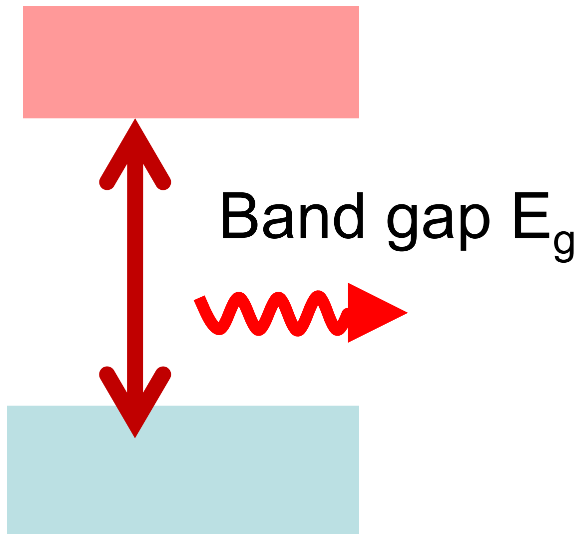
Lecture 2

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

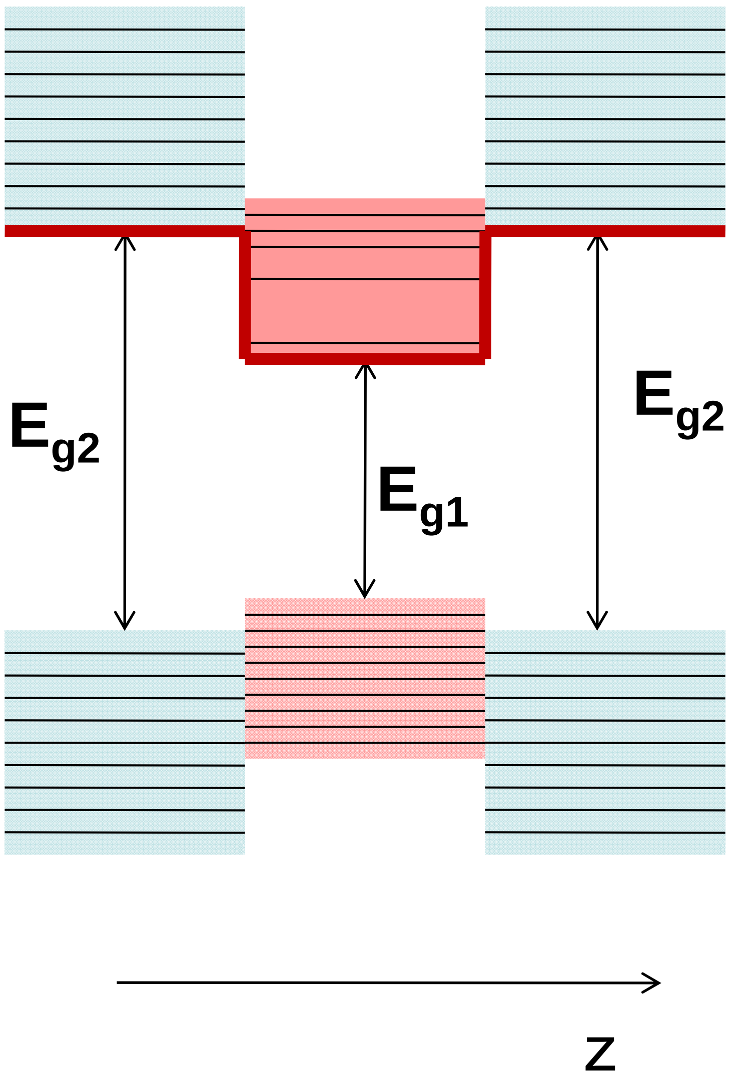
Near-infrared, red, blue
Just recently – green

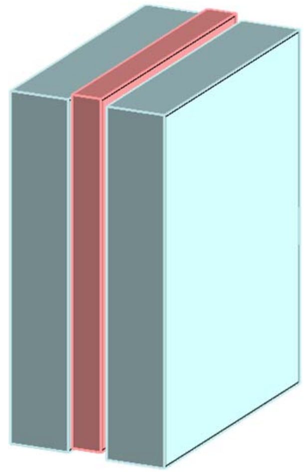
Mid-infrared: low-T operation, bad quality

Oscillator strengths, selection rules cannot be changed

Low density of states, low dg/dN

Quantum-confined electron gas



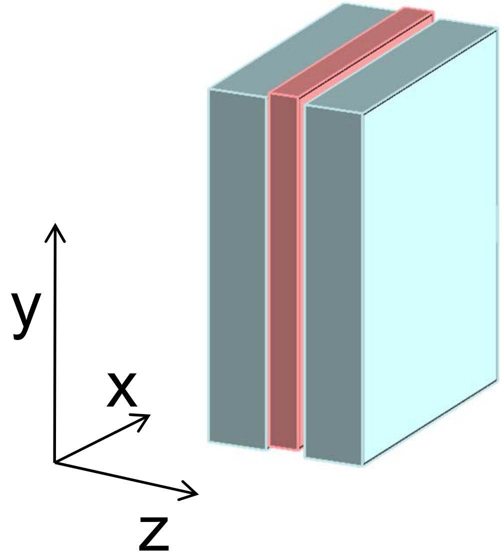


Envelope function approximation

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

(b) Seek the solution as

$$\psi(\mathbf{r}) = \frac{1}{A}\sum_n f_n(z) e^{ik_x x + ik_y y} u_{n0}(\mathbf{r})$$



***$f_n(z)$* – slowly varying envelope functions**

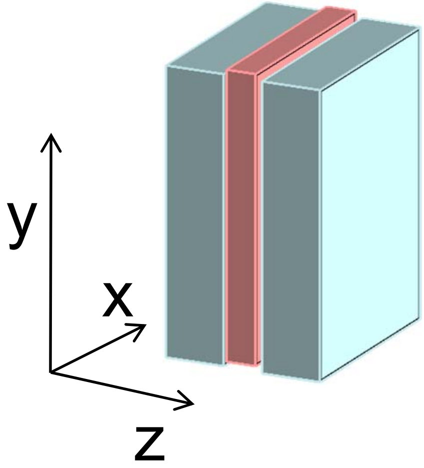
(c) Replace k_z with $-i\frac{\partial}{\partial z}$ **and solve the resulting differential matrix equation for the vector f*(z)***

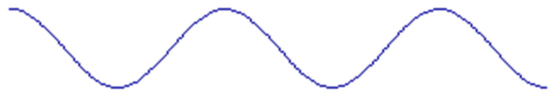
For a single band we may obtain effective mass approximation:

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

Continuity of *f* and its flux **Particle-in-a-box intuition**

Quantum wells

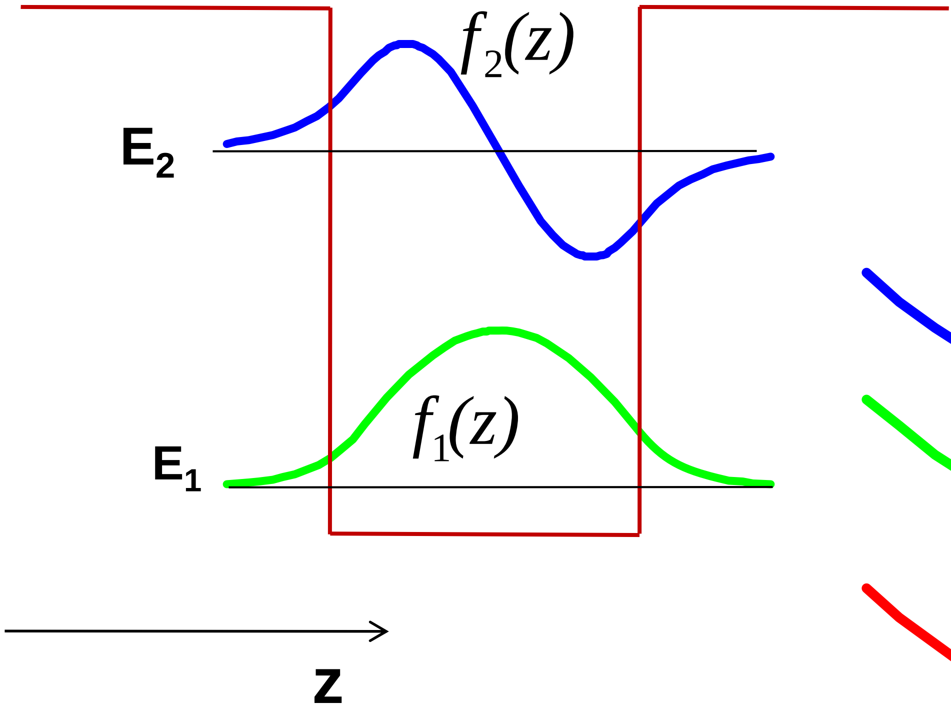




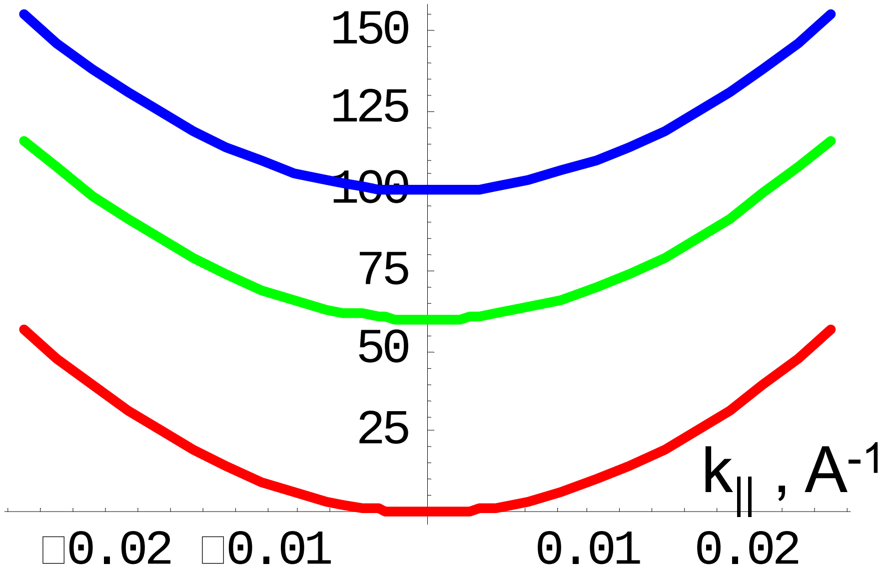
Bulk: $\psi = e^{i\mathbf{kr}} u_{\mathbf{k}}(\mathbf{r})$

Quantum well:

$$\psi = f(z) e^{ik_x x + ik_y y} u_{\mathbf{k}}(\mathbf{r})$$

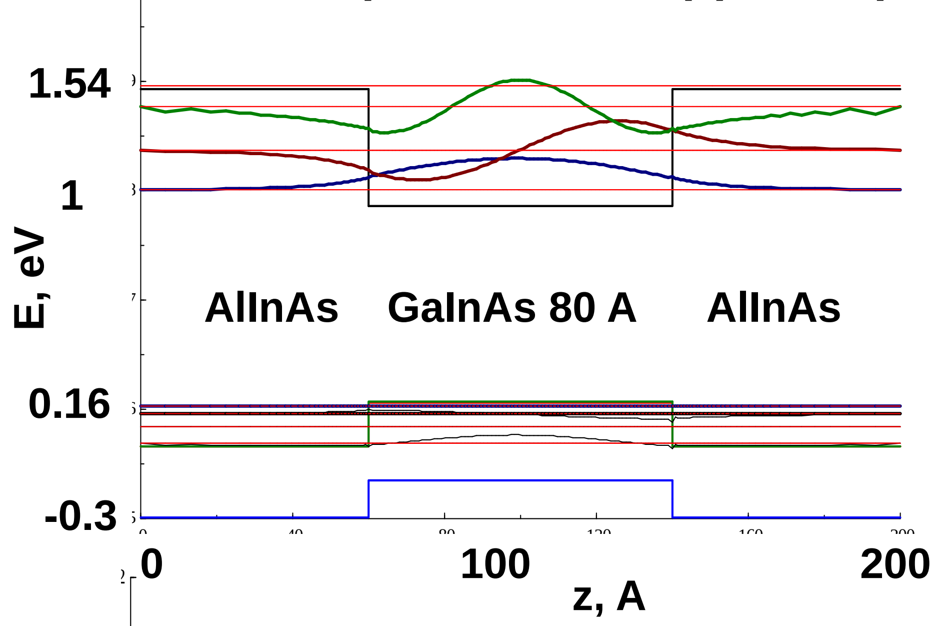


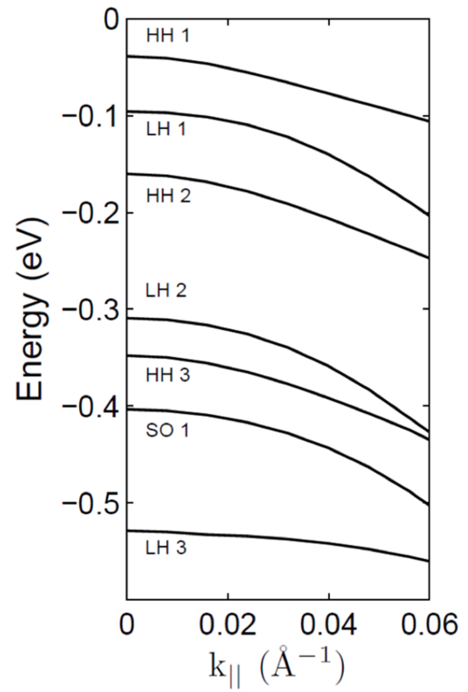
$$E_n \approx E_{n0} + \frac{\hbar^2 k_{\|}^2}{2m_n}; \quad n = 1,2,\ldots$$

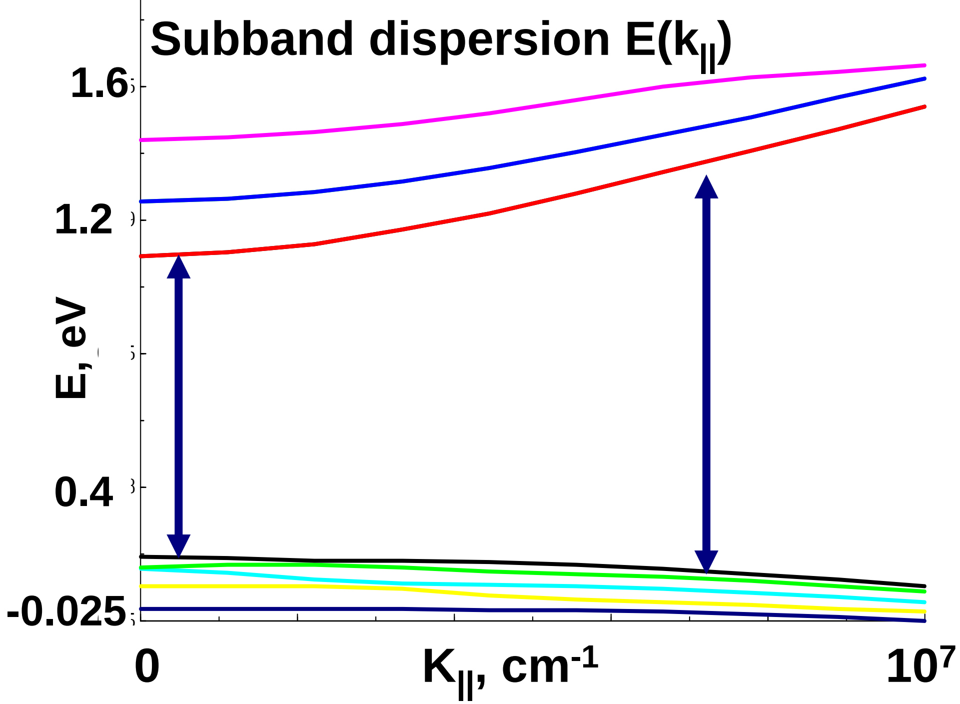


$$k_z = k_n; \quad n = 1,2,\ldots$$

Envelope functions f(z) and optical transitions





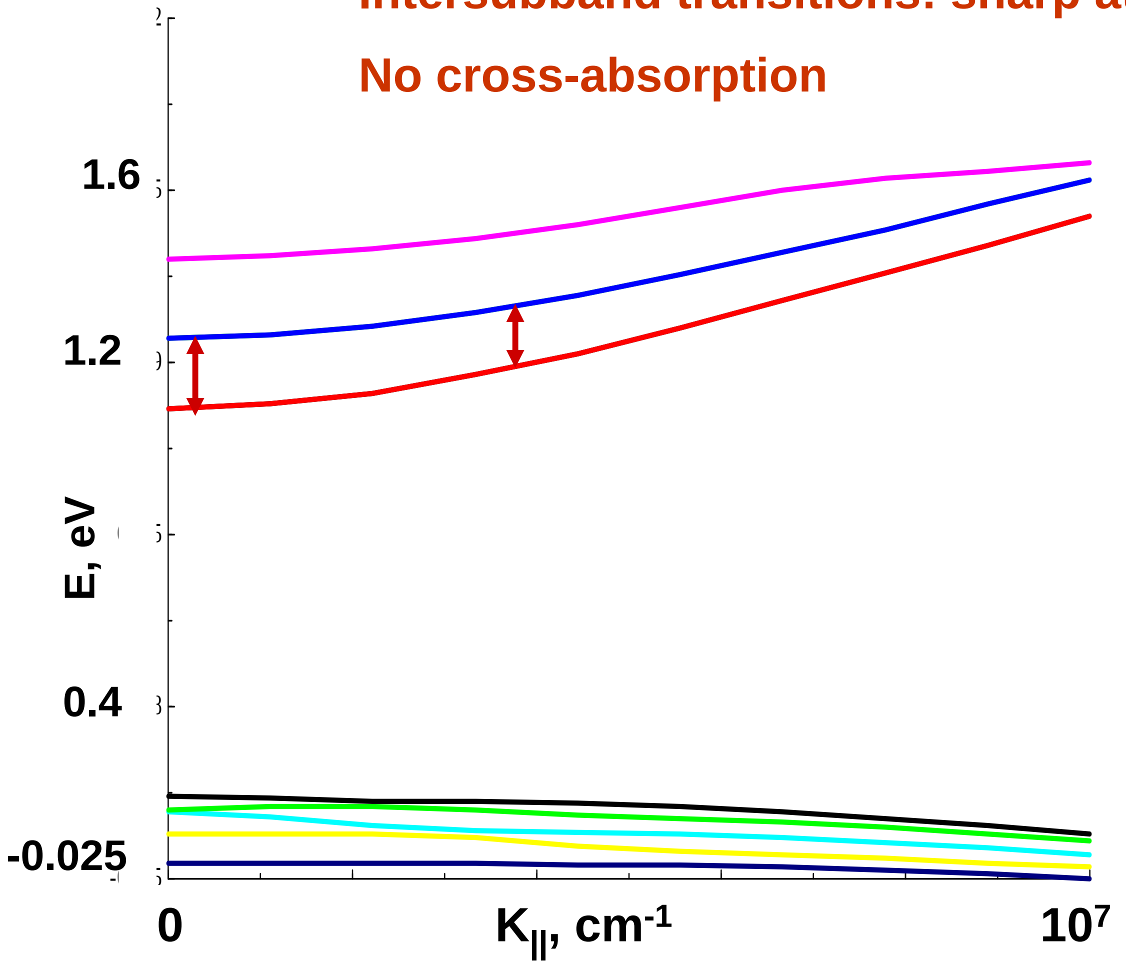


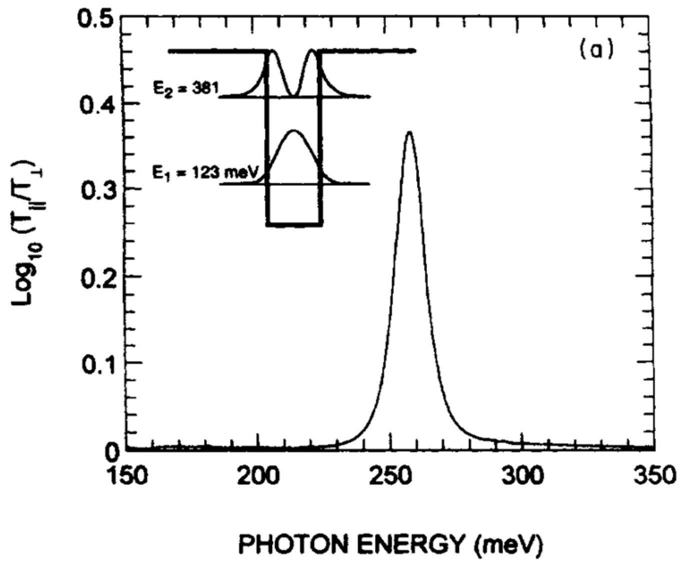
Interband transitions: similar to bulk materials, but better performance

Optical transitions in quantum wells

Intersubband transitions: sharp atomic-like lines

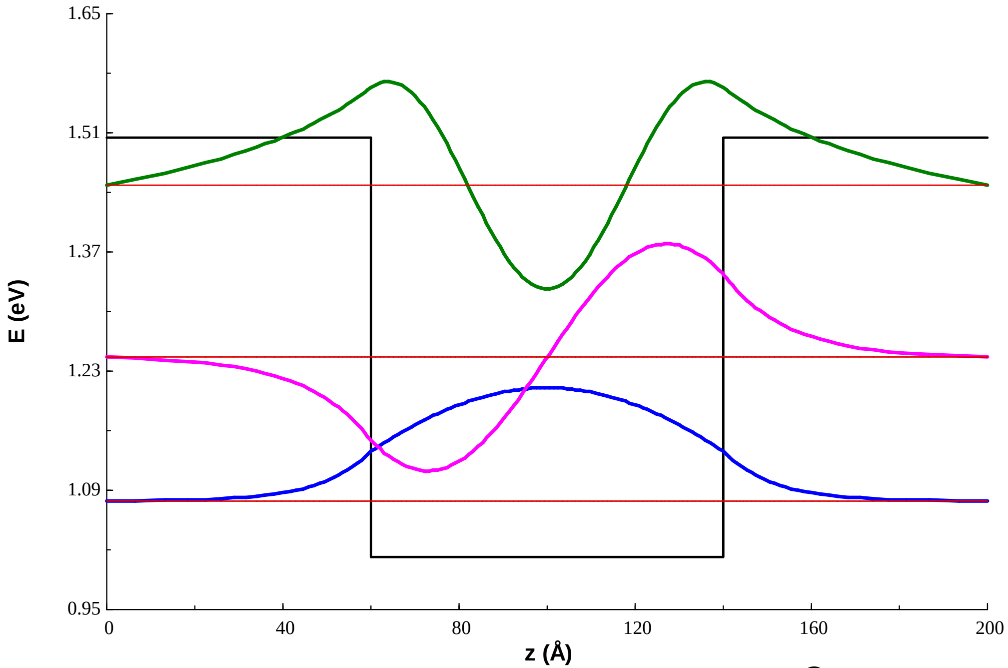
No cross-absorption





Line broadening ~ 10 meV due to interface roughness and non-parabolicity (in narrow-gap semiconductors)

Intersubband transitions: dipole moment

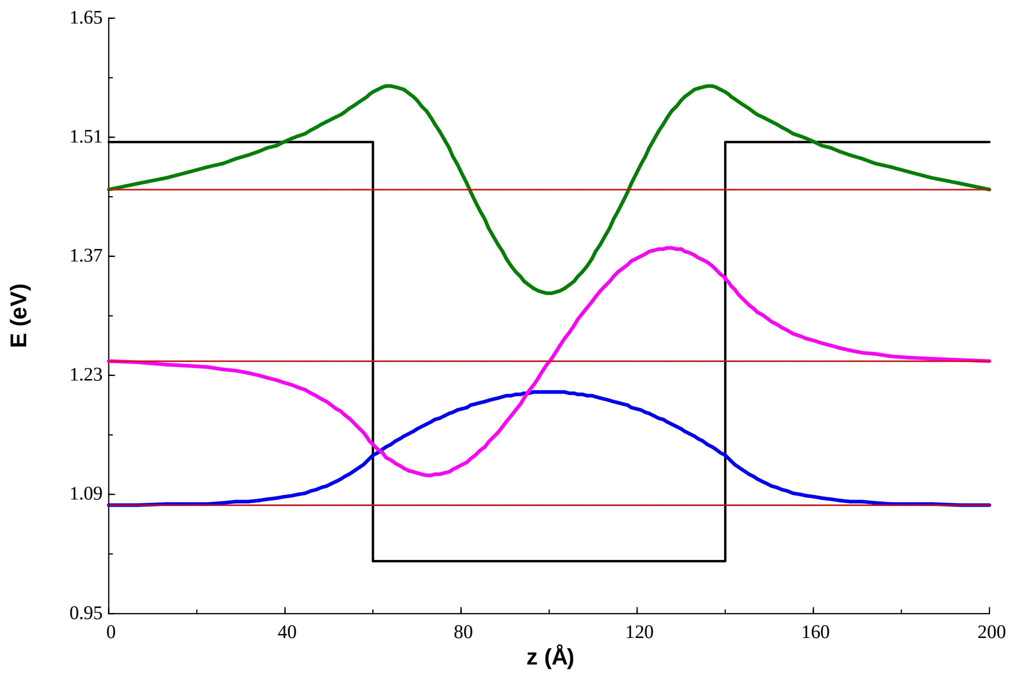


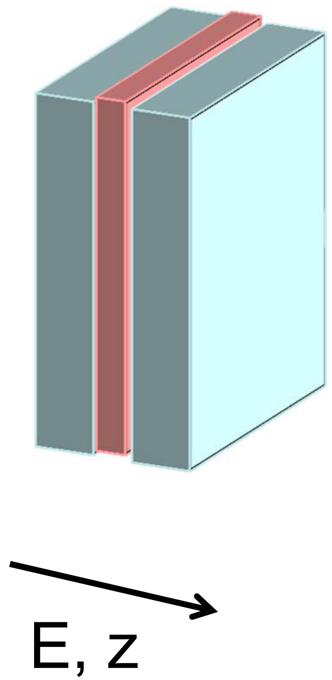
Dipole matrix element: $$z_{mn} \propto \int f_m^*(z) \frac{\partial}{\partial z} f_n(z)\, dz$$

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

Typical values ~ 10-100 A
Compare with atomic transitions ~ 0.2-0.5 A

Intersubband transitions: selection rules





- Only TM-polarization (E ⊥ QW plane)

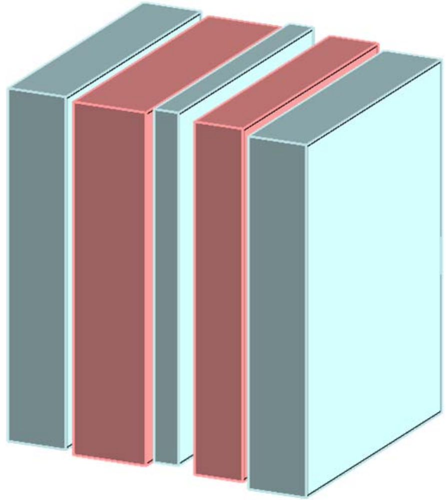
- Dipole matrix element: $$z_{mn} \propto \int f_m^*(z) \frac{\partial}{\partial z} f_n(z)\, dz$$

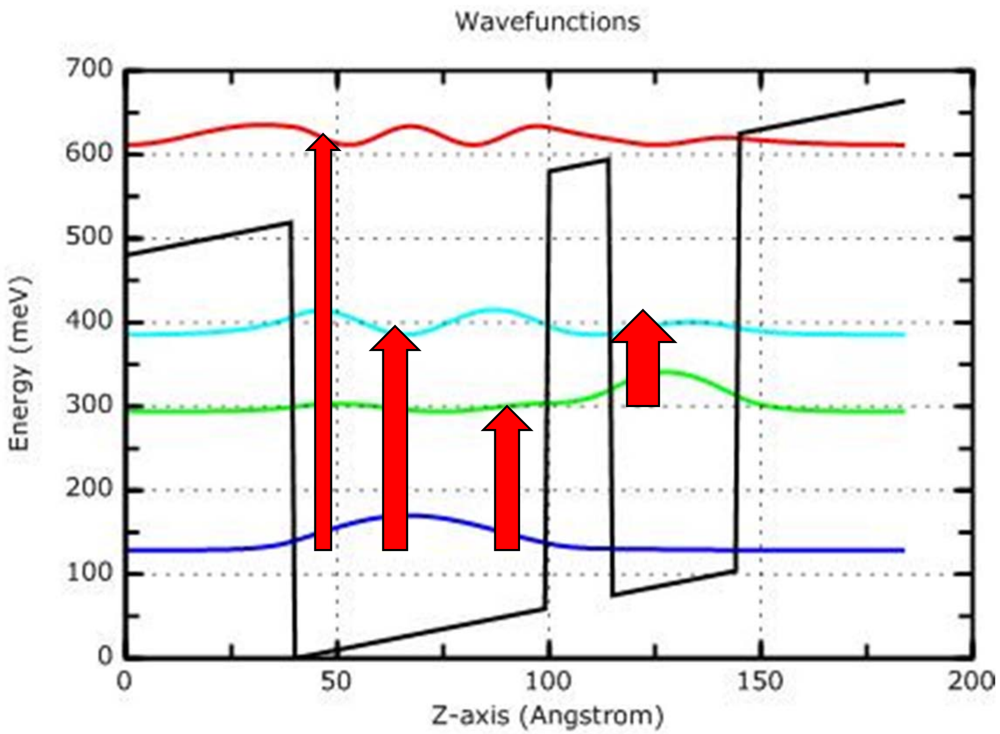
f_1 and f_3 are even -> $z_{13} = 0$

Build your own nanostructure:

E-field

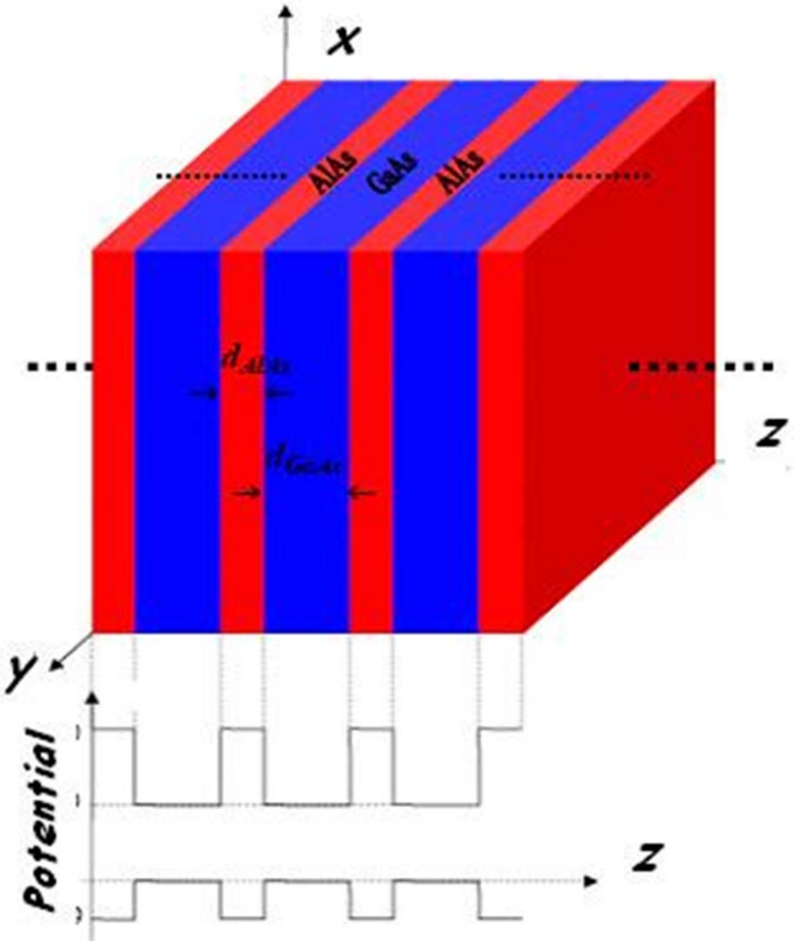
$$V(z) \Rightarrow V(z) + eEz$$





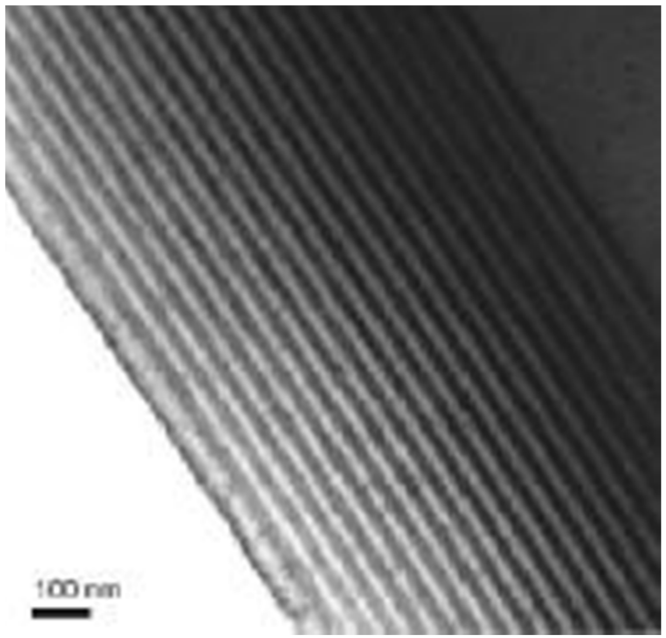
- Sharp resonances
- Tunable frequencies and oscillator strengths
- High-quality materials
- Indirect-gap semiconductors
- Coupling to other excitations: phonons, plasmons

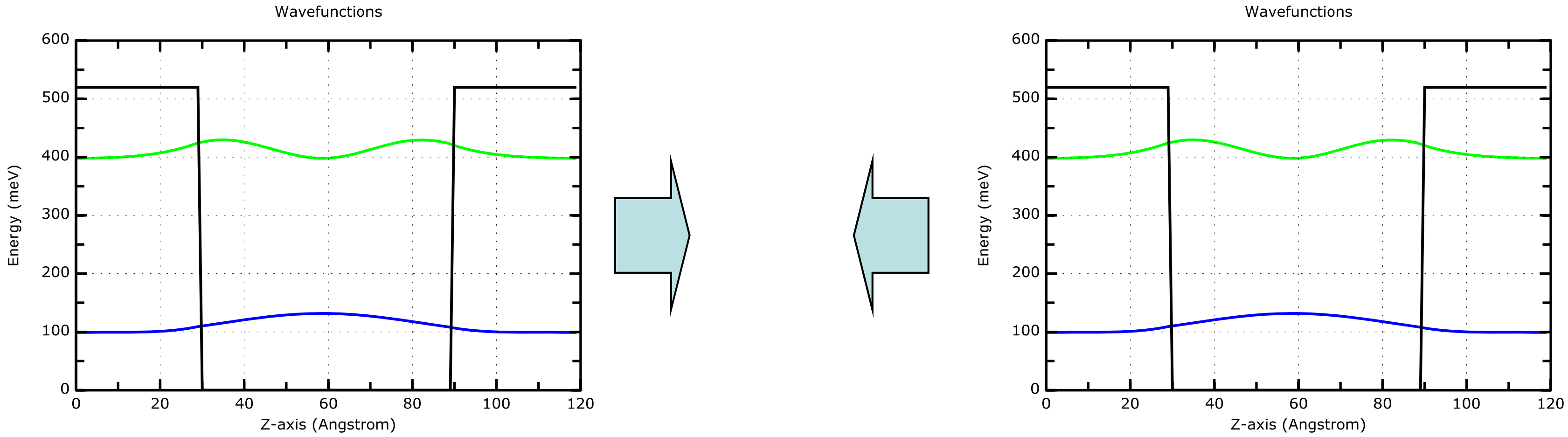
Superlattices



Periodic "super" potential superimposed on periodic lattice potential

Keldysh 1964; Esaki and Tsu 1970

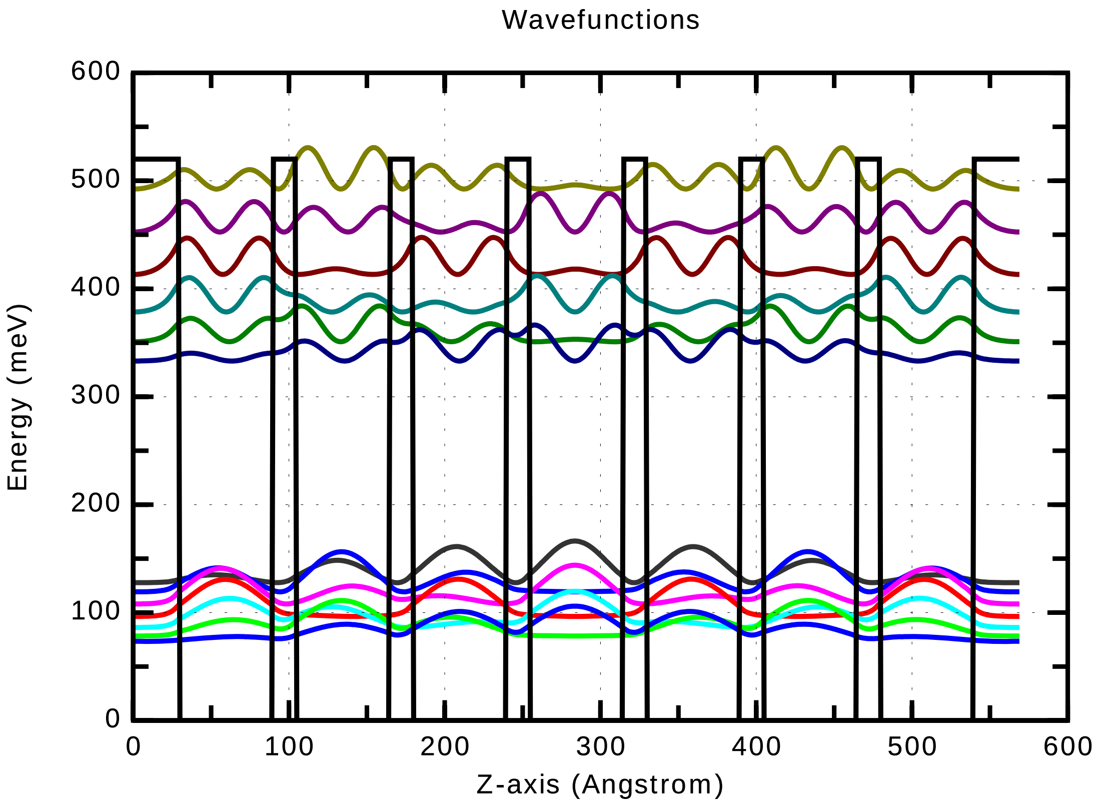




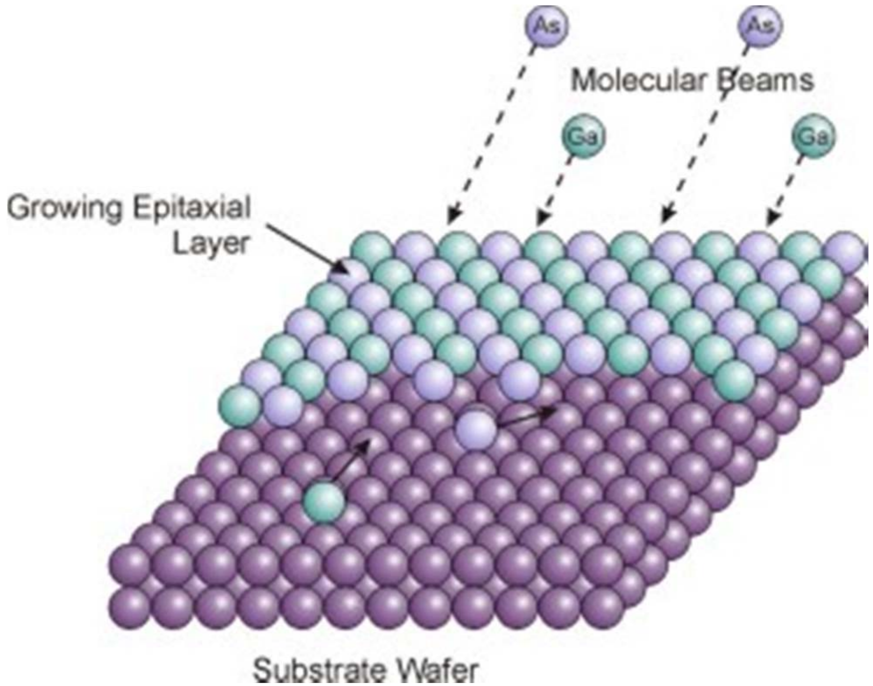
From discrete to quasi-continuous spectrum E(q)

minigap

miniband



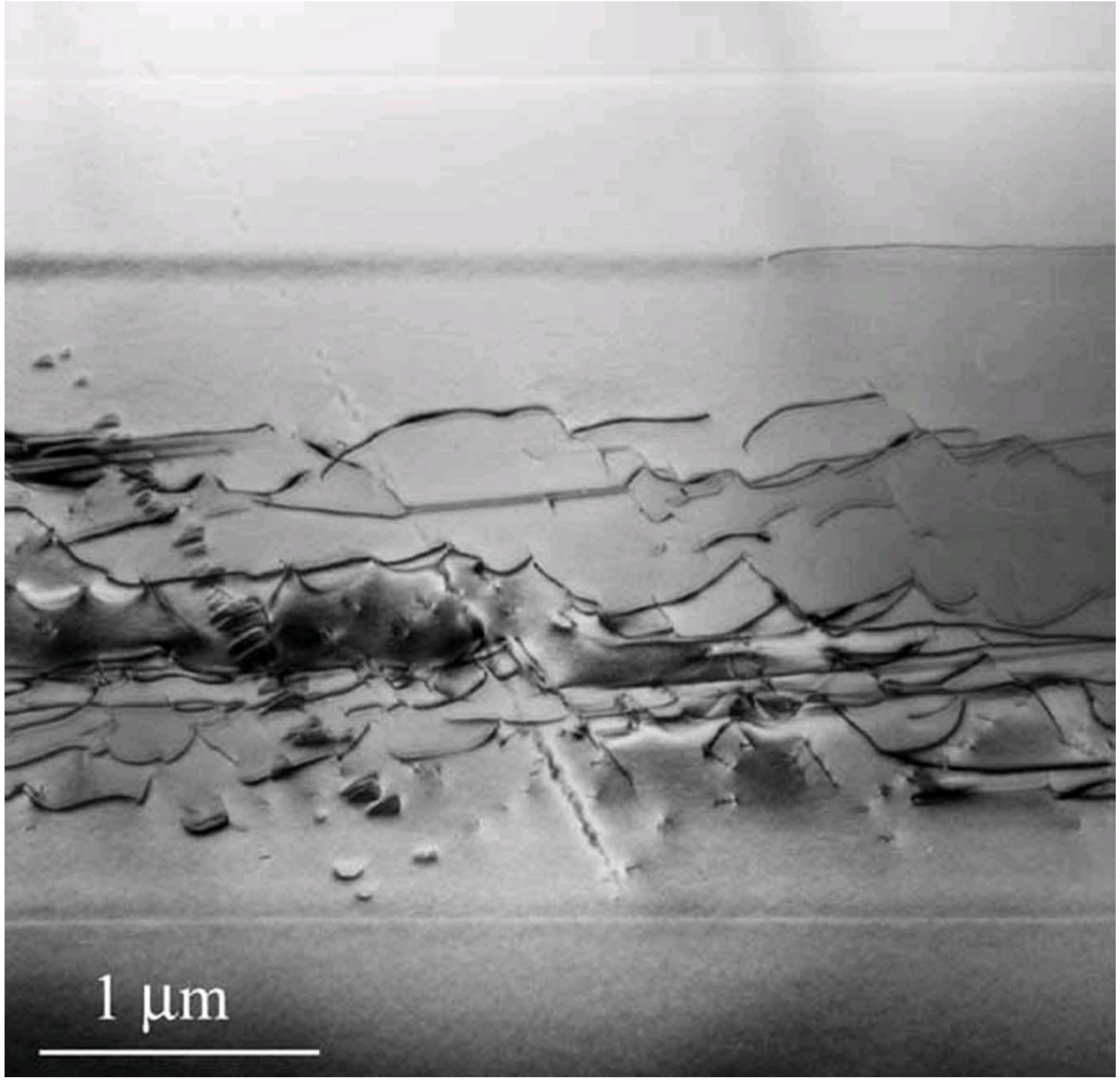
Molecular Beam Epitaxy

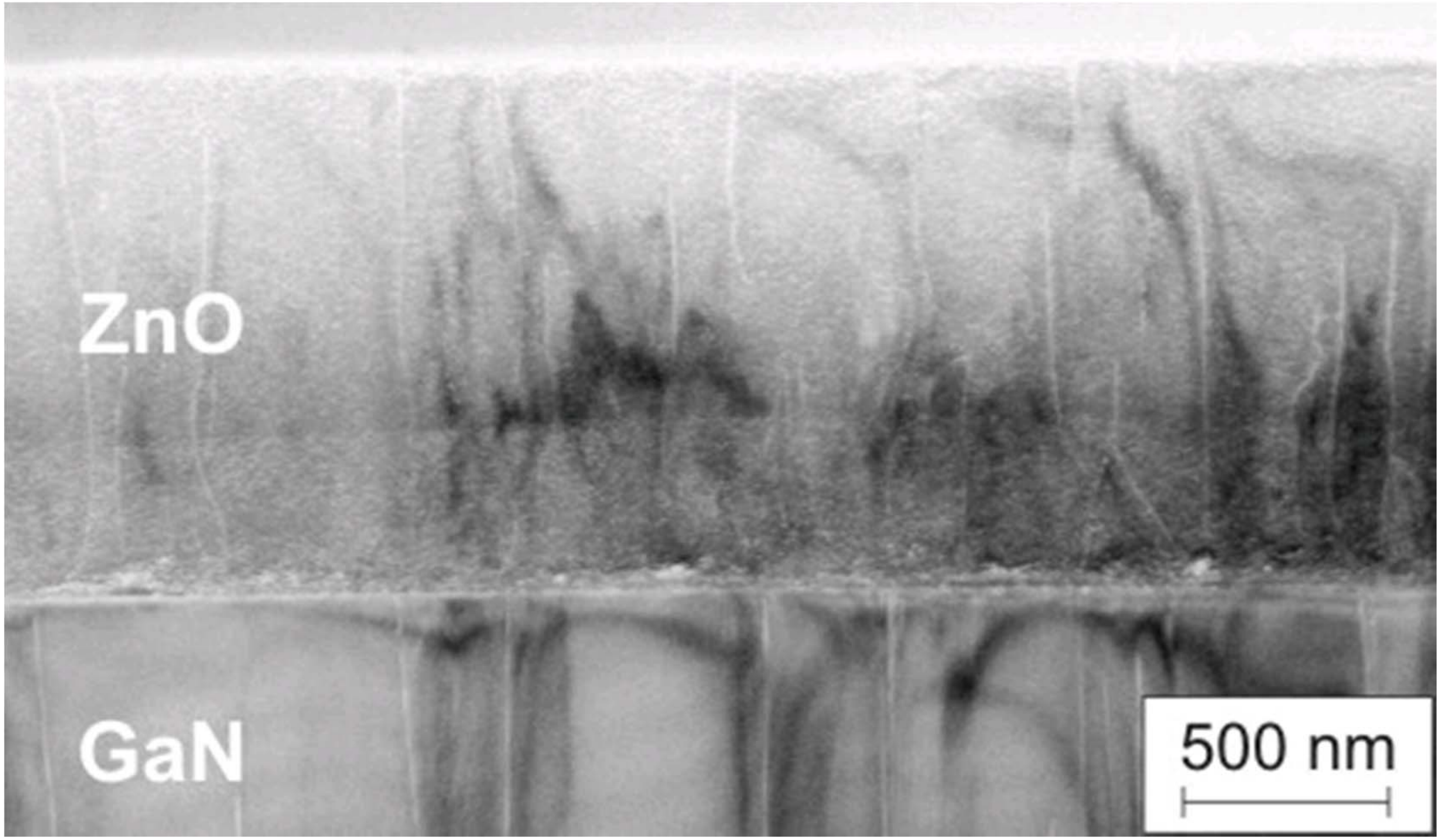


Growth rate 1 μm/hr or 1 atomic layer in 1 sec

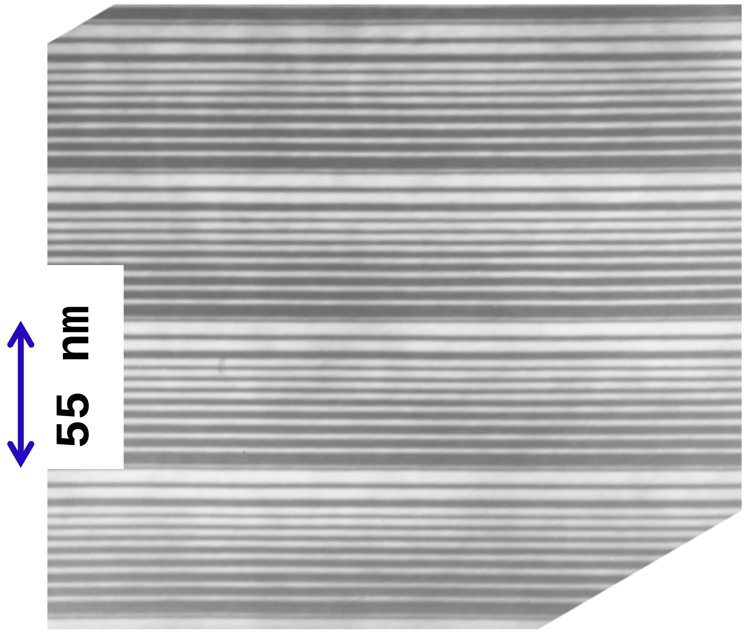
A. Cho, Bell Labs.

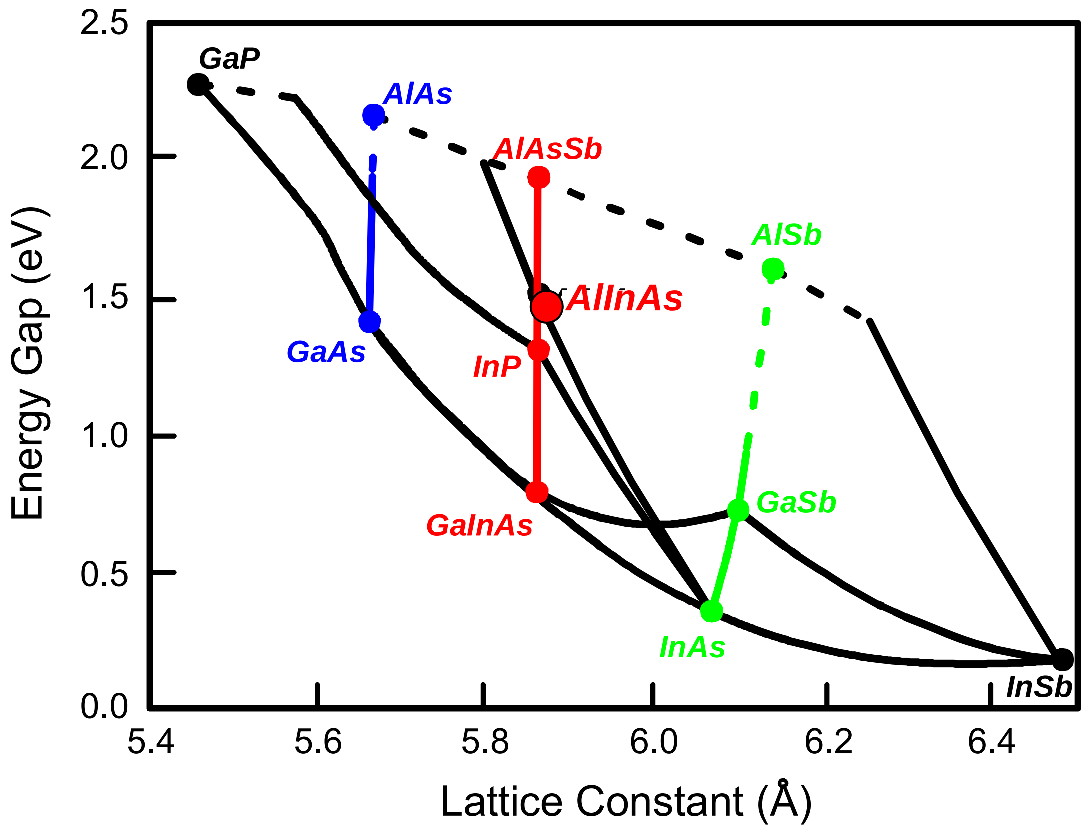
III-V semiconductor grown on Ge




ZnO
GaN
500 nm

Only materials with closely matching lattice periods and thermal expansion coefficients can be grown on top of each other without defects





$Ga_{0.47}In_{0.53}As/Al_{0.48}In_{0.52}As/$InP

$GaAs/Al_xGa_{1-x}As/$GaAs

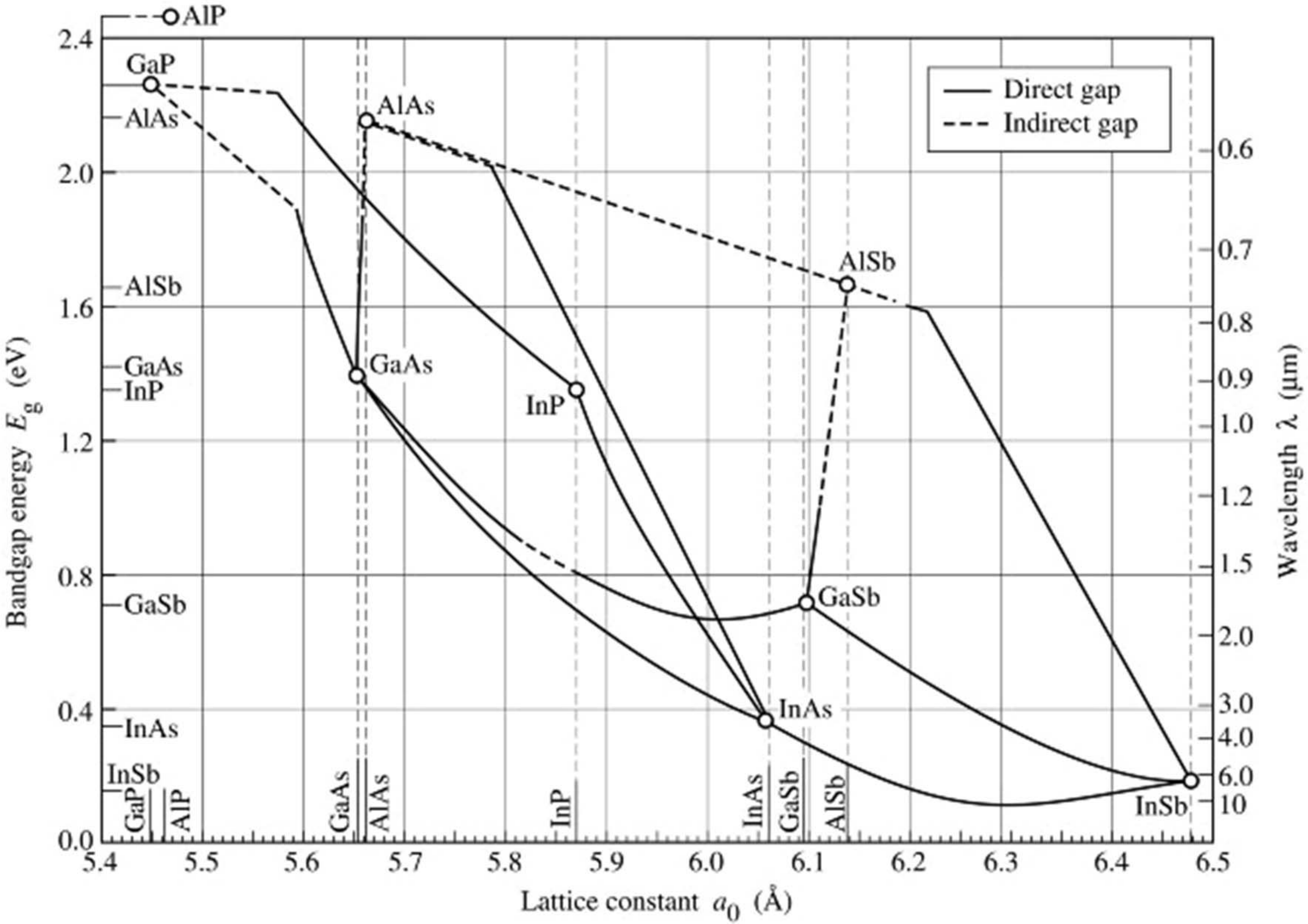
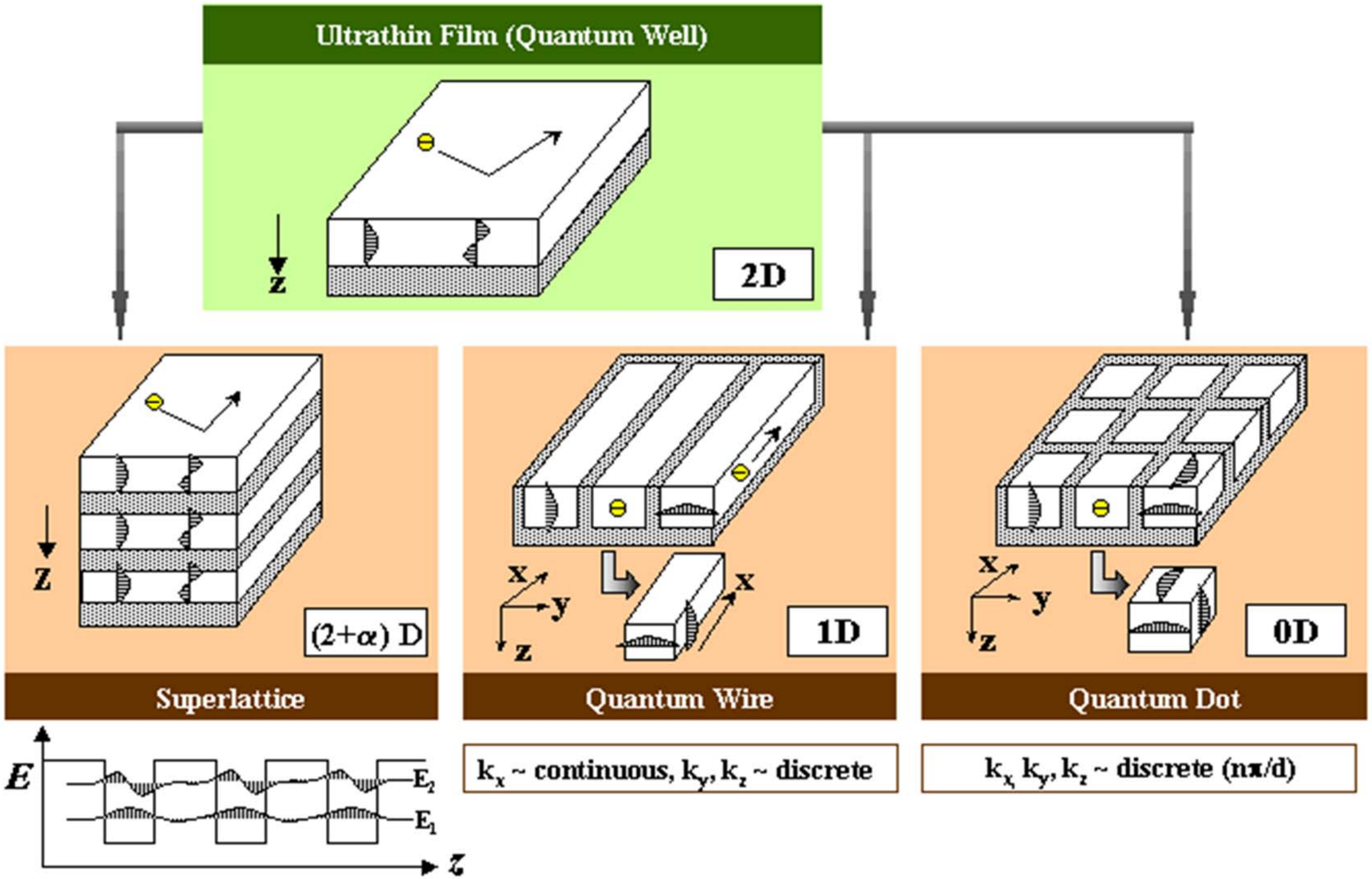


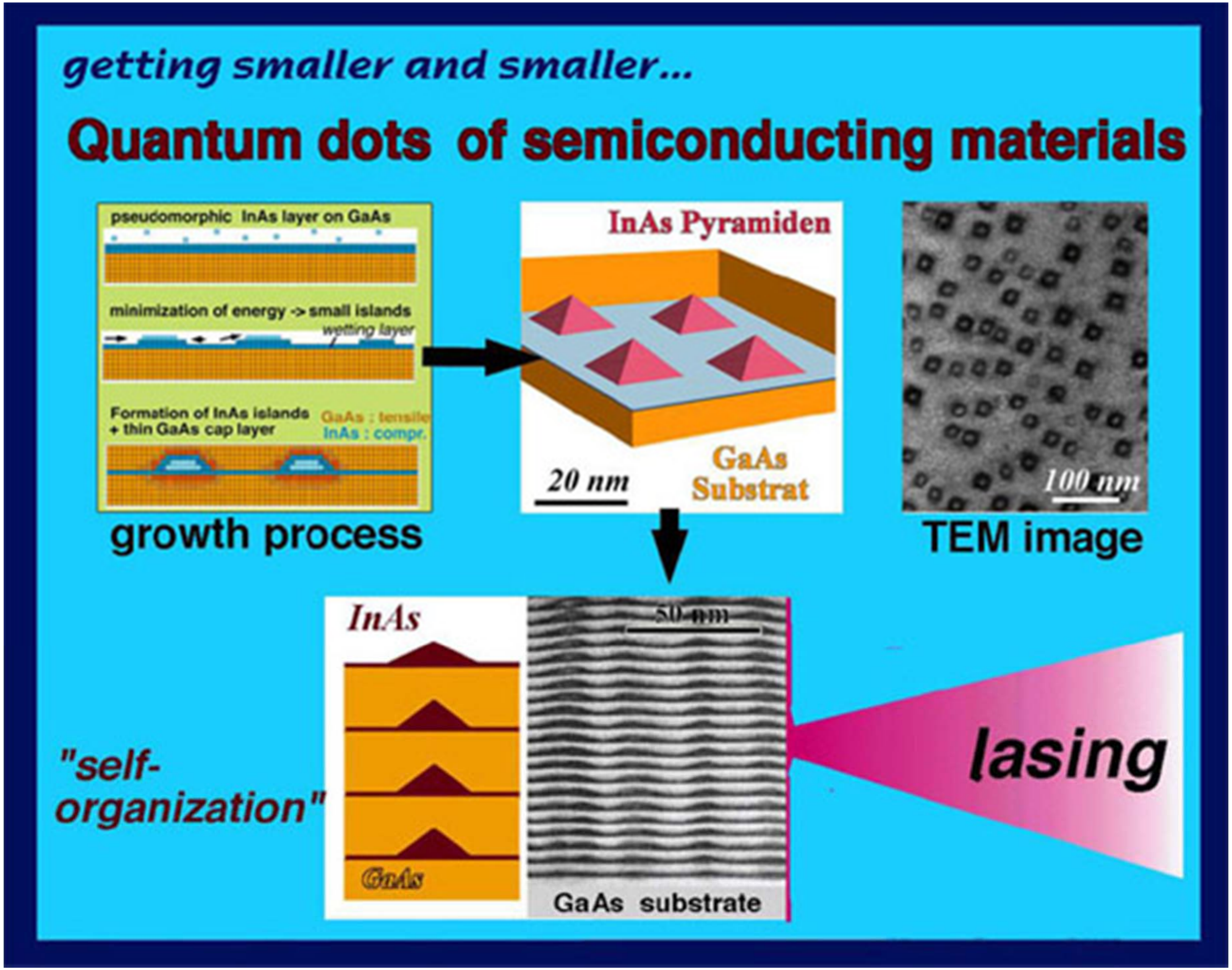
Fig. 7.6. Bandgap energy and lattice constant of various III-V semiconductors at room temperature (adopted from Tien, 1988).

GaAs/$Al_xGa_{1-x}As$; $Ga_xIn_{1-x}As_yP_{1-y}$/$Al_xIn_{1-x}As$ on InP; $InAs_{1-x}Sb$/$AlGa_{1-x}Sb$ on GaSb

Quantum wires and dots

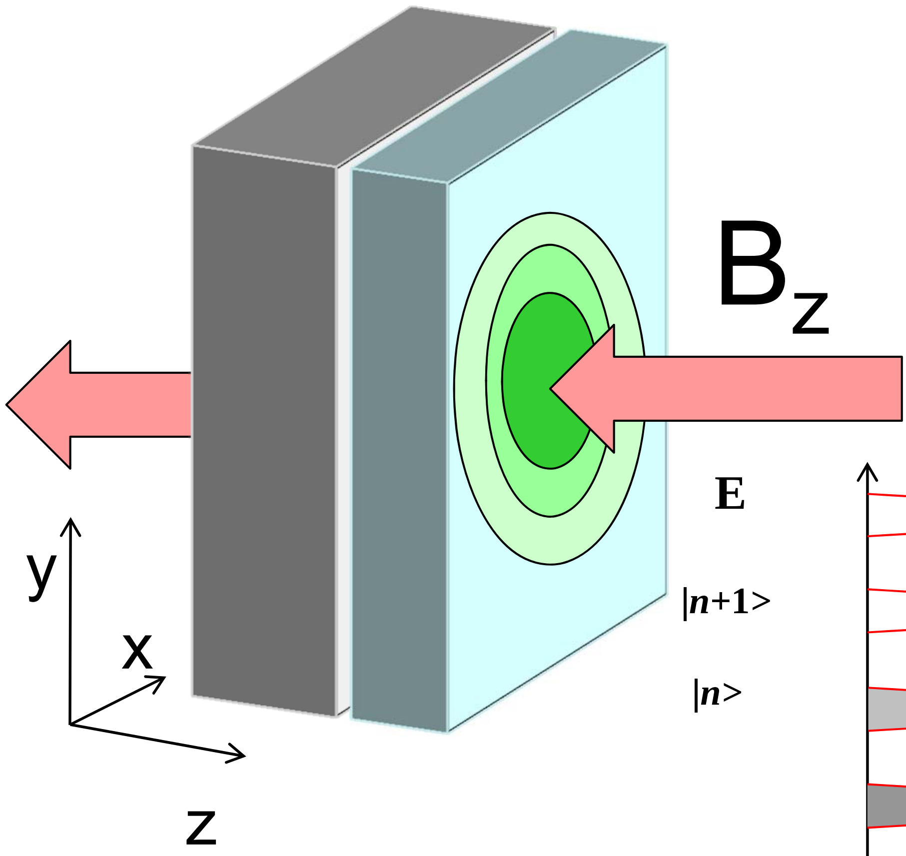


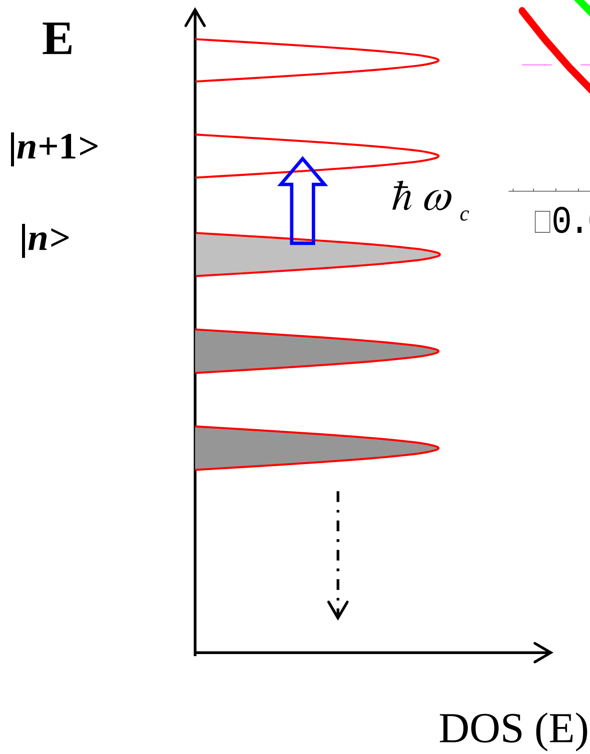
http://www.nanonet.go.jp/english/mailmag/2003/001a.html



http://www.mpi-halle.mpg.de/

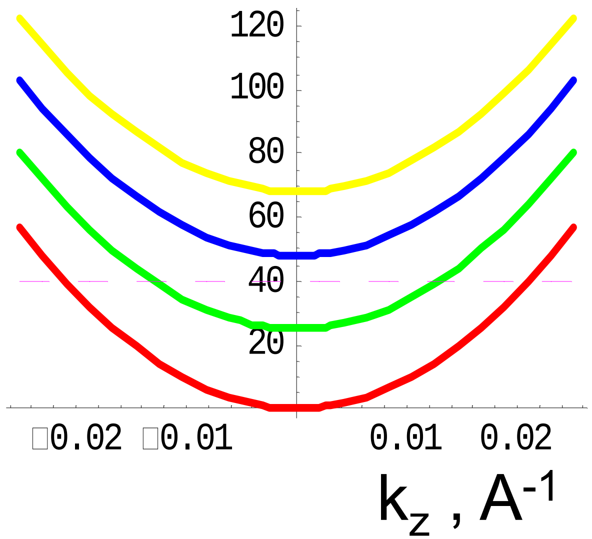
Magnetic quantum wires and dots





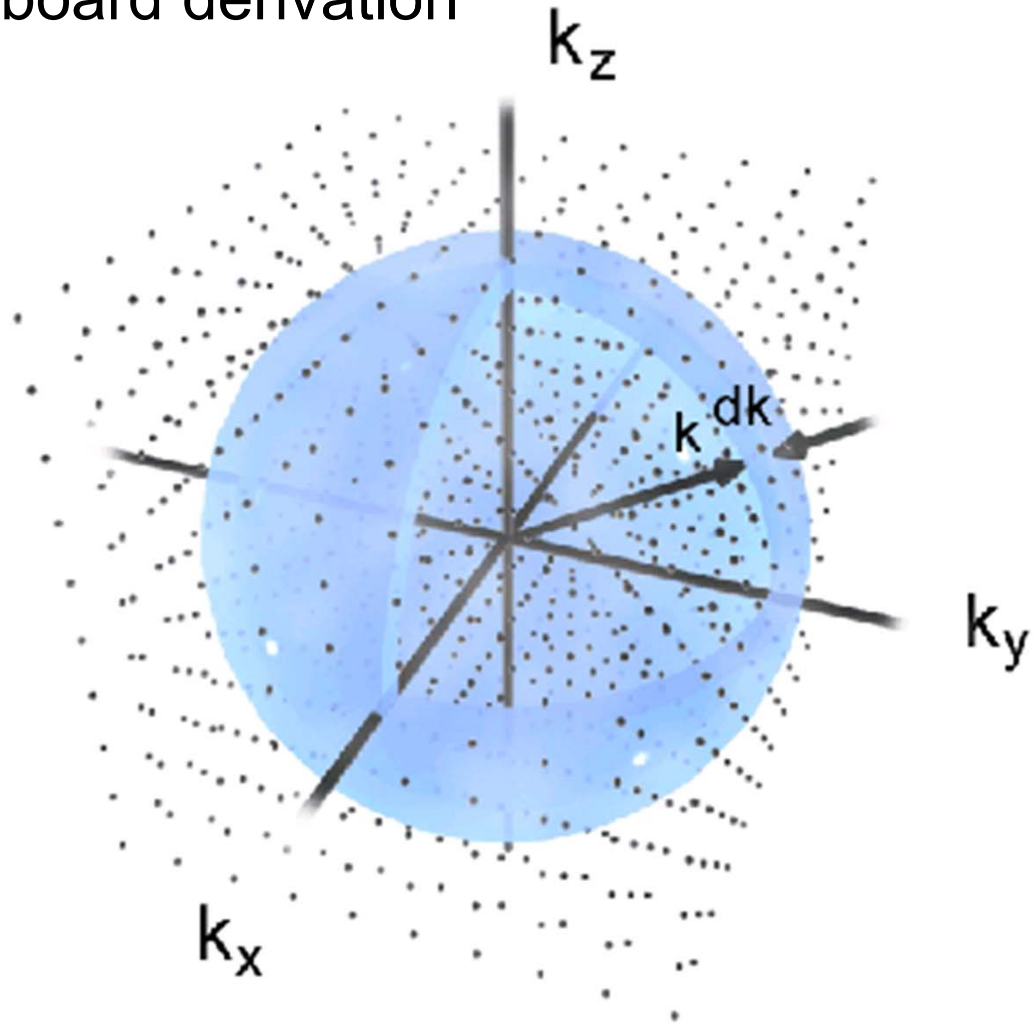
Landau levels:

$$E_n \approx \hbar\omega_B\left(n+\frac{1}{2}\right)+\frac{\hbar^2 k_z^2}{2m_n}; \quad n=0,1,2,\ldots$$

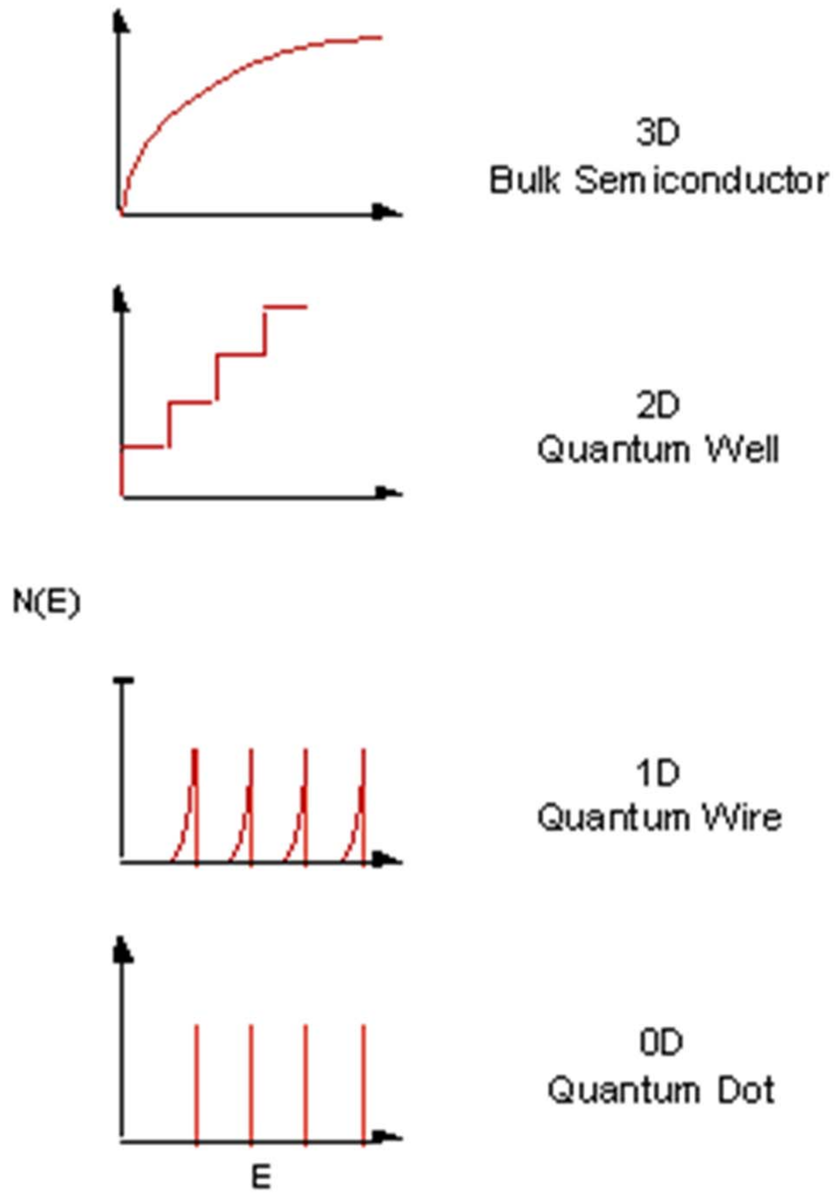


Electron states in semiconductor nanostructures

Blackboard derivation



Density of states



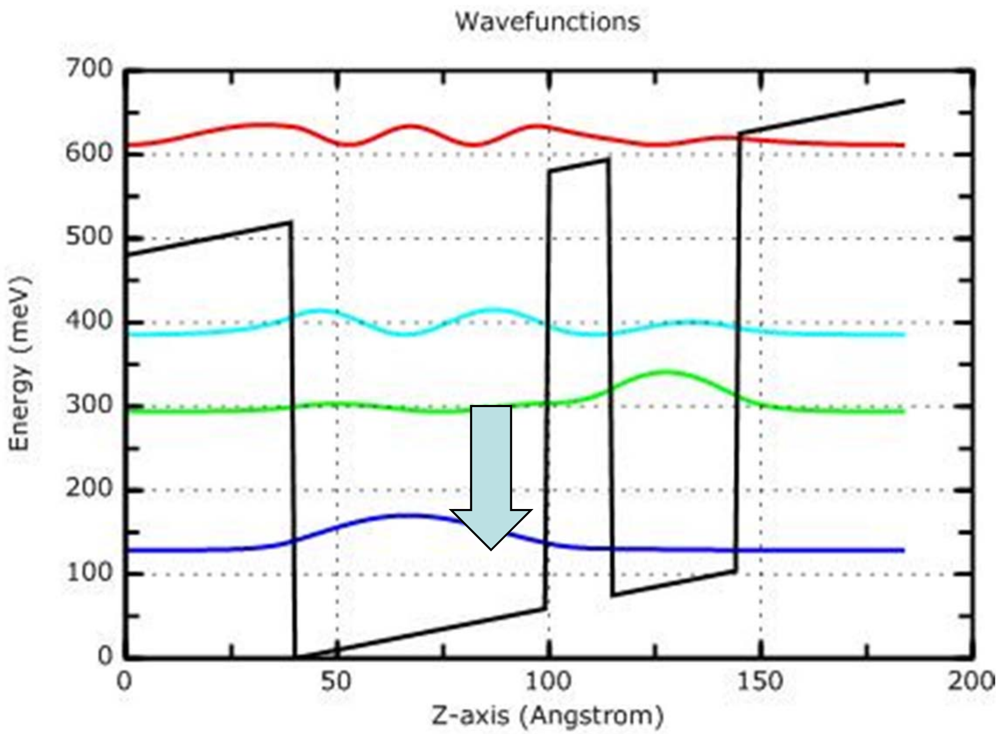
Quantum-confined electron gas has sharp, tunable resonances in "optics" (from terahertz to visible light)

How can we use it?

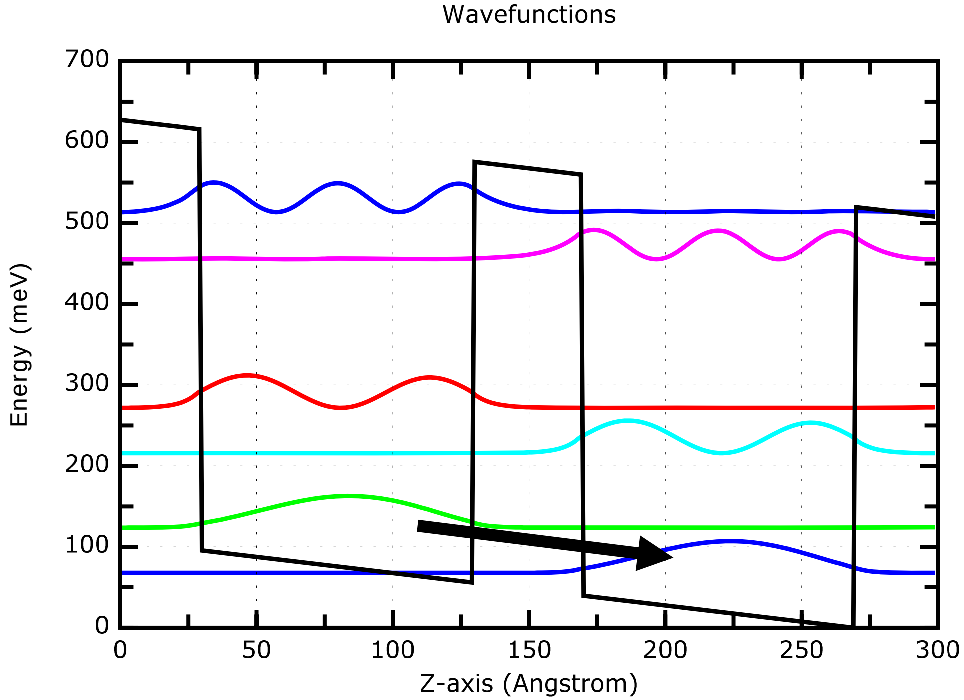
- Determine material parameters: effective masses, band offsets, g-factors, scattering rates
- Study new phenomena: Bloch oscillations, huge optical nonlinearities, BEC of excitons, entangled states, …
- Make new devices: lasers, detectors, transistors, memory, computers, etc.

How to get lasing between intersubband transitions?

Problem: ultrafast relaxation due to phonon emission

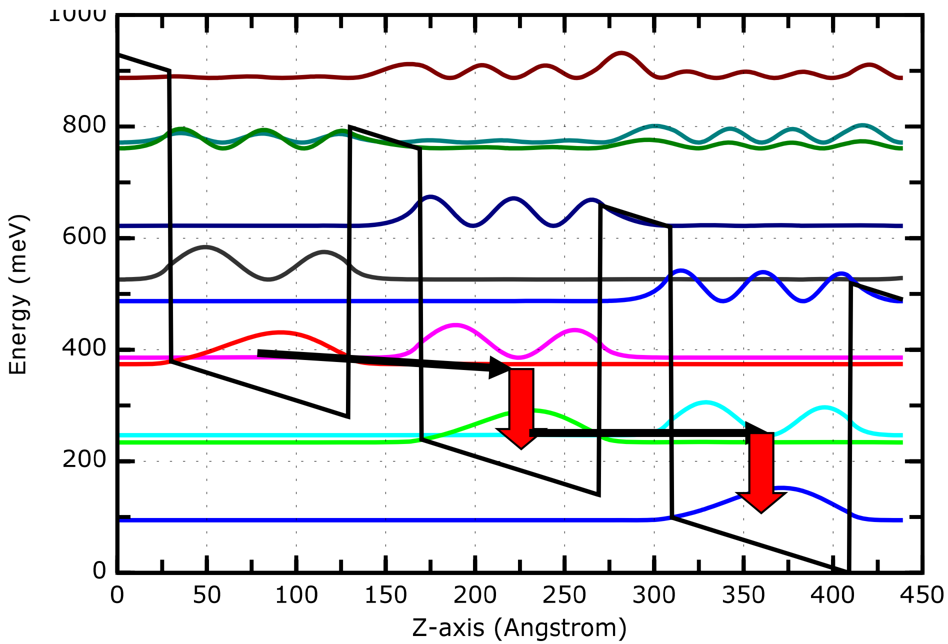


Superlattice laser: Kazarinov and Suris 1971



Electric field F = 40 kV/cm

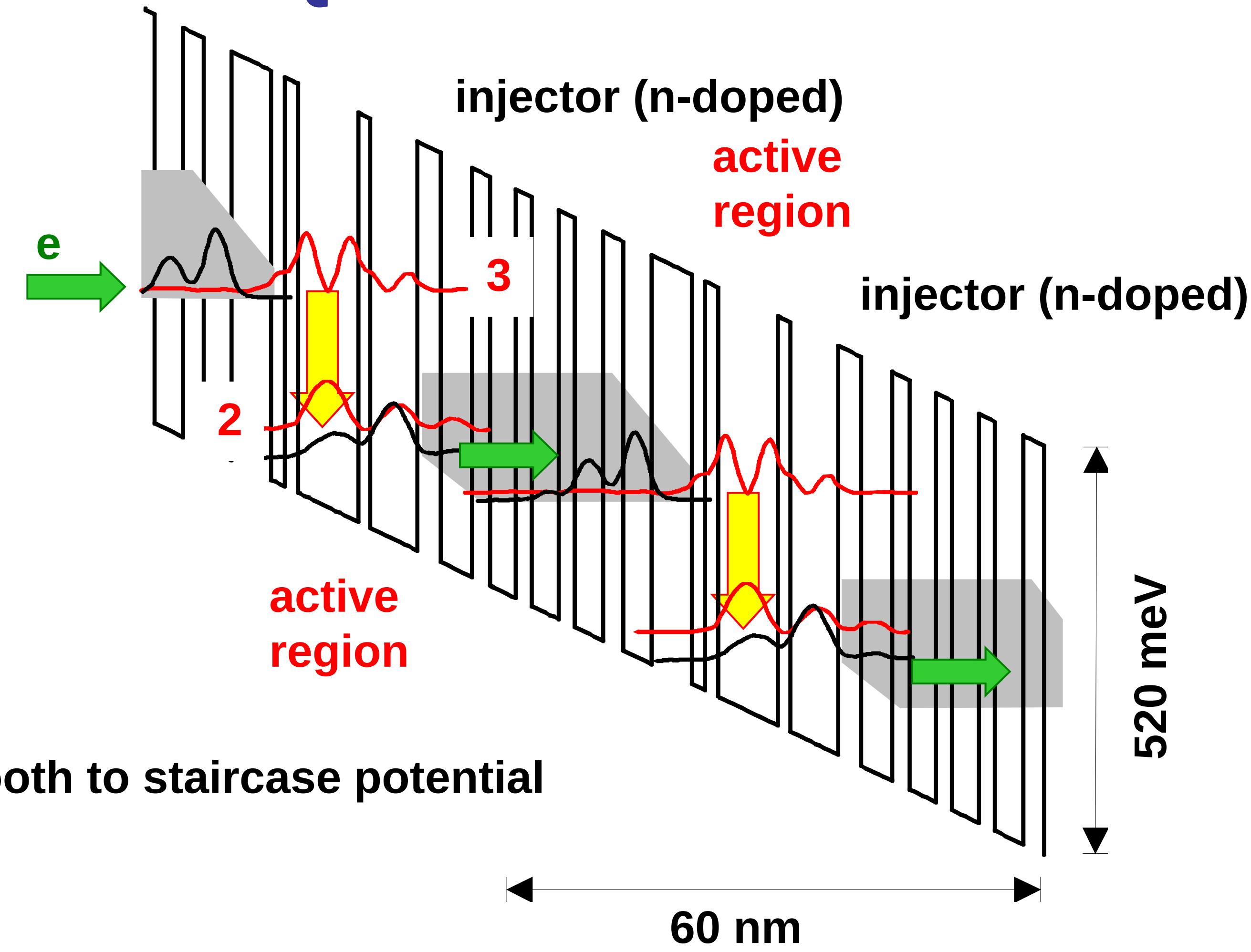
F = 100 kV/cm



Does not work due to domain formation and insufficient population inversion

J. Faist, F. Capasso, et al. *Science* 264, 553 (1994)

Quantum cascade lasers



From sawtooth to staircase potential

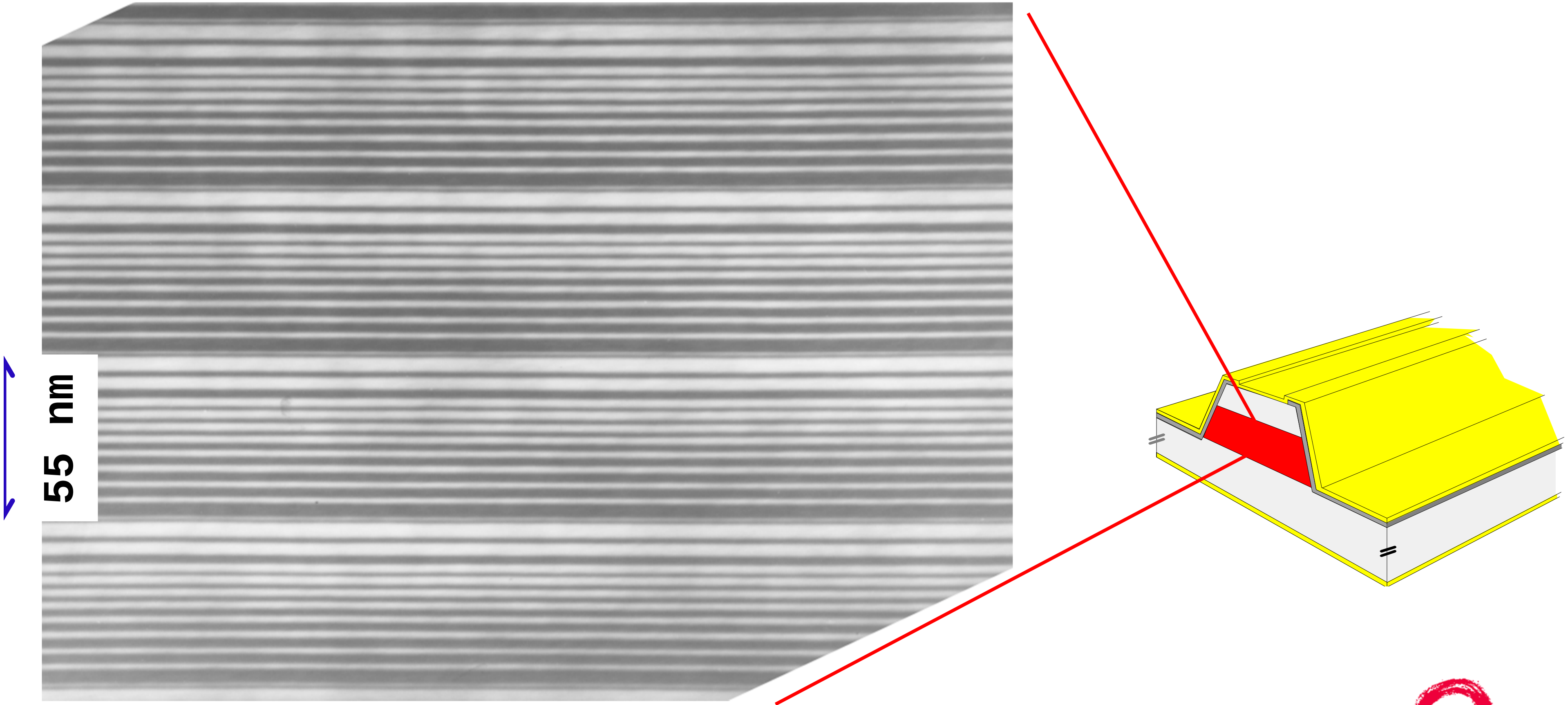
- **Control of lifetimes: phonons, tunneling; need t32 > t2**

$E_{21} = E_{phonon}$

$$k_{w,j} l_{w,j} + k_{b,j} l_{b,j} = \pi$$

- **Cascading: high power when t_stim approaches T_1**

Vertically stack 20-30 stages; sandwich them into the waveguide supporting a low-loss transverse EM mode



Mid-infrared (λ ~ 4-10 μm) Quantum cascade lasers are

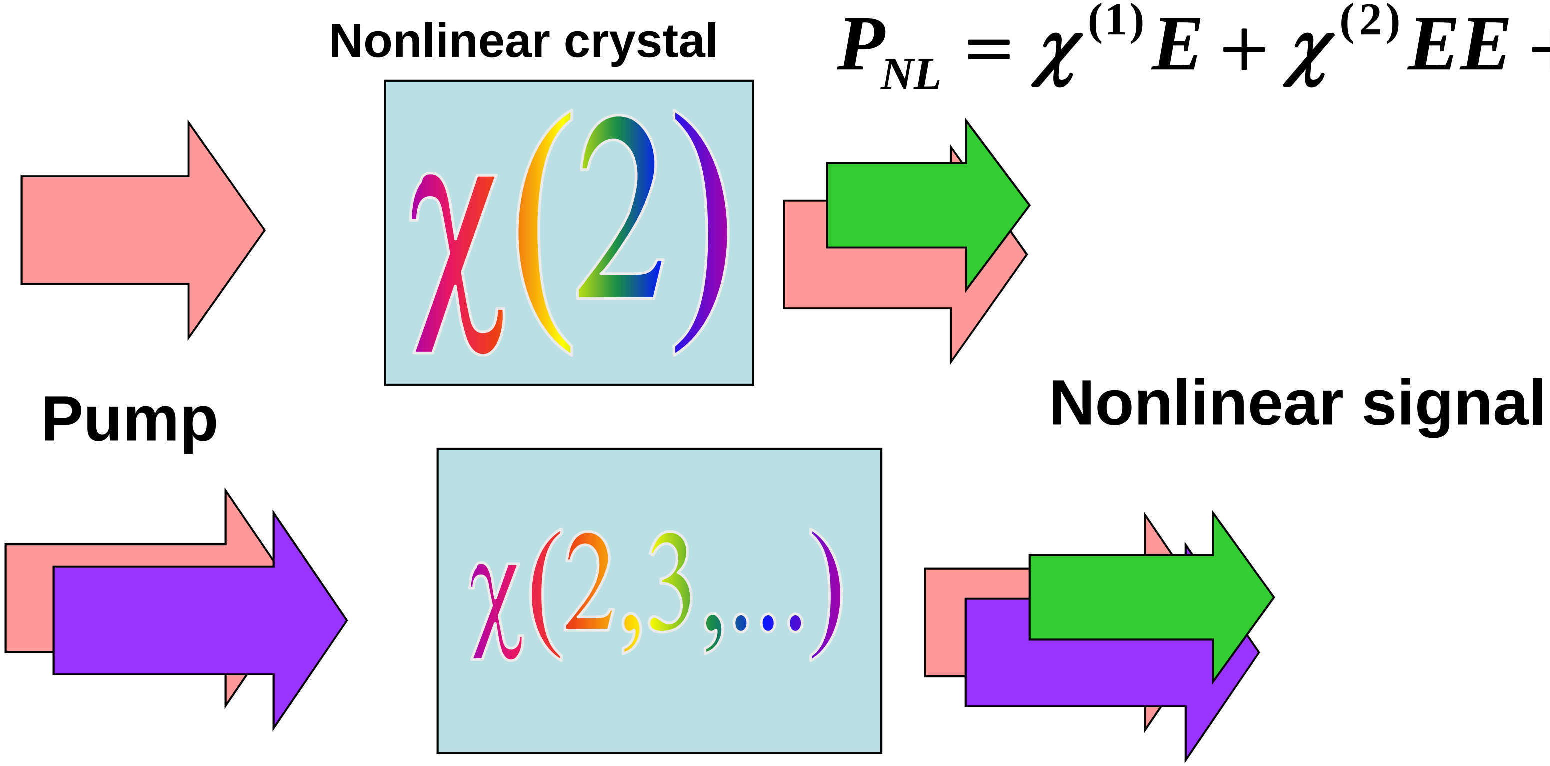
- Extremely powerful (P_{max} > 20 W)
- Operate at high temperatures T_{max} ~ 100 C
- Reliable, stable, etc.

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

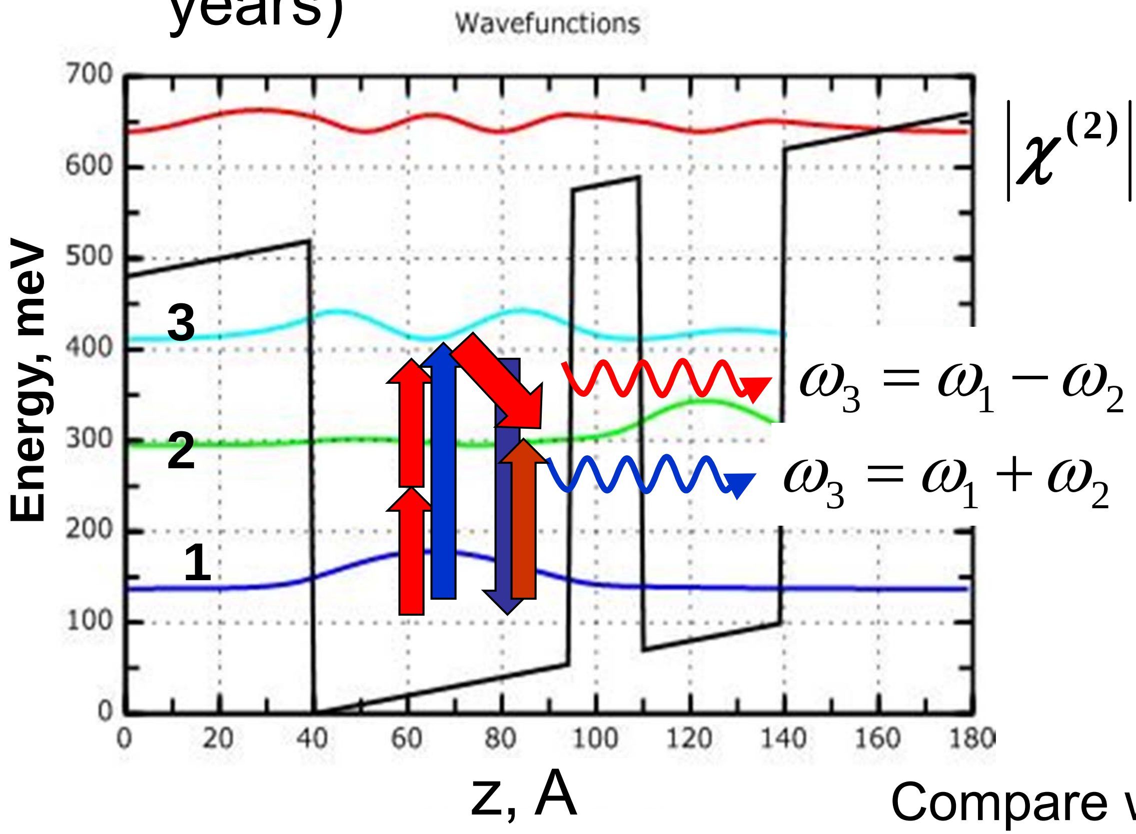
$$P_{NL} = \chi^{(1)}E + \chi^{(2)}EE + \ldots$$



- **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)



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

Δ_{ij} – detunings

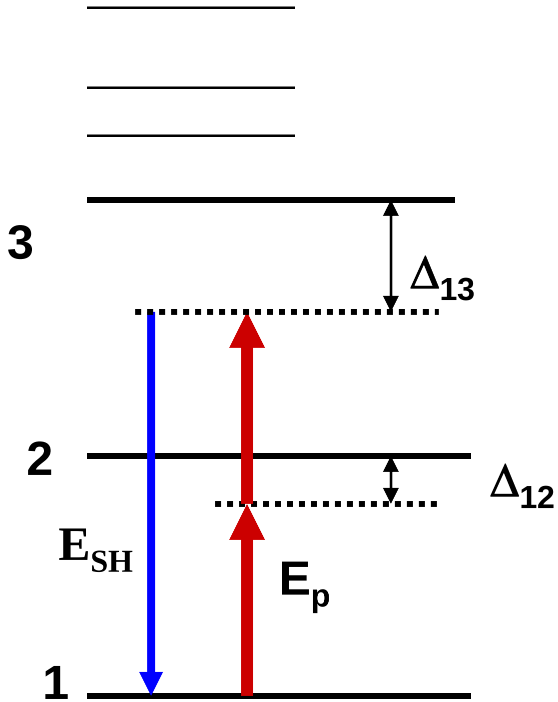
γ_{ij} – linewidths

d_{ij} – dipole moments

$$\left|\chi^{(2)}\right| \sim 10^4 - 10^6 \text{ pm/V}$$

Compare with 1-10 pm/V for bulk crystals

However, these advantages are usually inaccessible …



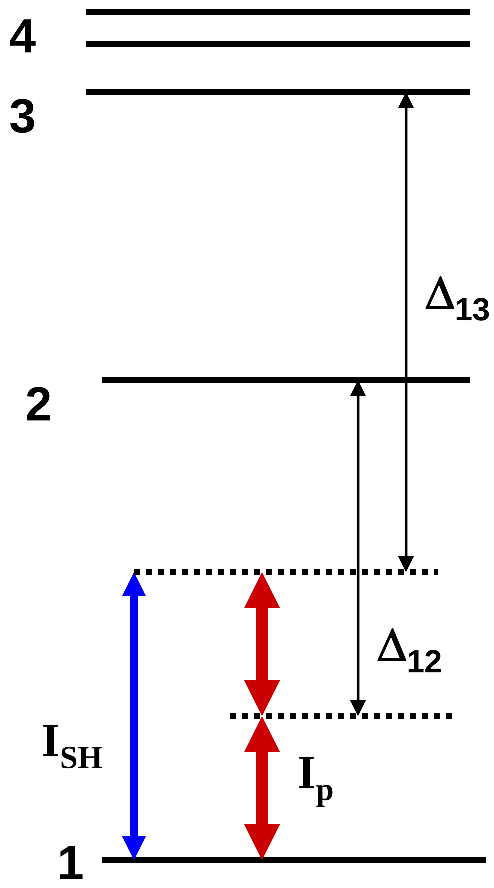
$$P_{NL} = \chi^{(1)}E + \chi^{(2)}EE + \ldots$$

Double resonance:

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

Resonance in absorption for both pump and the nonlinear signal:

$$\mathrm{Im}\left[\chi^{(1)}_{\mathrm{pump}}\right] \sim \frac{N_e d_{12}^2 \gamma_{12}}{\hbar(\gamma_{12}^2 + \Delta_{12}^2)}$$



Conventional nonlinear optics

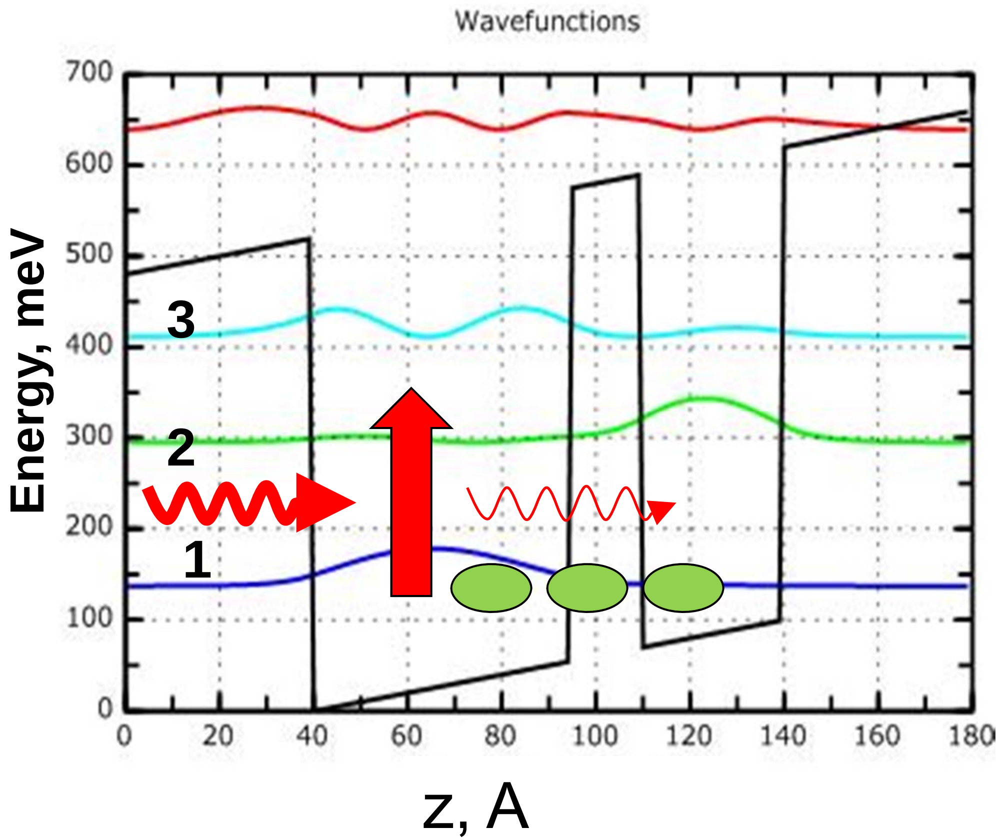
All detunings are large: $\Delta \sim \omega >> \gamma$;

All frequencies are in the transparency region of the NLO crystals

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

A way to get around resonant absorpti

Resonant optical nonlinearity is accompanied by resonant absorption

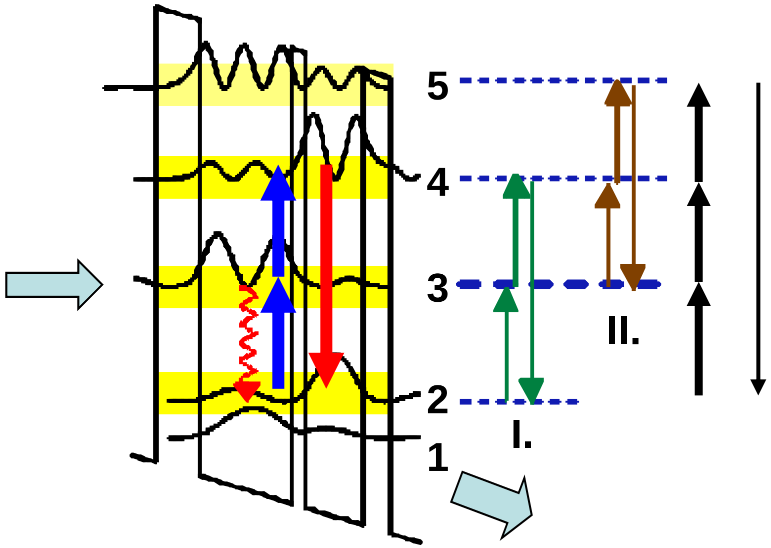


$$\left|\chi^{(2)}\right| \sim \frac{N_e d_{12} d_{13} d_{23}}{\hbar^2(\gamma_{12}^2 + \Delta_{12}^2)(\gamma_{13}^2 + \Delta_{13}^2)}$$

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:**

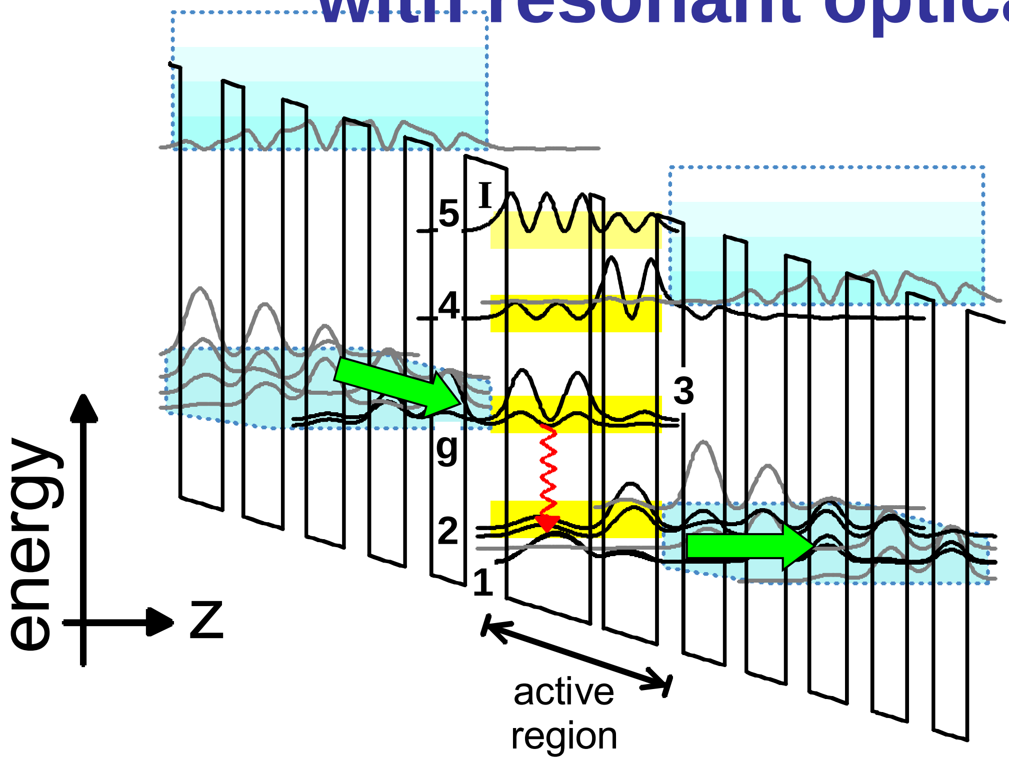
Laser field serves as a coherent optical pump for the nonlinear process

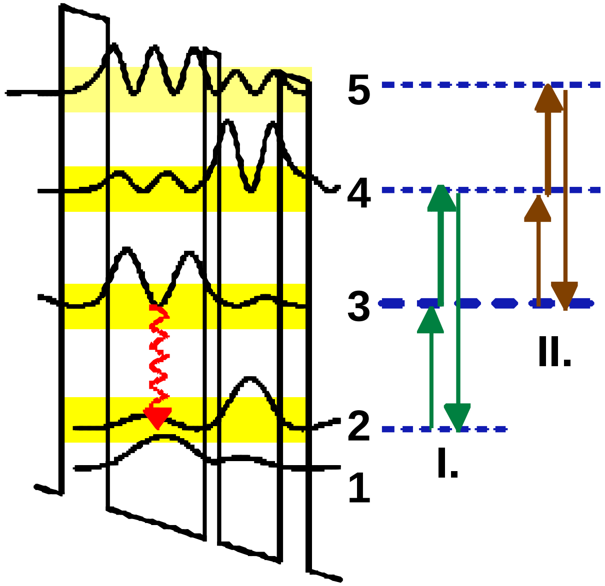
One can approach resonance since resonant absorption is compensated by laser gain

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





Second harmonic generation

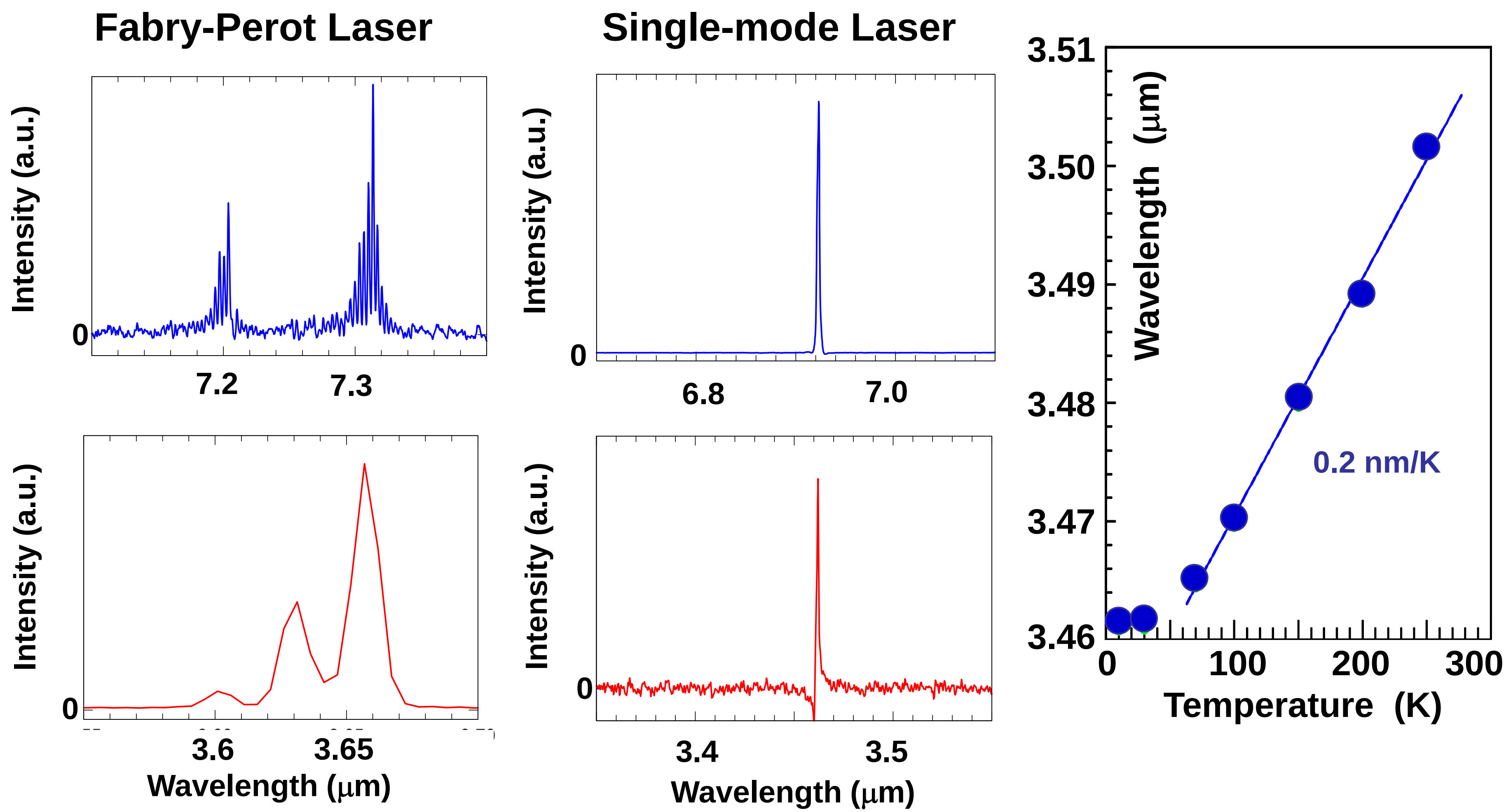
- Maximizing the product of dipoles $d_{23}d_{34}d_{24}$
- Quantum interference between cascades I and II

Milliwatt power in SHG:
O. Malis et al. 2004

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

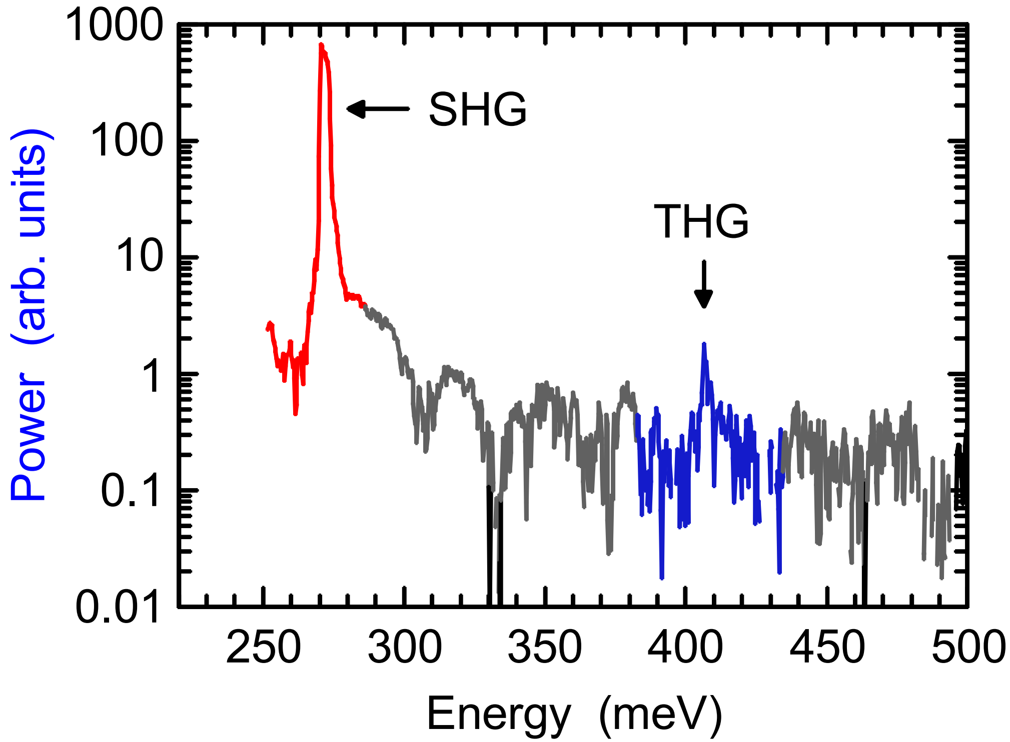
This is NOT sequential photon absorption/reemission!

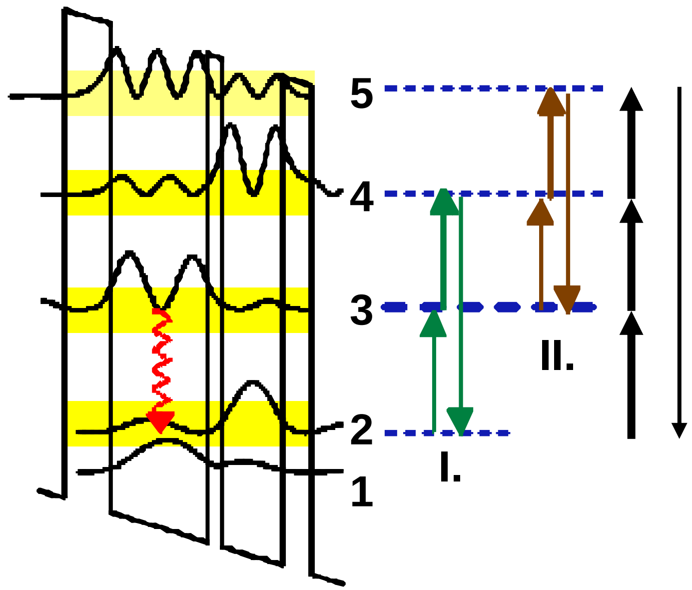
Single-mode and tunable SH emission



APL 84, 2751 (2004)

Third Harmonic Generation





Triple resonance

$\omega \rightarrow 2\omega \rightarrow 3\omega$

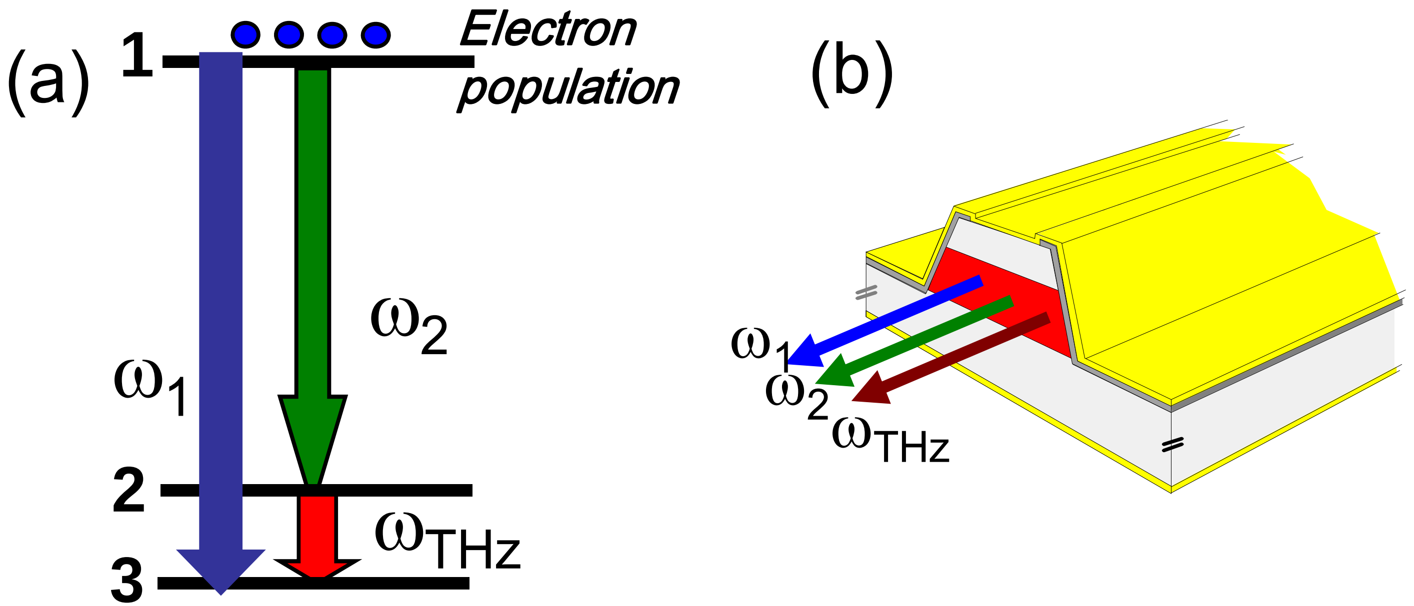
11.1 μm → 5.5 μm → 3.7 μm

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

$\chi^{(3)} \sim 10^{-7}$ **esu**

T. Mosely, A. Belyanin, C. Gmachl, Optics Express 12, 2972 (2004)

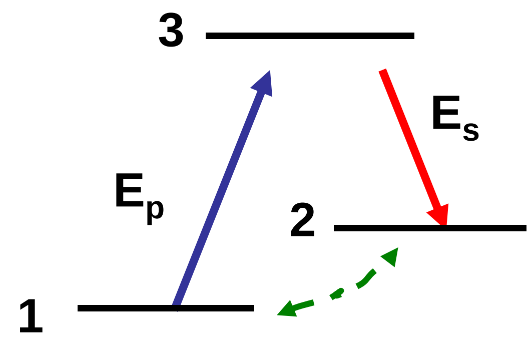
Difference frequency generation in two-wavelength QCLs



- 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



$$\frac{\partial E_s}{\partial t} \propto E_p \rho_{12} \propto |E_p|^2 E_s$$

- 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

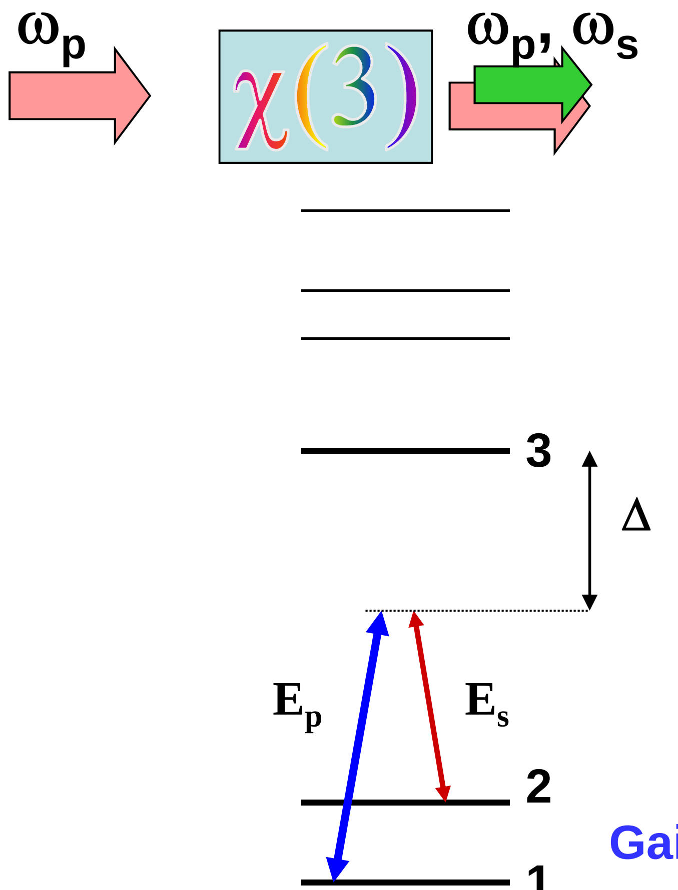
$$\frac{\partial \rho_{12}}{\partial t} \neq 0$$

Density matrix:

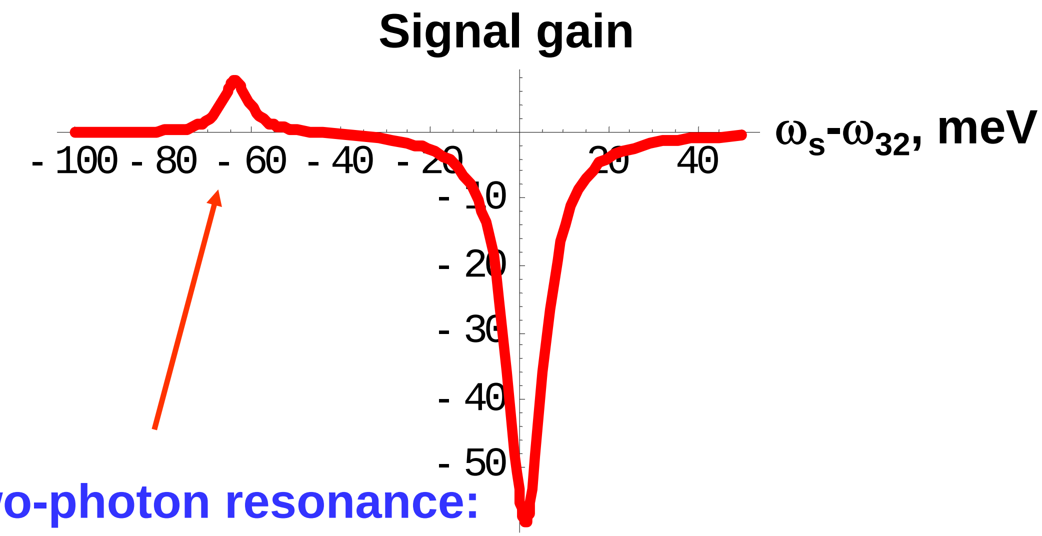
$$\begin{pmatrix} \rho_{11} & \rho_{12} & \rho_{13} \\ \rho_{21} & \rho_{22} & \rho_{23} \\ \rho_{31} & \rho_{32} & \rho_{33} \end{pmatrix}$$

"Quantum coherence"

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



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



Gain at two-photon resonance:

$\omega_p - \omega_s = \omega_{21}$

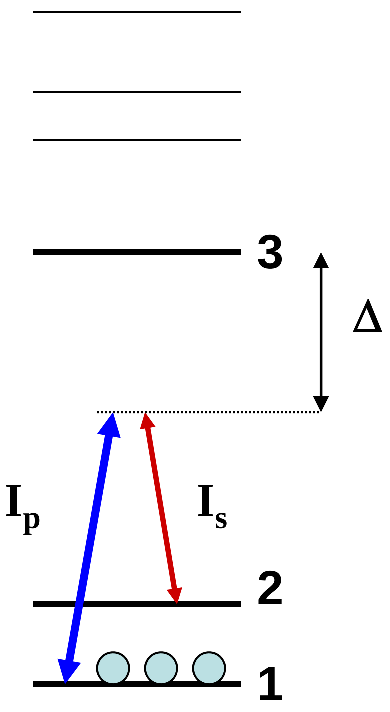
Stimulated Raman scattering

Raman inversion

Raman gain $\propto \dfrac{I_p(N_1 - N_2)}{\Delta^2 \gamma_{21}}$

$\Delta \sim \omega_s$

Raman decoherence rate



Raman coherence $\rho_{21} << 1$

(Except experiments by Sokolov, Harris et al.)

Propagation of coupled Raman and pump fields

Manley-Rowe relation: $\dfrac{I_p}{\omega_p} + \dfrac{I_s}{\omega_s} = \text{const}$

Approaching resonance

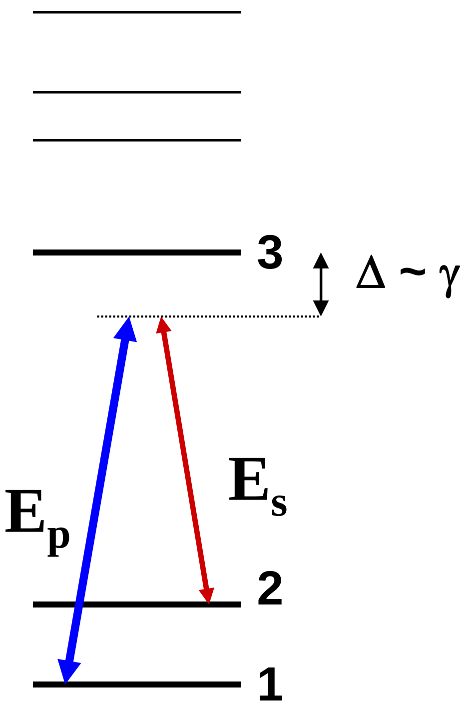
Both “good” and “bad” effects get enhanced

Real one-photon processes become important

Raman coherence ρ_{21} also increases

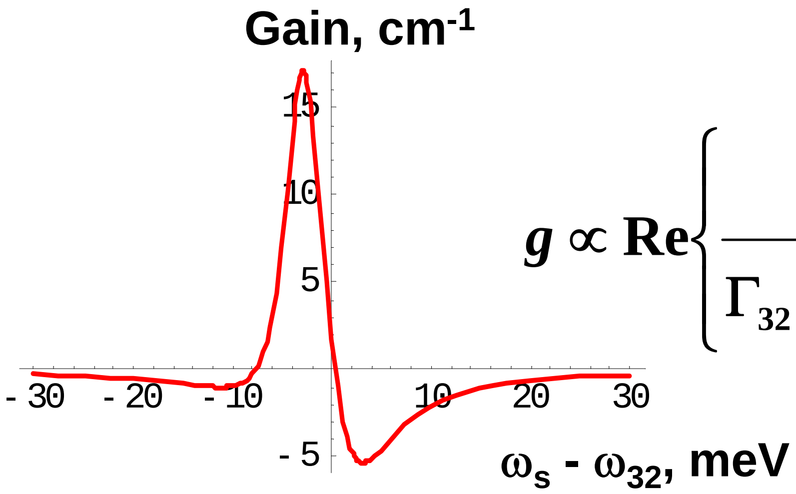
Raman gain increases strongly

Absorption is increased



$$\frac{\partial E_s}{\partial t} \propto E_p \rho_{12} \propto \left|E_p\right|^2 E_s$$

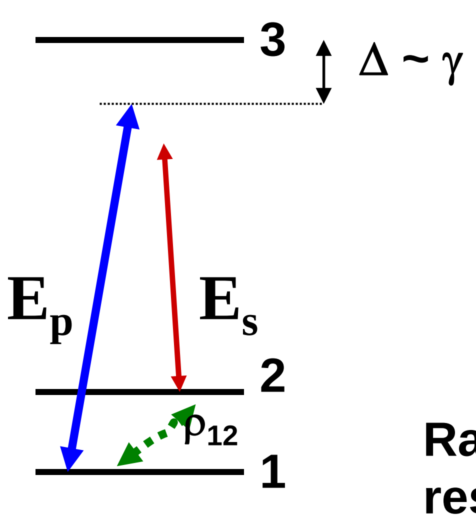
Stokes gain at arbitrary detuning



One-photon absorption

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

"Two-photon" gain



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

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

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

$$\gamma_{ij} \sim 5 - 10\ \mathrm{meV}$$

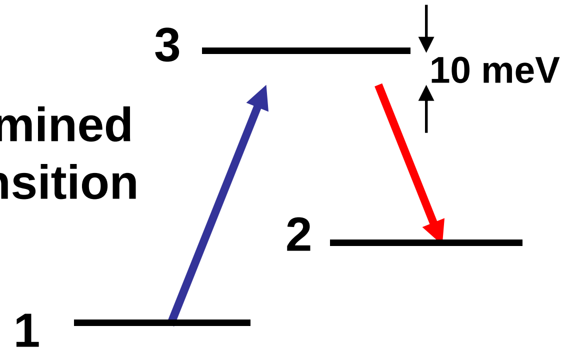
Rabi frequency $\Omega_p = \dfrac{d_{31} E_p}{\hbar}$

$$\Omega_s = \frac{d_{32} E_s}{\hbar}$$

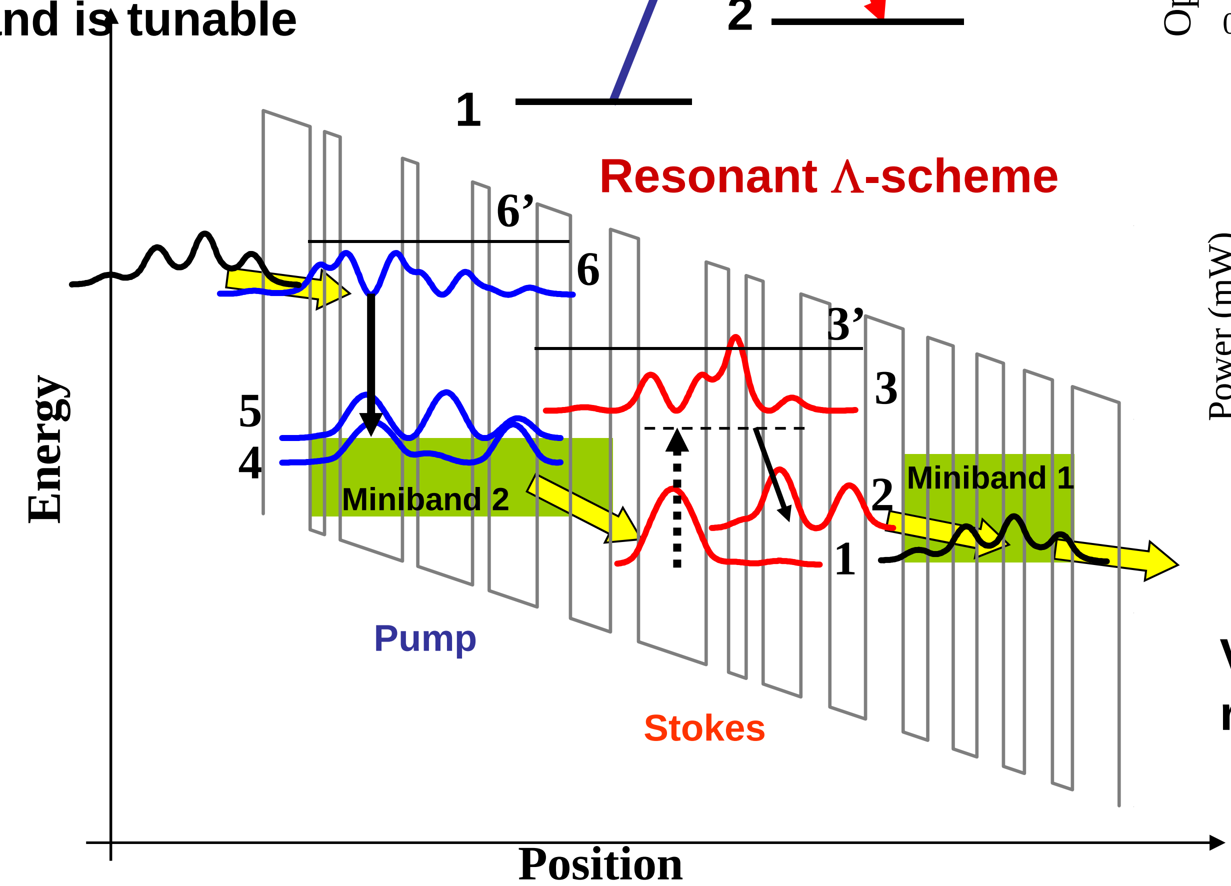
Raman gain is enhanced, but resonant absorption of the pump limits the interaction length

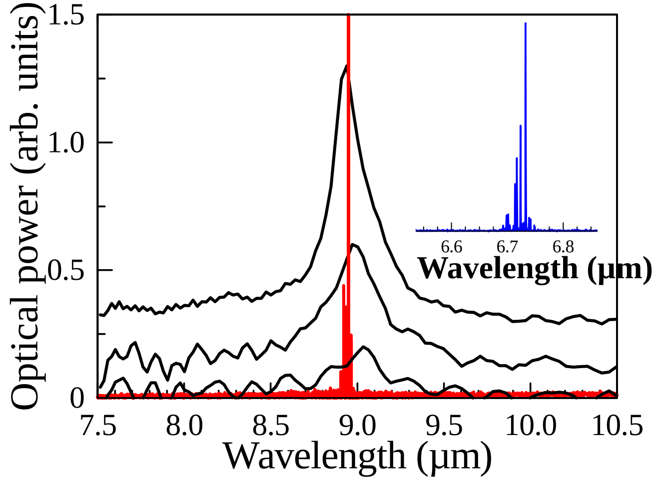
Mid-IR Raman injection laser

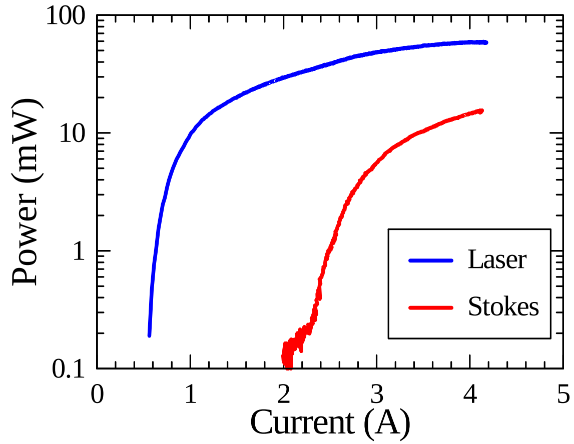
Raman shift is determined by intersubband transition and is tunable



Resonant Λ-scheme







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

40 mW Raman threshold
16 mW Stokes power

M. Troccoli, A. Belyanin, F. Capasso, Nature 433, 845 (2005)