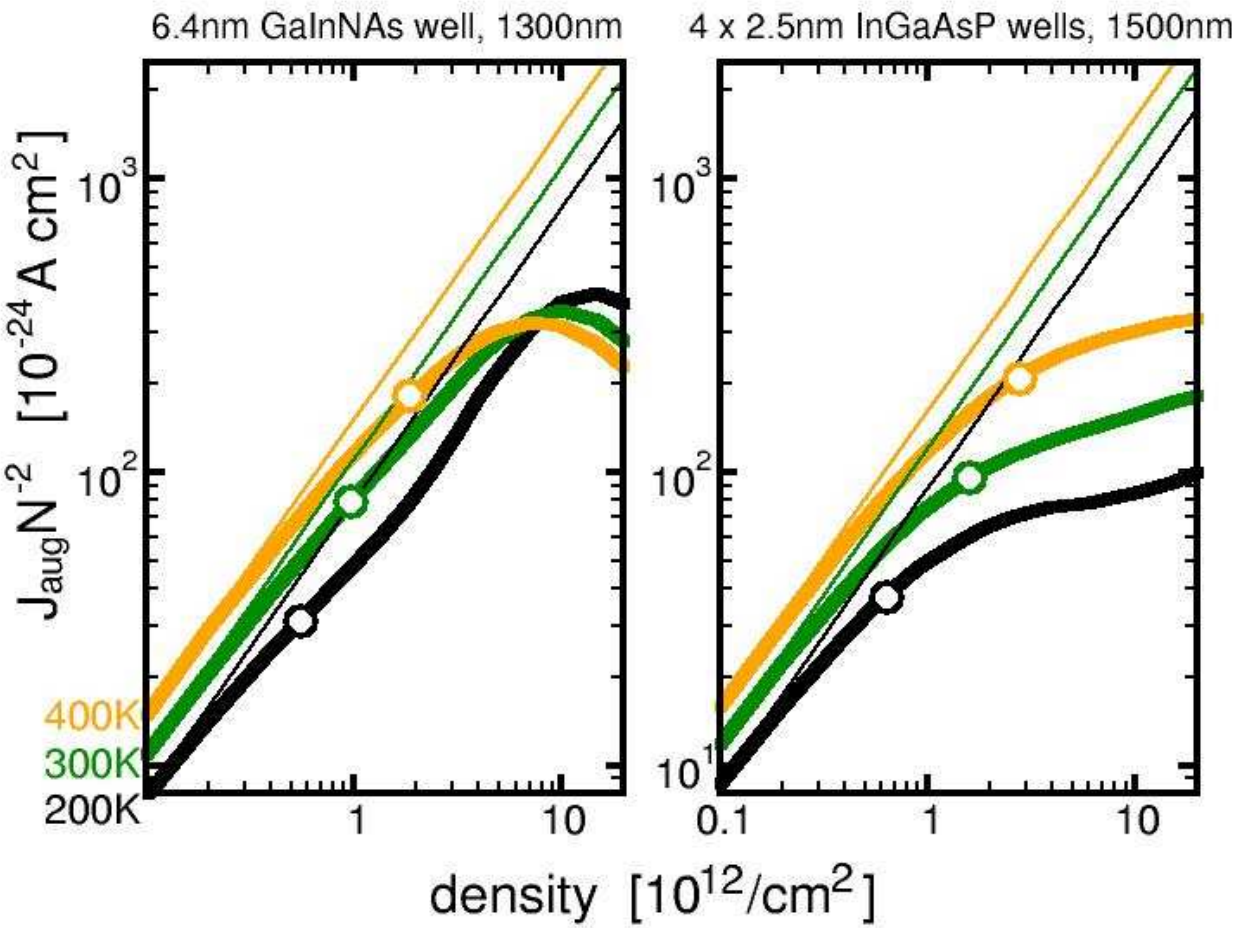
Impact Ionization

$$\frac{\partial}{\partial t} f_k^a|_{aug} = \frac{2\pi}{\hbar} \sum_{k_1,k_2,k_3,b=e,h} |W|^2 \left[f_{k_1}^b [1 - f_{k_2}^c][1 - f_{k_3}^d][1 - f_k^a] \right.$$

$$\left. -[1 - f_{k_1}^b] f_{k_2}^c f_{k_3}^d f_k^a \right] + \ldots$$

Auger Recombination

Auger Recombination: Examples



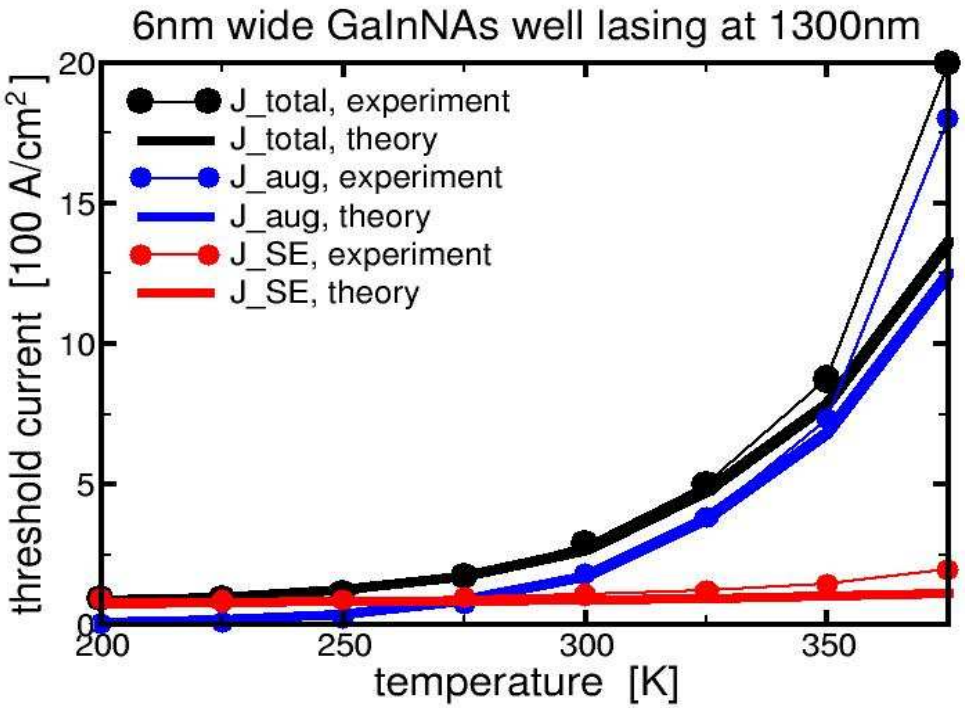
J_{aug} increases far less than cubic with N
sometimes even less than quadratic

Theory-Experiment Comparison I

Theoretical Procedure:

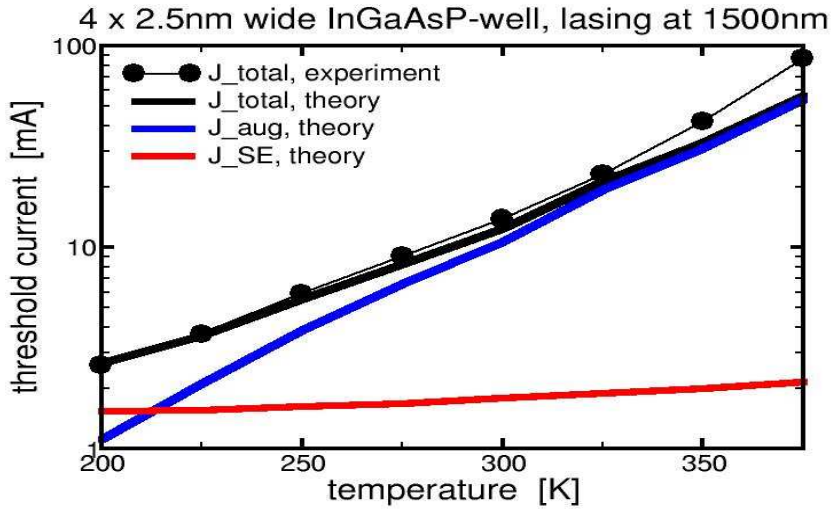
- calculate gain for various densities
- search for density that overcomes intrinsic losses (mirror losses) = threshold density
- calculate spontaneous emission and Auger recombination for this density

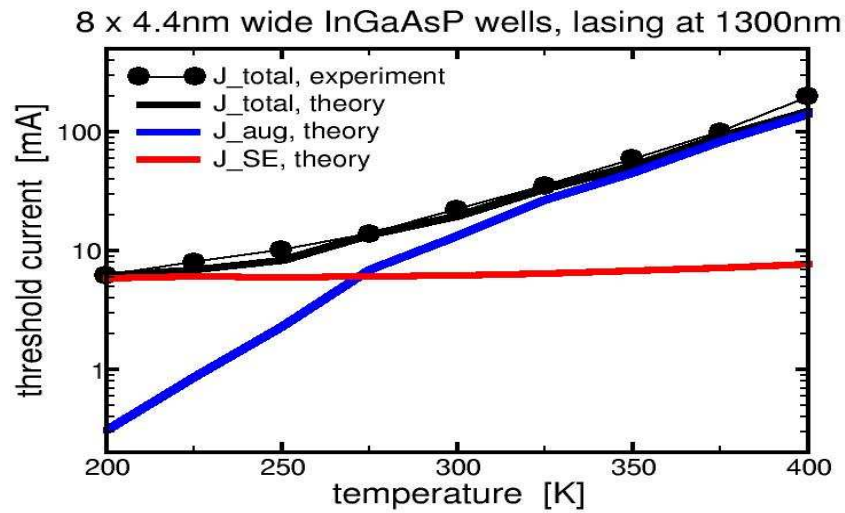
- **comparison with experimental data without adjustment**



(experimental data from R.Fehse, et al., IEEE J. Sel. Topics Quantum Electron. 8, 801 (2002))

Theory-Experiment Comparison II

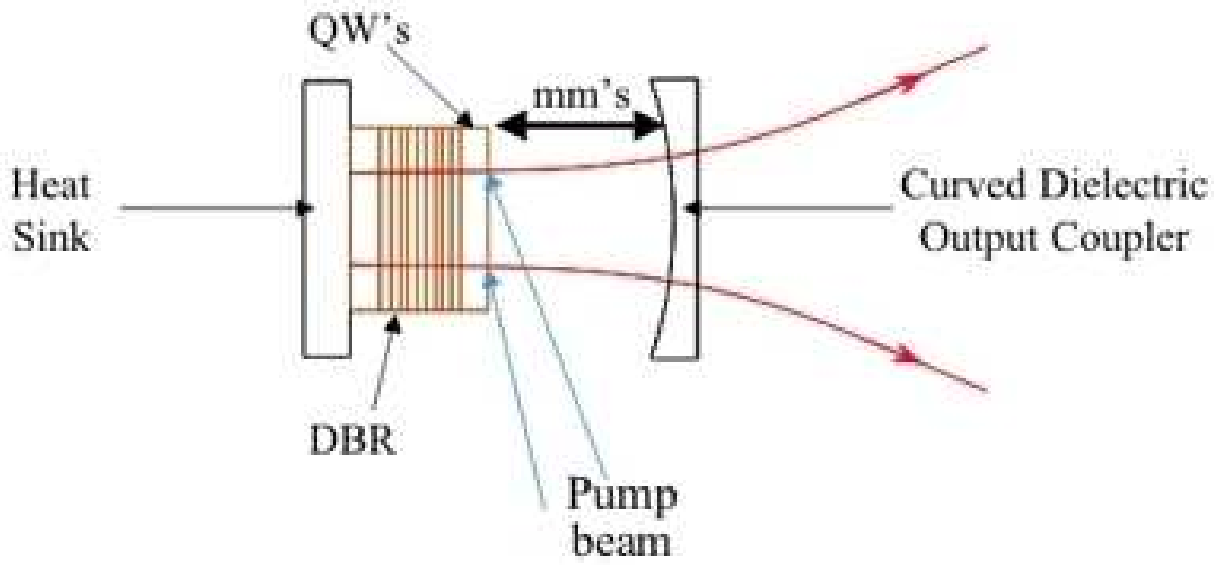




experimental data:
A.F. Phillips et al.,
IEEE J. Sel. Topics
Quantum Electron. 5,
401 (1999))

Laser Modelling: VECSEL

Vertical External Cavity Surface Emitting Laser



Numerical simulation issues:

- optical pumping = input of carriers and kinetic energy
- nonequilibrium carrier distributions, nonequilibrium gain
- dynamic response …..

Nonequilibrium Gain Simulations

- Semiconductor Bloch Equations

$$\left[i\hbar\frac{\partial}{\partial t} - \epsilon_k^e - \epsilon_k^h\right] P_k = \left[1 - f_k^e - f_k^h\right]\Omega_k + \frac{\partial}{\partial t}P_k|_{corr}$$

$$i\hbar\frac{\partial}{\partial t}\, f_k^a = -\Omega_k(t)P_k^* + \Omega_k^* P_k + \frac{\partial}{\partial t}f_k^a|_{corr}$$

- Reduced Wave Equation

$$\frac{dE(t)}{dt} = -\gamma_c E(t) + \frac{i\omega\Gamma}{\epsilon_B V_{qw}}\sum_k \mu_k^* P_k(t)$$

Full numerical solution way too time consuming

APPROACH

- Calculate rates for scattering/relaxation/dephasing
- Solve coupled equations
- Interesting situations: pulsed and/or CW excitation

Relaxation Rate Model for Scattering

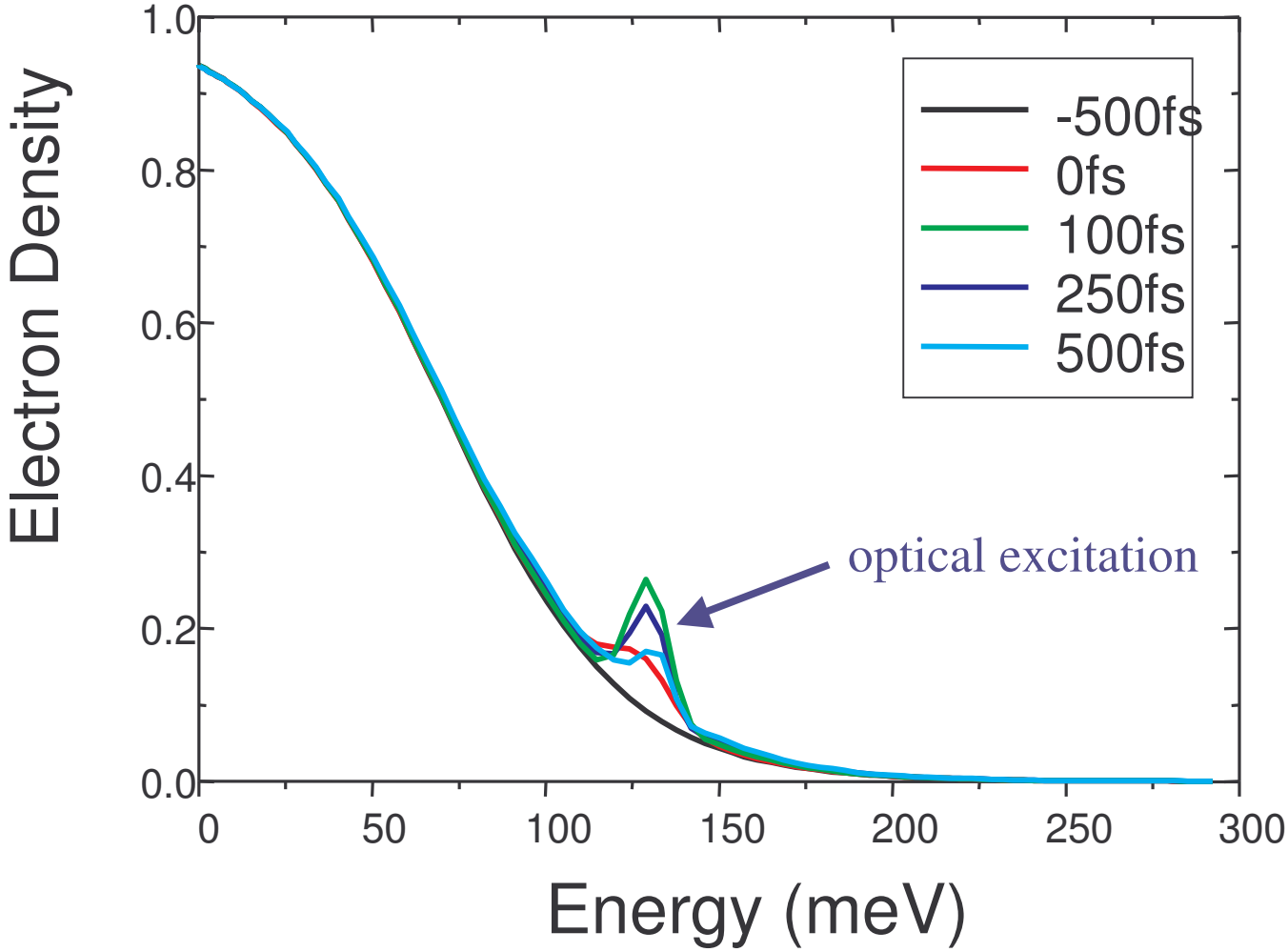
$$\frac{\partial}{\partial t} P_k|_{corr} = -\gamma_p P_k(t)$$

$$\frac{\partial}{\partial t} f_k^\alpha|_{corr} = -\gamma_{c-c}\left[f_k^\alpha - F_k^\alpha(\mu_p^\alpha, T_p)\right] - \gamma_{c-p}\left[f_k^\alpha - F_k^\alpha(\mu_l^\alpha, T_l)\right]$$

Parameters

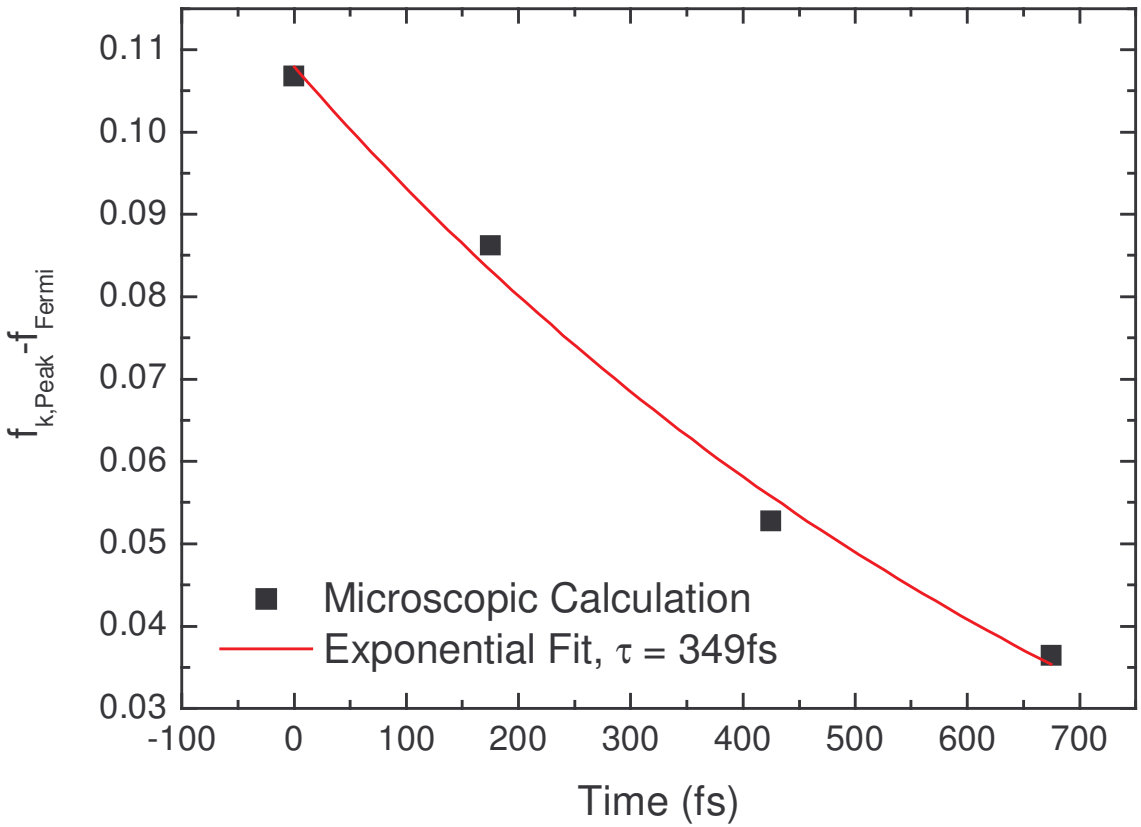
- Relaxation rates are obtained by fits to microscopic calculations
- $F_k^\alpha(\mu_p^\alpha, T_p)$ is calculated considering energy and particle conservation
- $F_k^\alpha(\mu_l^\alpha, T_l)$ is calculated considering particle conservation
- Carrier-carrier scattering is an order of magnitude faster than carrier-phonon scattering
- Band structure is taken from microscopic calculations

Carrier Dynamics



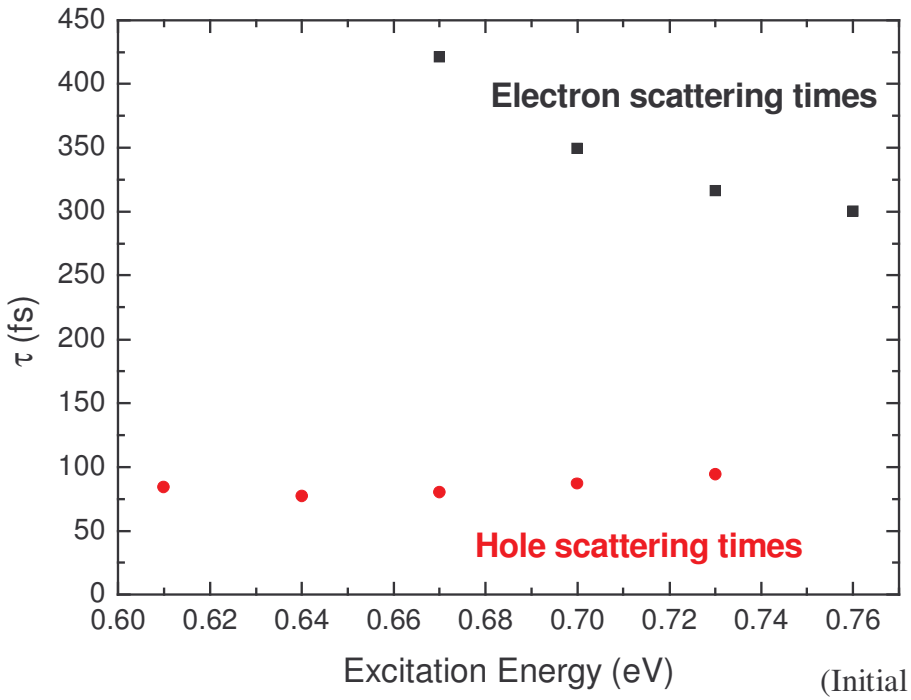
10nm $Ga_{0.764}In_{0.236}As$ QW between 20.3nm $GaN_{0.007}As_{0.993}$,
Excitation by a 50fs pulse at 1.35eV at t=0

Effective Carrier-Carrier Scattering Times



- simple evaluation
- exponential fits work well

Electron-Hole Scattering Times

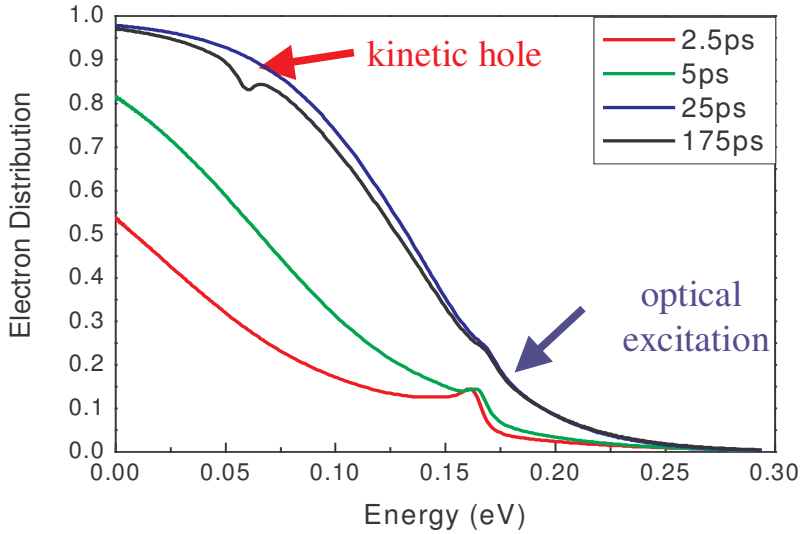


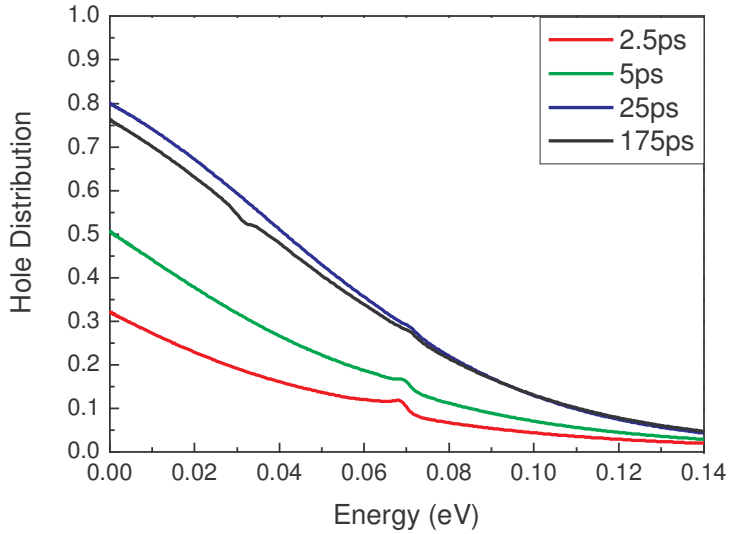
(Initial Density $2.0*10^{12}/cm^{-2}$)

- Hole scattering faster because of larger effective mass
- Variation of scattering times with excitation energy (band structure vs availability of scattering partners)
- Increased scattering time for lower densities (e.g. for $1.5 * 10^{12}$ cm^{-2} τ_e = 354 fs, τ_h = 94 fs, instead of τ_e = 349 fs, τ_h = 87 fs at $2.0 * 10^{12}$ cm^{-2})

Switch-On of Optically Pumped SCL

• **dynamics of electron and hole distributions**

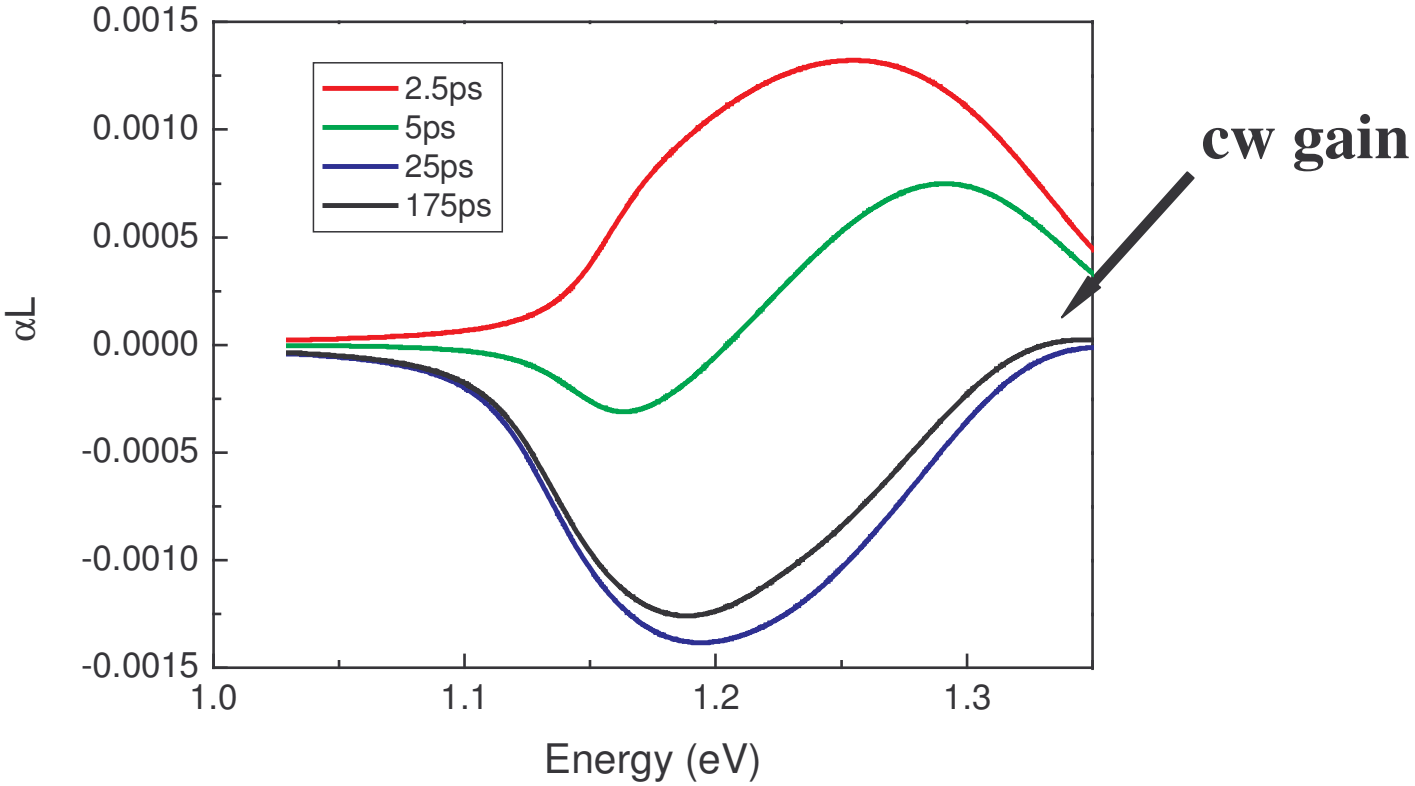




• distinct kinetic hole in electron distribution

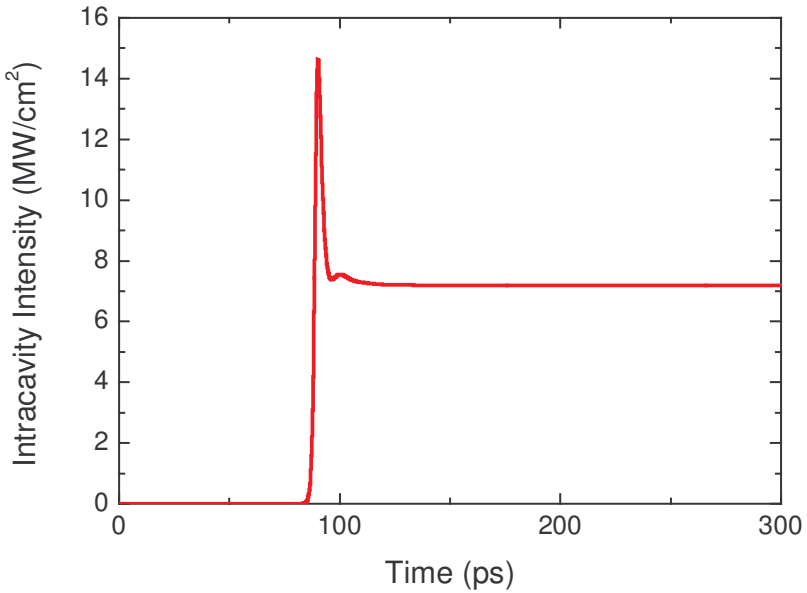
• small kinetic hole in hole distribution due to faster scattering time

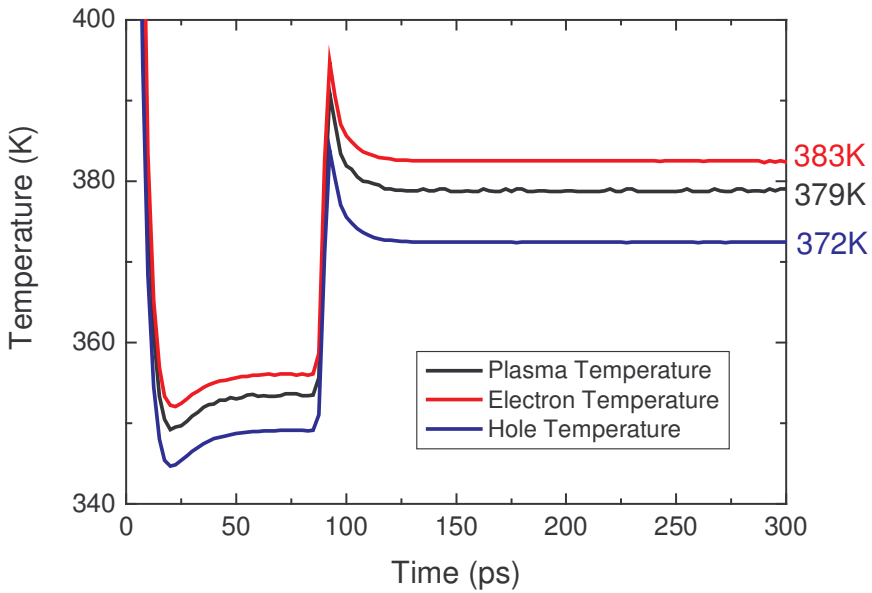
Gain Dynamics



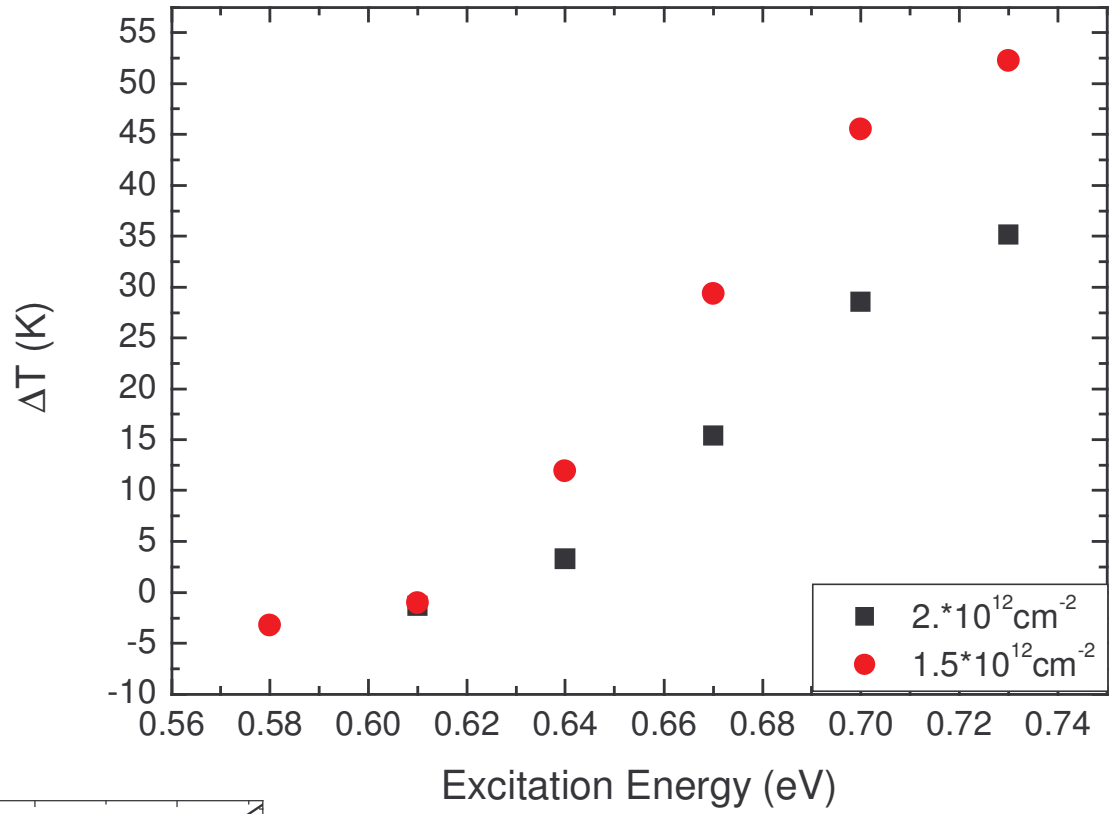
- initial absorption turns into gain
- transient gain overshoot

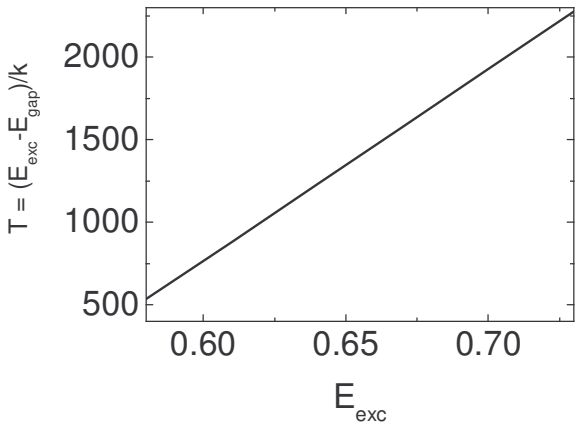
Carrier Temperature and Laser Intensity





Excitation Induced Temperature Change





- “hot”carriers (relative to the bandgap) can still cool down carrier distribution
- excitation energy = 0.61 eV => Excitation at energy chemical potential => $\Delta T=0$
- hole chemical potential always in the bandgap

Summary

- predictive microscopic theory
- application to semiconductor laser gain media
- luminescence, Auger loss rates
- nonequilibrium gain, laser dynamics

MANY CHALLENGES:

- non-classical properties (quantum optics, quantum information science, …)
- modified photonic environment (phot. x-tals, …)
- microscopic modelling of disorder

References and more informations:

http://www.physik.uni-marburg.de (look for: semiconductor theory)

http://www.nlcstr.com