8th International Quantum Cascade Laser School and Workshop (IQCLSW 2018)
Cassis, France

Modeling electron transport in quantum cascade lasers

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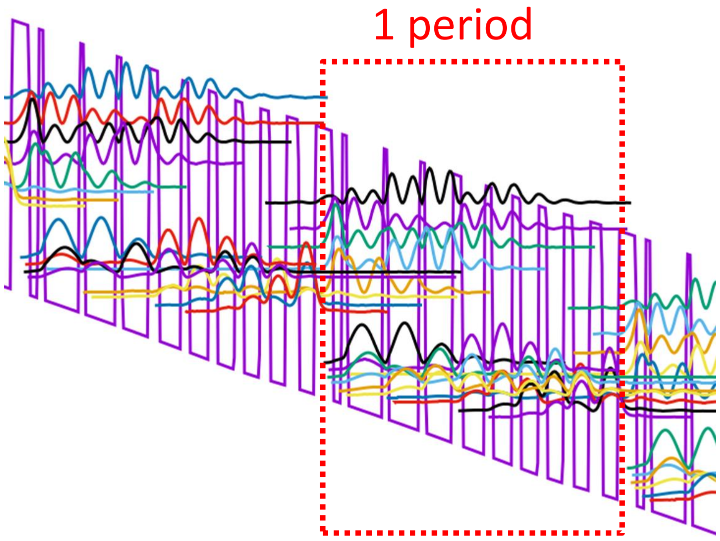
EC Horizon 2020 - FET Open

nextnano
Software for semiconductor nanodevices

Electron transport in QCLs



Essential ingredients to model electron transport in QCLs?

- Quantum confinement
- Scattering processes

Other ingredients:

- Coherent effects? (tunneling vs hopping between eigenstates)
- Broadening effects? is energy conserved for each scattering event?

Outline

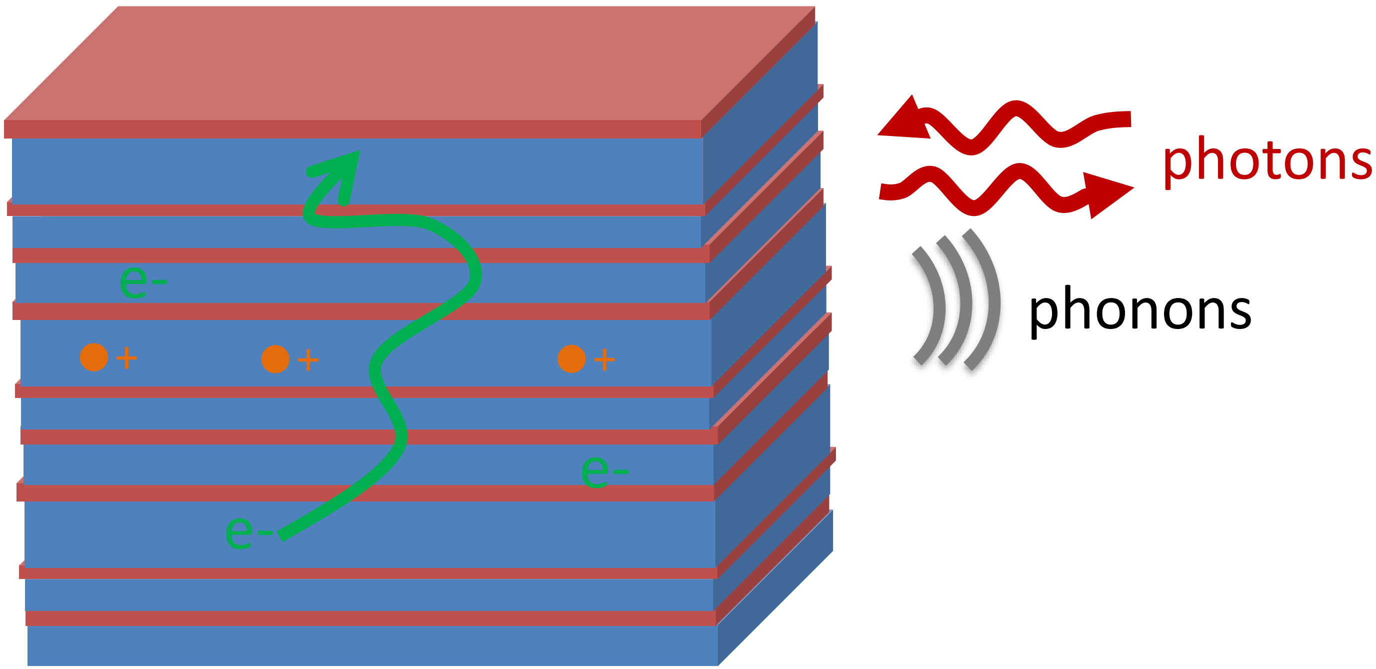
- **Essential ingredients for modeling QCLs: electronic structure and scattering processes**

- Different formalisms from semi-classical to quantum transport
  - Rate equation for populations
  - Density matrix
  - Non-equilibrium Green's functions (NEGF)

- Development of a commercial NEGF simulator: nextnano.QCL

- New physical insights QCLs

Hamiltonian of an electron in a QCL



Hamiltonian of a single charge carrier (3D problem)

$$H = H_{\mathrm{e}}^{\mathrm{3D}} + H_{\text{e-e}} + H_{\text{e-phonon}} + H_{\text{e-photon}}$$

Hamiltonian of an electron in a QCL

$$H = H_{\text{e}}^{\text{3D}} + H_{\text{e-e}} + H_{\text{e-phonon}} + H_{\text{e-photon}}$$

- Separate into an exactly solvable part and a scattering part (treated in perturbation)

$$H = H_0 + H_{scatt}$$

$$H_0 = H_e^{\text{ideal}} + H_{\text{e-e}}^{\text{mean-field}}$$

$$H_e^{\text{ideal}} = \frac{\hat{p}_z^2}{2m^*} + \hat{V}(z) - eF\hat{z} + \frac{\hat{p}_x^2 + \hat{p}_y^2}{2m^*}$$

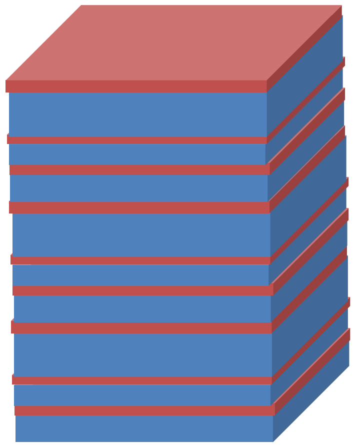
1D Schrödinger equation

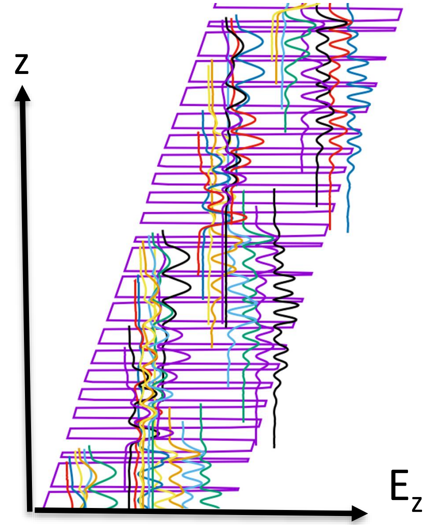
2D free motion

$$H_{scatt} = H_e^{\text{disorder}} + H_{\text{e-e}}^{\text{scatt}} + H_{\text{e-phonon}} + H_{\text{e-photon}}$$

- Interface roughness
- Alloy disorder
- Charged impurity scattering

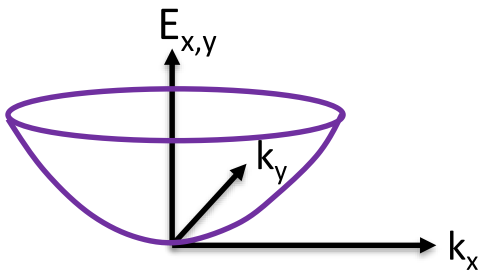
Ideal case: 1D and 2D motions decoupled





$$H_e(z) = \frac{\hat{p}_z^2}{2m^*} + \hat{V}(z) - eF\hat{z}$$

Eigenstates = Wannier-Stark states

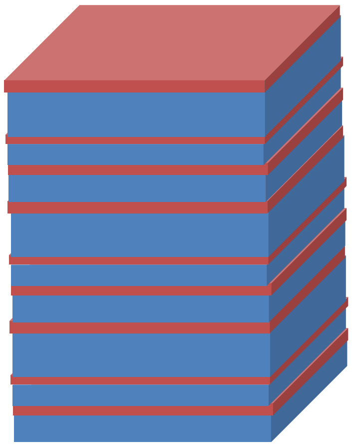


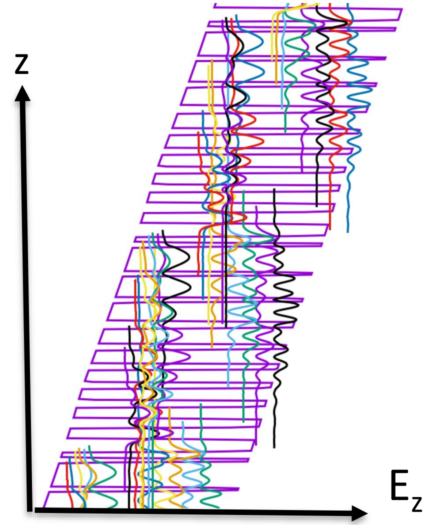
$$E_\perp = \hbar^2 \frac{(\hat{k}_x^2 + \hat{k}_y^2)}{2m^*}$$

Free in-plane motion: subbands

If no scattering: no transport, only Bloch oscillations

Scattering procees





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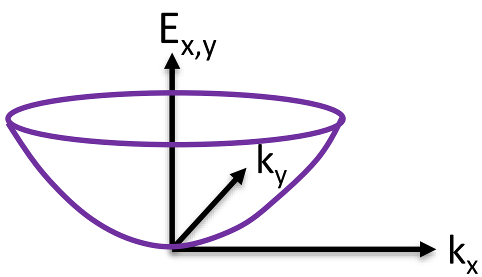
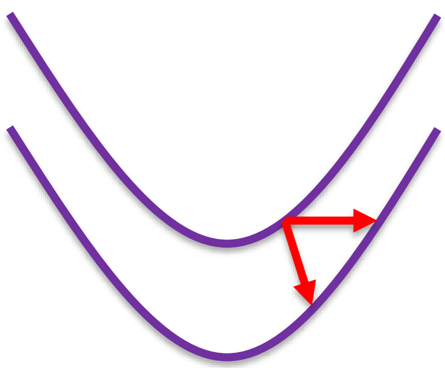


$$E_\perp = \hbar^2 \frac{(\hat{k}_x^2 + \hat{k}_y^2)}{2m^*}$$

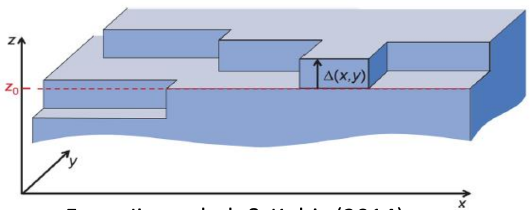
Free in-plane motion: subbands



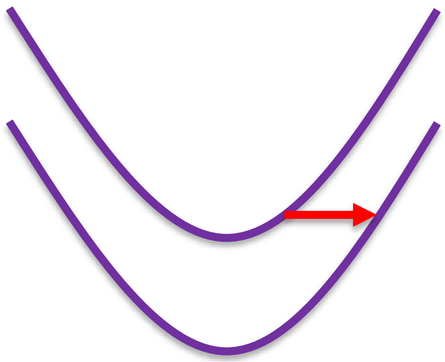
Scattering processes couple the 1D and 2D motions

Non-radiative scattering processes

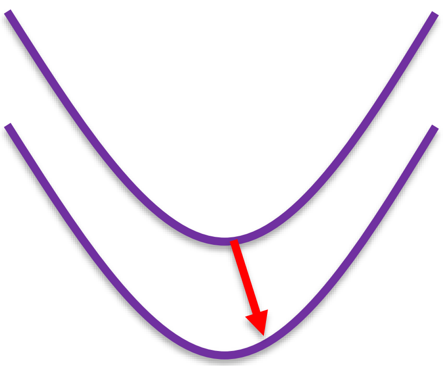
- Disorder effects that breaks the 2D invariance induces **elastic scattering** processes
    - Charged impurities (ionized dopants)
    - Alloy disorder
    - Rough interfaces



From Jirauschek & Kubis (2014)

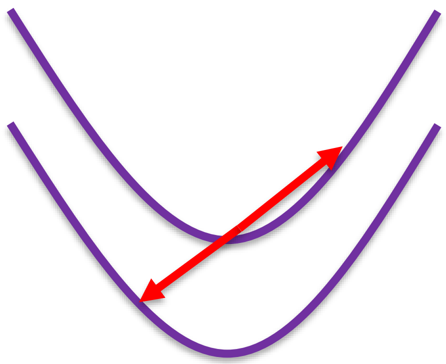


- Coupling to phonons: **inelatic scattering** processes
    - **Optical phonons**
    - Acoustic phonons (usually very weak)



Non-radiative scattering processes

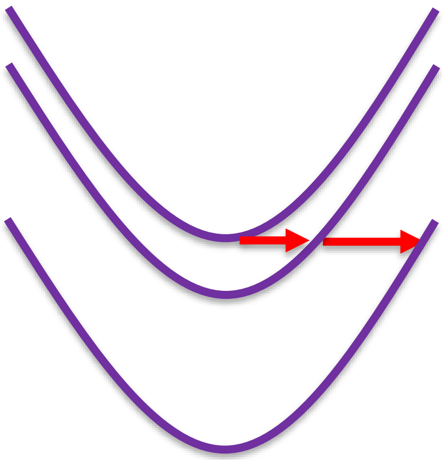
- Electron-electron scattering



- Conservation of total energy and total momentum: inelastic process for a given electron but energy conservation in total

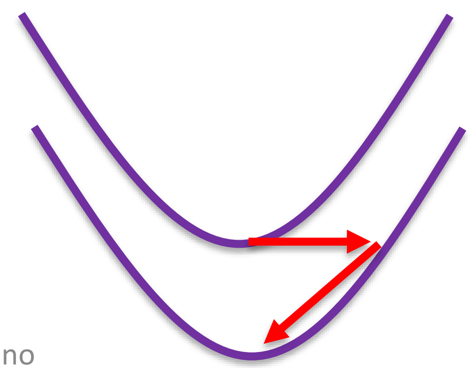
Non-radiative scattering processes

Elastic scattering processes alone?



infinitely increasing electron temperature

- Combination of elastic and inelastic scattering processes



Intersubband elastic process
+ intrasubband inelastic process

Outline

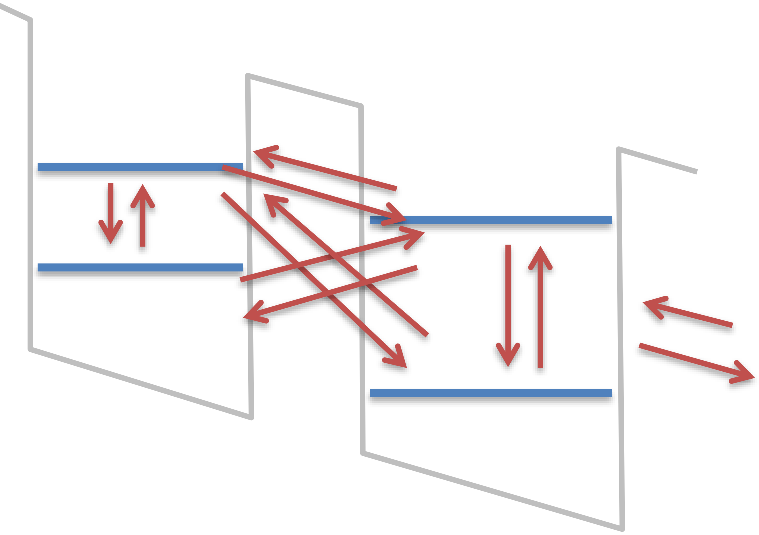
- Essential ingredients for modeling QCLs: electronic structure and scattering processes

- **Different formalisms from semi-classical to quantum transport**
  - **Rate equation for populations**
  - **Density matrix**
  - **Non-equilibrium Green's functions (NEGF)**

- Development of a commercial NEGF simulator: nextnano.QCL

- New physical insights QCLs

Rate equations for populations



$$\frac{dn_i}{dt} = \sum_j \frac{n_j}{\tau_{j\to i}} - n_i \sum \frac{1}{\tau_{i\to j}}$$

$$= 0$$ in steady state

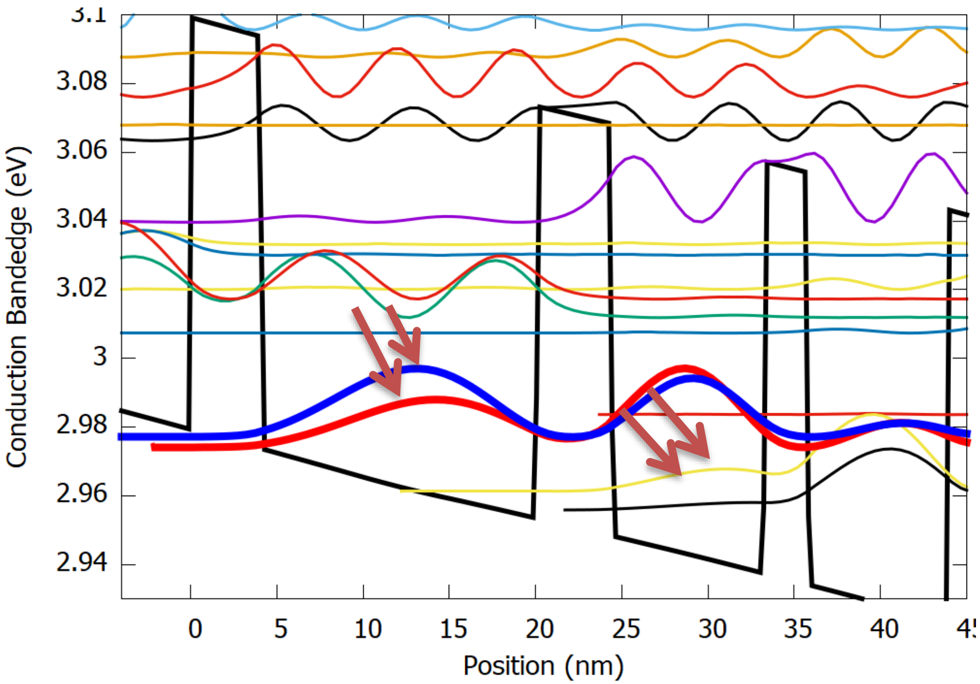
Scattering rates can be calculated using Fermi Golden rule.
For elastic processes:

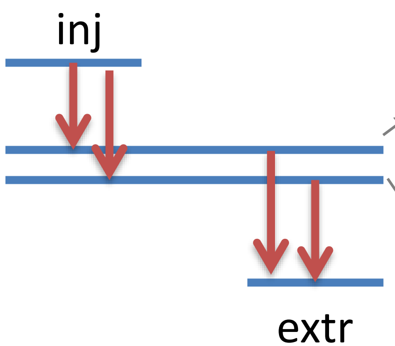
$$\frac{1}{\tau^{\text{elastic}}_{(i,k)\to j}} = \frac{2\pi}{\hbar} \sum_{k'} \langle i, k | H^{\text{elastic}}_{\text{scatt}} | j, k' \rangle \delta \left(E_i + \frac{\hbar^2 k^2}{2m^*} - E_j - \frac{\hbar^2 k'^2}{2m^*} \right)$$

- Convenient expression of scattering rate
- Ensemble Monte-Carlo method can be used
- Fast simulations

Rate equations for populations

… **but** problem for describing resonant tunneling





$$|\mathrm{Upper}\rangle = \frac{|\mathrm{Left}\rangle - |\mathrm{Right}\rangle}{\sqrt{2}}$$

$$|\mathrm{Lower}\rangle = \frac{|\mathrm{Left}\rangle + |\mathrm{Right}\rangle}{\sqrt{2}}$$

$$\tau_{\mathrm{inj}\to\mathrm{Upper}} = \tau_{\mathrm{inj}\to\mathrm{Lower}} = 2\tau_{i\to\mathrm{Left}}$$

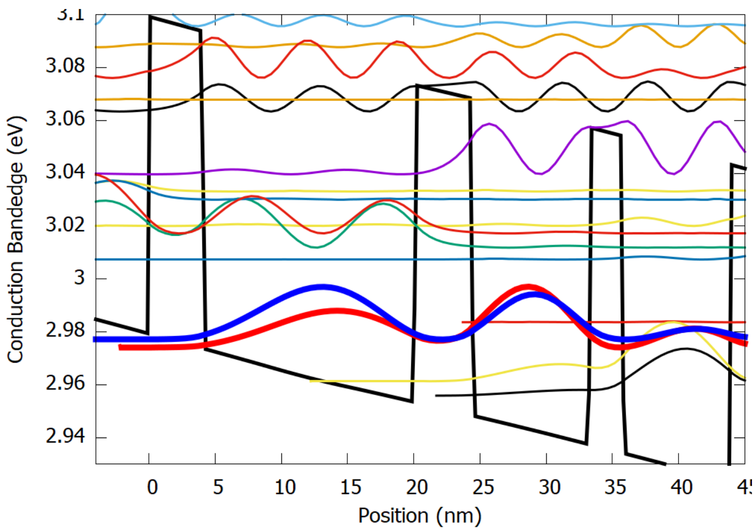
Transport time = injection time + extraction time

Tunneling time does not depend on the barrier thickness!

Rate equation approach works only if tunneling processes faster than scattering processes

Resonant tunneling

Wannier-Stark states

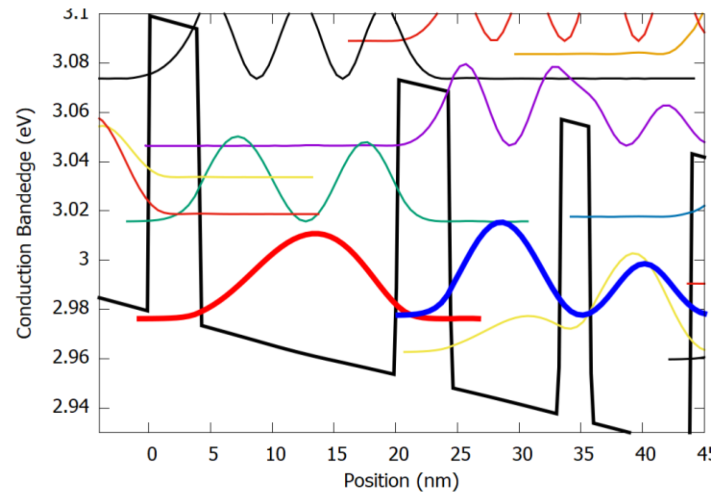


$$|\text{Upper}\rangle = \frac{|\text{Left}\rangle - |\text{Right}\rangle}{\sqrt{2}}$$

$2\hbar\Omega$

$$|\text{Lower}\rangle = \frac{|\text{Left}\rangle + |\text{Right}\rangle}{\sqrt{2}}$$

Tight-binding states (decoupling states from left and right from the barrier)



$|\text{Left}\rangle$ $|\text{Right}\rangle$

$\hbar\Omega$

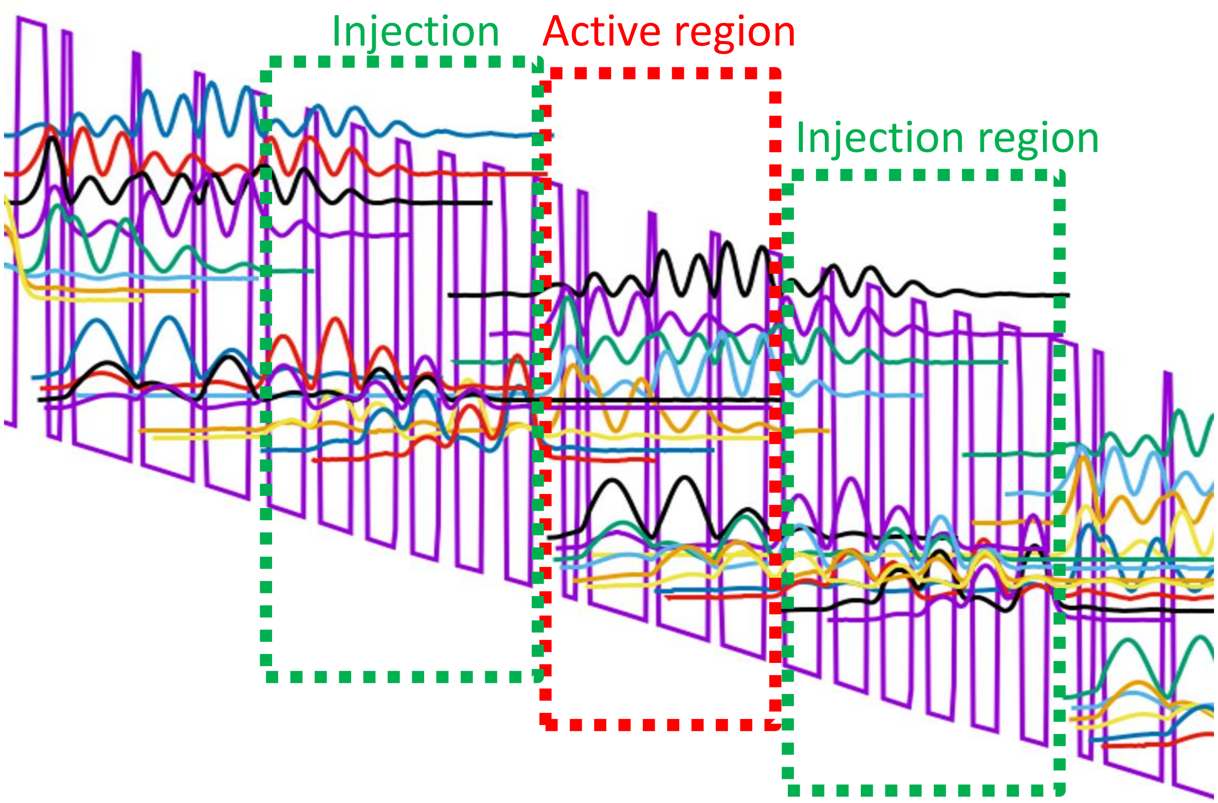
Tunneling rate = Ω in the coherent case

= $2\pi\hbar\Omega^2\rho(E)$ in the incoherent case (Fermi golden rule)

Hybrid approach

Spatially decoupling the wavefunctions into different modules:
- rate equation for populations inside each module
- tunneling rate between modules

Kazarinov and Suris, 1972



Limitation: arbitrary distinction needed between tunneling and scattering processes

Density matrix

- Basis invariance using equations for the full density matrix

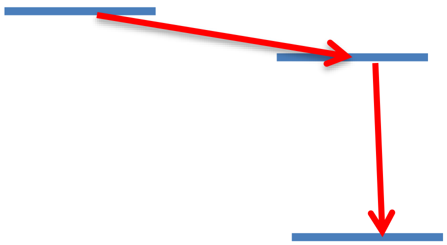
$$-i\hbar\frac{\partial\rho}{\partial t} = [\rho, H]$$

Two existing approaches:

- Lindblad equation with phenonenological parameters for dephasing
  Williams, Kumar
- Perturbative treatment of scattering processes
  Iotti & Rossi, Terrazi et al

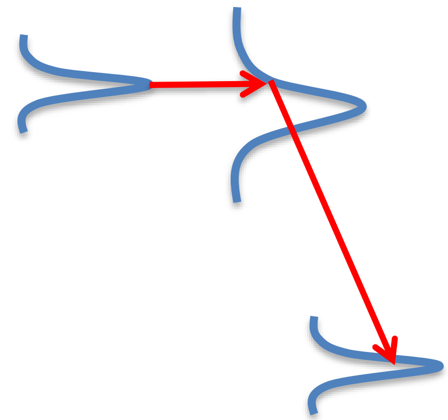
Time-energy uncertainty

Sequential scattering processes
(rate equation / density matrix)



Energy conservation is enforced for each scattering process

Green's functions: energy-resolved description



Account for high-order processes

But we know that $\delta t \delta E \geq \frac{\hbar}{2}$

Non-equilibrium Green's functions (NEGF)

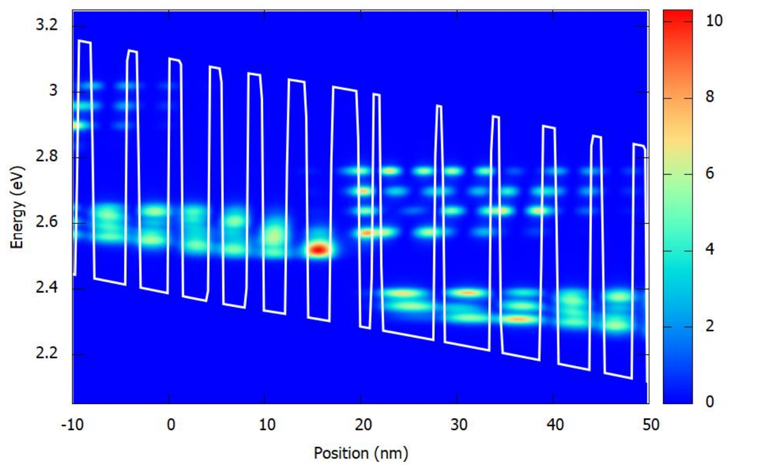
In steady-state transport, two independent quantities

Retarded Green's function:

$$G^R(E) = \frac{1}{E - H_0 - \Sigma^R(E)}$$

Imaginary part →

Spectral function (i.e. density of states)

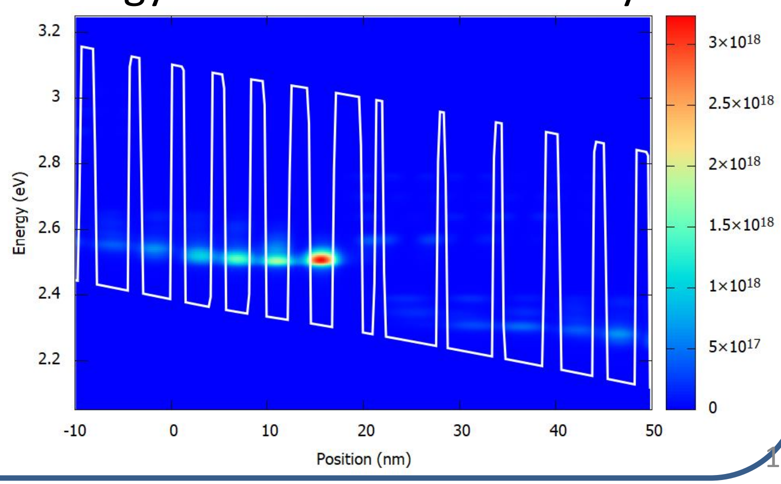


Lesser Green's function:
energy-resolved density matrix

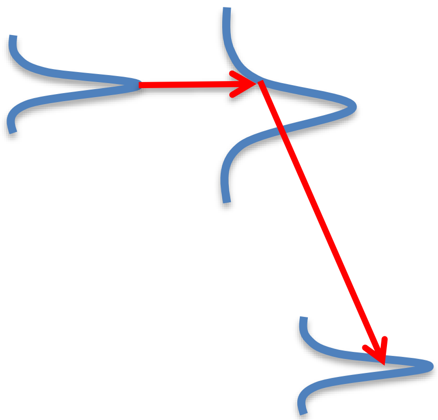
$$\rho(E) = -\frac{i}{2\pi} \int dE G^<(E)$$

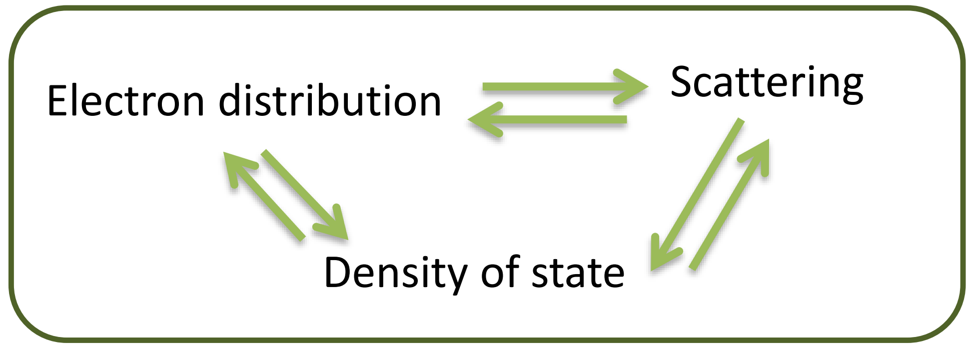
→

Energy-resolved carrier density



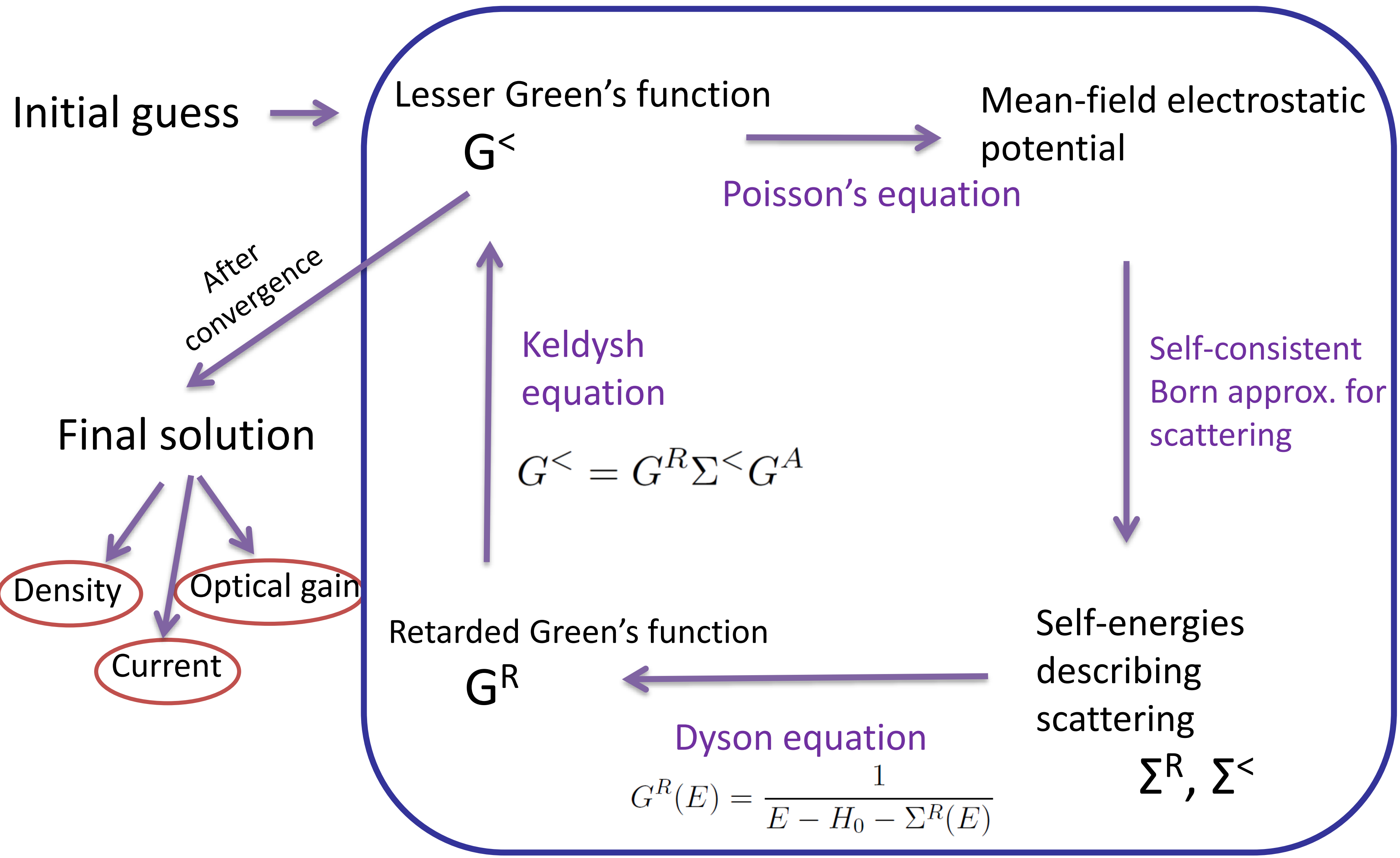
Coupling between Green's functions





- The density of state and the electron distribution needs to be solved self-consistently

Solving self-consistent NEGF equations



Implementation of NEGF in QCLs: Lee & Wacker (2002)

Linewidths of radiative transitions

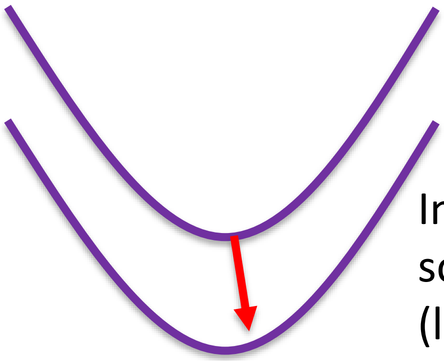
Broadening of radiative transitions:

T. Ando (1985):

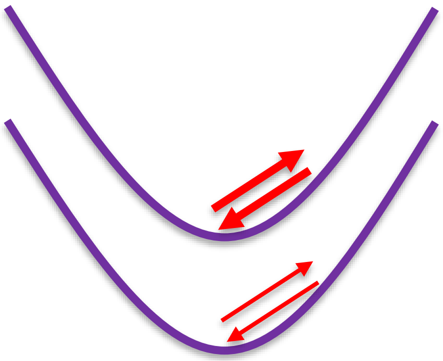
$$\Gamma_{op}(E)=\frac{1}{2}[\Gamma_{intra}(E)+\Gamma_{inter}(E)],$$

$$\Gamma_{intra}(E)=2\pi\sum_{k'}\langle|(0k'|H_1|0k)-(1k'|H_1|1k)|^2\rangle\delta(\varepsilon(k)-\varepsilon(k'))|_{E=\varepsilon(k)},$$

$$\Gamma_{inter}(E)=2\pi\sum_{k'}\langle|(0k'|H_1|1k)|^2\rangle\delta(\varepsilon(k)-\varepsilon(k')+E_{10})|_{E=\varepsilon(k)},$$



Intersuband scattering (lifetime limit)



Pure dephasing arises from the difference in intrasubband scattering.

- Linewidth can be smaller than individual subband broadening if intrasubband processes are correlated
- NEGF: self-consistent calculation of gain needed to account for these correlation effects

Outline

- Essential ingredients for modeling QCLs: electronic structure and scattering processes

- Different formalisms from semi-classical to quantum transport
  - Rate equation for populations
  - Density matrix
  - Non-equilibrium Green's functions (NEGF)

- **Development of a commercial NEGF simulator: nextnano.QCL**

- New physical insights QCLs

nextnano.QCL

Input file:

- Heterostructure geometry
- Material parameters
- Simulation parameters (energy grid, …)

Electronic structure

- Effective mass approximation with non-parabolicity
- Wurzite materials (piezo and pyro-electric effects)
- Group IV materials

Scattering processes

- Charged impurities
- Interface roughness
- Alloy disorder
- Electron-electron
- Optical phonons
- Acoustic phonons

NEGF solver

Simulation results

- Physical observables (current density, gain)
- Analysis in different basis

Input file

nextnanomat - C:\Results\THz_QCL_GaAs_AlGaAs_Amanti09\THz_QCL_GaAs_AlGaAs_Amanti09.xml

File Edit Run View Tools Help

Input | Template | Run | Output | New Template

THz_QCL_GaAs_AlGaAs_Amanti09.xml

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      <Alloy_Composition>0.15</Alloy_Composition>
      <Alias>barrier</Alias>
      <Effective_mass_from_kp_parameters>yes</Effective_mass_from_kp_parameters>
    </Material>

    <NonParabolicity>yes</NonParabolicity>
    <UseConductionBandOffset>yes</UseConductionBandOffset>
  </Materials>


<Superlattice>
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      <Layer>
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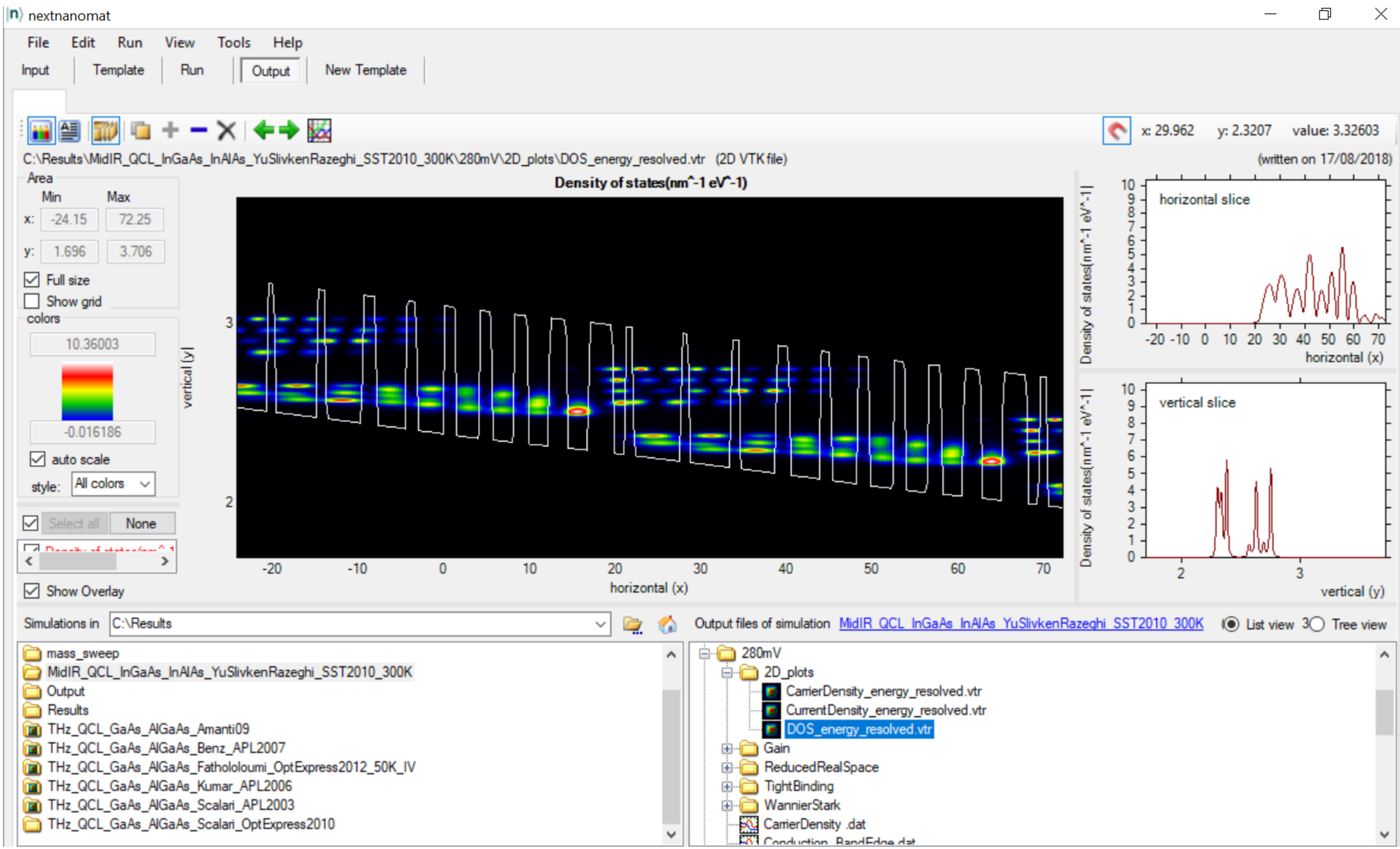
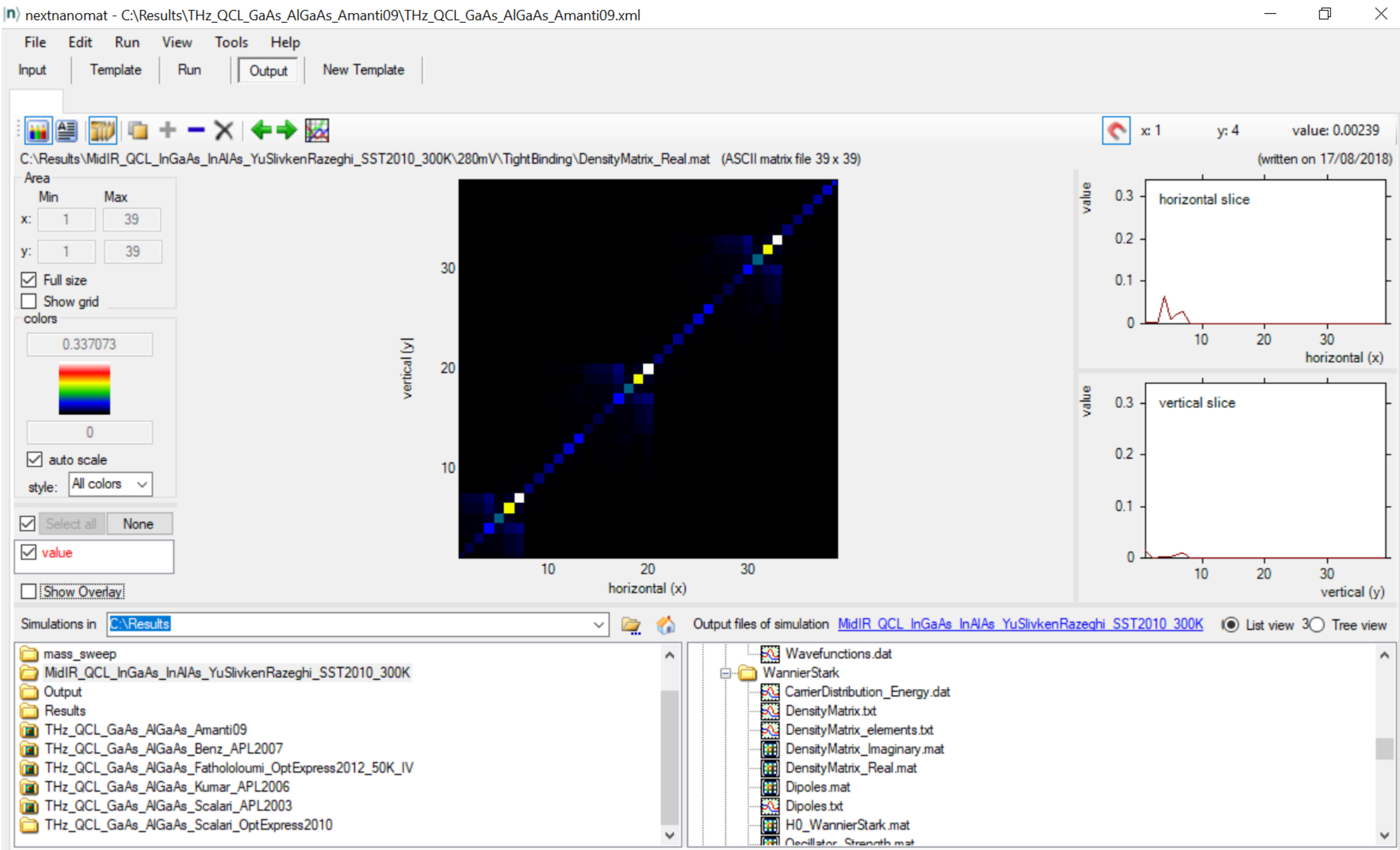
Ln 70 Col 10 input file for nextnano.QCL

Visualization of results

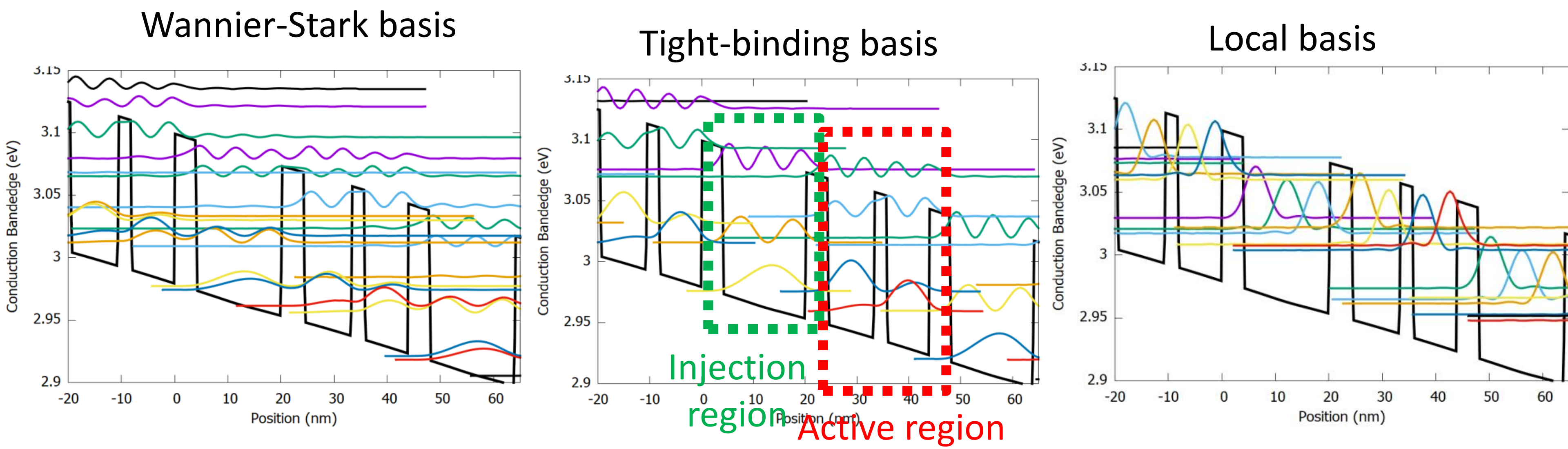


Visualization of results



Analysis of results

- Analysis of populations, density matrix, oscillator strengths etc in different basis

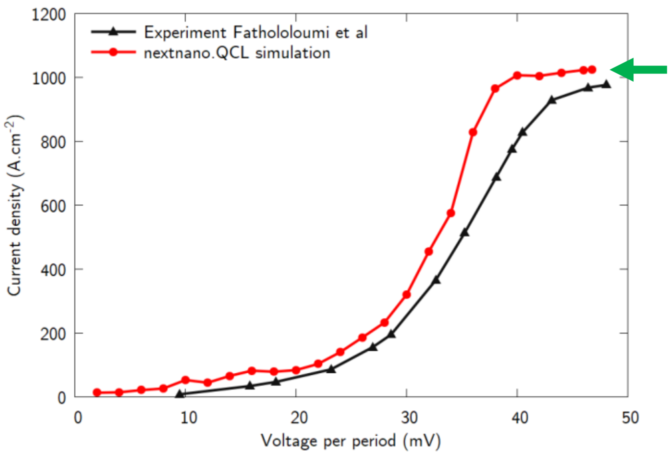


➢ Analysis of the physics in the more adapted/intuitive basis

Comparison with experimental data

THz QCL of Fathololoumi et al (record temperature of 200 K)

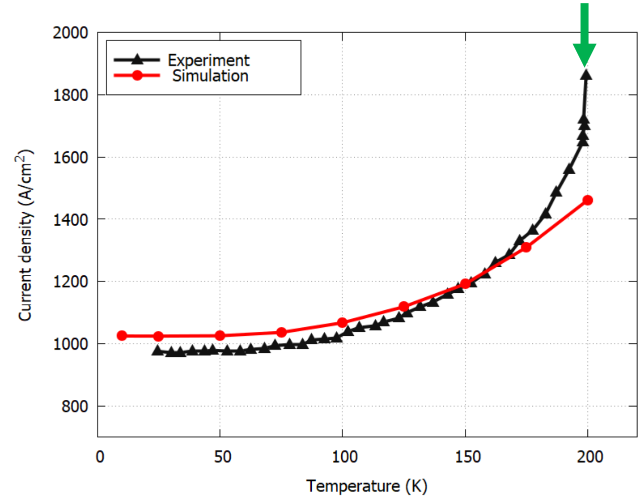
Current-voltage characteristics



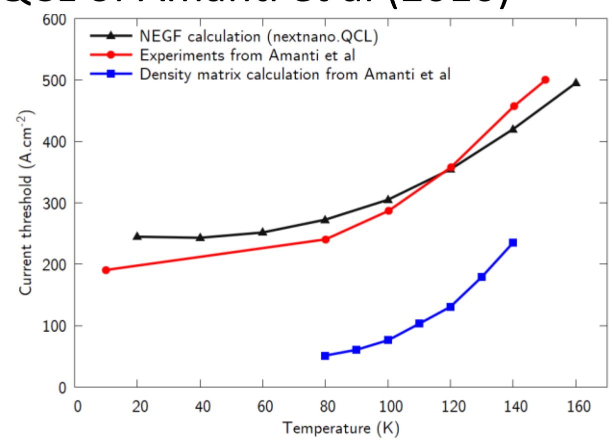
Lasing threshold assuming cavity losses of 27/cm

Current threshold vs temperature

Maximum operating temperature



THz QCL of Amanti et al (2010)



No phenomenological fitting parameter

Only material parameters: Conduction band offsets, interface roughness

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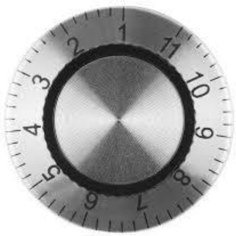
- **New physical insights QCLs**

Analyse the physics

Input file: Possibility to tune individual scattering processes



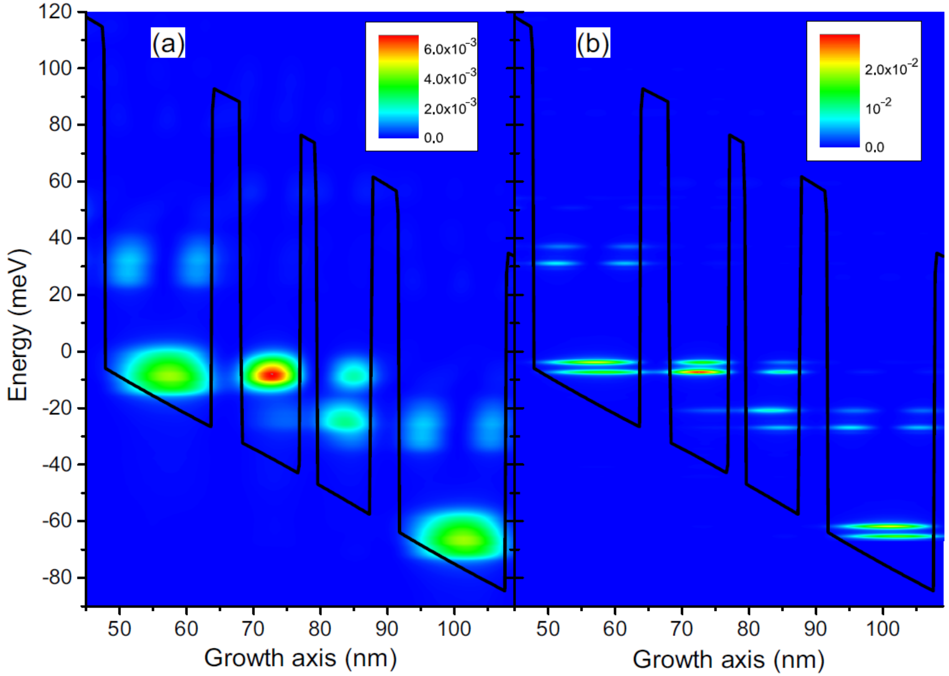
- LO-phonons
- Charged impurities
- Interface roughness
- …

➢ New physical insights

Coulomb scattering: a major source of broadening

Full calculation

Without e-impurity nor e-e scattering

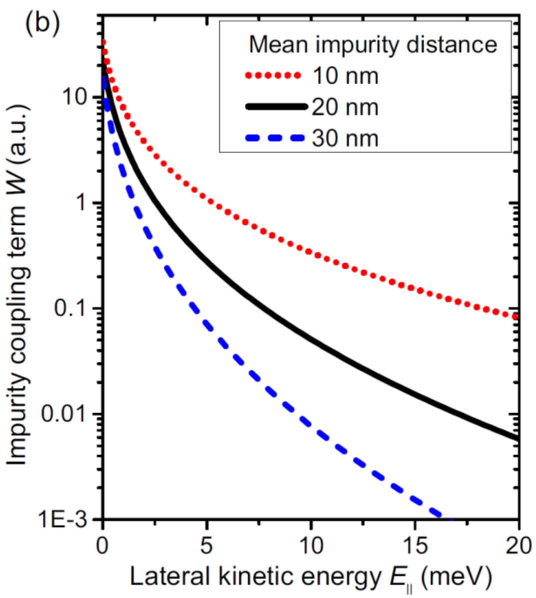


- Coulomb scattering processes are a dominant source of dephasing
- Transition from coherent to incoherent tunneling

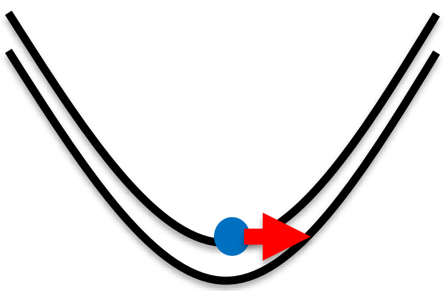
Ionized impurities

Coulomb potential created by a ionized impurity

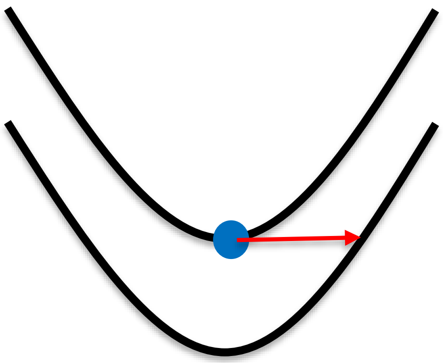
$$V_{\mathbf{r}_\mathrm{d}}^{(\mathrm{c})}(\mathbf{r}) = -\frac{e^2}{4\pi\,\varepsilon_0\varepsilon_\mathrm{s}|\mathbf{r}-\mathbf{r}_\mathrm{d}|}\exp\left(-\frac{|\mathbf{r}-\mathbf{r}_\mathrm{d}|}{\lambda_\mathrm{s}}\right)$$



Efficient scattering between closed subbands



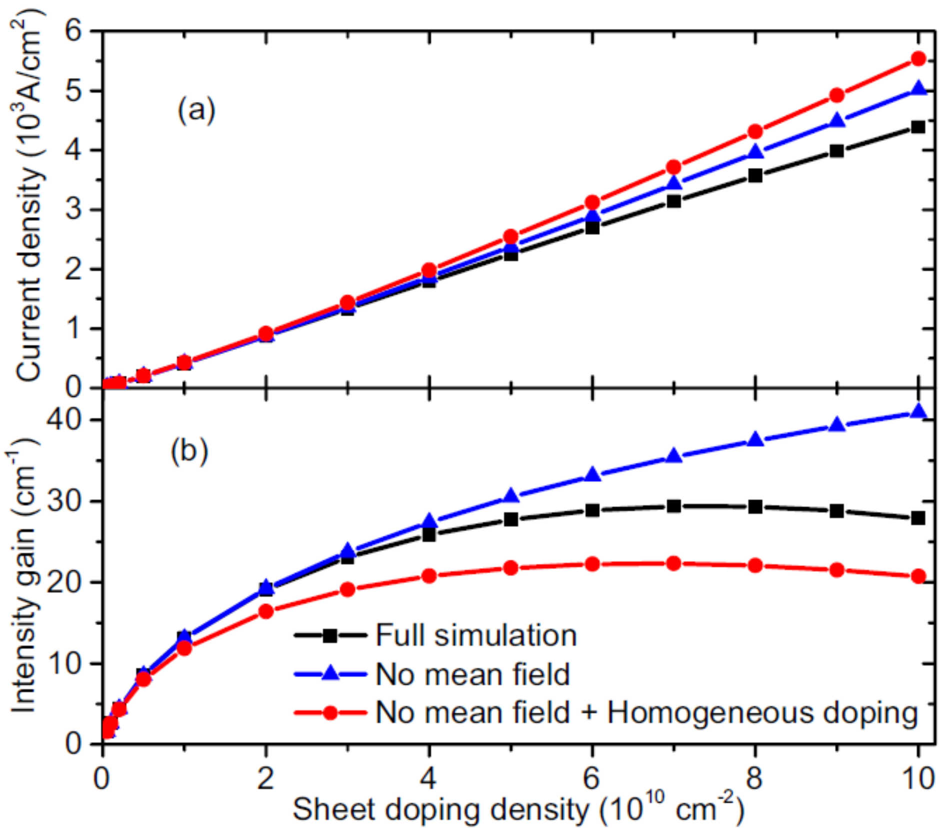
Slow scattering between distant subbands



Influence of doping density on THz QCL

Current

Gain



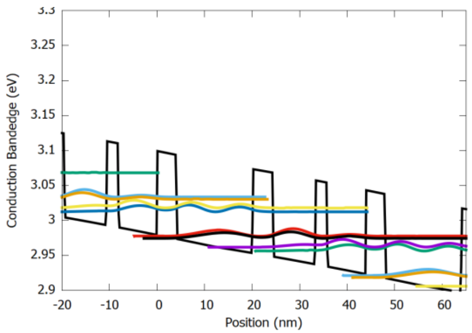
T. Grange, PHYSICAL REVIEW B **92**, 241306(R) (2015)

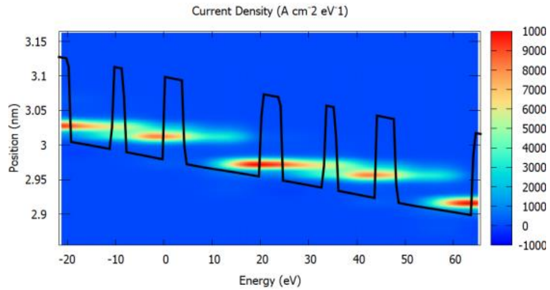
- Explanation of the contrasting influence of doping density on current (linear) and gain (non-linear)

Leakage into the continuum

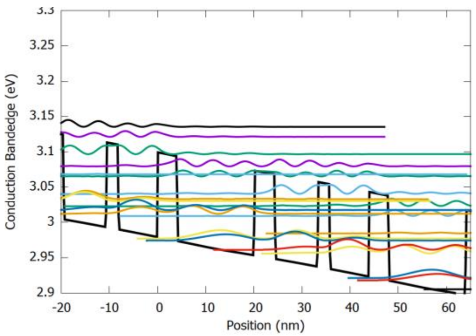
- ➢ Tune the number of minibands considered in the simulation

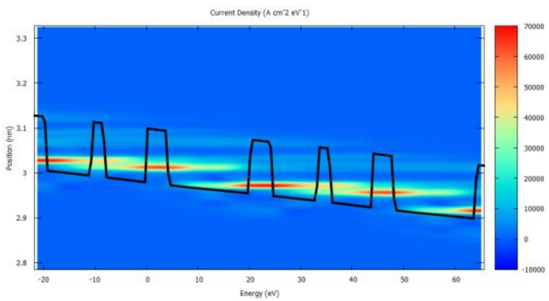
4 minibands



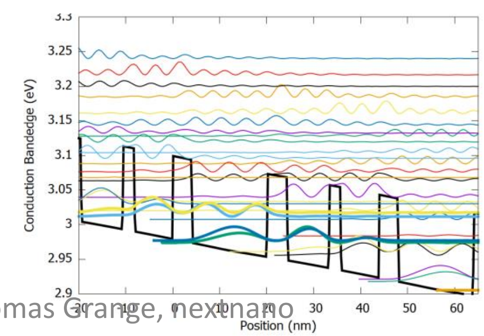


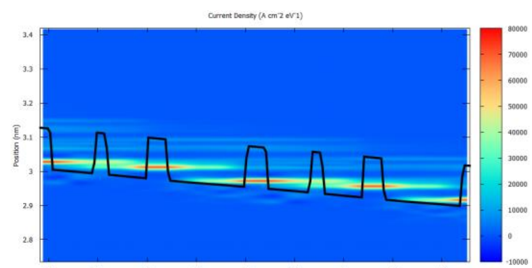
7 minibands



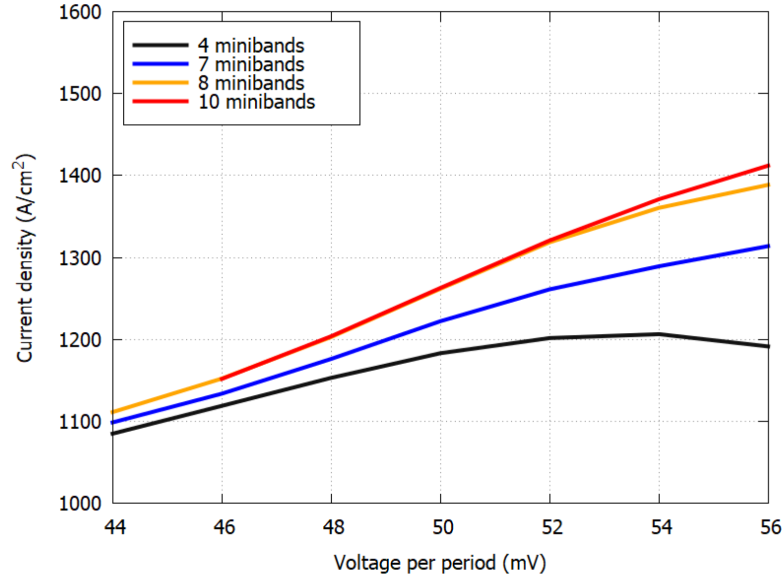


10 minibands





Current-voltage characteristics

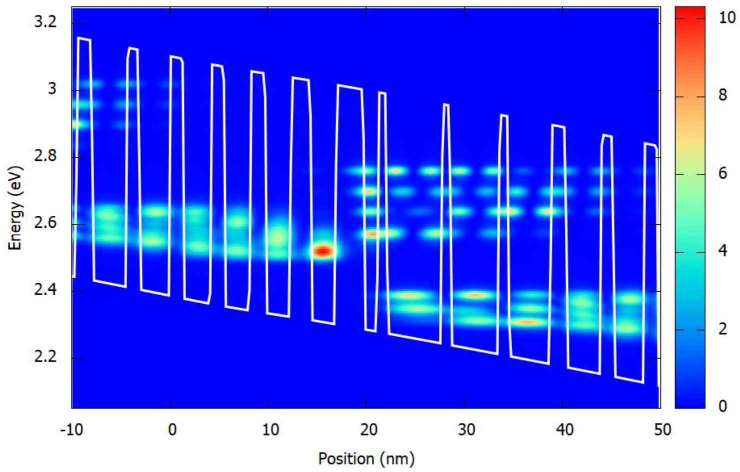


- ➢ Clear identification of the leakage into the high-energy states

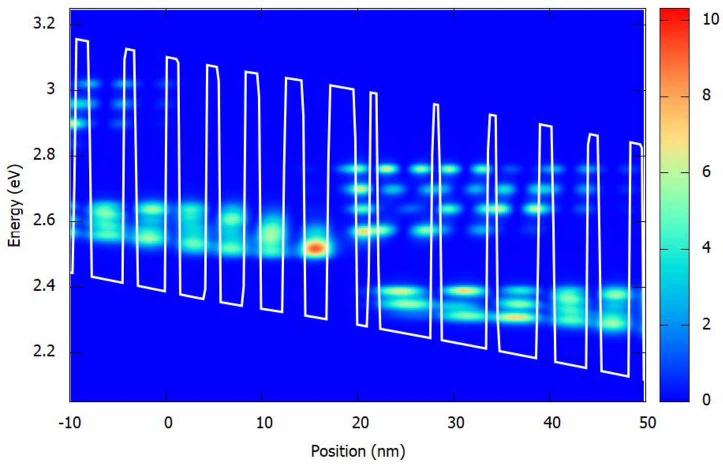
Influence of interface roughness

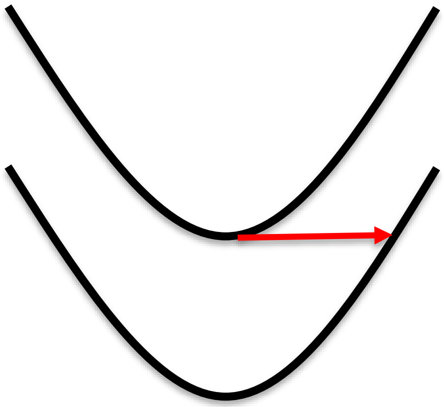
Impact of interface roughness on mid-infrared QCL

Design of Yu et al, SST 2010

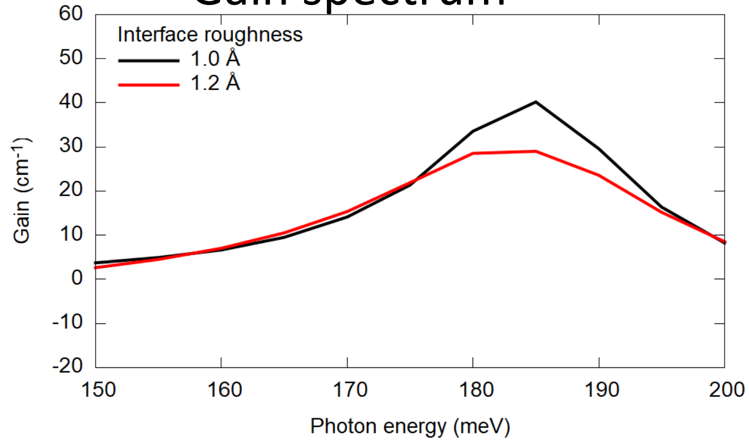


Increasing interface roughness





Gain spectrum



- Gain in MIR QCL very sensitive to interface roughness

Decoupling THz transitions from LO phonons?

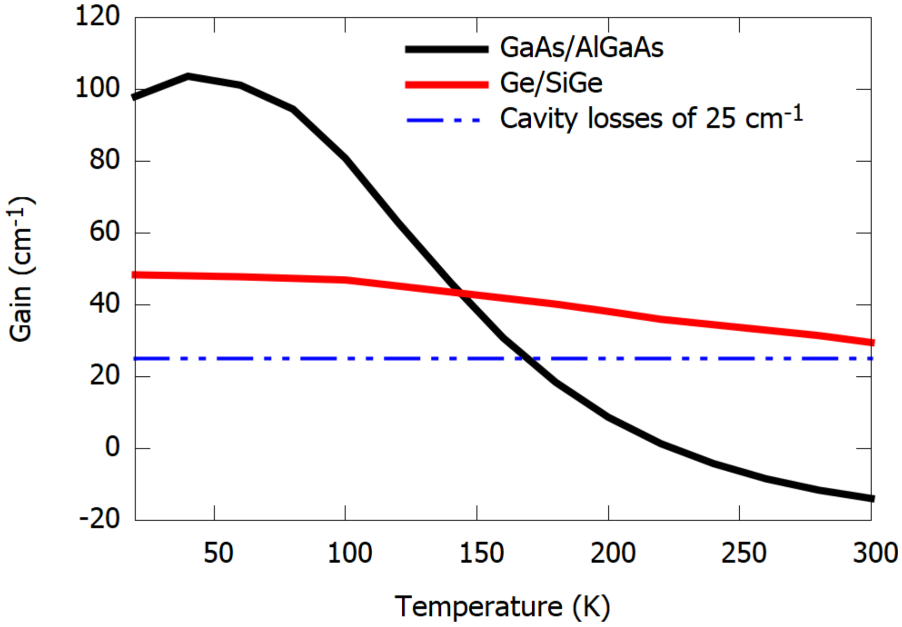
Decoupling THz radiative transition from optical phonons. Two possible strategies:

- Using non-polar materials: no polar (Fröhlich) coupling in **group IV materials** (Ge/SiGe)

- Using a material with a high optical phonon energy (e.g. **GaN**)

Ge/SiGe THz QCL?

Temperature dependence of gain : GaAs/AlGaAs vs Ge/SiGe



- Increasing temperature robustness with decreasing coupling to optical phonons

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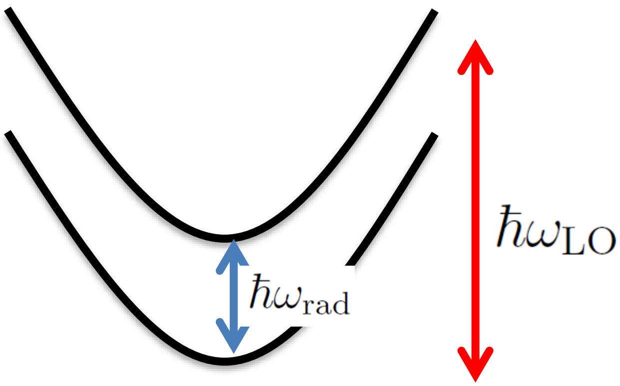
See posters:

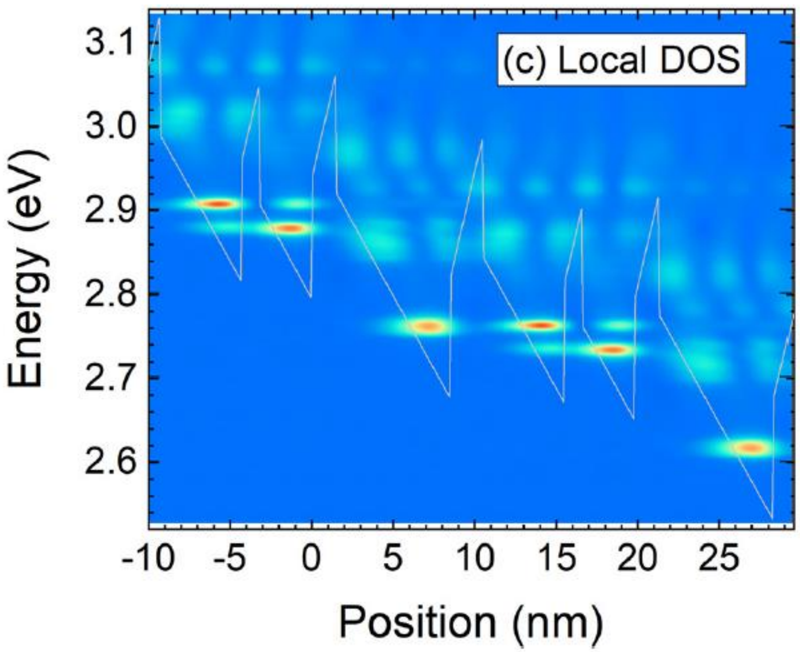
- D. Stark
- C. Ciano
- T. Grange

Optical phonons in GaN



- Large LO-phonon energy (90 meV)
- **But** Fröhlich constant 16 times stronger than in GaAs

Is LO-phonon induced broadening a limitation?



K Wang et al, APL 2018

Broadening in GaN THz QCL is **not** limited by LO-phonon

See talk of Ke Wang (Friday)

Summary

- Different transport models available for QCLs from semiclassical to fully quantum
- NEGF allows an accurate description of both quantum transport and scattering processes
- Predictive simulations with nextnano.QCL
  www.nextnano.com/nextnano.QCL
- Explore new material systems and new physics: tuning optical phonons with group IV and nitride materials
- Further improvement: transport under lasing action

Acknowledgements

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