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

\[ P = \sum_k d_{cv}^* P_k + c.c. \]

two-band model, one QW subband

\[
\begin{pmatrix}
\langle a^+_k a_k \rangle & \langle b_{-k} a_k \rangle \\
\langle a^+_k b^+_{-k} \rangle & \langle b^+_k b_k \rangle
\end{pmatrix}
= 
\begin{pmatrix}
f^c_k & P_k \\
P^*_k & f^v_k
\end{pmatrix}
\]

\(P_k\) ... e-h-pair transition amplitude
\(f^c_k, f^v_k\) ... occupation probabilities
Hamiltonian

\[ H = H_0 + H_{\text{Coul}} + H_{\text{dip}} \]

- \( H_0 \) single particle (band structure)
- \( H_{\text{Coul}} \) Coulomb interaction between carriers
- \( H_{\text{dip}} \) dipole interaction with optical field
Calculation of Optical Response

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

structure of resulting equations:

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

infinite hierarchy of many-body correlations

systematic CLUSTER expansion
Schematics of Cluster Expansion

- two-point level (uncorrelated single particles)

\[ i\hbar \frac{\partial}{\partial t} \langle \{1\}^\dagger \{1\} \rangle = T [\langle \{1\}^\dagger \{1\} \rangle] + V [\langle \{2\}^\dagger \{2\} \rangle] \]

\[ = T [\langle \{1\}^\dagger \{1\} \rangle] + V [\langle \{2\}^\dagger \{2\} \rangle_s] + V [\Delta \langle \{2\}^\dagger \{2\} \rangle] \]

- four-point level (two particle correlations)

\[ i\hbar \frac{\partial}{\partial t} \Delta \langle \{2\}^\dagger \{2\} \rangle = T [\Delta \langle \{2\}^\dagger \{2\} \rangle] + V [\langle \{3\}^\dagger \{3\} \rangle] \]

\[ = T [\Delta \langle \{2\}^\dagger \{2\} \rangle] + V [\langle \{3\}^\dagger \{3\} \rangle_s] + V [\langle \{3\}^\dagger \{3\} \rangle_D] + V [\Delta \langle \{3\}^\dagger \{3\} \rangle] \]
Semiconductor Bloch Equations

\[
\begin{align*}
\left[i\hbar \frac{\partial}{\partial t} - \epsilon_k^e - \epsilon_k^h \right] P_k &= [1 - f_k^e - f_k^h] \Omega_k + \frac{\partial}{\partial t} P_k \big|_{\text{corr}} \\
\frac{i\hbar}{\partial t} f_k^a &= -\Omega_k(t) P_k^* + \Omega_k^* P_k + \frac{\partial}{\partial t} f_k^a \big|_{\text{corr}}
\end{align*}
\]

HF field renormalization

\[
\Omega_k(t) = d_{cv} E^{QW}(t) + \sum_{k'} V_{k-k'} P_{k'}(t)
\]

HF energy renormalization

\[
\epsilon_k^a(t) = \epsilon_k^a - \sum_{k'} V_{k-k'} f_{k'}^a(t)
\]

- **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

\[
\frac{\partial}{\partial t} f_{k}^{a}(t) |_{corr} = \sum_{k}^{in,a} (t)[1 - f_{k}^{a}(t)] - \sum_{k}^{out,a} (t) f_{k}^{a}(t)
\]

Coulomb scattering

phonon scattering
Coulomb Scattering I

\[
\frac{\partial}{\partial t} f^a_k(t)|_{corr} = \Sigma^{in,a}_k(t)[1 - f^a_k(t)] - \Sigma^{out,a}_k(t)f^a_k(t)
\]

scattering rates:

\[
\frac{\partial}{\partial t} f^a_k|_{corr} \approx \sum_{k_1,k_2,k_3,b=e,h} |W|^2 f^a_{k_1} f^b_{k_2} [1 - f^b_{k_3}] [1 - f^a_k] - \ldots
\]
Coulomb Scattering II

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

quasi-equilibrium: \[
\frac{\partial}{\partial t} f_k^a|_{\text{corr}} = 0
\]
detailed balance

Fermi-Dirac distribution: \[
f_k^a = \Rightarrow \quad F_k^a = \frac{1}{e^{(E_k - \mu_a)/k_BT} + 1}
\]
Excitation Induced Dephasing

\[
\left[ i\hbar \frac{\partial}{\partial t} - \varepsilon^e_k(t) - \varepsilon^h_k(t) \right] P_k(t) - \left[ 1 - f^e_k(t) - f^h_k(t) \right] \Omega_k(t) = i \left[ \Gamma_k(t) P_k(t) + \sum_{k'} \Gamma_{k,k'}(t) P_{k'}(t) \right]
\]

- \( \text{Re} \ \Gamma_k(t) \) ... diagonal dephasing
  \( \Rightarrow \) generalized \( T_2 \) time
- \( \text{Im} \ \Gamma_k(t) \) ... diagonal energy shift
  \( \Rightarrow \) generalized band-gap shift
- \( \text{Re} \ \Gamma_{k,k'}(t) \) ... off-diagonal dephasing
- \( \text{Im} \ \Gamma_{k,k'}(t) \) ... 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 \textit{et al.}, PRL 77, 5257 (1996).
Density Dependent Absorption


- In Transmission

- Detuning \((h\omega - E_G)/E_B\)

- experiment: InGaAs/GaAs QW
- EID first observed in 4-wave mixing, Wang et al. PRL 71, 1261 (1993)
• $\Delta = (\hbar \omega - E_G)/E_B$
• gain of two-band bulk material
• nondiagonal scattering contributions $\rightarrow$ lineshape modification, no absorption below the gap

Hughes, Knorr, Koch, Binder, Indik, and Moloney
Optical Gain: Theory and Experiment

InGaAs QW

In- and out scatterings
Lorentzian (T₂=100 fs)

Photon Energy (eV)
Current 0-20mA… Density 0.6-3.0×10¹²cm⁻²

exp: C. Ellmers et al., theory: A. Girndt et al. Marburg

Detuning
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

8nm InGaAs/AlGaAs

Detuning Δ
Current 0-20mA… Density 0.6-3.0×10¹²cm⁻²
exp: C. Ellmers et al., theory: A. Girndt et al. Marburg

6.8 nm InGa₄P/(Al₁Ga₃)₄In0.5P₁.49

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

\[ i\hbar \frac{\partial}{\partial t} B_{q_z q_\parallel} = \hbar \omega_q B_{q_z q_\parallel} + F_q \sum_k b_{-k} a_{k+q_\parallel} - F_q^* \sum_k a_{k-q_\parallel}^\dagger b_{-k}^\dagger \]

- polarization operator dynamics

\[ i\hbar \frac{\partial}{\partial t} b_{-k} a_{k+q_\parallel} = \left( \tilde{E}_{k+q_\parallel}^e + \tilde{E}_{k}^h \right) b_{-k} a_{k+q_\parallel} + \sum_{k' \neq k, k''} \left[ V_{k'-k} b_{-k} \left( a_{k'+k''-k}^\dagger a_{k''} + b_{k'+k''-k}^\dagger b_{k''} \right) a_{k'+q_\parallel} + \ldots \right] \]

\[ - \sum_{q_\parallel'} a_{k+q_\parallel'}^\dagger \sum_{q_z} F_{q_z}^* B_{q_z q_\parallel'} a_{k+q_\parallel} + \ldots \]

- Coulomb hierarchy problem (as in semiclassical theory)

- photon-carrier coupling hierarchy

- Cluster expansion to treat hierarchies on equal footing
Semiconductor Luminescence Equations II

\[ i\hbar \frac{\partial}{\partial t} \Delta \langle B^\dagger_q B_{q'} \rangle = \hbar (\omega_{q'} - \omega_q) \Delta \langle B^\dagger_q B_{q'} \rangle + \sum_k (F_q \Pi^*_k,q - \text{c.c.}) \]

photon assisted interband polarization \( \Pi_{k,q} = \Delta \langle B^\dagger_q b_{-k} a_{k+q,||} \rangle \)

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

\[ + \left. \frac{\partial \Pi_{k,q}}{\partial t} \right|_{\text{co}l} + \Omega_{k,q}^{\text{coh}} + \Omega_{k,q}^{\text{stim}} + F_q S_{k,q} \]

incoherent source \( S = \langle a^\dagger b^\dagger a b \rangle = f^e f^h + \Delta \langle a^\dagger b^\dagger a b \rangle \)

coherent source \( \Omega^{\text{coh}} \propto P, E \)

feedback (cavity) \( \Omega^{\text{stim}} \)

excitonic signatures \( (1 - f^e - f^h) \sum V \Pi \)
Quasi Stationary: Luminescence Elliott Equation

\[ I_{PL}(\omega) \propto \text{Im} \left[ \sum_{\nu} \frac{\phi^r_{\nu}(r = 0)}{E_{\nu} - \hbar \omega - i\gamma_{\nu}} \sum_{k,k'} \left( \phi^l_{\nu}(k) \right)^* \langle a_{k}^{\dagger}, a_{k} b_{k'}^{\dagger}, b_{k} \rangle \right] \]

- \[ \langle N_{\nu} \rangle_{\text{corr}} \propto \sum_{k,k'} \langle a_{k}^{\dagger}, a_{k} b_{k'}^{\dagger}, b_{k} \rangle = \langle N_{\nu} \rangle_{\text{plasma}} + \Delta \langle N_{\nu} \rangle \]

with
- \[ \langle N_{\nu} \rangle_{\text{plasma}} \propto \sum_{k} f_{k}^{e} f_{k}^{h} \]
- \[ \Delta \langle N_{\nu} \rangle \text{ "true" excitons} \]
- excitonic resonances independent of source term
- relative peak height depends on populations

PRL 92, 067402 (2004)
Gain/Absorption and Luminescence

5nm $\text{Ga}_{0.8}\text{In}_{0.2}\text{As/GaAs}$ 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, …

Luminescence Analysis II

- sample with three quantum wells
- nominal composition: 5nm Ga(0.8)In(0.2)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 \(10^{12}\text{cm}^2\)

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 \([10^{12}/\text{cm}^2]\)

- 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
Classical Parametrization of Loss Current $J_{\text{loss}}$:

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

- **Defect-recombination**: Usually negligible in high quality crystal growth.
- **Spontaneous emission**: Absent in optically pumped devices.
- **Auger recombination**: Usually dominant.
- **Non-capture, escape**: 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

\[ J_{\text{spon}} = eR_{\text{spon}} = e \int d\omega \ I_{PL}(\omega) \]

6.4nm wide GaInNAs-well, lasing at 1300nm

\[ J_{\text{spon}} N^{-1} \text{[10^{-12} A]} \] vs. density \( [10^{12}/\text{cm}^2] \)

- Microscopic
- \( B(N=0.1) \times N^2 \)

\( J_{\text{spon}} \) increases only linear with \( N \) at high densities
Auger Recombination I

\[ J_{aug}^e = eR_{aug}^e = e \sum_{k,i} \frac{\partial}{\partial t} f_k^i \bigg|_{aug} \]

Inclusion of particle number non-conserving terms:

\[ H_V = 2 \sum \left[ a_{i_1,s,k+q}^{\dagger} b_{j_1,s',k'}^{\dagger} b_{j_2,-s,-k} b_{j_3,s',k'} V_{s,s',s',-s}^{i_1,j_3,j_1,j_2}(q) 
+ b_{j_3,s,k+q}^{\dagger} b_{j_2,-s',-k'} b_{j_1,s,k} a_{i_1,s',k'} V_{s',s,s',s}^{j_2,j_1,j_3,i_1}(q) \right] - a \leftrightarrow b \]
Auger Recombination II

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

$$\frac{\partial}{\partial t} f^a_k|_{aug} = \frac{2\pi}{\hbar} \sum_{k_1,k_2,k_3,b=e,h} |W|^2 \left[ f^b_{k_1} [1 - f^c_{k_2}] [1 - f^d_{k_3}] [1 - f^a_k] ight]$$

$$- [1 - f^b_{k_1} f^c_{k_2} f^d_{k_3} f^a_k] + ...$$
Auger Recombination: Examples

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

Theory-Experiment Comparison I

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

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

• 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

Electron Density

Energy (meV)

optical excitation
Effective Carrier-Carrier Scattering Times

- \( f_{k, \text{Peak}} - f_{\text{Fermi}} \)

- Simple evaluation

- Exponential fits work well

\( t = 349 \text{fs} \)
**Electron-Hole Scattering Times**

- 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\(^{-2}\) \(\tau_e = 354\) fs, \(\tau_h = 94\) fs, instead of \(\tau_e = 349\) fs, \(\tau_h = 87\) 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
• initial absorption turns into gain

• transient gain overshoot
Carrier Temperature and Laser Intensity

- Intracavity Intensity (MW/cm²) vs. Time (ps)
- Temperature (K) vs. Time (ps)

Graphs showing the relationship between intracavity intensity and time, as well as temperature and time, under different conditions.
• “hot” carriers (relative to the bandgap) can still cool down carrier distribution
• excitation energy = 0.61 eV $\Rightarrow$ Excitation at energy chemical potential $\Rightarrow \Delta T=0$
• 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