Lecture 2

Electron states and optical properties of semiconductor nanostructures
Bulk semiconductors

Band-gap slavery: only light with photon energy equal to band gap can be generated.

Very few semiconductors are suitable

Near-infrared, red, blue
Just recently – green

Mid-infrared: low-T operation, bad quality

Oscillator strengths, selection rules cannot be changed

Low density of states, low $dg/dN$
Quantum-confined electron gas
Envelope function approximation

(a) Add quantum-well potential $U(r)$ to the bulk Hamiltonian $H_0$

(b) Seek the solution as

$$\psi(r) = \frac{1}{A} \sum_n f_n(z) e^{ik_xx + ik_yy} u_{n0}(r)$$

$f_n(z)$ – slowly varying envelope functions

(c) Replace $k_z$ with $-i \frac{\partial}{\partial z}$ and solve the resulting differential matrix equation for the vector $f(z)$

For a single band we may obtain effective mass approximation:

$$-\frac{\hbar^2}{2m_{eff}(z, E)} \frac{d^2 f(z)}{dz^2} + \left( \frac{\hbar^2 k_{||}^2}{2m_{eff}} + U(z) \right) f(z) = Ef(z)$$

Continuity of $f$ and its flux  Particle-in-a-box intuition
Quantum wells

Bulk: \( \psi = e^{i k r} u_k(r) \)

Quantum well:
\[
\psi = f(z)e^{ik_x x + ik_y y} u_k(r)
\]

\[
E_n \approx E_{n0} + \frac{\hbar^2 k_{||}^2}{2m_n}; \quad n = 1, 2, ...
\]

\( k_z = k_n; \quad n = 1, 2, ... \)
Envelope functions $f(z)$ and optical transitions

Interband transitions: similar to bulk materials, but better performance
Optical transitions in quantum wells

Intersubband transitions: sharp atomic-like lines
No cross-absorption

Line broadening ~ 10 meV due to interface roughness and non-parabolicity (in narrow-gap semiconductors)
Intersubband transitions: dipole moment

Dipole matrix element: \( z_{mn} \propto \int f_m^*(z) \frac{\partial}{\partial z} f_n(z) \, dz \)

\[ f_1 \sim \frac{1}{\sqrt{L_z}} \cos k_z z, \quad f_2 \sim \frac{1}{\sqrt{L_z}} \sin k_z z; \quad \Rightarrow z_{12} \sim L_z \]

Typical values ~ 10-100 Å
Compare with atomic transitions ~ 0.2-0.5 Å
Intersubband transitions: selection rules

- Only TM-polarization (E \perp QW plane)

- Dipole matrix element:

\[ z_{mn} \propto \int f_m^*(z) \frac{\partial}{\partial z} f_n(z) \, dz \]

\[ f_1 \text{ and } f_3 \text{ are even } \rightarrow z_{13} = 0 \]
Build your own nanostructure:

\[ V(z) \Rightarrow V(z) + eEz \]

- Sharp resonances
- Tunable frequencies and oscillator strengths
- High-quality materials
- Indirect-gap semiconductors
- Coupling to other excitations: phonons, plasmons
Superlattices

Periodic “super” potential superimposed on periodic lattice potential

Keldysh 1964; Esaki and Tsu 1970
From discrete to quasi-continuous spectrum $E(q)$

- minigap
- miniband
Molecular Beam Epitaxy

Growth rate 1 $\mu$m/hr or 1 atomic layer in 1 sec

A. Cho, Bell Labs.
III-V semiconductor grown on Ge
Only materials with closely matching lattice periods and thermal expansion coefficients can be grown on top of each other without defects.
GaAs/Al$_x$Ga$_{1-x}$As; Ga$_x$In$_{1-x}$As$_y$P$_{1-y}$/Al$_x$In$_{1-x}$As on InP; InAs$_{1-x}$Sb/AlGa$_{1-x}$Sb on GaSb

Fig. 7.6. Bandgap energy and lattice constant of various III-V semiconductors at room temperature (adopted from Tien, 1988).
Quantum wires and dots

Quantum dots of semiconducting materials

- **growth process**

- **TEM image**

- "self-organization"

- **lasing**

http://www.mpi-halle.mpg.de/
Magnetic quantum wires and dots

Landau levels:

\[ E_n \approx \hbar \omega_B \left( n + \frac{1}{2} \right) + \frac{\hbar^2 k_z^2}{2m_n}; \quad n = 0, 1, 2, \ldots \]
Electron states in semiconductor nanostructures

Blackboard derivation
Density of states

3D Bulk Semiconductor

2D Quantum Well

N(E)

1D Quantum Wire

0D Quantum Dot
Quantum-confined electron gas has sharp, tunable resonances in “optics” (from terahertz to visible light)

How can we use it?

• Determine material parameters: effective masses, band offsets, g-factors, scattering rates
• Study new phenomena: Bloch oscillations, huge optical nonlinearities, BEC of excitons, entangled states, …
• Make new devices: lasers, detectors, transistors, memory, computers, etc.
How to get lasing between intersubband transitions?

Problem: ultrafast relaxation due to phonon emission
Superlattice laser: Kazarinov and Suris 1971

Wavefunctions

Electric field $F = 40 \text{ kV/cm}$

Does not work due to domain formation and insufficient population inversion
Quantum cascade lasers

- Control of lifetimes: phonons, tunneling; need $t_{32} > t_2$
- Cascading: high power when $t_{stim}$ approaches $T_1$

$E_{21} = E_{phonon}$

$k_{w,j} l_{w,j} + k_{b,j} l_{b,j} = \pi$
Vertically stack 20-30 stages; sandwich them into the waveguide supporting a low-loss transverse EM mode
Mid-infrared ($\lambda \sim 4\text{-}10\ \mu\text{m}$)
Quantum cascade lasers are

- Extremely powerful ($P_{\text{max}} > 20\ W$)
- Operate at high temperatures $T_{\text{max}} \sim 100\ ^\circ\text{C}$
- Reliable, stable, etc.
Problems with lasers:

• Lasers are not widely tunable, do not cover all wavelengths of interest, can operate CW at room-T only in the narrow spectral range, cryogenic at very short and very long wavelengths

• Nonlinear optical sources (OPO etc.) flexible and tunable, but they are bulky and expensive

• Is it possible to combine the advantages of both types of sources??
Nonlinear Optics

\[ P_{NL} = \chi^{(1)} E + \chi^{(2)} EE + \ldots \]

- Need high-power external laser pump
- Nonlinearity is small in the transparency region
- Bulky and costly lab equipment
Resonant nonlinear optics with nanostructures

Coupled quantum well structures can be designed to have huge resonant optical nonlinearity (known for 30 years)

\[ |\chi^{(2)}| \sim \frac{N_e d_{12} d_{13} d_{23}}{\hbar^2 (\gamma_{12}^2 + \Delta_{12}^2)(\gamma_{13}^2 + \Delta_{13}^2)} \]

- \( \Delta_{ij} \) – detunings
- \( \gamma_{ij} \) – linewidths
- \( d_{ij} \) – dipole moments

\[ |\chi^{(2)}| \sim 10^4 - 10^6 \text{ pm/V} \]

Compare with 1-10 pm/V for bulk crystals
Double resonance:

\[ P_{NL} = \chi^{(1)}E + \chi^{(2)}EE + \ldots \]

Resonance in absorption for both pump and the nonlinear signal:

\[
\left| \chi^{(2)} \right| \sim \frac{N_e d_{12} d_{13} d_{23}}{\hbar^2 (\gamma_{12}^2 + \Delta_{12}^2)(\gamma_{13}^2 + \Delta_{13}^2)}
\]

\[
\text{Im}\left[ \chi_{\text{pump}}^{(1)} \right] \sim \frac{N_e d_{12}^2 \gamma_{12}}{\hbar (\gamma_{12}^2 + \Delta_{12}^2)}
\]
Conventional nonlinear optics

All detunings are large: $\Delta \sim \omega >> \gamma$;

All frequencies are in the transparency region of the NLO crystals

- Absorption and nonlinearity are small;
- Need high power pump

$\omega \rightarrow 2\omega$
A way to get around resonant absorpti

Resonant optical nonlinearity is accompanied by resonant absorption

\[ |\chi^{(2)}| \sim \frac{N_e d_{12} d_{13} d_{23}}{\hbar^2 (\gamma_{12}^2 + \Delta_{12}^2)(\gamma_{13}^2 + \Delta_{13}^2)} \]

Solution: create the nonlinear medium with gain

This leads to nonlinear quantum cascade lasers
Integration of injection lasers with resonant electronic nonlinearities

We deal with semiconductors

Let’s try to inject electrons, create population inversion and generate the optical pump right inside the nonlinear structure

Active nonlinear medium:
Laser field serves as a coherent optical pump for the nonlinear process
One can approach resonance since resonant absorption is compensated by laser gain
The tightest possible confinement and mode purity
No problem with external pump; an injection-pumped device
Monolithic integration of quantum-cascade lasers with resonant optical nonlinearities

- Maximizing the product of dipoles $d_{23}d_{34}d_{24}$
- Quantum interference between cascades I and II

$\chi^{(2)} \sim 10^5$ pm/V in the mid-IR
$\chi^{(2)} \sim 10^6$ pm/V in the THz

This is NOT sequential photon absorption/reemission!

Milliwatt power in SHG:
O. Malis et al. 2004
Single-mode and tunable SH emission

Fabry-Perot Laser

Single-mode Laser

Intensity (a.u.)

Wavelength (µm)

Temperature (K)

Wavelength (µm)

0.2 nm/K

APL 84, 2751 (2004)
Third Harmonic Generation

\[ \omega \rightarrow 2\omega \rightarrow 3\omega \]

11.1 \(\mu\)m \(\rightarrow\) 5.5 \(\mu\)m \(\rightarrow\) 3.7 \(\mu\)m

\[ \frac{P(3\omega)}{P(2\omega)} \sim 10^{-2} \]

\[ \chi^{(3)} \sim 10^{-7} \text{ esu} \]

T. Mosely, A. Belyanin, C. Gmachl, Optics Express 12, 2972 (2004)
Difference frequency generation in two-wavelength QCLs

- Make a powerful mid-IR QCL emitting at two modes
- Provide strong nonlinearity for frequency mixing process
- Design a low-loss, phase-matched waveguide for all three modes

\[ \omega_{THz} = \omega_1 - \omega_2, \quad k_{THz} = k_1 - k_2 \]
Raman lasing and other coherent nonlinear phenomena

- Triply resonant Raman lasing
- Lasing without inversion
- “Slow light”, intersubband polaritons, mixing with phonons, plasmons, ... 
- Beyond semiclassical picture: squeezing, entanglement
- Beyond rate approximation: instabilities, superfluorescence

\[ \rho_{12} \neq 0 \]

Density matrix:
\[
\begin{pmatrix}
\rho_{11} & \rho_{12} & \rho_{13} \\
\rho_{21} & \rho_{22} & \rho_{23} \\
\rho_{31} & \rho_{32} & \rho_{33}
\end{pmatrix}
\]

“Quantum coherence”
In most Raman amplifiers and lasers, both pump and Raman fields are very far from one-photon resonance

- Very large detuning $\Delta$ to avoid absorption
- No real transitions to upper state 3
- Raman shift $\omega_{21}$ is fixed to be the phonon frequency

Gain at two-photon resonance:
$\omega_p - \omega_s = \omega_{21}$
Stimulated Raman scattering

Raman gain

\[ I_p (N_1 - N_2) \propto \frac{\Delta^2 \gamma_{21}}{\Delta} \]

Raman coherence \( \rho_{21} \ll 1 \)

(Except experiments by Sokolov, Harris et al.)

Propagation of coupled Raman and pump fields

Manley-Rowe relation:

\[ \frac{I_p}{\omega_p} + \frac{I_s}{\omega_s} = \text{const} \]
Approaching resonance

Both “good” and “bad” effects get enhanced

- Real one-photon processes become important
- Raman coherence $\rho_{21}$ also increases
- Raman gain increases strongly
- Absorption is increased

$$\frac{\partial E_s}{\partial t} \propto E_p \rho_{12} \propto |E_p|^2 E_s$$
Stokes gain at arbitrary detuning

One-photon absorption

\[ g \propto \text{Re} \left\{ \frac{\omega_s d_{32}^2}{\Gamma_{32} + |\Omega_p|^2 / \Gamma_{21}^*} \left[ \frac{|\Omega_p|^2 (n_1 - n_3)}{\Gamma_{21}^* \Gamma_{31}^*} - (n_2 - n_3) \right] \right\} \]

\[ \Gamma_{32} = \gamma_{32} + i(\omega_{32} - \omega_s) \]

\[ \Gamma_{31} = \gamma_{31} + i(\omega_{31} - \omega_p) \]

\[ \Gamma_{21} = \gamma_{21} + i(\omega_{21} - (\omega_p - \omega_s)) \]

\[ \gamma_{ij} \sim 5 - 10 \text{ meV} \]

Raman gain is enhanced, but resonant absorption of the pump limits the interaction length.

Gain, cm\(^{-1}\)

\[ \omega_s - \omega_{32}, \text{ meV} \]

\[ \Delta \sim \gamma \]

\[ \vec{E}_p \quad \vec{E}_s \]

Rabi frequency

\[ \Omega_p = \frac{d_{31} E_p}{\hbar} \]

\[ \Omega_s = \frac{d_{32} E_s}{\hbar} \]
Mid-IR Raman injection laser

Raman shift is determined by intersubband transition and is tunable

Very large Raman gain at resonance: ~ $10^{-4}$ cm/W

40 mW Raman threshold
16 mW Stokes power