Computational methods for calculating the bandstructure of THz QCLs

Alex Valavanis

Institute of Microwaves and Photonics,
University of Leeds

July 16, 2014

Overview

The Schrödinger equation

QCLs: Formulating the problem

Numerical solutions

The Schrödinger wave equation

- An expression of energy conservation:

$$\left[-\frac{\hbar^2}{2m}\frac{\partial^2}{\partial z^2} + V(z)\right]\psi(z) = E\psi(z) \tag{1}$$

- Kinetic energy:

$$\hat{T}\psi(z) = -\frac{\hbar^2}{2m}\frac{\partial^2}{\partial z^2}\psi(z) \tag{2}$$

- Potential energy:

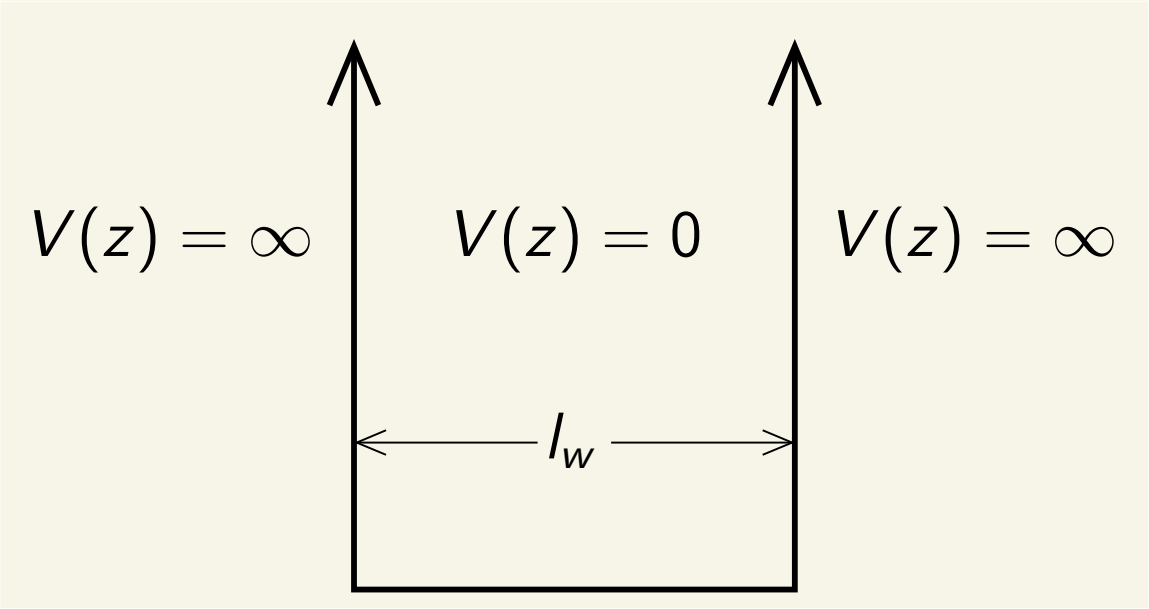
$$\hat{V}\psi(z) = V(z)\psi(z) \tag{3}$$

- Total energy (Hamiltonian):

$$\hat{H}\psi(z) = [\hat{T} + \hat{V}]\psi(z) = E\psi(z) \tag{4}$$

Example: infinite quantum well

For *very* simple systems, we can sometimes find an analytical solution.



The infinite well represents an electron trapped between impenetrable barriers. Inside the well, $V(z) = 0$:

$$-\frac{\hbar^2}{2m}\frac{\partial^2}{\partial z^2}\psi(z) = E\psi(z) \tag{5}$$

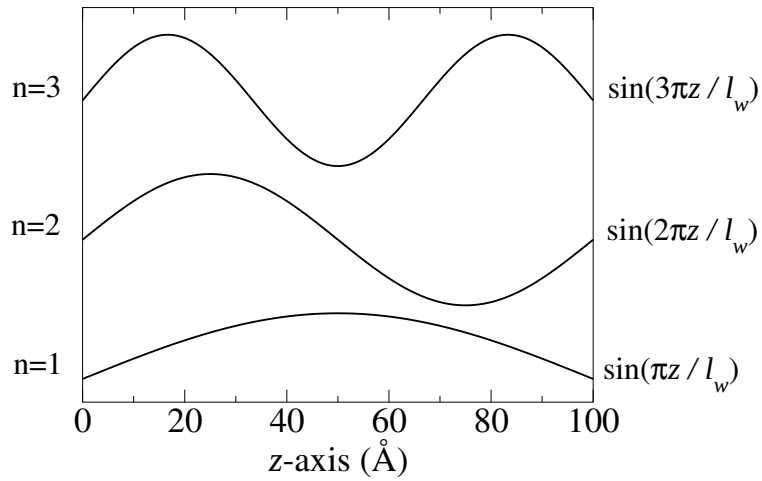
Solution for infinite quantum well

- Inside the well, electron is free:

$$\psi(z) = A \sin kz + B \cos kz \quad (6)$$

- Wavefunctions must decay to zero at boundaries: $\psi(0) = \psi(l_w) = 0$.
- Therefore $B = 0$.
- Only valid solutions are sine waves with:

$$\psi_n(z) = A \sin\left(\frac{\pi n}{l_w} z\right) \quad (7)$$



Corresponding energies:

$$E_n = \frac{\hbar^2 \pi^2 n^2}{2 m l_w^2} \quad (8)$$

- Probability of finding electron in region (a, b) is:

$$P(a,b) = \int_a^b \psi^*(z)\psi(z)\ \mathrm{d}z \tag{9}$$

- Probability of finding electron *somewhere* in well must be 1. Can use this to find the amplitude of wavefunction: $A = \sqrt{(2/l_w)}$
- Therefore, complete expression for wavefunction in infinite well is:

$$\psi_n(z) = \sqrt{\frac{2}{l_w}} \sin\left(\frac{\pi n z}{l_w}\right) \tag{10}$$

Toward more complex systems. . .

There is **no analytical solution** for complex systems like THz QCLs. We can, however, use some "ingredients" of the solution:

- Specify a potential profile $V(z)$ and effective mass m^*
- Specify boundary conditions $\psi(0)$ and $\psi(\infty)$
- Find amplitude of wavefunctions at each point!

A numerical (sampling) approach is needed!

Overview

The Schrödinger equation

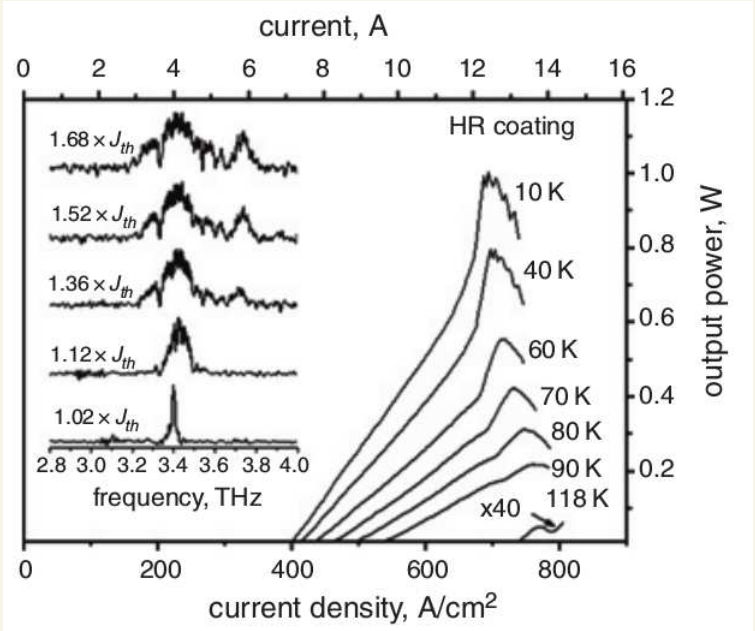
QCLs: Formulating the problem

Numerical solutions

Example: World record 1 W THz QCL

Recently demonstrated (at Leeds!): THz QCL with 1 W output power.

- 4 GaAs wells with $Al_{0.16}Ga_{0.84}As$ barriers
- Layer thicknesses (barriers in bold, $3 \times 10^{16}\,cm^{-3}$ doped layer underlined): **52**/103/**17**/107.5/**36**/88/**39.5**/<u>172</u> Å
- External electric field of $7.6\,kV\,cm^{-1}$.
- Periodic system



L. Li *et al., Electron. Lett.* **50**, 309 (2014)

Confining potential

- Band-offset between GaAs and $Al_{0.16}Ga_{0.84}As$ is given by:

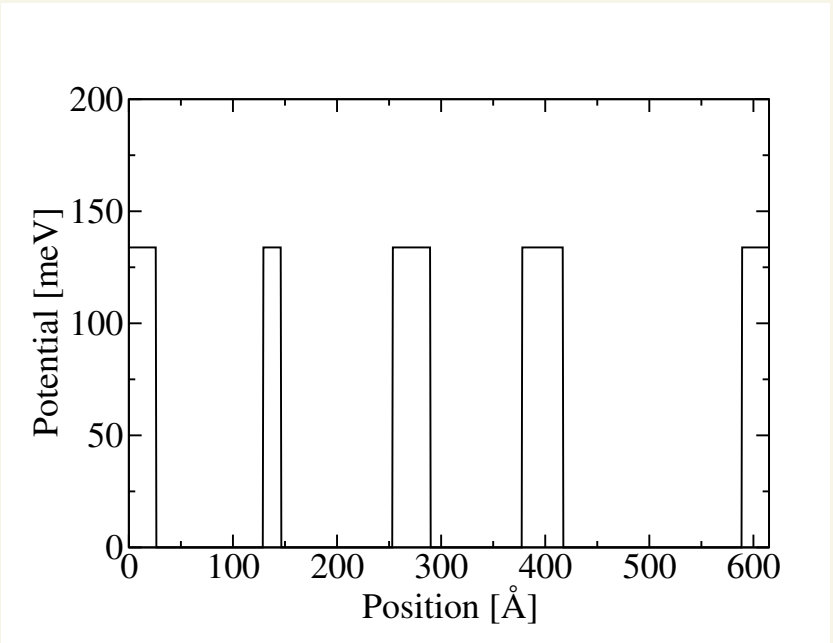
$$\Delta V_{CB} = 0.67 \times E_g \qquad (11)$$

where bandgap is:

$$E_g = (1.426 + 1.247x)\,\text{eV} \quad (12)$$

and $x = 0.16$ is alloy composition

- Therefore, can compute confining potential using $\Delta V_{CB} = 133.8\,\text{meV}$
- Wells are strongly coupled!

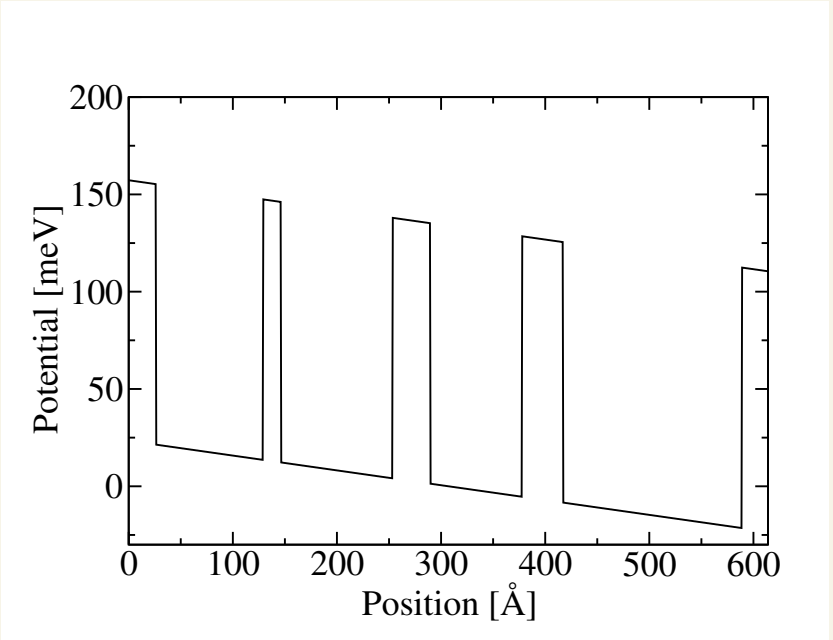


Effect of bias voltage

Now, we can add on the effect of the external electric field. The simplest model assumes that the field inside the structure is constant (and identical to the external field):

$$V(z) \rightarrow V(z) + qF(z - z_0) \quad (13)$$

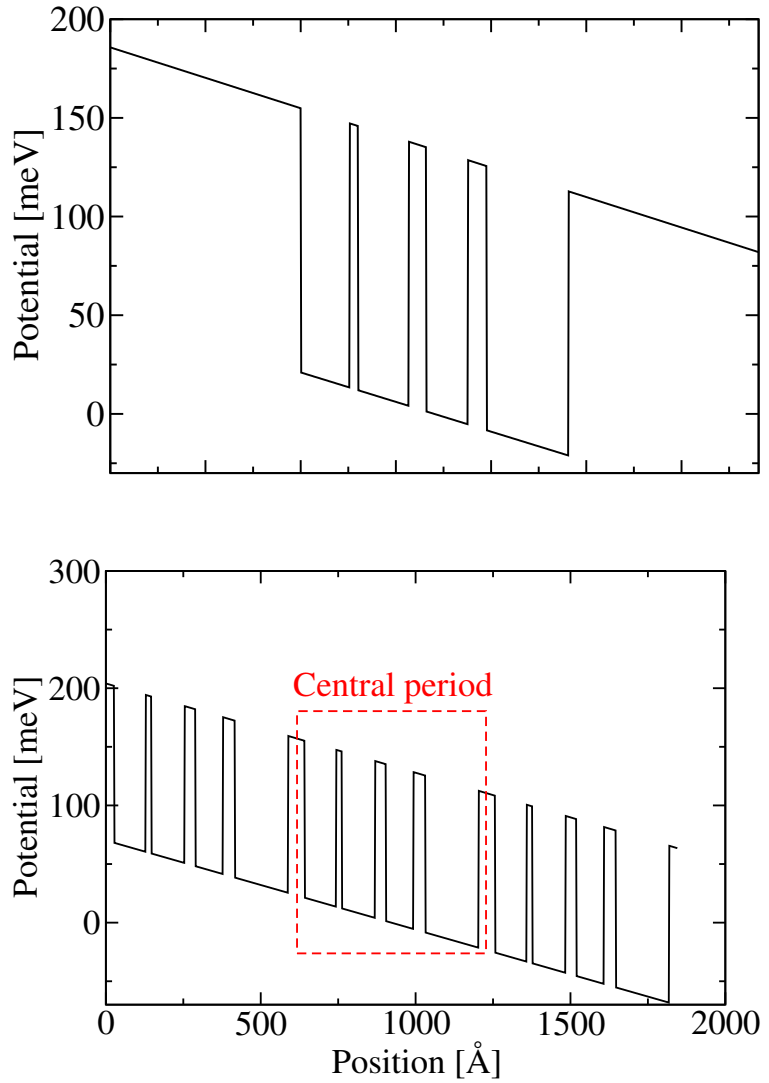
This gives a constant potential gradient throughout the structure



Note that this does not account for internal charge or variation in permittivity between materials! We'll address this later using Poisson's equation.

Boundary conditions for QCLs

- QCLs are *multi-period devices*.
- Therefore, wavefunctions do not decay to zero at edge of period!
- **Option 1**: "pad" the structure to allow "soft" decay into barrier. Used for density-matrix solvers
- **Option 2**: Solve several periods of structure to allow "leakage" between periods. Used for rate equations



Overview

The Schrödinger equation

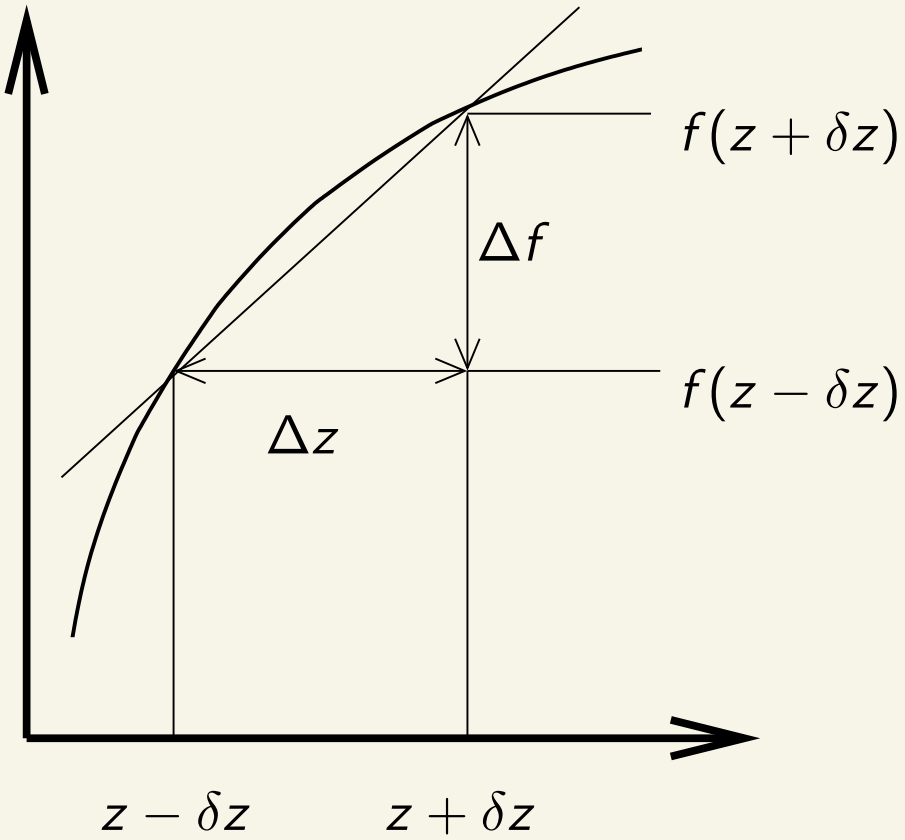
QCLs: Formulating the problem

Numerical solutions

The finite-difference approximation

We can solve differential equations numerically using the finite-difference (FD) approximation:

$$\frac{\mathrm{d}f}{\mathrm{d}z} \approx \frac{\Delta f}{\Delta z} = \frac{f(z+\delta z) - f(z-\delta z)}{2\delta z} \tag{14}$$



Repeat the process to find second derivative...

$$\frac{\mathrm{d}^2 f}{\mathrm{d}z^2} \approx \frac{\frac{\mathrm{d}f}{\mathrm{d}z}\Big|_{z+\delta z} - \frac{\mathrm{d}f}{\mathrm{d}z}\Big|_{z-\delta z}}{2\delta z} \tag{15}$$

$$\therefore \frac{\mathrm{d}^2 f}{\mathrm{d}z^2} \approx \frac{f(z+2\delta z) - 2f(z) + f(z-2\delta z)}{(2\delta z)^2} \tag{16}$$

Can tidy up by making transform $2\delta z \to \delta z$

$$\frac{\mathrm{d}^2 f}{\mathrm{d}z^2} \approx \frac{f(z+\delta z) - 2f(z) + f(z-\delta z)}{(\delta z)^2} \tag{17}$$

We can use a neater notation, $f(z + i\delta z) = f_i$:

$$\left.\frac{\mathrm{d}f}{\mathrm{d}z}\right|_i \approx \frac{f_{i+1} - f_{i-1}}{2\delta z} \tag{18}$$

$$\left.\frac{\mathrm{d}^2 f}{\mathrm{d}z^2}\right|_i \approx \frac{f_{i+1} - 2f_i + f_{i-1}}{(\delta z)^2} \tag{19}$$

It is now possible to split a function $f(z)$ up into a set of samples f_i and compute the derivatives at each point.

Discretizing the Schrödinger equation

Start from analytical form:

$$-\frac{\hbar^2}{2m^*}\frac{\partial^2}{\partial z^2}\psi(z) + V(z)\psi(z) = E\psi(z) \tag{20}$$

Replace $\psi(z)$ and $V(z)$ with samples at points i:

$$-\frac{\hbar^2}{2m^*}\left.\frac{\partial^2\psi}{\partial z^2}\right|_i + V_i\psi_i = E\psi_i \tag{21}$$

Now substitute FD approximation for the derivative:

$$-\frac{\hbar^2}{2m^*}\left[\frac{\psi_{i+1} - 2\psi_i + \psi_{i-1}}{(\delta z)^2}\right] + V_i\psi_i = E\psi_i \tag{22}$$

The "shape" of the wave function at any point is determined by the mass, energy and confining potential

Can rearrange the discrete SE by gathering together wave function samples at each point:

$$-\frac{\hbar^2}{2m^*(\delta z)^2}\psi_{i+1} + \left[\frac{\hbar^2}{m^*(\delta z)^2} + V_i\right]\psi_i - \frac{\hbar^2}{2m^*(\delta z)^2}\psi_{i-1} = E\psi_i \tag{23}$$

More compact notation:

$$a_i\,\psi_{i-1} + b_i\,\psi_i + c_i\,\psi_{i+1} = E\psi_i, \tag{24}$$

where:

$$a_{i+1} = c_i = -\frac{\hbar^2}{2m^*(\delta z)^2} \quad \text{and} \quad b_i = \frac{\hbar^2}{m^*(\delta z)^2} + V_i \tag{25}$$

$$a_i \psi_{i-1} + b_i \psi_i + c_i \psi_{i+1} = E\psi_i, \tag{26}$$

Now, taking the standard boundary conditions,[1] $\psi_0 = \psi_{N+1} = 0$
We have a system of linear equations for wave function at each point:

$$\begin{aligned} b_1\psi_1 + c_1\psi_2 &= E\psi_1 \\ a_2\psi_1 + b_2\psi_2 + c_2\psi_3 &= E\psi_2 \\ &\dots \\ a_{N-1}\psi_{N-2} + b_{N-1}\psi_{N-1} + c_{N-1}\psi_N &= E\psi_{N-1} \\ a_N\psi_{N-1} + b_N\psi_N &= E\psi_N \end{aligned} \tag{27}$$

[1] recall that simulation domain for a QCL includes "padding" or multiple periods

This system of equations can be represented more succinctly in matrix form:

$$\mathsf{H}\,\boldsymbol{\psi} = E\,\boldsymbol{\psi} \tag{28}$$

where H is a matrix containing all the coefficients:

$$\mathsf{H} = \begin{pmatrix} b_1 & c_1 & 0 & \cdots & 0 \\ a_2 & b_2 & c_2 & \cdots & 0 \\ 0 & \ddots & \ddots & \ddots & 0 \\ \vdots & \cdots & a_{N-1} & b_{N-1} & c_{N-1} \\ 0 & \cdots & 0 & a_N & b_N \end{pmatrix} \tag{29}$$

and $\boldsymbol{\psi}$ is a column-vector containing all the samples of the wave function in their correct order:

$$\boldsymbol{\psi}^T = \{\psi_1, \psi_2, \cdots, \psi_N\} \tag{30}$$

Matrix formulation: summary

We have converted the Schrödinger equation into a matrix eigenvalue problem:

$$\mathrm{H}\,\boldsymbol{\psi} = E\,\boldsymbol{\psi} \tag{31}$$

- Eigenvalues: E
- Eigenvectors: ψ

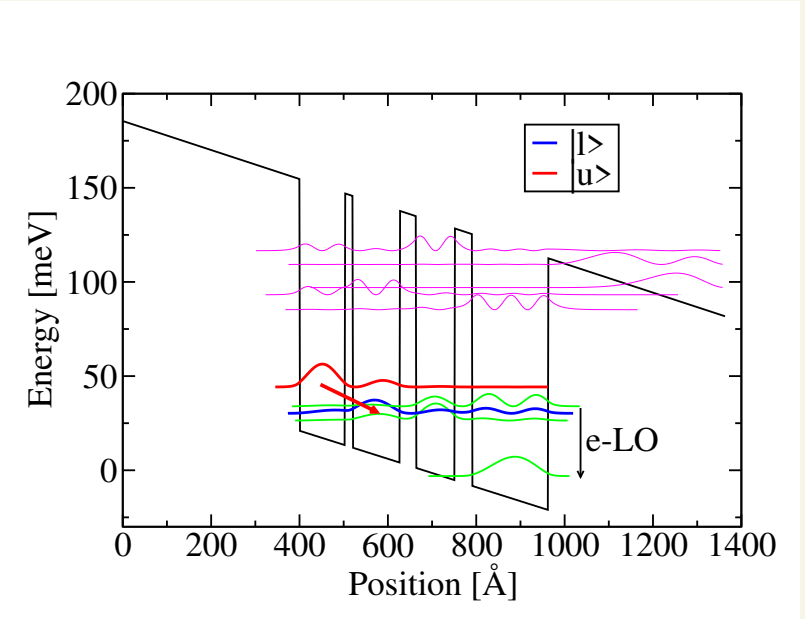
H is symmetrical and tridiagonal. This problem can be solved extremely rapidly as follows:

1. Compute the potential $V(z)$ and take samples at a set of points V_i
2. Create the Hamiltonian matrix H by using the appropriate value of a_i, b_i or c_i for each element.
3. Send the matrix to an eigenvalue solver. Free software libraries such as the *GNU Scientific Library*, *LAPACK* or *Armadillo* include excellent solvers!

Solution for 1 W THz QCL

Using padded structure:

- Lasing transition: $\langle u|l\rangle = 14.5\,\text{meV}$ (3.63 THz)
  - (Expected 3.4 THz)
- Extraction transition: 38.4 meV
  - (Expected 36 meV)



High-energy states (pink) include unbound continuum states.
Physically realistic?
Solver gives pretty good results, although energies are slightly high.
Is something missing?

Variable effective mass

Effective mass is larger in $Al_{0.16}Ga_{0.84}As$ barriers. This could explain the overestimated energy!

$$m^* = (0.067 + 0.083x)\, m_0 = 0.080 m_0 \tag{32}$$

Can define a spatially dependent effective mass for the QCL $m^*(z)$.

$$-\frac{\hbar^2}{2}\frac{\partial}{\partial z}\frac{1}{m^*(z)}\frac{\partial}{\partial z}\psi(z) + V(z)\psi(z) = E\psi(z) \tag{33}$$

After applying FD approximation, matrix elements become:

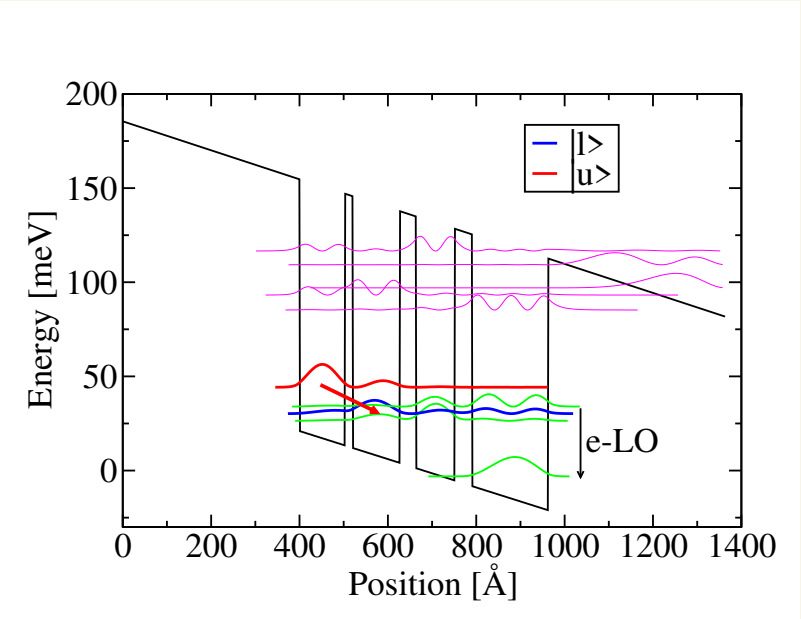
$$a_{i+1} = c_i = -\frac{\hbar^2}{2m^*_{i+\frac{1}{2}}(\delta z)^2} \tag{34}$$

$$b_i = \frac{\hbar^2}{2(\delta z)^2}\left(\frac{1}{m^*_{i+\frac{1}{2}}} + \frac{1}{m^*_{i-\frac{1}{2}}}\right) + V_i \tag{35}$$

- H is still a tridiagonal symmetric matrix, so the solution method is exactly the same!
- The half-integer masses ($m^*_{i+\frac{1}{2}}$ etc.) are just the average of the adjacent samples

Solution for 1 W THz QCL (variable mass)

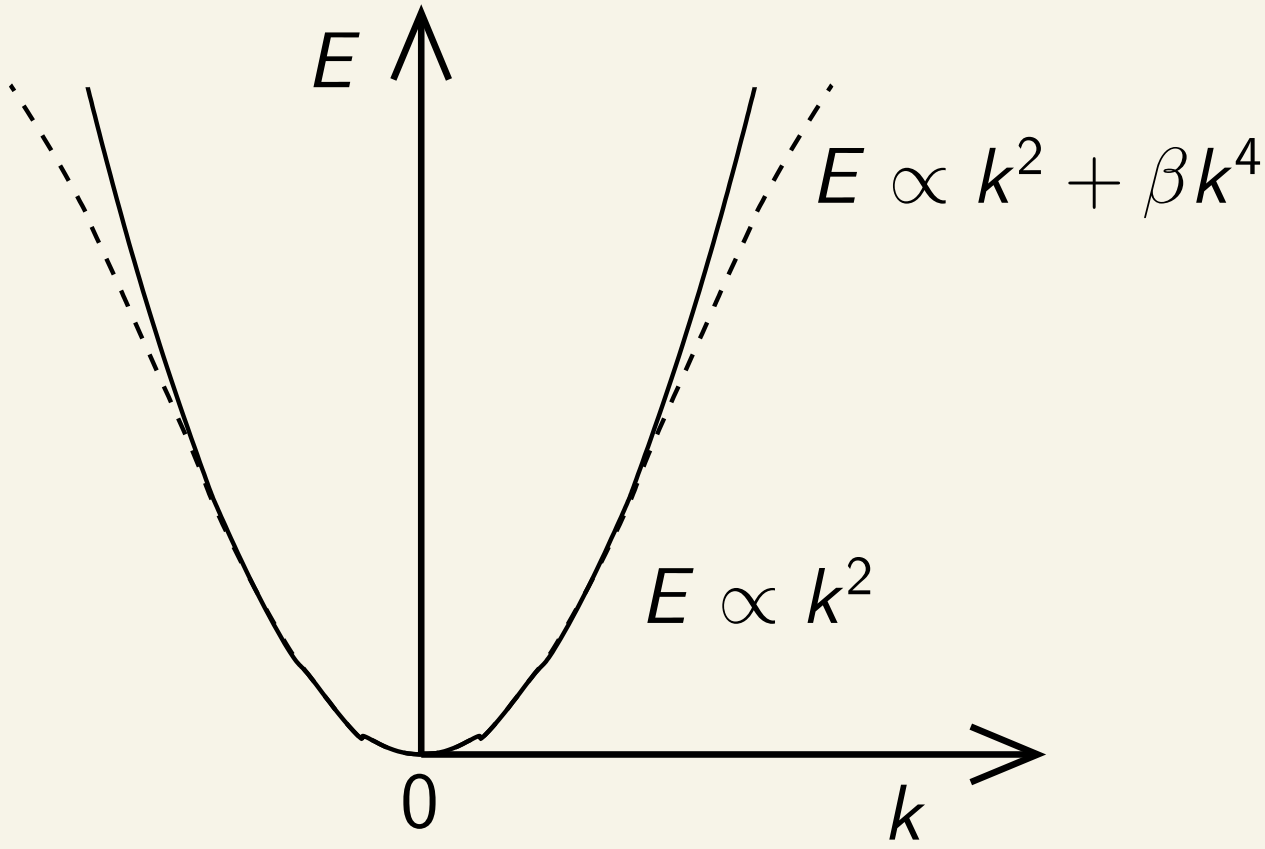
- Lasing transition: $\langle u|l\rangle = 14.0\,\text{meV}$ (3.5 THz)
  - (Expected 3.4 THz)
- Extraction transition: 37.1 meV
  - (Expected 36 meV)



Variable effective mass gives better results with no penalty in speed of solution!

Band nonparabolicity effects

As electrons gain energy, their dispersion is influenced by the valence band as well as the conduction band.



Band nonparabolicity effects

Dispersion is modelled more accurately by energy-dependent effective mass:

$$m^*(E) = m^*(0)[1 + \alpha(E - V)] \tag{36}$$

where $\alpha \approx 1/E_g$

- $\alpha = 0.7\,\mathrm{eV}^{-1}$ in GaAs. Not very important for low-energy states
- . . . but very significant effect for high-energy electrons or narrow-gap materials (InSb: $\alpha = 5.6\,\mathrm{eV}^{-1}$)

Non-parabolic effects result in an energy-dependent Hamiltonian:

$$-\frac{\hbar^2}{2}\frac{\partial}{\partial z}\frac{1}{m^*(z,E)}\frac{\partial}{\partial z}\psi(z)+V(z)\psi(z)=E\psi(z) \tag{37}$$

- Masses and matrix elements $a_i(E)$, $b_i(E)$ and $c_i(E)$ now energy dependent!
- Cannot solve *nonlinear* matrix eigenvalue problem directly:

$$\mathsf{H}(E)\,\boldsymbol{\psi}=E\,\boldsymbol{\psi} \tag{38}$$

- Instead, iteratively search for energies for which:

$$\det\left[E\mathsf{I}-\mathsf{H}(E)\right]=0 \tag{39}$$

Other approaches include:

- linearizing the eigenvalue problem[2]
- applying a Taylor expansion for $1/m^*(E)$[3]
- two-band models[4]

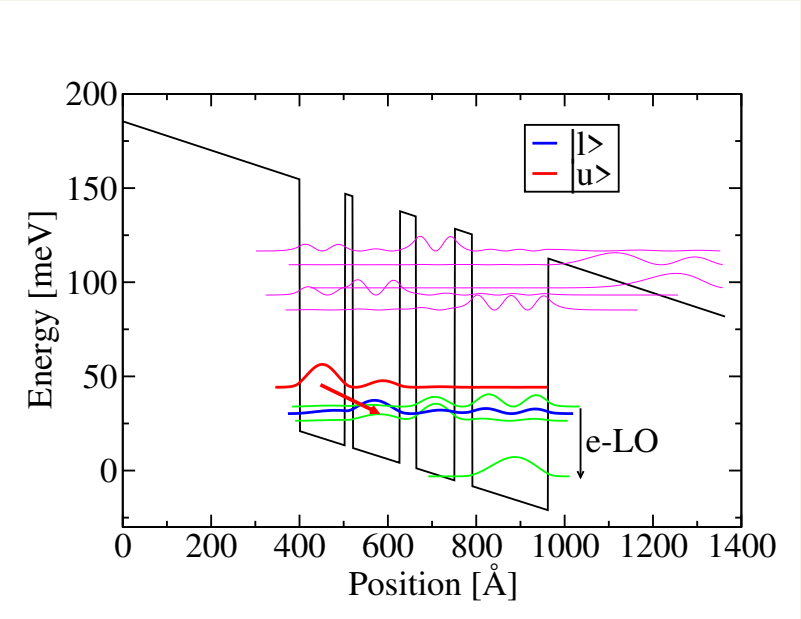
Alternatively, use multi-band $\mathbf{k} \cdot \mathbf{p}$ model

---

Cooper et al., DOI: 10.1063/1.3512981
Le et al., DOI: 10.1002/mop.23976
Ma et al., DOI: 10.1063/1.4817795

Solution for 1 W THz QCL (variable mass)

- Lasing transition: $\langle u|l\rangle = 14.2\,\text{meV}$ (3.5 THz)
  - (Expected 3.4 THz)
- Extraction transition: 36.7 meV
  - (Expected 36 meV)



Non-parabolic effective mass improves result for high-energy phonon transition without affecting low-energy lasing transition

Poisson's equation

Poisson's equation including variable permittivity $\epsilon(z)$:

$$\frac{\partial}{\partial z}\left[\epsilon(z)\frac{\partial}{\partial z}V(z)\right] = -\rho(z) \tag{40}$$

where $\rho(z)$ is charge profile (accounts for modulation doping).

$$\rho(z) = q\left(\sum_{i=1}^{n} N_i \psi_i^*(z)\psi_i(z) - d(z)\right) \tag{41}$$

where $\sum_{i=1}^{n} N_i = N$.

Poisson's equation

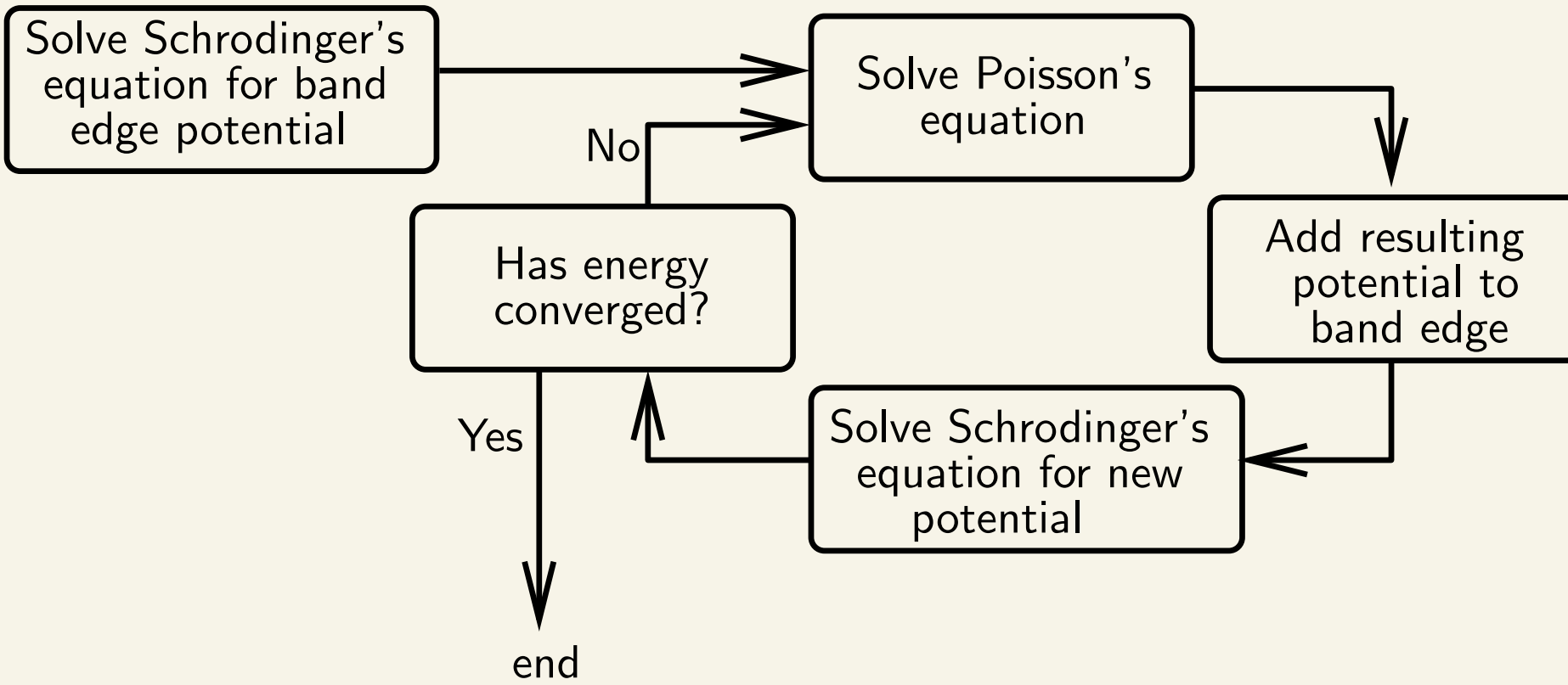
Need to know populations of subbands N_i

- For a full calculation, we need electron transport model. Beyond the scope of this lecture!
- Approximate solution: ignore high-energy (pink) states
- Assume equal population in low-energy states

Self-consistent solutions

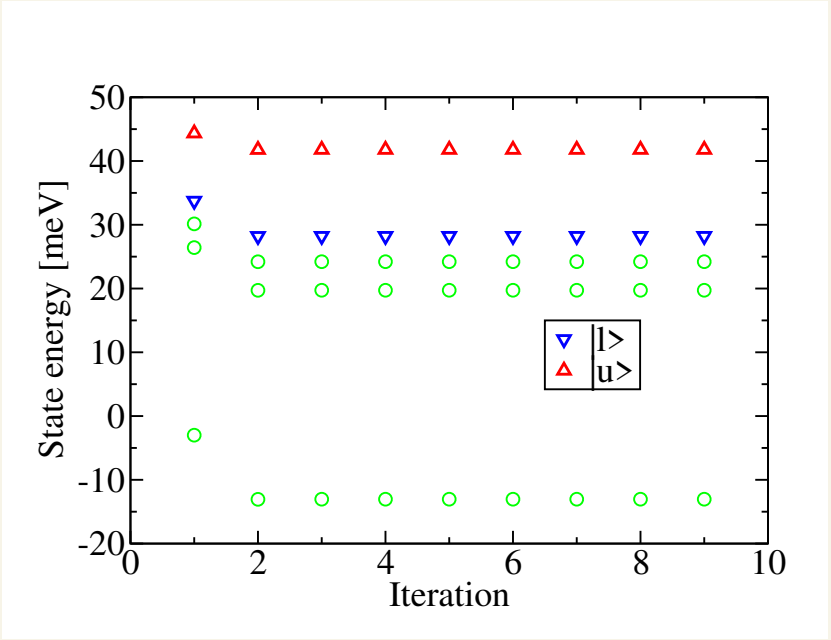
Poisson and Schrödinger solutions are intrinsically coupled...

- Poisson equation calculates potential using electron distribution.
- Schrödinger equation calculates electron distribution using potential.
- Need to solve iteratively



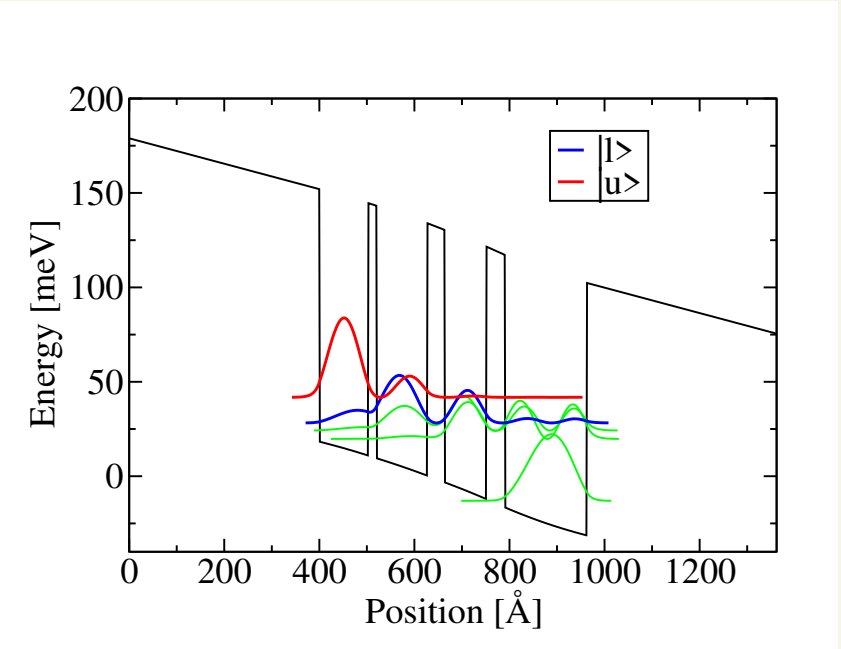
Self-consistent solutions

- Self-consistent solution performed iteratively.
- Convergence within 3 iterations (a bit slower than simple Schrödinger solution!)
- Full transport model can take much longer! Care needed to avoid instability.



Solution for 1 W THz QCL (variable mass)

- Band bending effects appear
- Lasing transition: $\langle u|l\rangle = 13.6\,\text{meV}$ (3.4 THz)
  - (Expected 3.4 THz)
- Extraction transition: 37.2 meV
  - (Expected 36 meV)

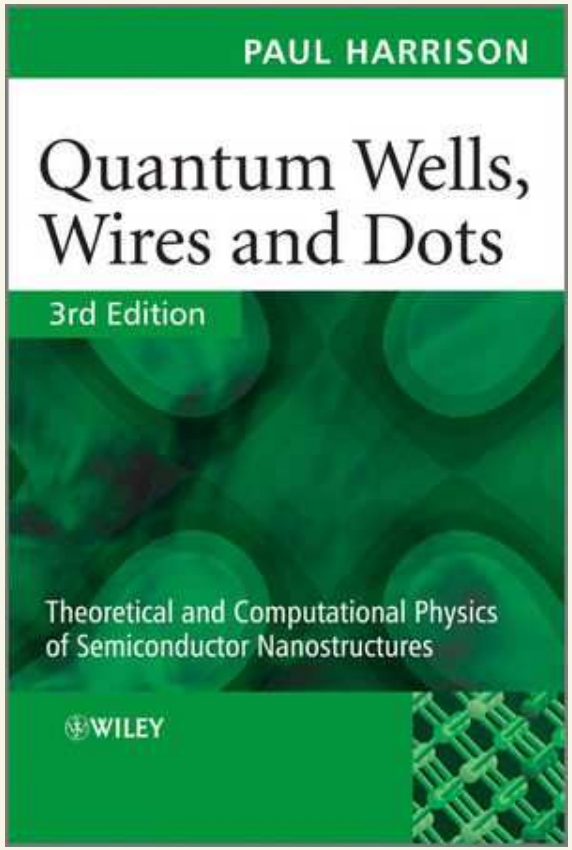


Self-consistent (non-parabolic) solver gives correct result for lasing transition! Slower, but probably worth it!

Shameless advertising!

All these numerical methods are discussed in full in the 4th edition of "Quantum Wells, Wires and Dots", P. Harrison and A. Valavanis, Wiley (expected 2015).

- New free and open source software library (beta testers wanted!)
- New end-of-chapter examples
- Thermal modelling
- Non-parabolic dispersion
- Density-matrix calculations



Free software example

QWWAD simulation suite is based on simple scripts, which tie together programs that solve specific physical problems.
The full non-parabolic, self-consistent solver is implemented using the following code snippets

```
# Define padded QCL structure: width, alloy, doping
cat > s.r << EOF
400     0.16   0
103     0      0
17      0.16   0
107.5   0      0
36      0.16   0
88      0      0
39.5    0.16   0
172     0      3e16
400     0.16   0
EOF
```

sourceforge.net/projects/qwwad

```
# Generate samples of alloy potential at each point
find_heterostructure

# Compute potential, mass and permittivity at each
    point
efxv

# Find an estimate of the field effect (assume no
    charge)
find_poisson_potential --uncharged --field 7.6
```

sourceforge.net/projects/qwwad

```
# Solve Poisson and Schroedinger equations iteratively
for l in `seq 1 9`; do

  # Use non-parabolic SE solver, with max. 5 subbands
   efss --solver matrix-taylor-nonparabolic --nst-max 5

   densityinput  # Estimate subband populations
   chargedensity # Compute charge profile

   find_poisson_potential --field 7.6
done
```

sourceforge.net/projects/qwwad

Conclusions

- Numerical methods are required for solving the Schrödinger equation in QCLs
- Boundary conditions are not trivial for QCLs!
- Matrix methods are fast, accurate and reliable
- It is important to account for spatially varying (and nonparabolic) effective mass
- Self-consistent Poisson–Schrödinger solution is also important
- Our free software is available online. . . textbook coming soon!

Acknowledgements

Thanks to my colleagues and the students in the IMP "quantum well"

- Paul Harrison, Sheffield Hallam University
- Jonny Cooper, Andrew Grier, Zoran Ikonić, Rob Kelsall and Dragan Indjin, Inst. Microwaves and Photonics