Short wavenlength Quantum Cascade Lasers: Physics, materials and applications

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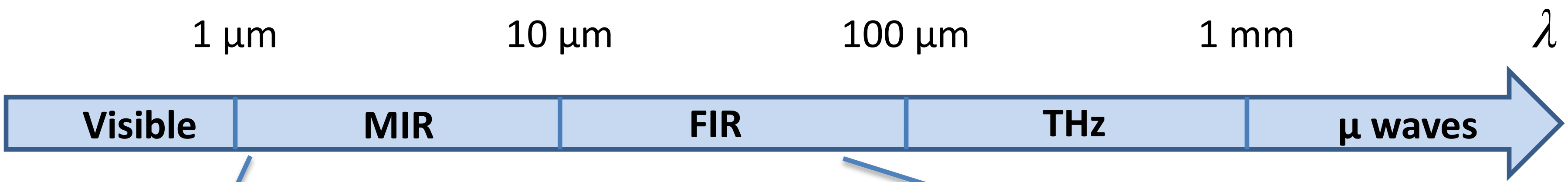
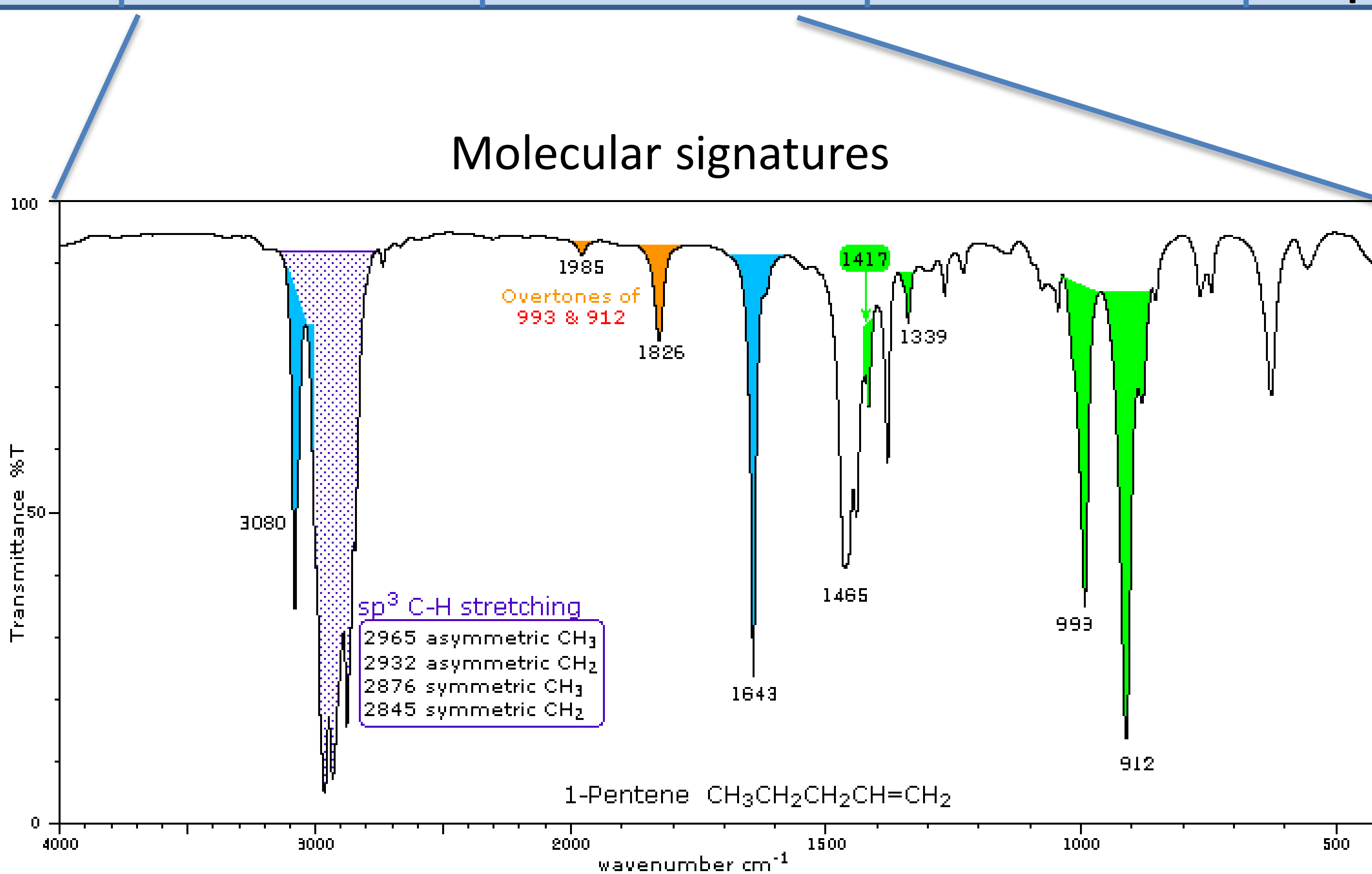
1 - MOTIVATIONS

Motivations: Molecular IR spectroscopy



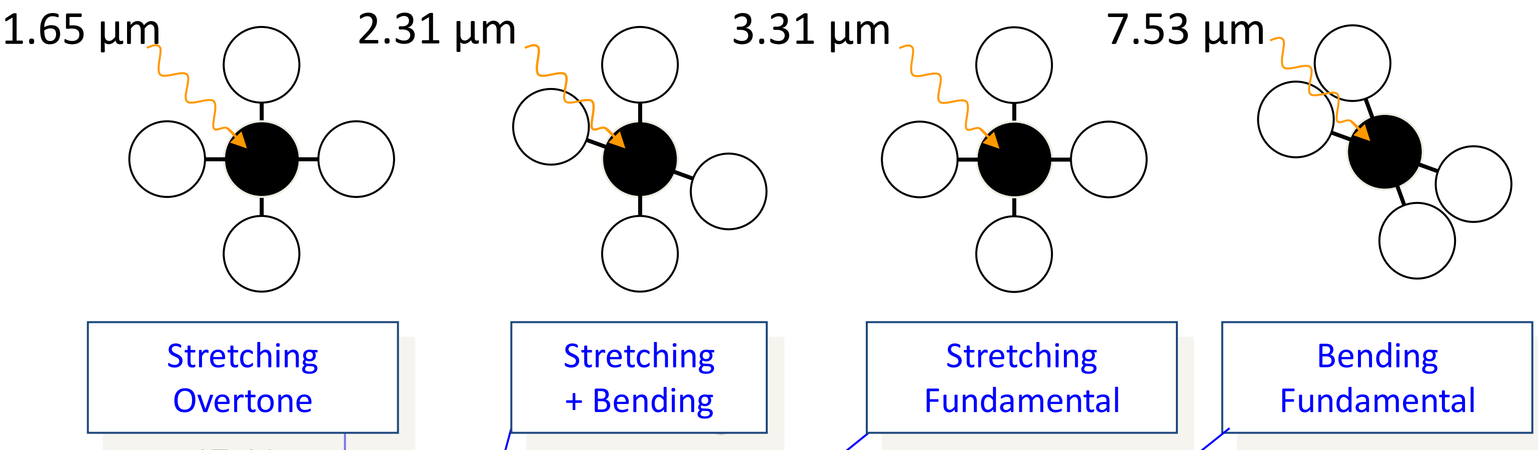
Molecular signatures

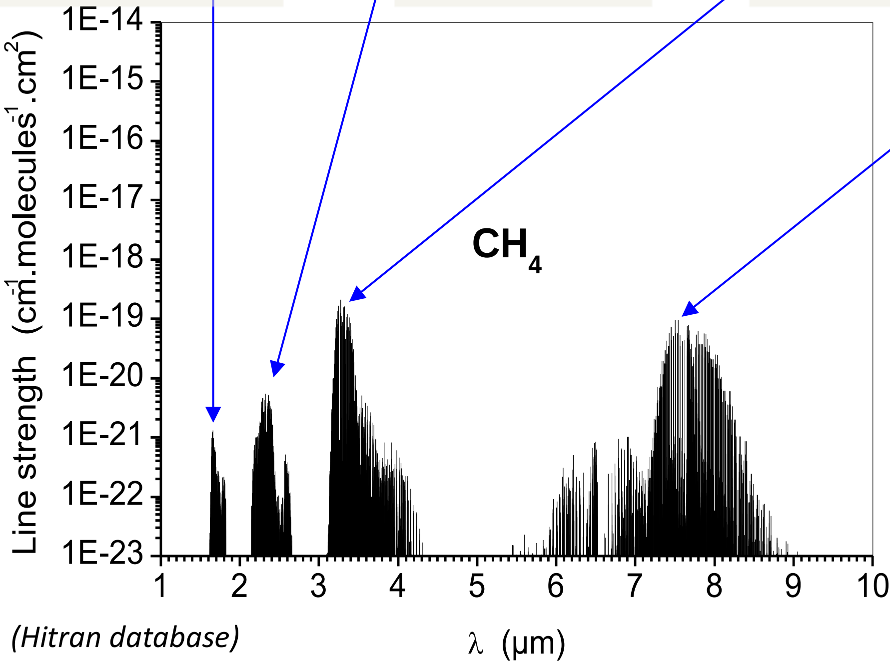


Applications in the whole IR spectrum

An important wavelength is λ=3.3 μm

3.3 µm : important wavelength for CH_4 and hydrocarbons





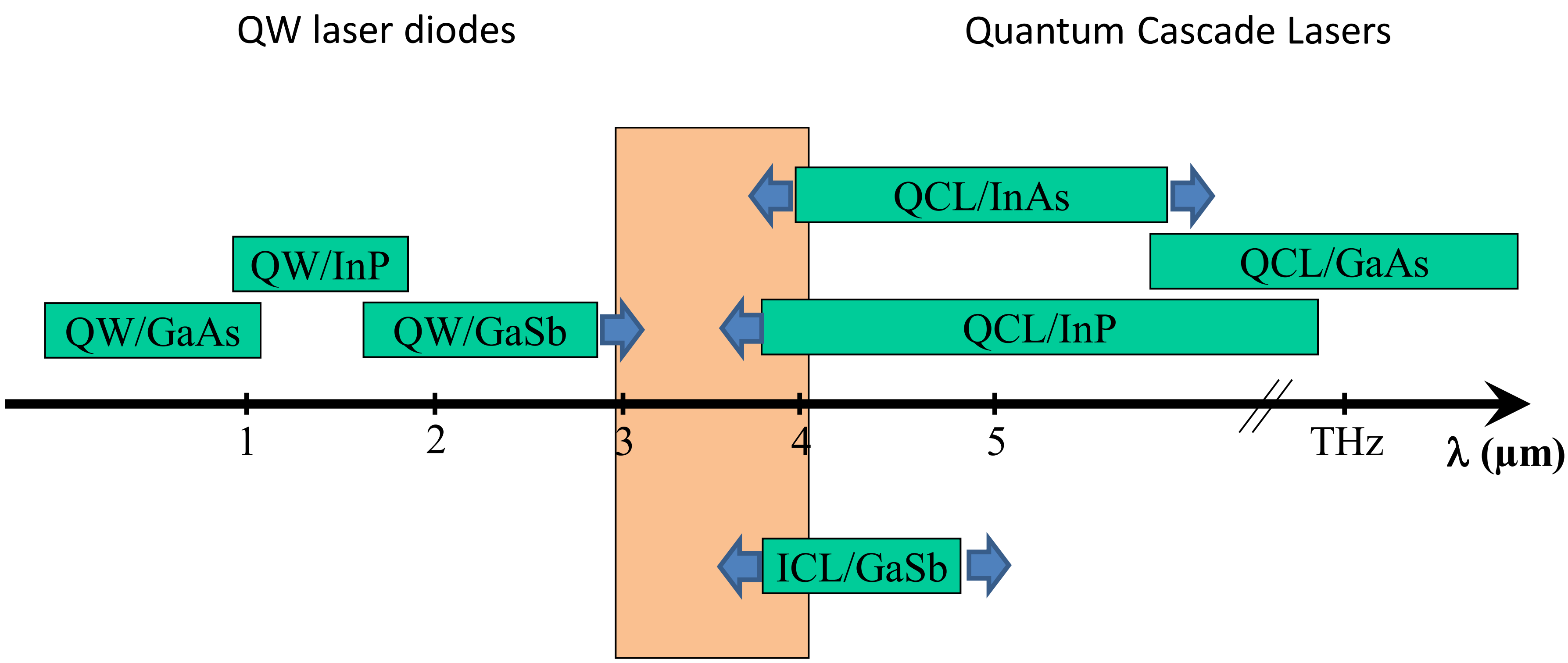
3.31 µm :

. Fundamental vibration

. Maximum of absorption

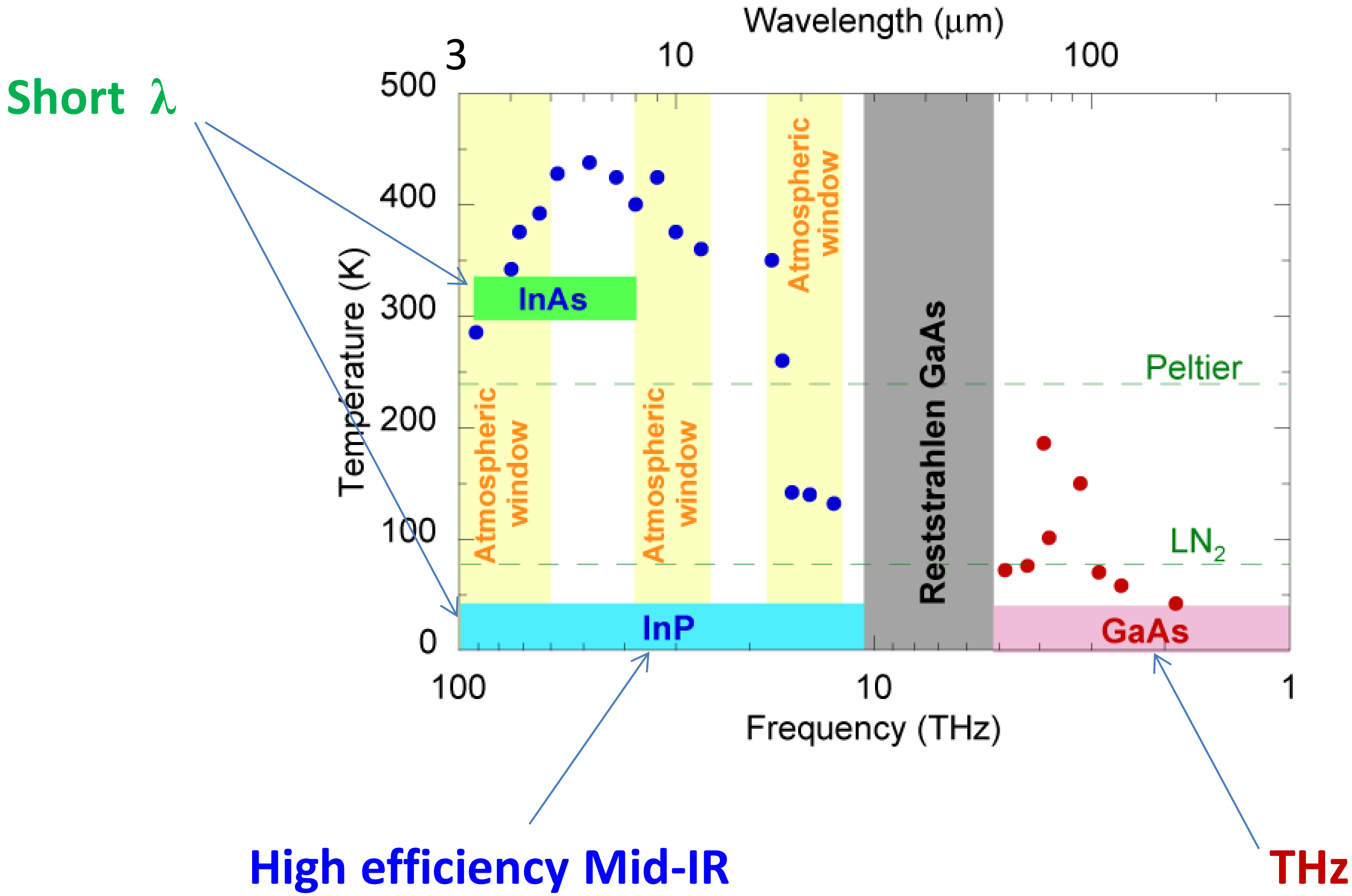
Applications

Semiconductor laser sources



(There was) a mid-IR gap in the 3-4 µm region

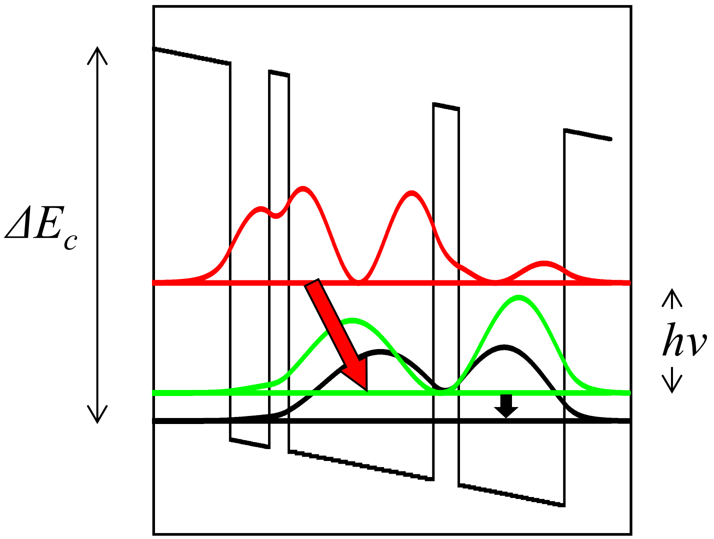
QCLs



A very large spectral coverage

Versatile technology

Energy scale



• QCL standard :
λ= 4– 12 µm ($h\nu$ = 100 – 300 meV)

• THz range :
λ= 60 – 150 µm ($h\nu$ = 8 – 20 meV)

• **Short wavelength :**
λ= 2.5 – 4 µm ($h\nu$ = 300 – 500 meV)

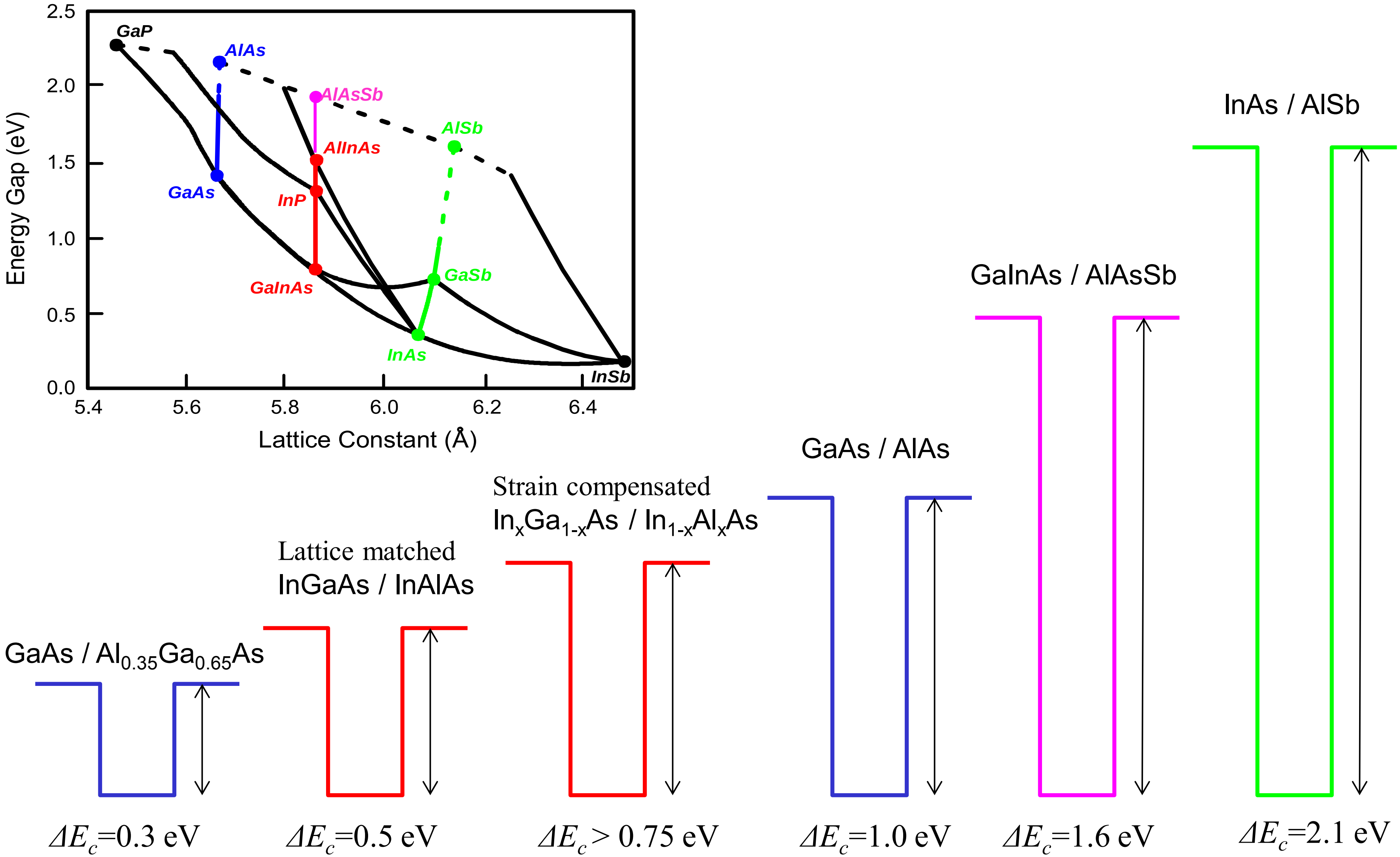
➢ The photon energy depends only on QWs design

➢ It is limited towards short wavelength by the finite depth (ΔE_c) of the QWs

A typical value of $\Delta E_c = 2.h\nu$ is required
New materials are needed

2 - MATERIALS

Large band offset materials



Effective QW depth

GaAs-based materials

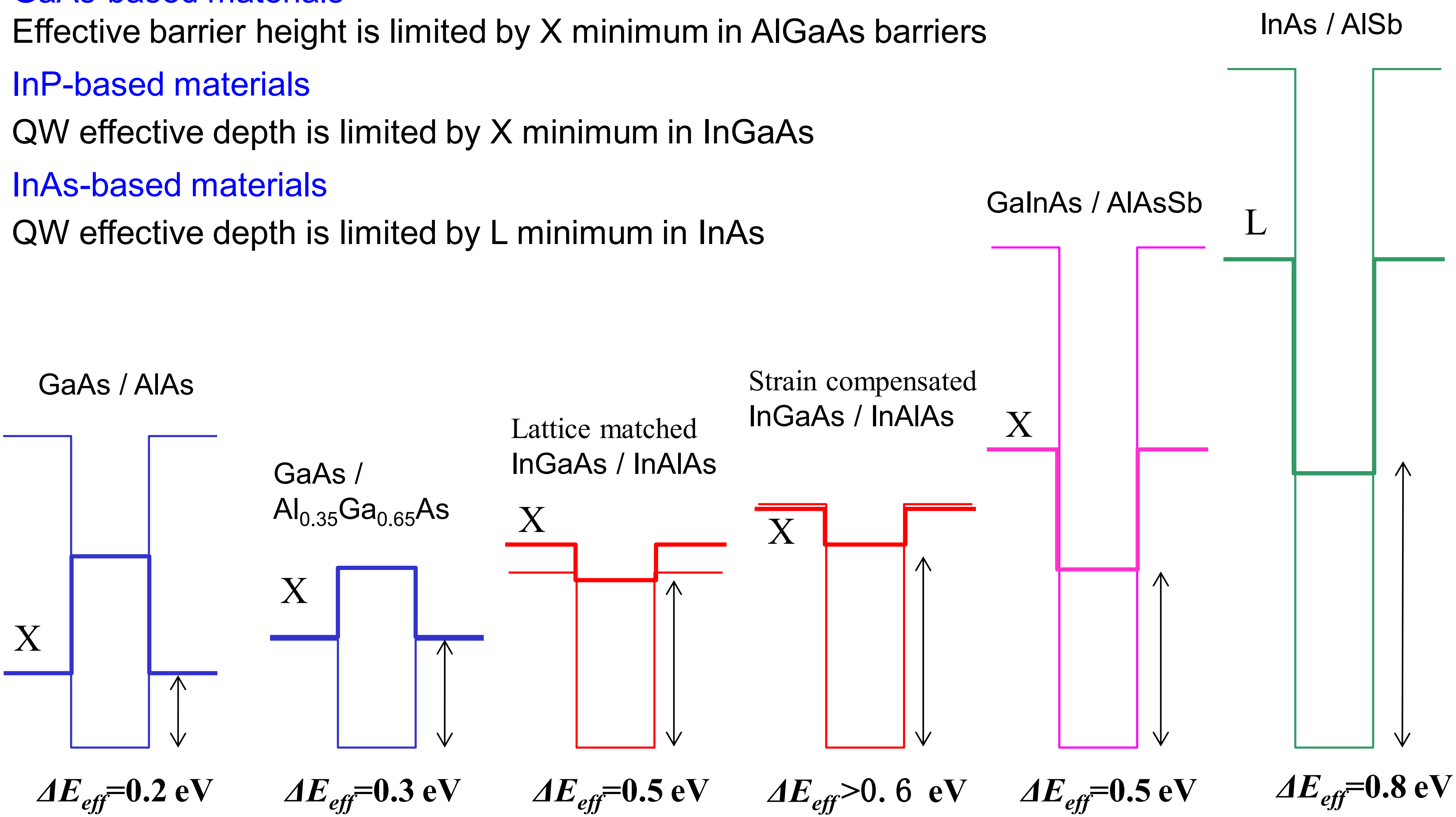
Effective barrier height is limited by X minimum in AlGaAs barriers

InP-based materials

QW effective depth is limited by X minimum in InGaAs

InAs-based materials

QW effective depth is limited by L minimum in InAs



Three material systems have been successfully developed for short λ QCLs

on InP substrate

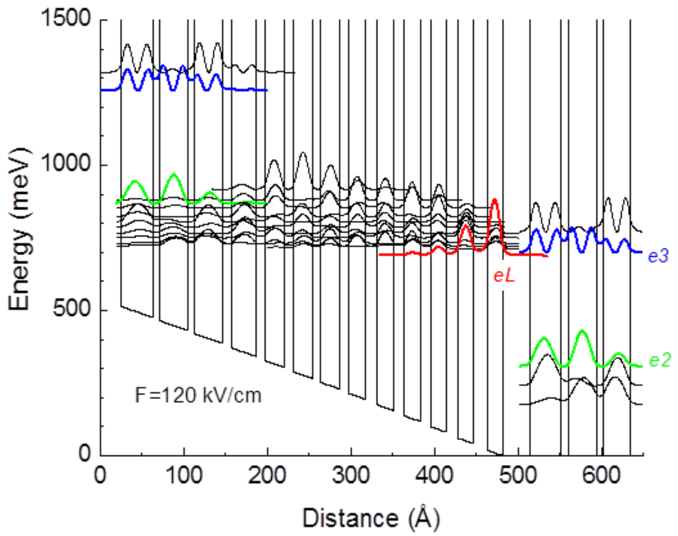
Strain-compensated
InGaAs / Al(In)As
ΔE_c= 0.7 – 1.4 eV

Lattice matched
InGaAs / AlAsSb
ΔE_c= 1.6 eV

on InAs substrate

Lattice matched
InAs / AlSb
ΔE_c= 2.1 eV

Main difficulties: growth control and active region design



3 – PHYSICS ISSUES

Intersubband optical gain

$$G = \Gamma \frac{e^2 \hbar f_{32}}{\varepsilon_0 n c m_0 2\gamma L_p} (n_3 - n_2)$$

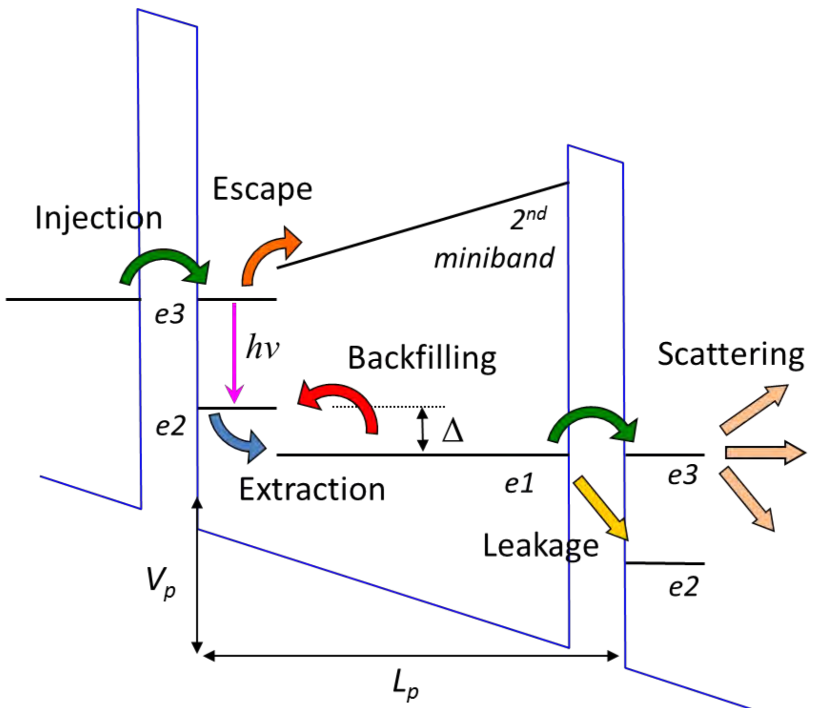
Oscillator strength (f_{32})

Transition linewidth (2γ)

Population inversion ($n_3 - n_2$)

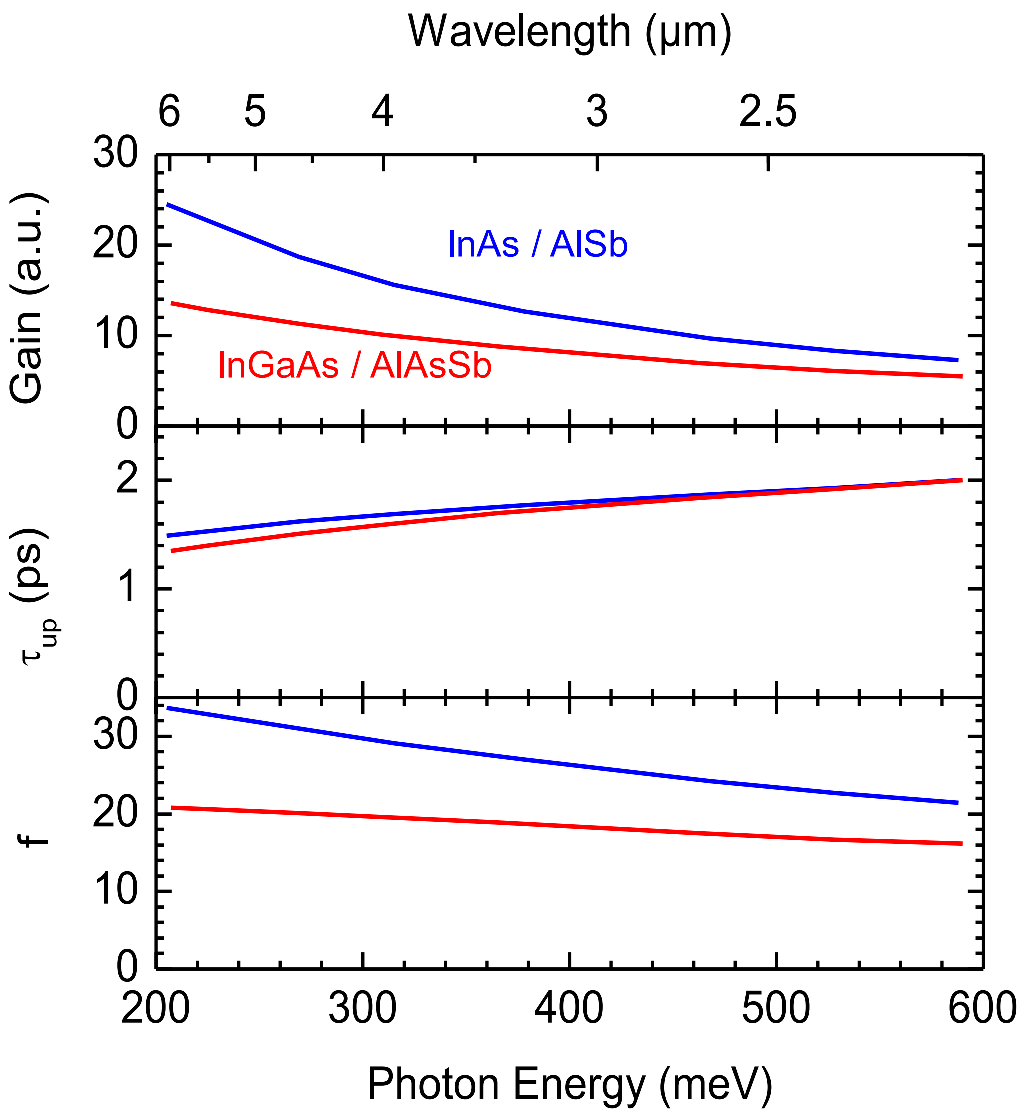
$$n_3 - n_2 = \frac{\eta_i}{e} \tau_3 \left(1 - \frac{\tau_2}{\tau_{32}}\right) \cdot J$$

Simplified 3 levels model



a more realistic picture

A first simple evaluation

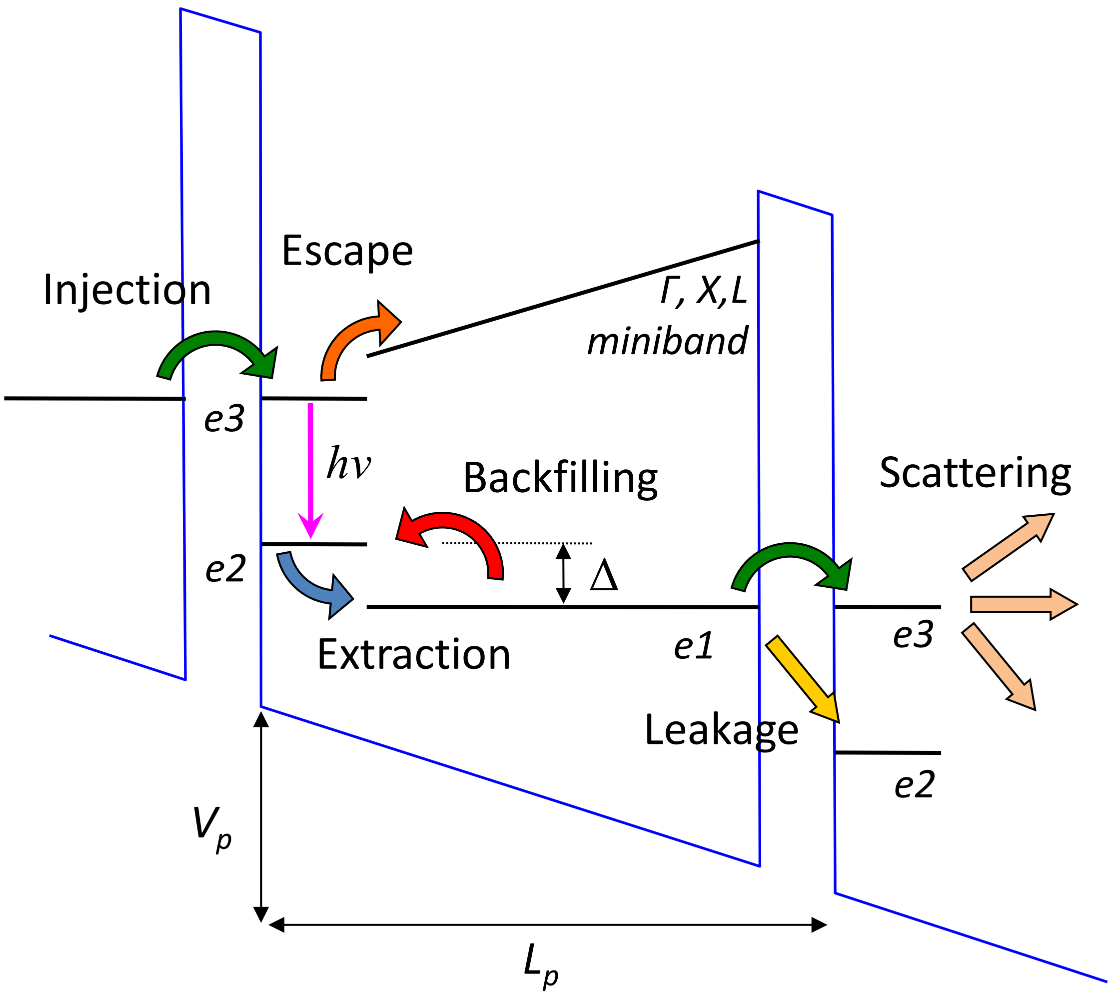


$$g \propto \frac{f \cdot \tau_{up}}{2\gamma}$$

no fundamental limitation at short λ

but gain is reduced

Specificities of short λ QCLs



large ΔE_c

high energy states: e3 ~ ΔE_{eff}

non parabolicity

narrow wells, thin barriers

large V_p , high field

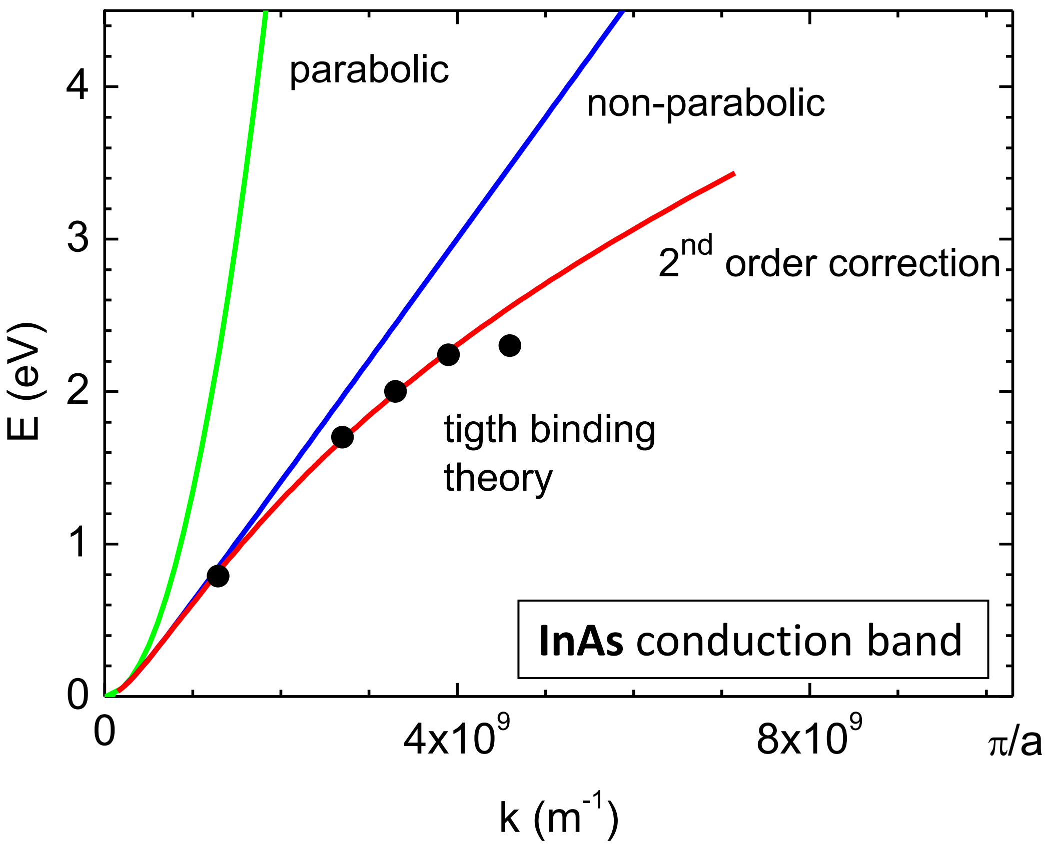
interface roughness

subband broadening >> $\hbar\Omega_{tun}$

lateral valleys

$h\nu$ >> kT
Δ >> kT
$h\nu$ >> $\hbar\omega_{LO}$

Band nonparabolicity



$$m(E) = m^* \cdot \left[1 + \frac{(E - E_c)}{E_{eff}} \right]$$

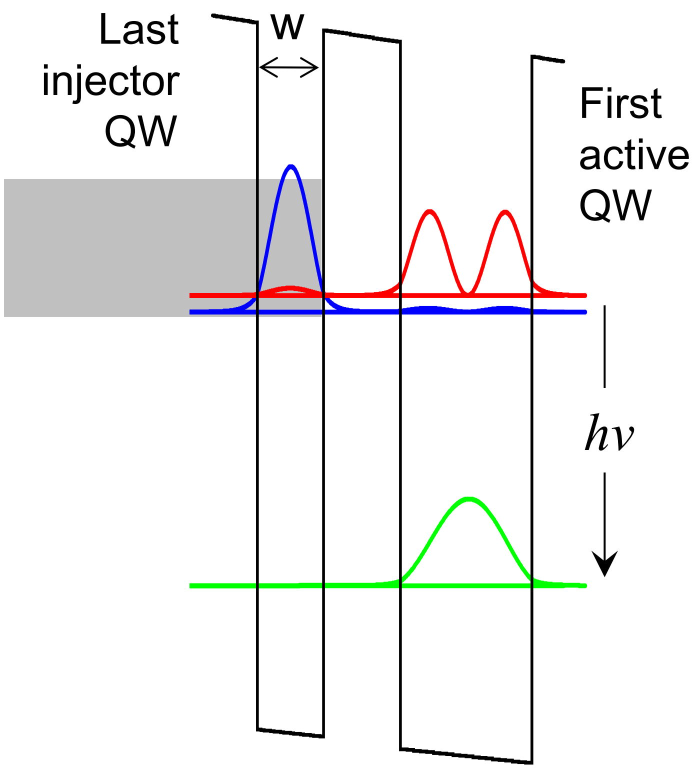
$$E_{eff} \approx E_g \approx 0.4\, eV$$

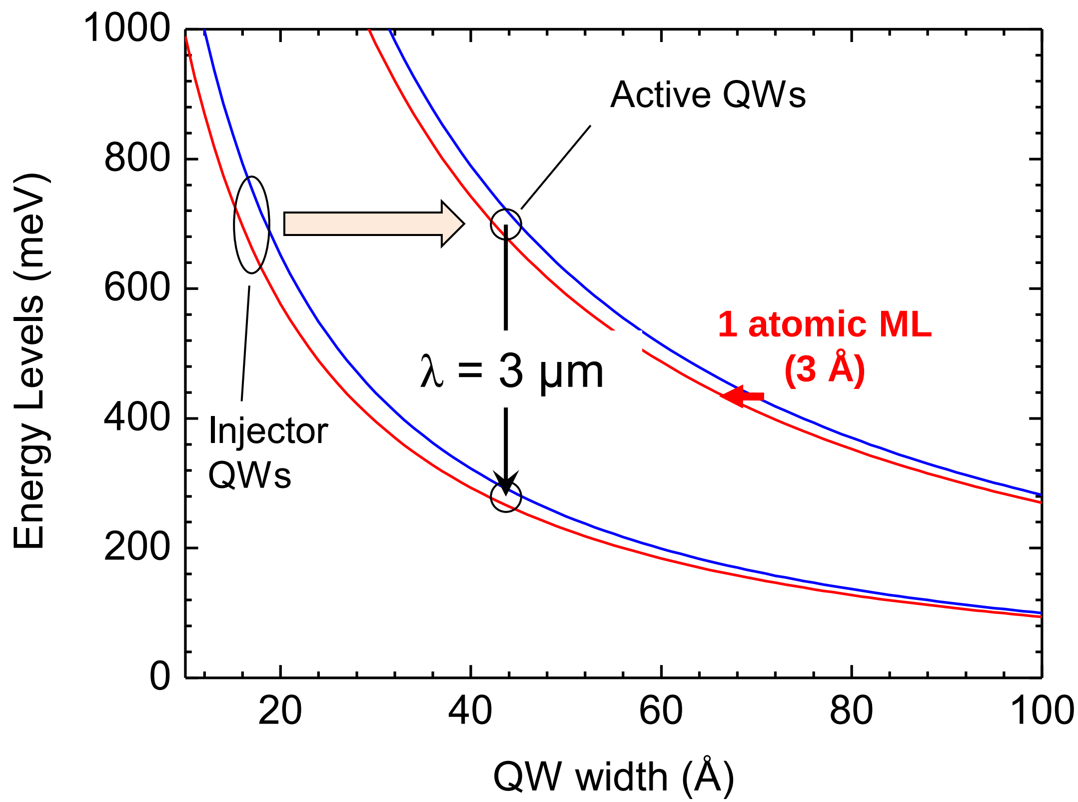
high confinement energies

strong non-parabolicity effects

second order terms may be required

Narrow quantum wells



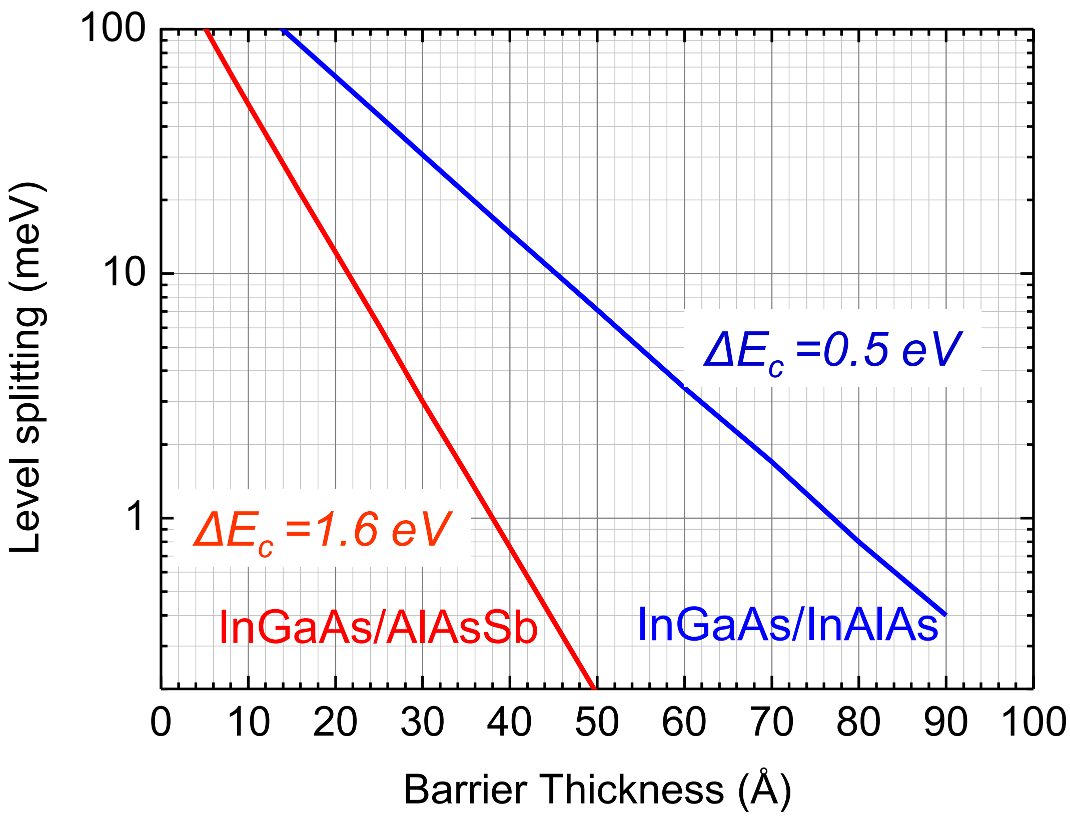


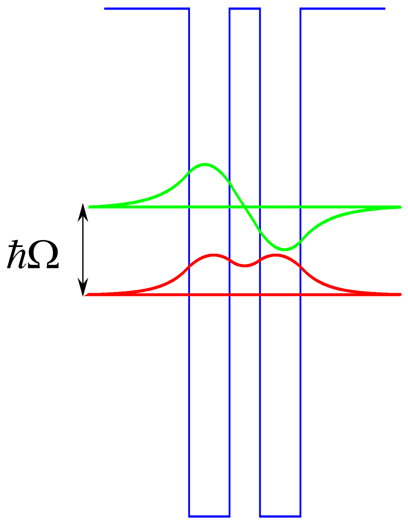
Particularly in injector

subbands broadening

importance of interface roughness

Thin barriers

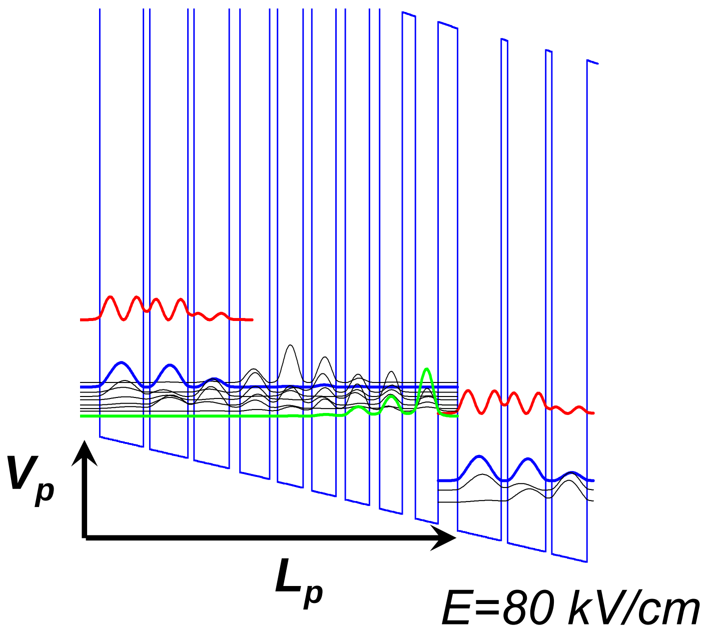




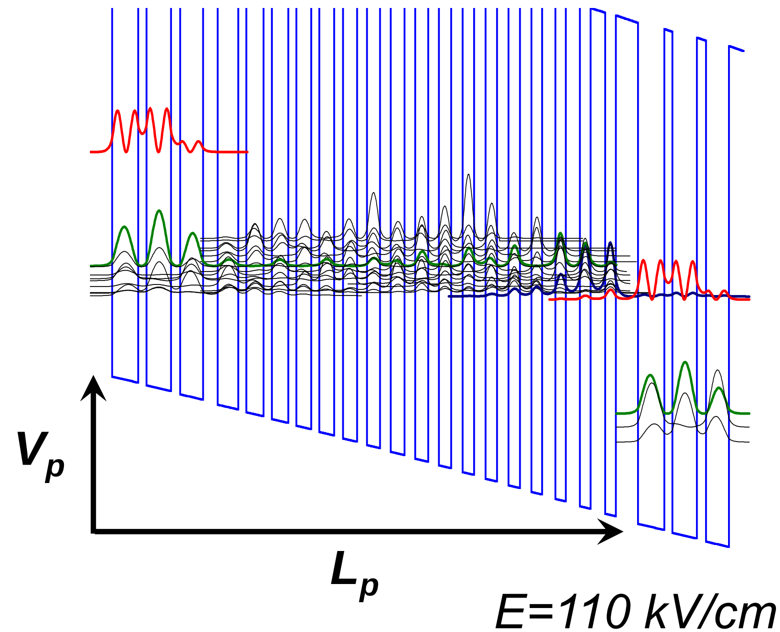
because of larger ΔE_c

More QWs per period

λ=4.5 μm – 6 injector QWs



λ=2.5 μm – 17 injector QWs



Higher V_p and thinner QWs

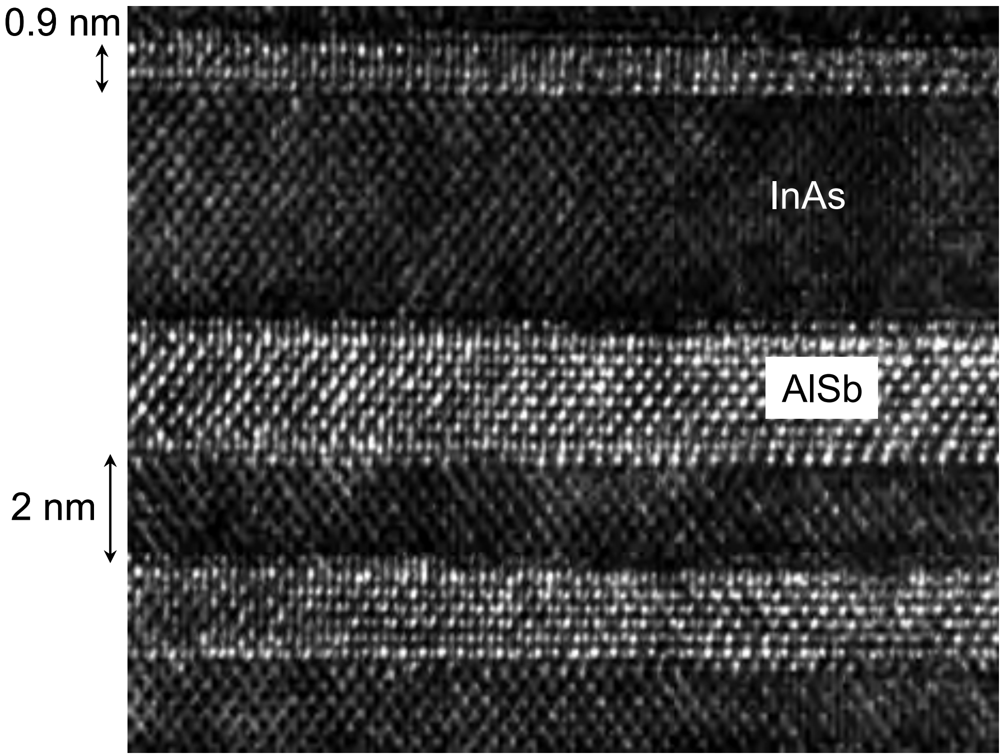
➔ Large Number of QW :

Increases the growth time and difficulty

Best solution : increase the electric field as much as possible

also: smaller tuning rate for injection

Importance of interfaces



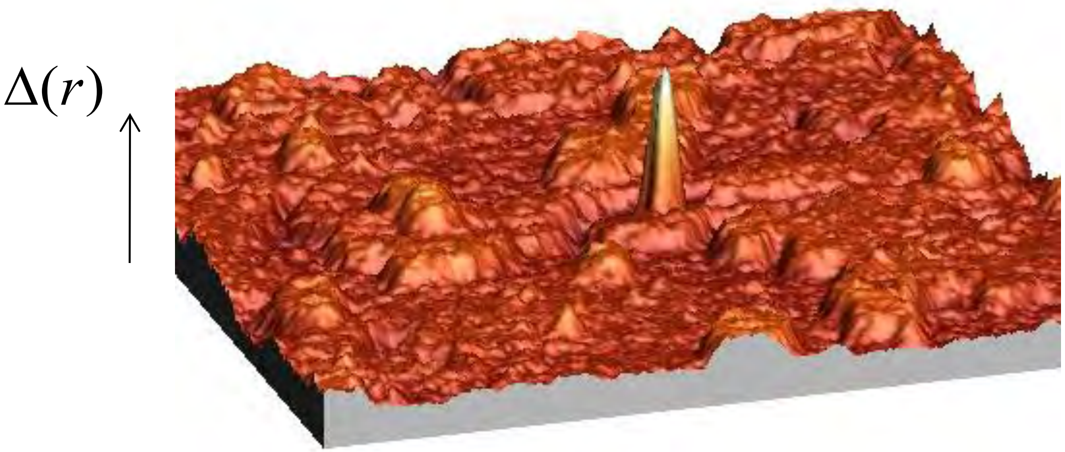
TEM image : Anne Ponchet, CEMES, Toulouse

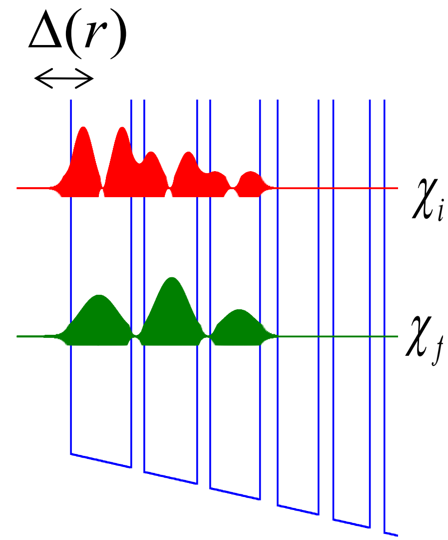
control of layer thicknesses
better than 1 ML (0.3 nm)

interface roughness

Interface roughness scattering

Image AFM, (1 µm x 1 µm x 1 nm),
M. Ramonda, Univ. Montpellier 2





$$PSD(q) = \frac{1}{S}\left|\int_S \exp(iqr)\Delta(r)\,dr\right|^2$$

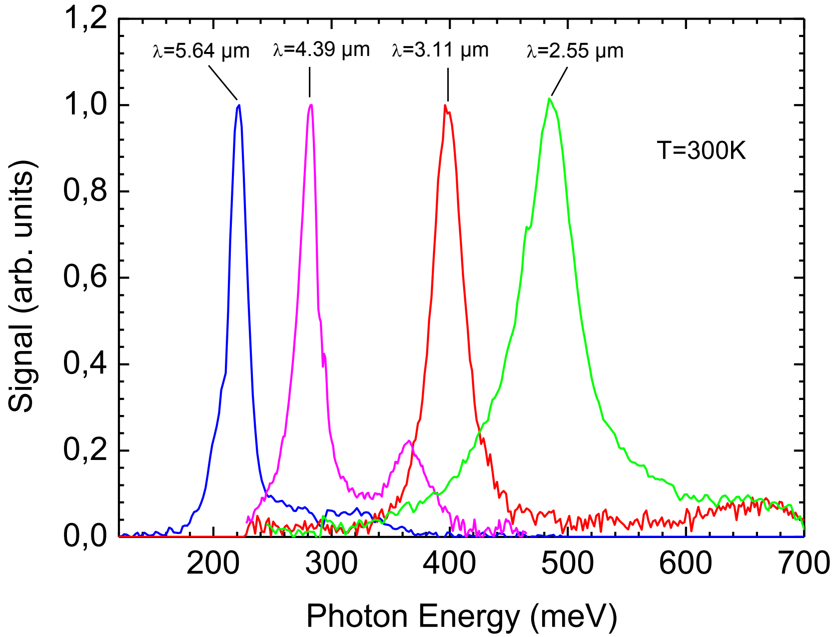
$$\frac{1}{\tau_{if}} = \frac{2m^*}{\hbar^3}\Delta E_c^2 \sum_n \left|\chi_i(z_n)\chi_f(z_n)\right|^2 PSD(q_{if})$$

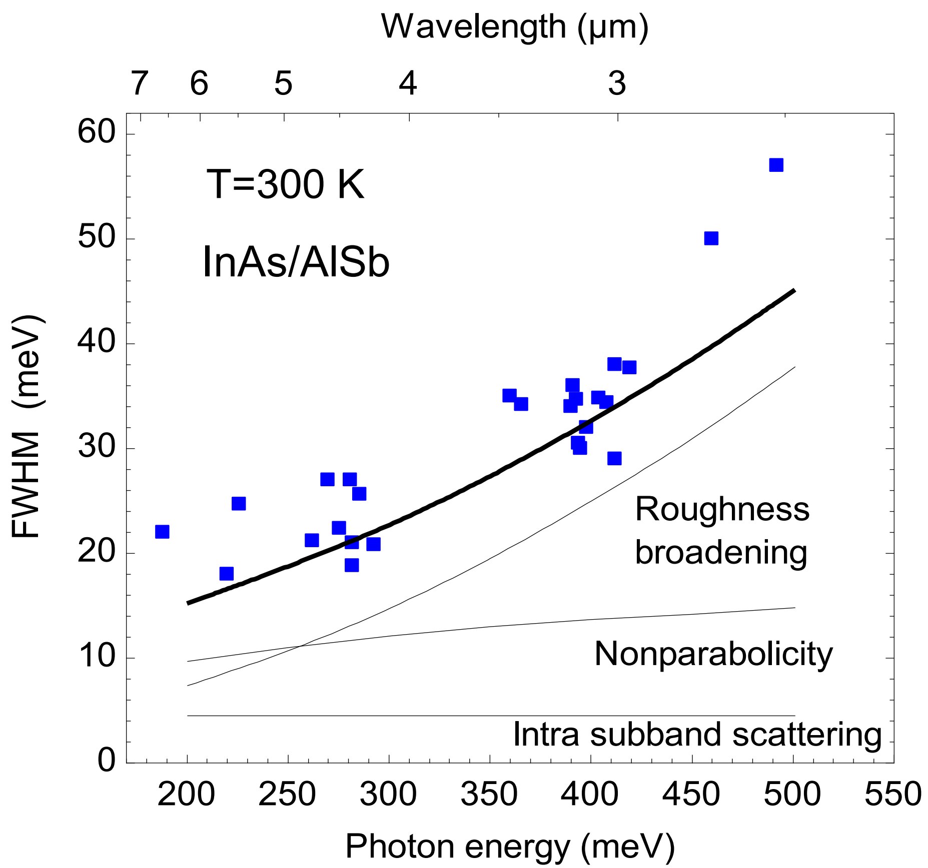
$$\cong \frac{\Delta^2\Lambda^2}{2}\int_0^{\pi} e^{-\frac{q^2\Lambda^2}{4}}d\theta$$

stronger interface overlap and barrier height for short λ

ISB transition broadening

FWHM=10% *hv*





D. Barate et al. (Montpellier)
APL 87 (5), 051103, (2005)

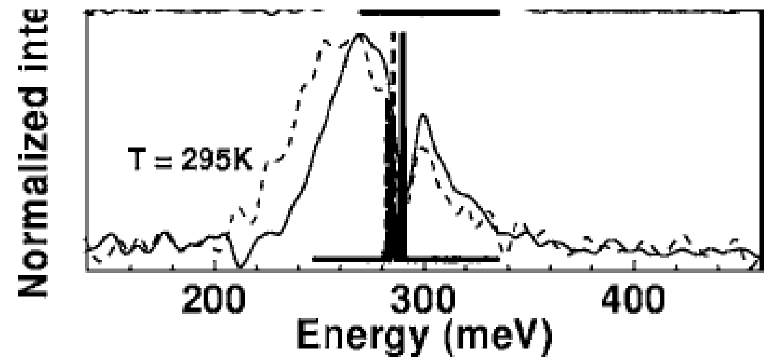
Reduction of intersubband gain : $g \propto \frac{1}{2\gamma}$

Reduction of tunnel injection efficiency

FWHM (300 K)

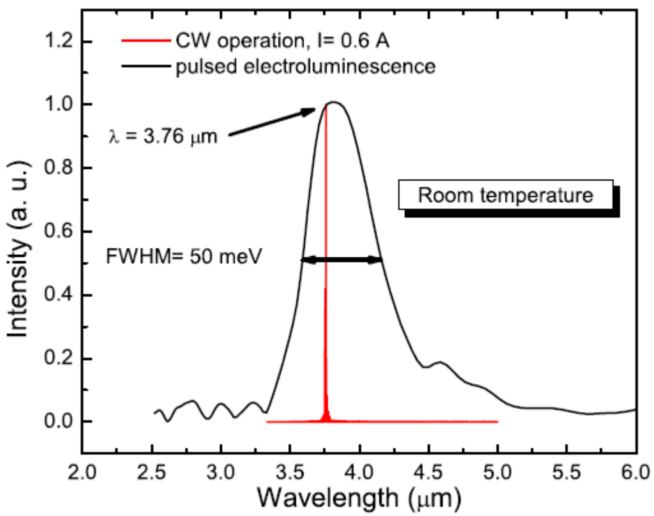
InGaAs/AlAsSb **(20% *hv*)**

Revin et al., (Sheffield), APL. 91, 051123, (2007)



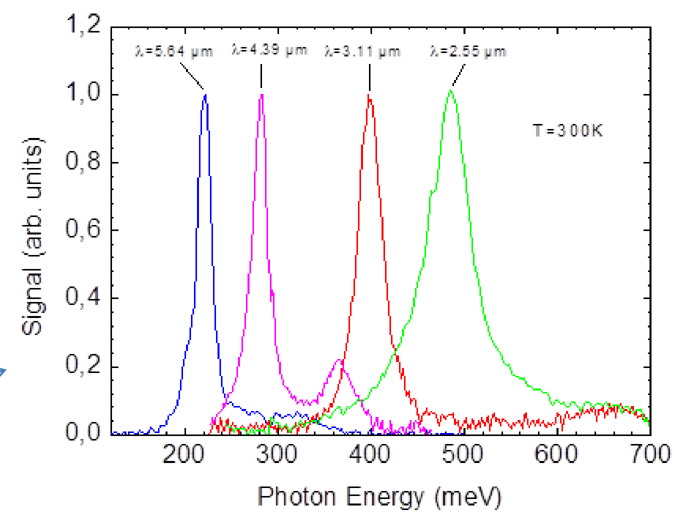
Strained InGaAs/AlInAs **(15% *hv*)**

Bandyopadhyay et al. (NWU), APL, 97, 131117, (2010)



InAs/AlSb **(10% *hv*)**

Barate et al. (Montpellier) APL 87, 051103, (2005)

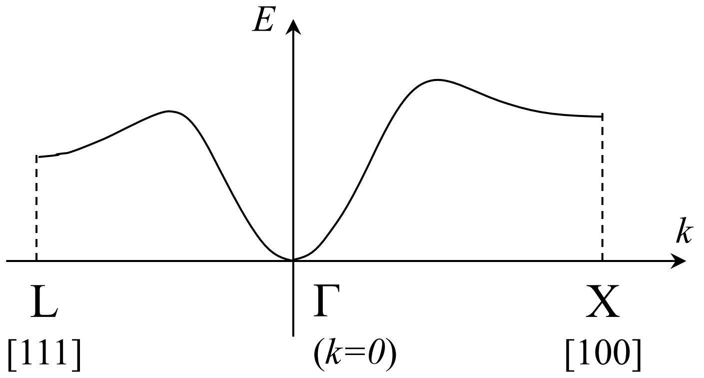


Lattice matched binary compounds

Lateral valleys

Bulk material
conduction band dispersion

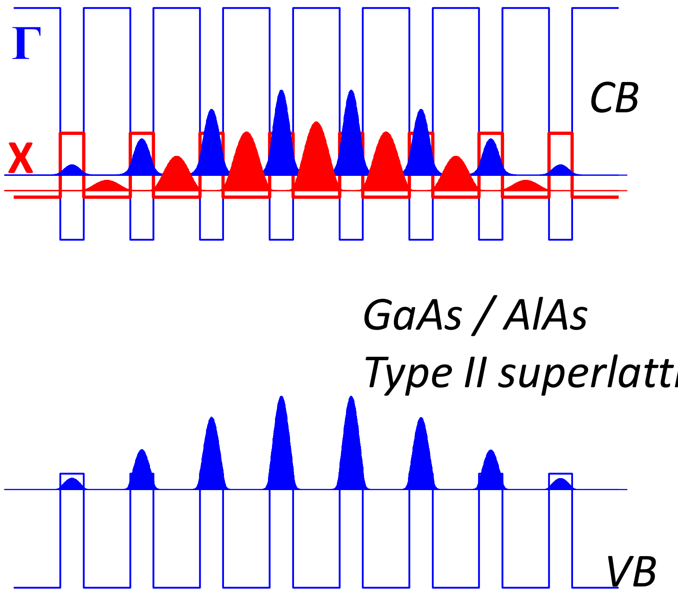
Γ, X or L minima



Heterostructure states
(envelop function theory)

Each minimum is treated independently,
with given band potential and effective mass

Confined states originating from different valleys



Transfer to X-states in QWs

$\Gamma \to X$ transfer time

Very efficient transfer : $\tau_{\Gamma X} \approx 0.1$ ps
but depends on wavefunction overlap

$X \to \Gamma$ transfer time

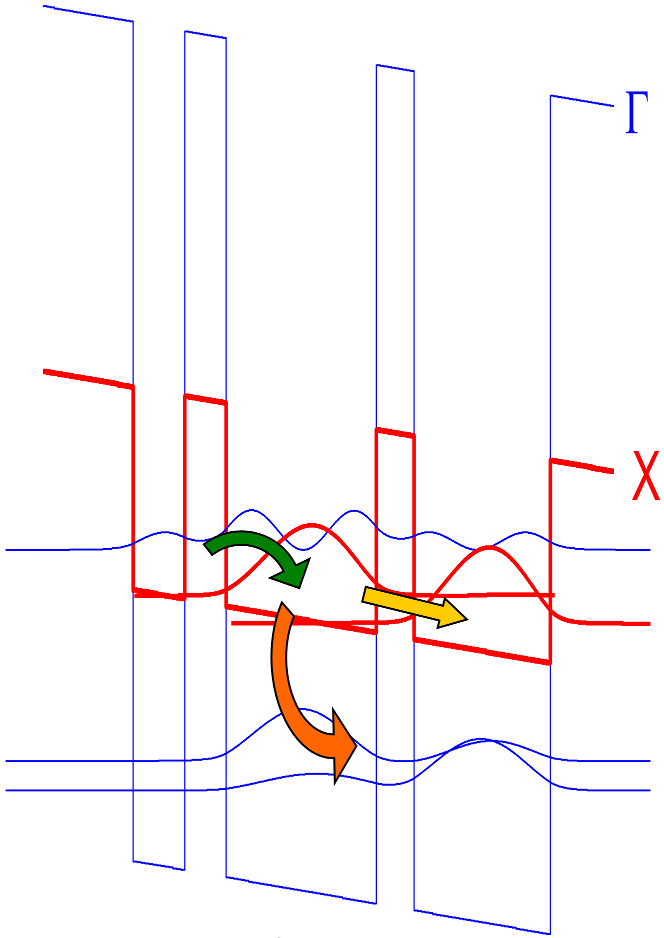
Limited by the small final density of states
($D \approx m^*$)

$$\tau_{X \to \Gamma} \approx \frac{6 \cdot m^*(X)}{m^*(\Gamma)} \tau_{\Gamma \to X} \approx 50 \times \tau_{\Gamma \to X}$$

- Localization (similar to deep levels)
- X (or L) miniband transport

Consequences for QCLs :

ISB emission still possible
Reduction of excited state lifetime
Reduction of injection efficiency

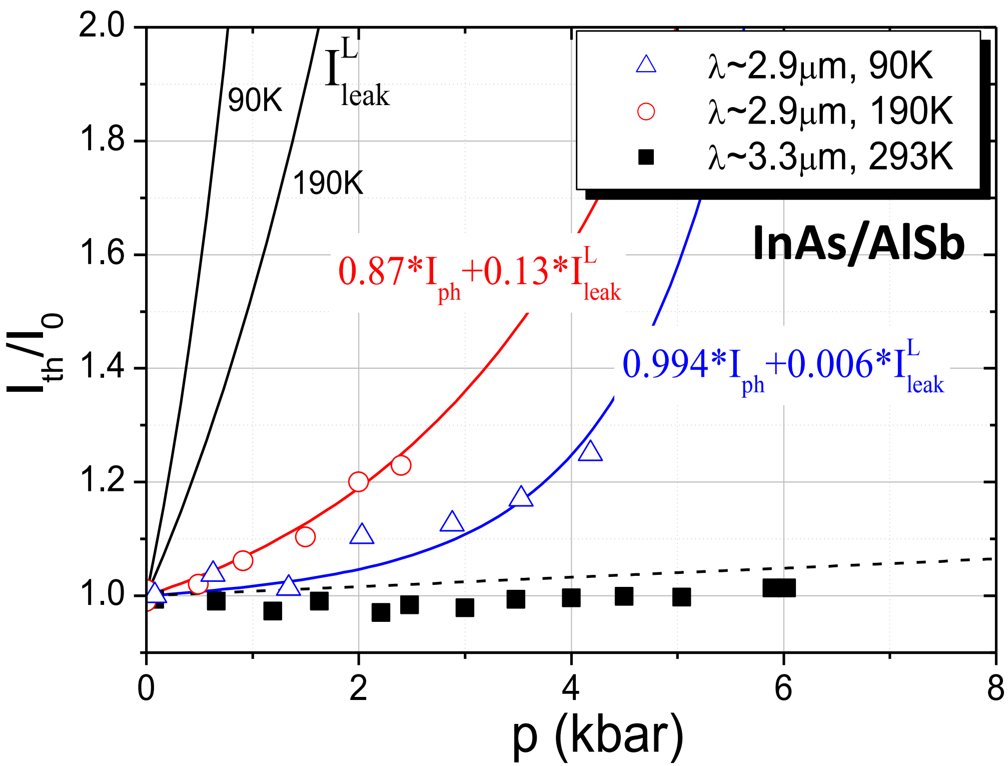


⇒ **Reduced gain**

Evidence of Γ- L scattering : hydrostatic pressure study

J_{th} vs. hydrostatic pressure
I.P. Marko et al. (Univ. Surrey)

$$I^L_{leak} \propto \exp\left(-\frac{d(E_L - E_\Gamma)}{dp}\frac{p}{kT}\right)$$

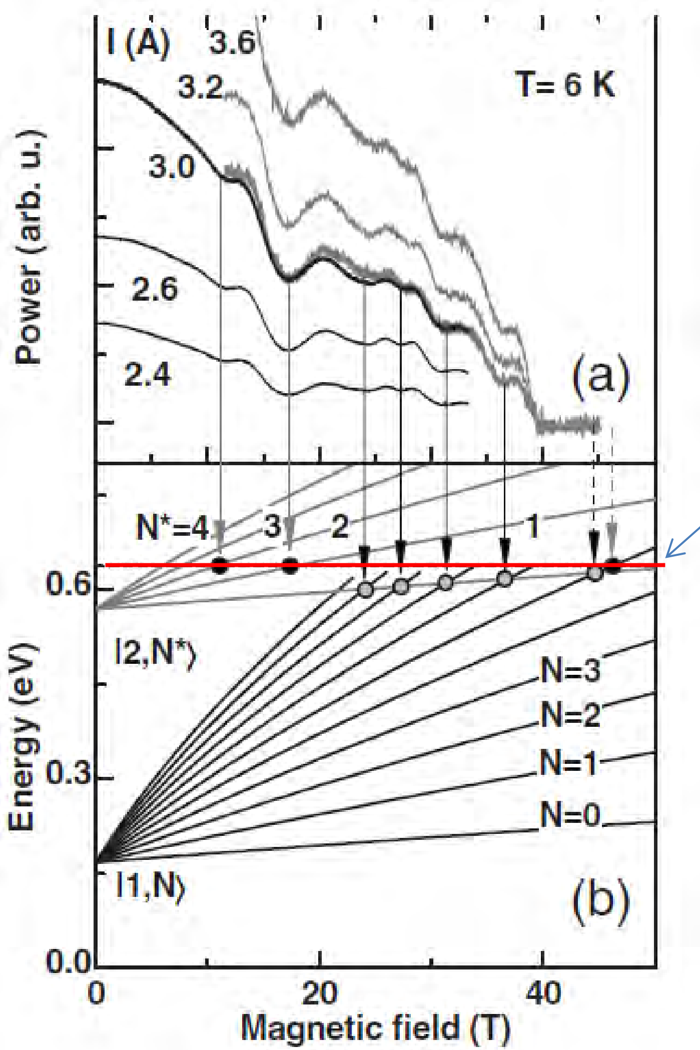


- no effect of pressure in λ=3.3 µm QCLs
- in λ=2.9 µm QCLs: **Thermally activated Γ-L scattering**

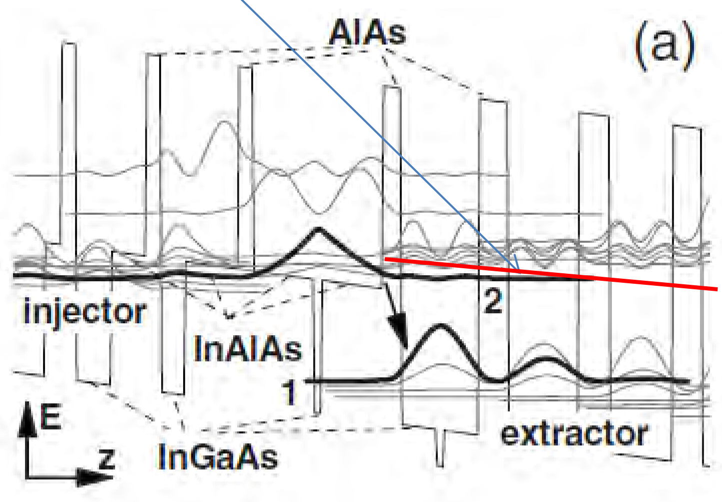
But not very strong (13% of J_{th} at 190 K)

Evidence of Γ- L scattering : magnetic field study

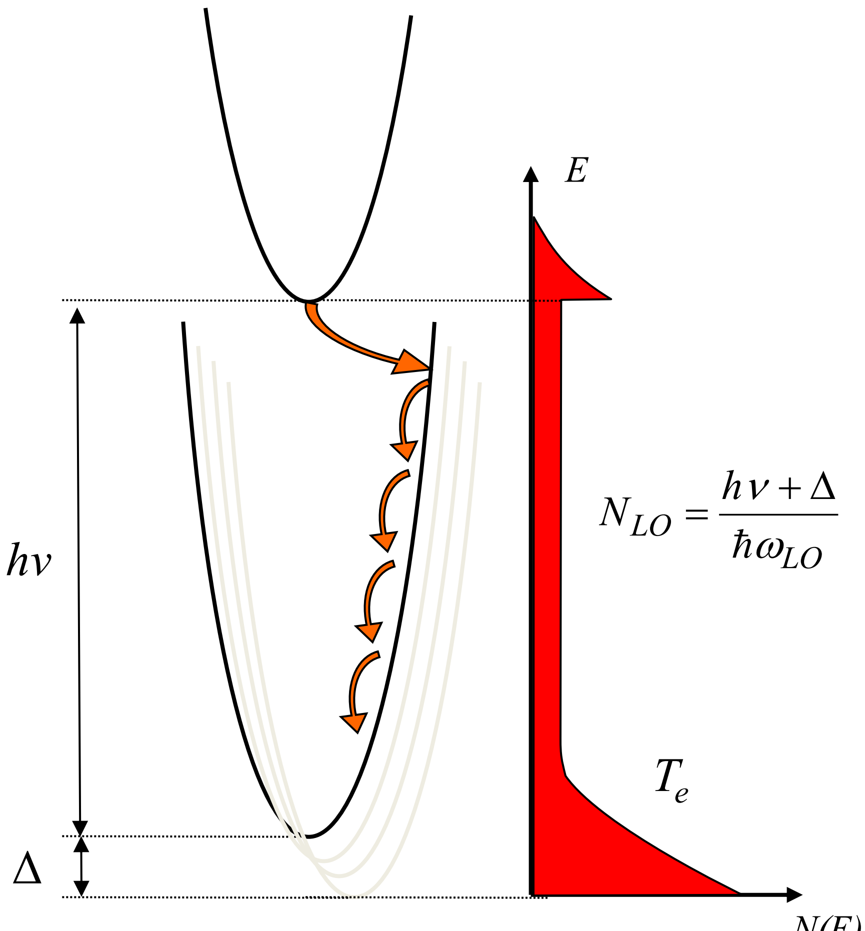
Semtsiv et al. (Berlin / Thallahassee)



L / X state 630 meV above Γ



Hot electrons



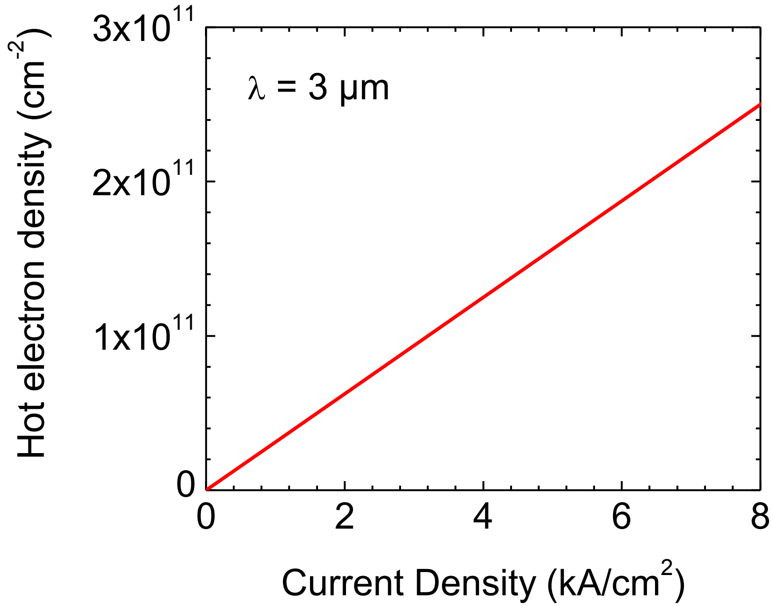
$\tau_{LO} \approx 0.3$ ps

Energy relaxation time

$$\tau_{hot} = N_{LO} \cdot \tau_{LO}$$

Hot electron density per period

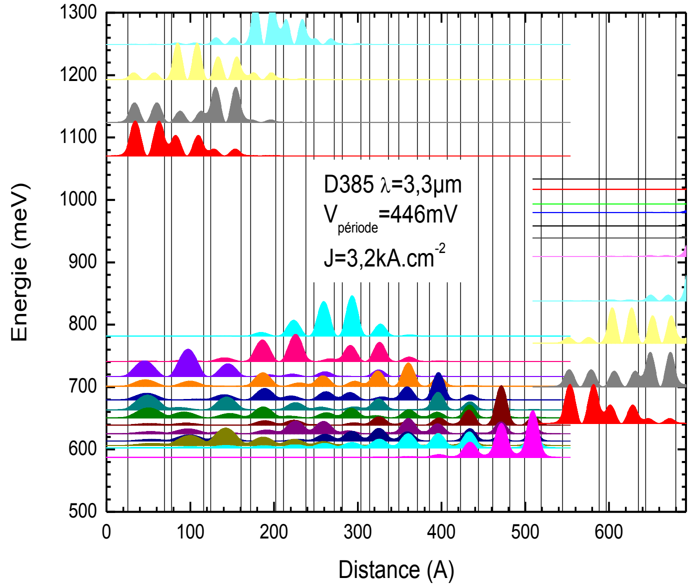
$$n_{hot} = \frac{J}{e}\tau_{hot}$$



Hot electron density can be comparable to average doping density per period

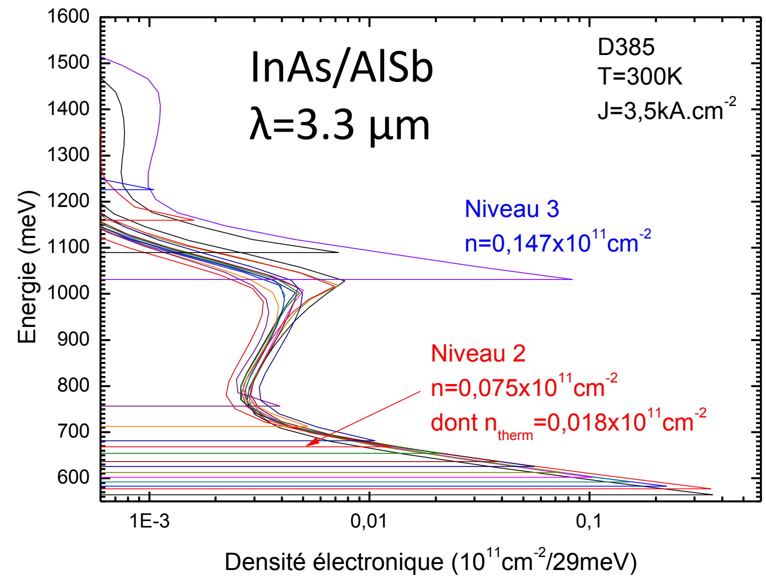
Energy-resolved rate equations model

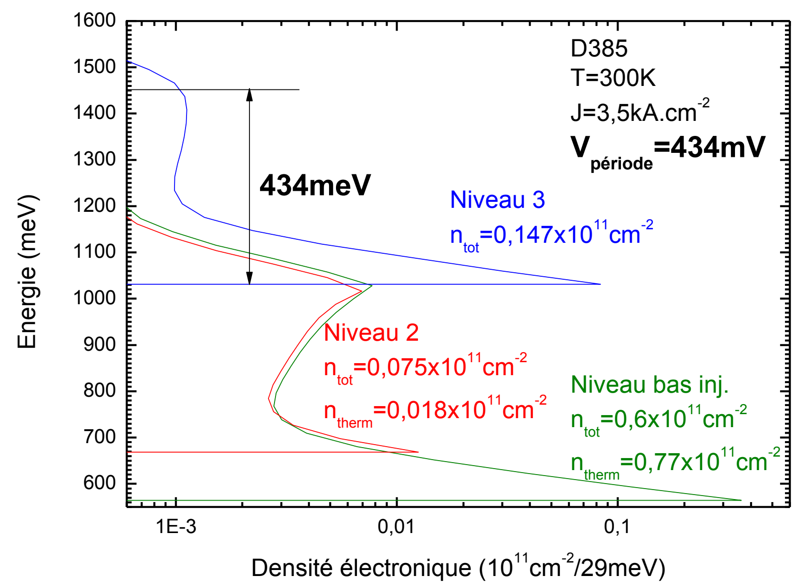
P. Laffaille, PhD thesis, Montpellier (2013)

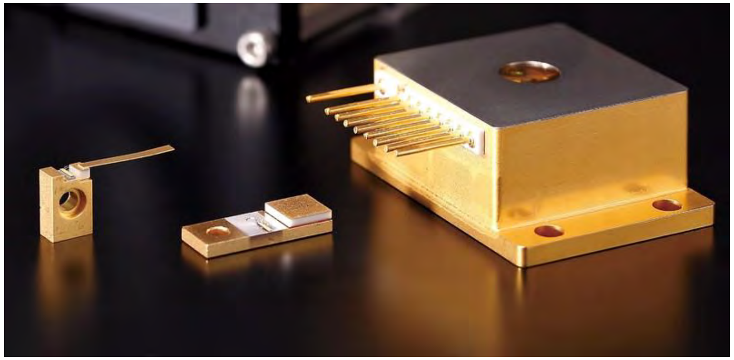


$n_{hot} \approx n_s$

but $n_{hot}(e2) \approx n_s / 10$





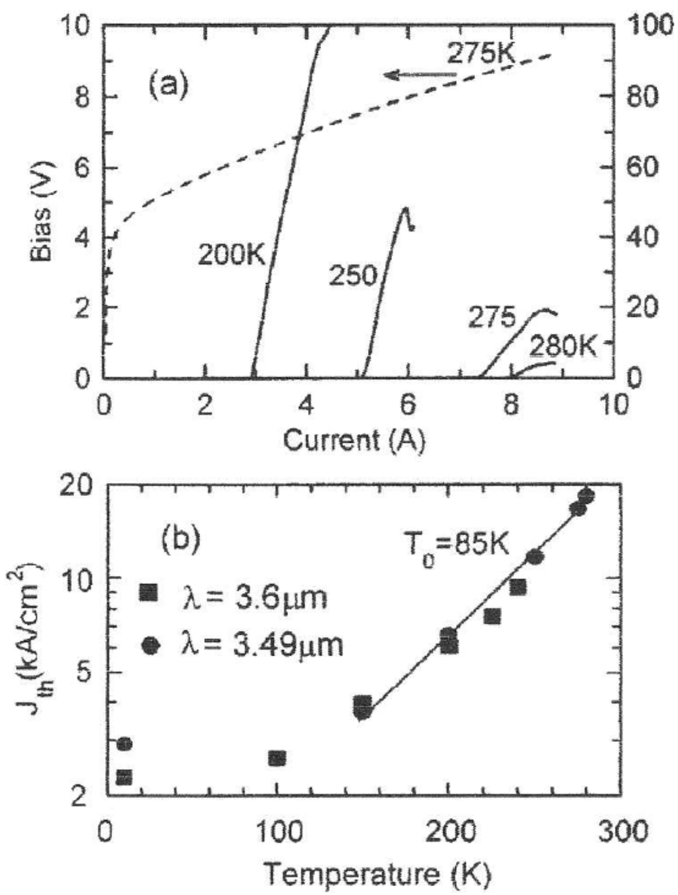


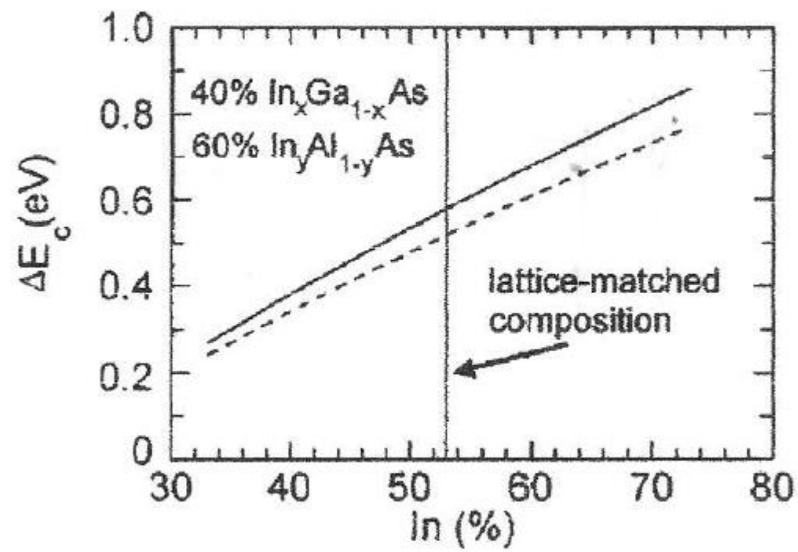
4 – SHORT λ QCLs

- InGaAs / AlInAs
- InGaAs / AlAsSb
- InAs AlSb

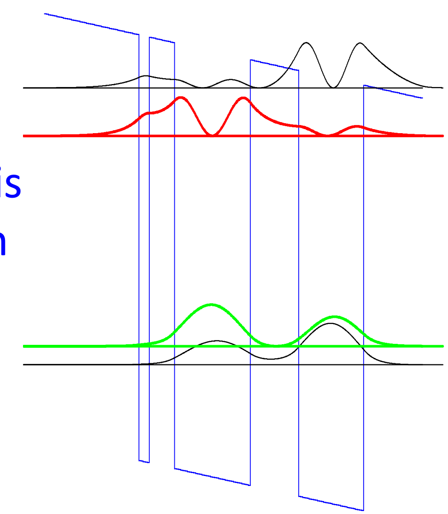
First experimental result of a short λ QCL

Strain compensated InGaAs/InAlAs
[Faist et al. APL 72, 680, (1998)]

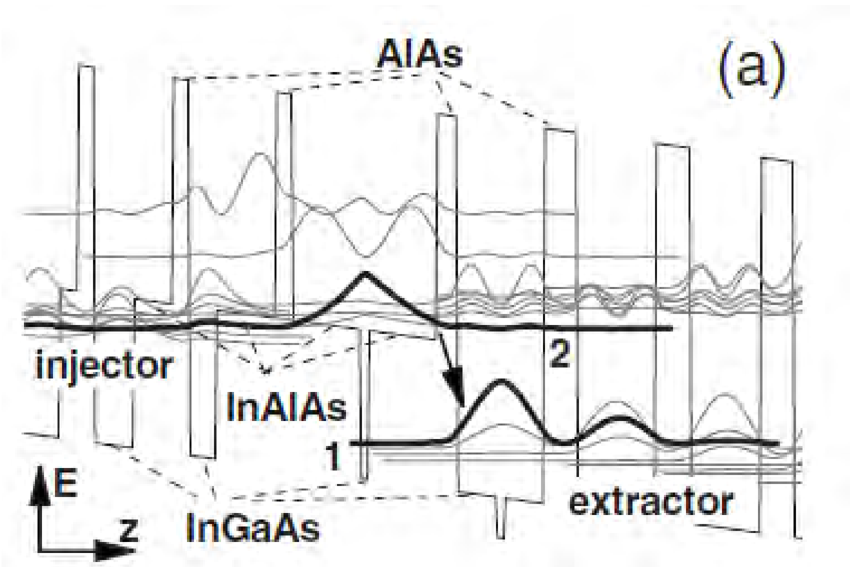


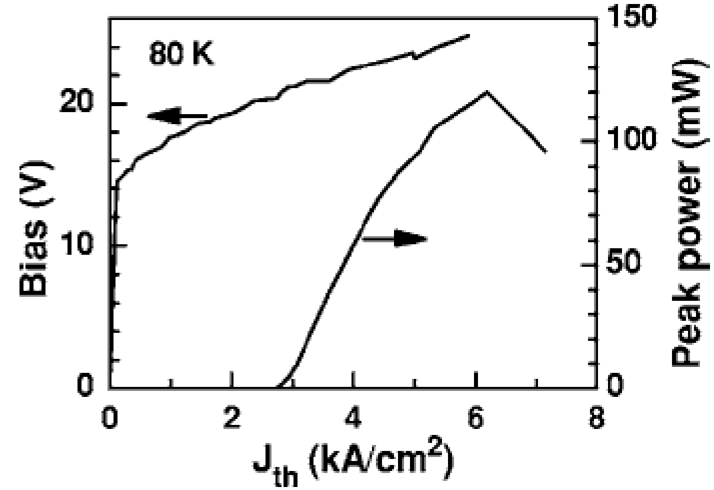


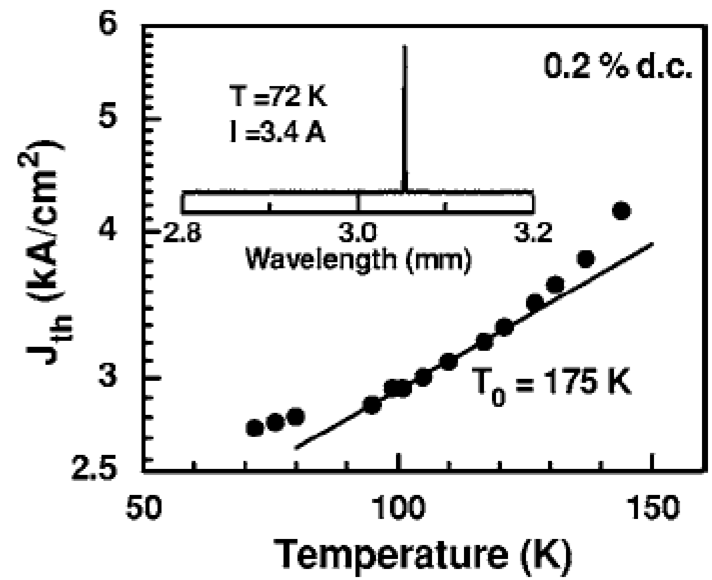
This is the limit for this material composition InGaAs (70%)



Composite barriers AlAs-InAlAs







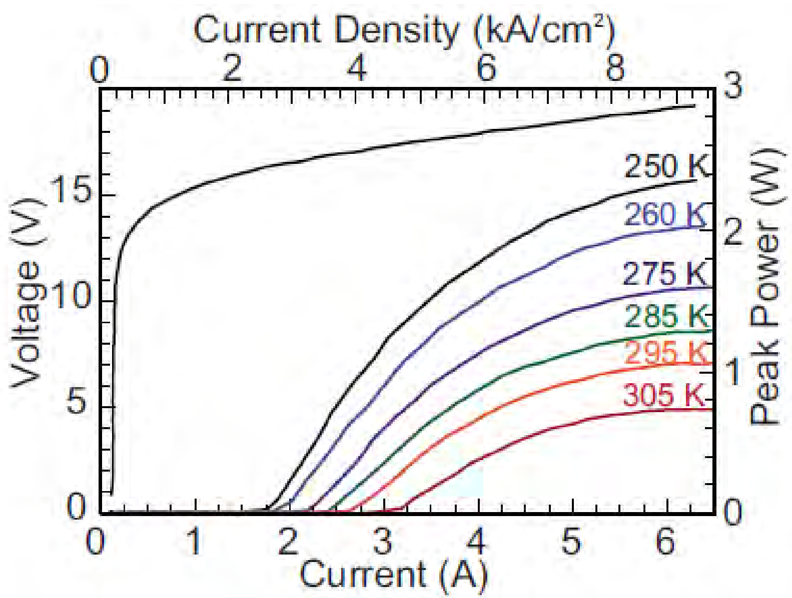
InGaAs(73%) / InAlAs(55%) / AlAs

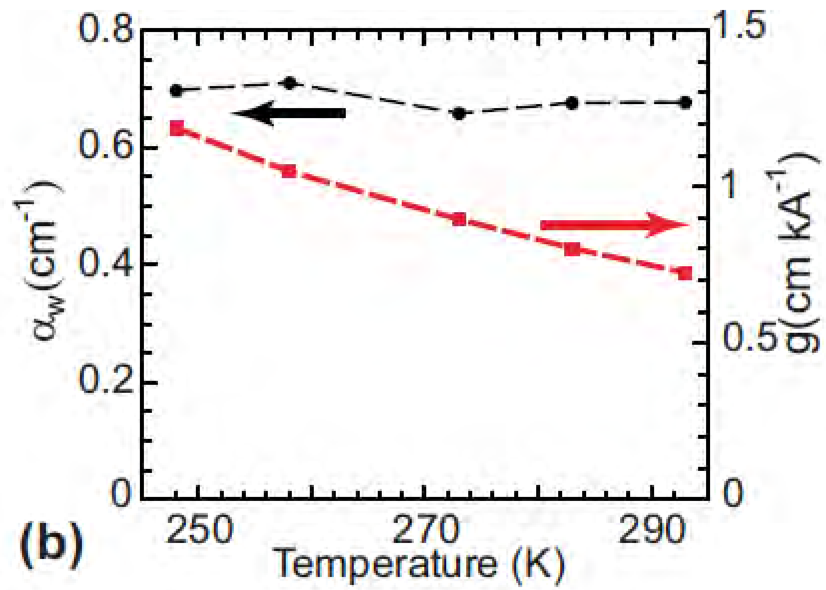
λ=3.05 µm , ΔE_c= 1.4 eV

Semtsiv et al., (Berlin), APL. **90**, 051111, (2007)

Above room temperature operation (λ=3.3 µm)

InGaAs(72%) / InAlAs(53%) / AlAs
λ=3.3 µm, ΔE_c= 1.4 eV

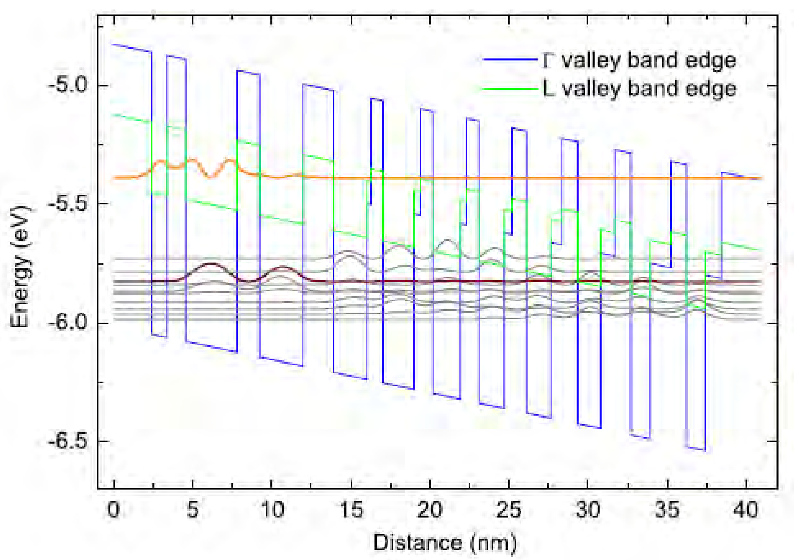


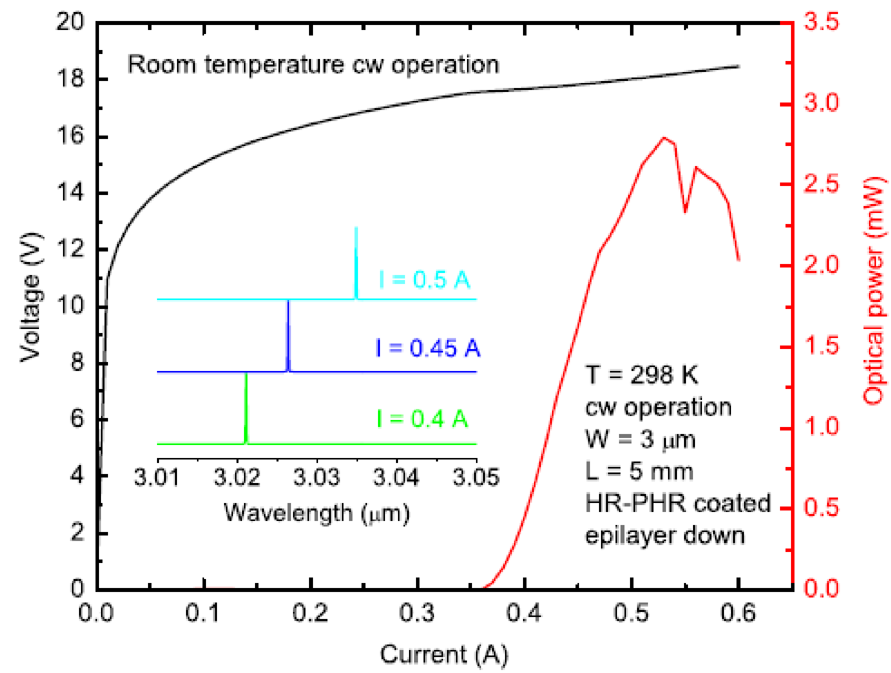


But low gain !

Bismuto et al., (ETHZ), APL. **98**, 191104, (2011)

Highly strained materials: RT, CW





InGaAs(79%) / InAlAs(89%)

λ=3.02 μm, ΔE_c= 1.2 eV

Bandyopadhyay et al. (NWU), APL, 101, 241110, (2012)

Watt level RT, CW operation above 3.3 μm

Milestones - Strained InGaAs/AlInAs QCLs

First Strain compensated InGaAs/InAlAs QCL (λ=3.5 μm)
Faist et al., (Neuchatel) ,APL 72, 680, (1998)

Room temperature (λ=4 μm) composite AlAs barriers
Semtsiv et al., (Berlin), APL. 85, 1478, (2004)

Room temperature CW (λ=3.8 μm)
Yu et al., (NorthWestern U.), APL. 88, 251118, (2006)

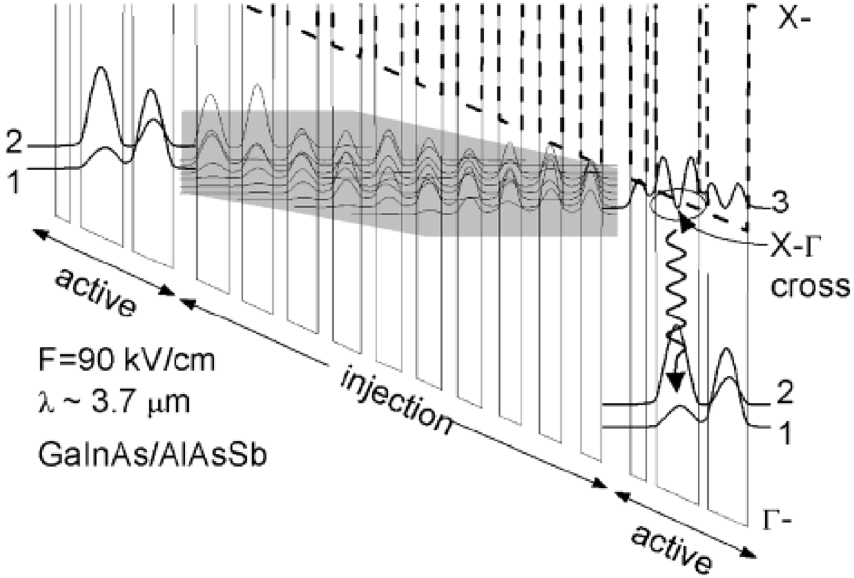
Short wavelength (λ=3.05 μm) composite AlAs barriers
Semtsiv et al., (Berlin), APL. 90, 051111, (2007)

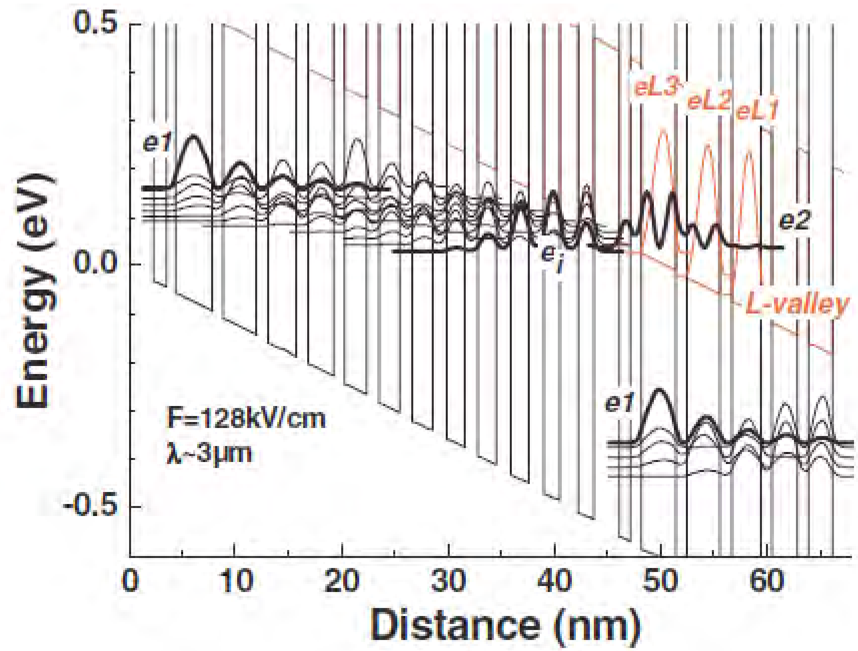
Room temperature (λ=3.3 μm) composite AlAs barriers
Bismuto et al., (ETHZ), APL. 98, 191104, (2011)

High power Room temperature CW (λ=3.4 μm)
Bandyopadhyay et al. (NorthWestern U.), APL, 100, 212104, (2012),

Room temperature CW (λ=3.02 μm) Composite InAlAs barriers
Bandyopadhyay et al. (NorthWestern U.), APL, 101, 241110, (2012)

InGaAs/AlAsSb designs





Yang et al. (Freiburg) APL 88, 121127, (2006)

Revin et al. (Sheffield) APL 90, 021108, (2007)

Lattice matched

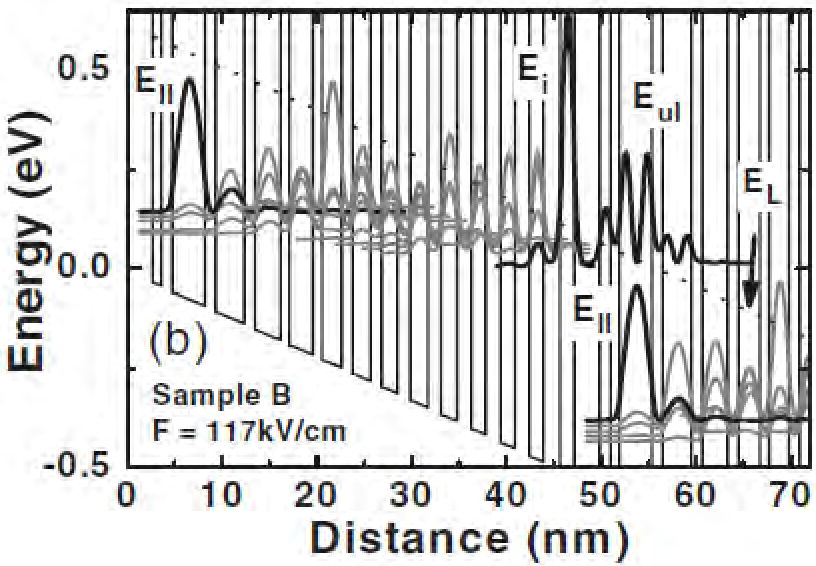
λ=3.7 µm, 300 K

Strain compensated materials
InGaAs(67%) / AlAsSb

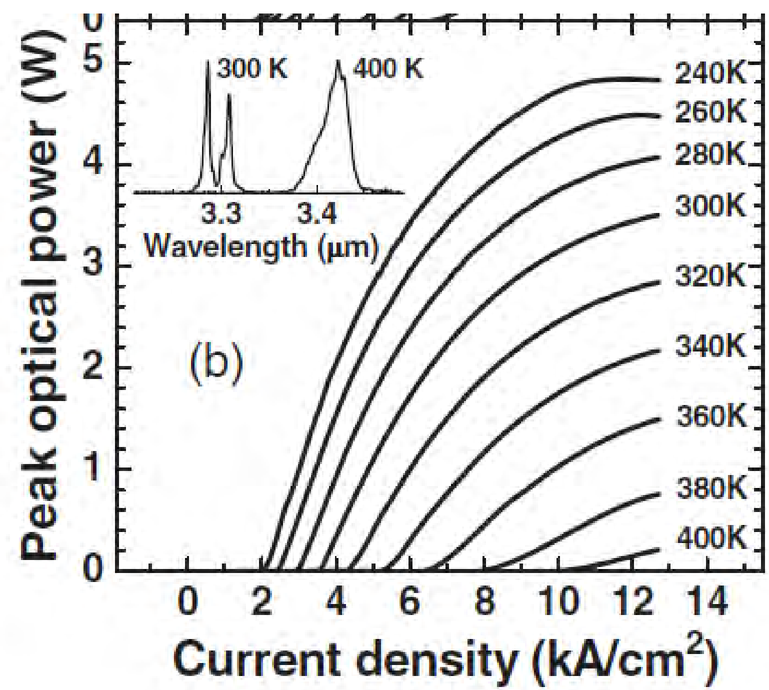
λ=3.1 µm, RT
but J_{th}=19 kA/cm²

Large ΔE_c , but the (strain dependent) lateral valley sets the limit

InGaAs/AlAsSb QCL @ λ=3.3 μm



Commin et al. (Sheffield), APL 97, 031108, (2010)



Strain compensated materials
InGaAs(70%) / AlAsSb / AlAs

λ=3.3 μm, J_{th}(300K)=3.6 kA/cm²

AlAs barriers in the active QWs improved interface quality

Milestones - InGaAs/AlAsSb QCLs on InP

First GaInAs/AlAsSb on InP substrate (λ=4.3 μm)
Revin et al. APL 85, 3992, (2004)

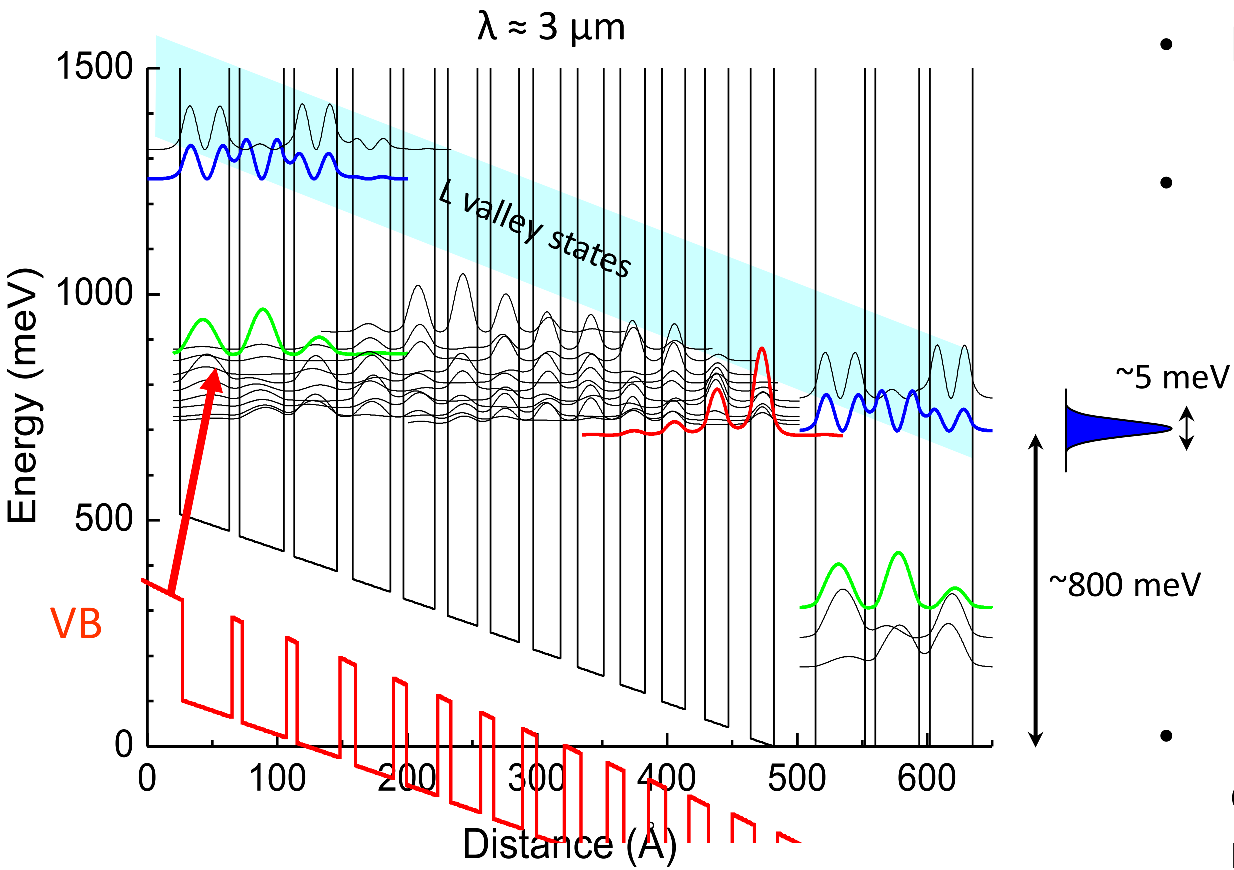
Room temperature (λ=3.7 μm)
Yang et al. APL 88, 121127, (2006)

Record short λ (λ=3.05 μm)
Revin et al. APL 90, 021108, (2007)

Room temperature (λ=3.1 μm) strain compensated
Revin et al. APL 94, 031106, (2009)

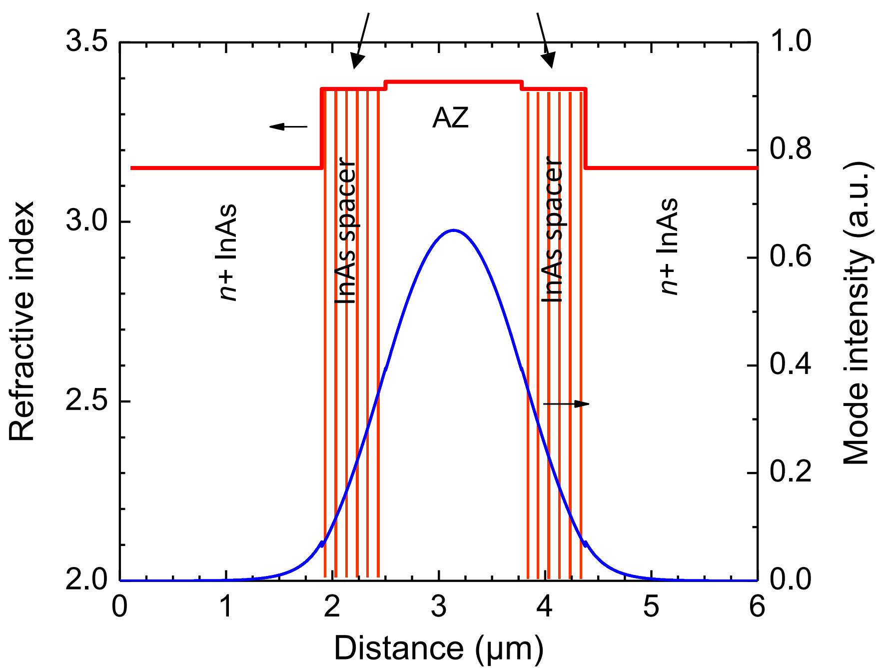
Tmax=400 K and high peak power (λ=3.3 μm) AlAs barriers
Commin et al., APL 97, 031108, (2010)

InAs/AlSb QCLs



- Lateral valleys (L)
- Interband absorption
- Design:
  control electron levels and resonant tunneling injection

Waveguide for short λ InAs/AlSb QCLs

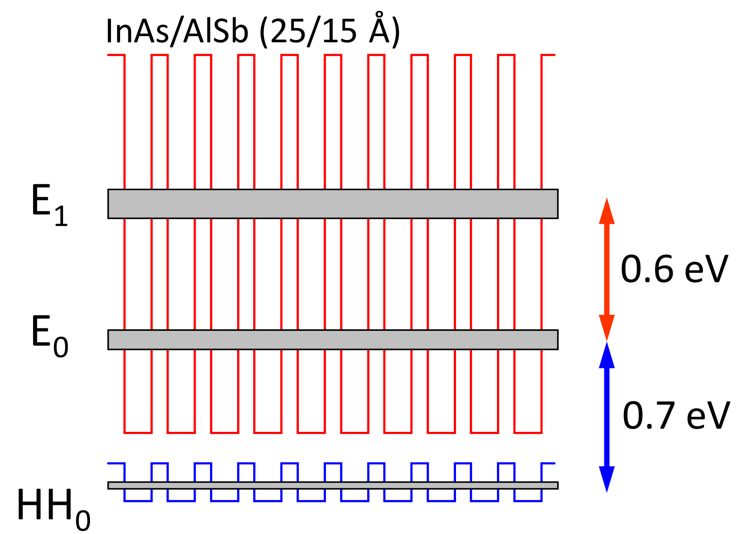


InAs spacers replaced with InAs/AlSb superlattices

InAs plasmon enhanced waveguide successfully used for λ>4.5 μm

BUT: InAs bandgap (300K) = 0.35 eV

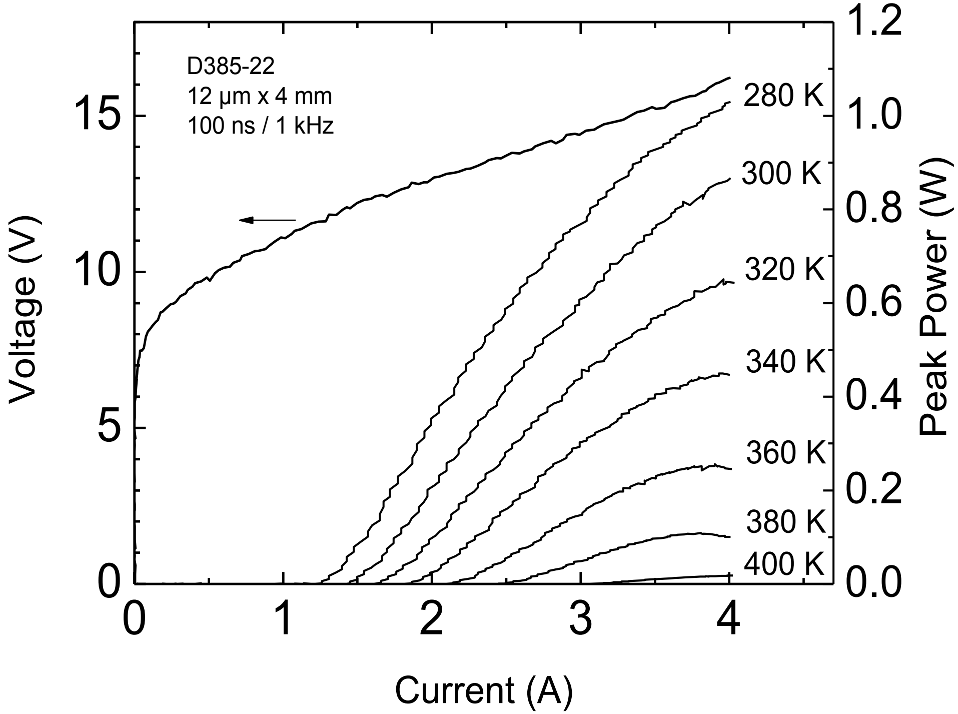
➔ Cutoff ≈ 3.6 μm



SL transparent for IB and ISB transitions

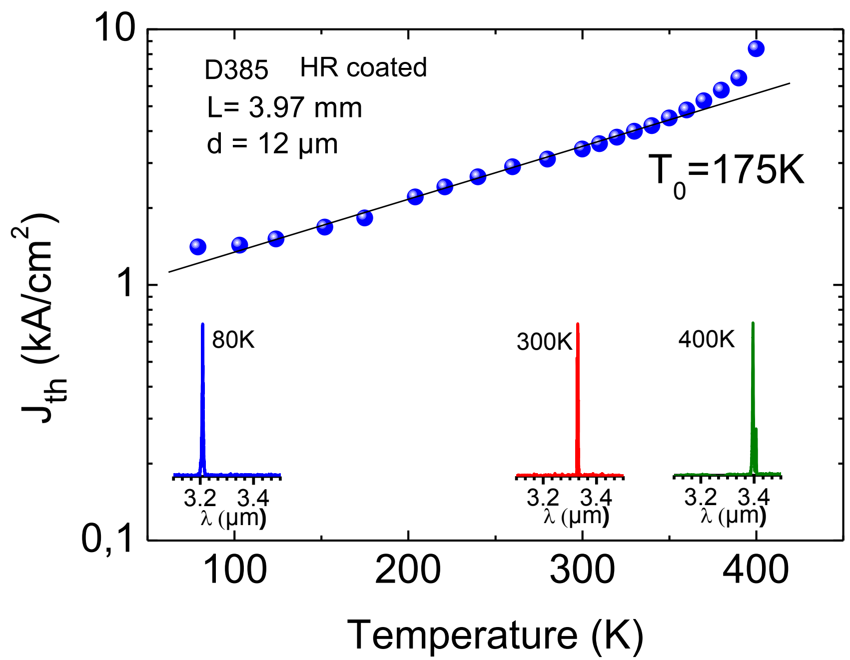
InAs/AlSb QCLs @ λ=3.3 μm

Devenson et al. (Montpellier), APL 91, 141106, (2007)



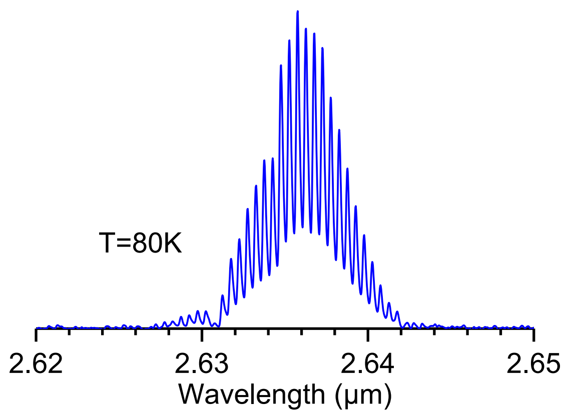
Room temperature :
λ= 3.33 μm, J_{th}=3.0 kA/cm², P > 1 W

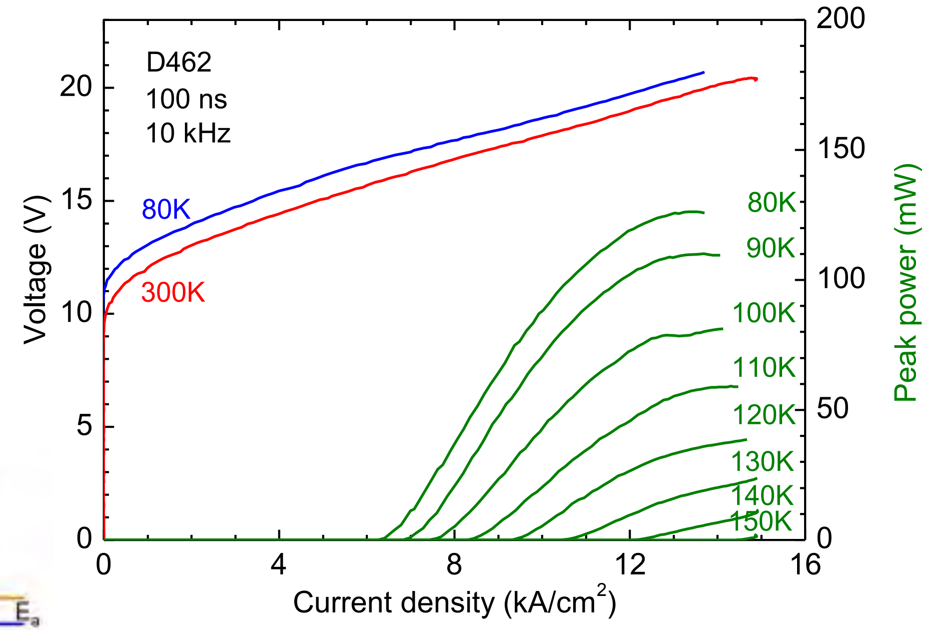
T_{max} > 400 K

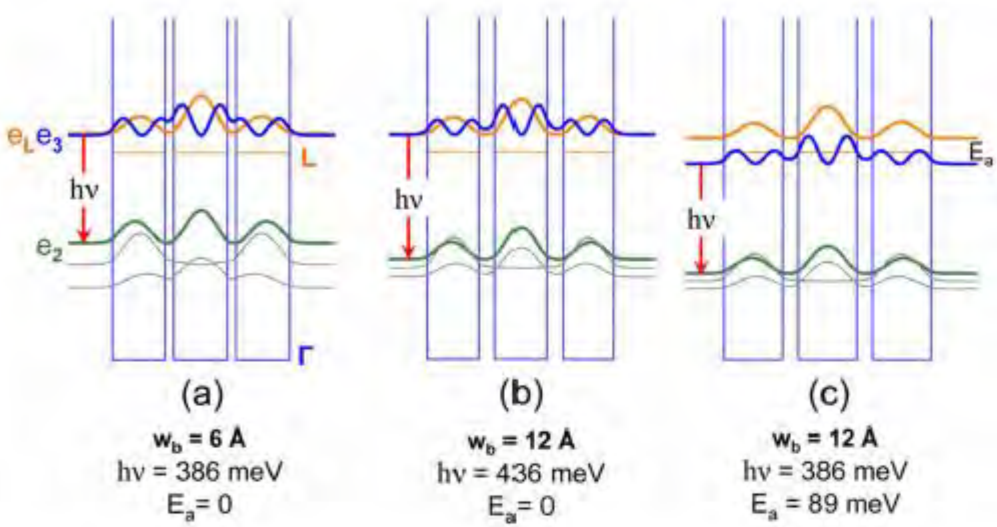


High temperature operation, up to 150 °C
an advantage compared to interband diodes

The short wavelength frontier of QCLs : 2.6 µm (hν= 470 meV)







Increased localisation of Γ states

$J_{th} \approx 6$ kA/cm^2

$T_{max} = 155$ K

Cathabard et al., (Montpellier), APL 96, 141110, (2010)

Milestones - InAs/AlSb QCLs

First InAs/AlSb on InAs substrate (λ=10 μm)
Ohtani et al., (Tohoku), APL 82, 1003, (2003)

Room temperature (λ=4.5 μm)
Teissier et al., (Montpellier), APL 85, 167, (2004)

Short λ QCL (λ=3.1 μm)
Devenson et al. (Montpellier), APL 89, 191115, (2006)

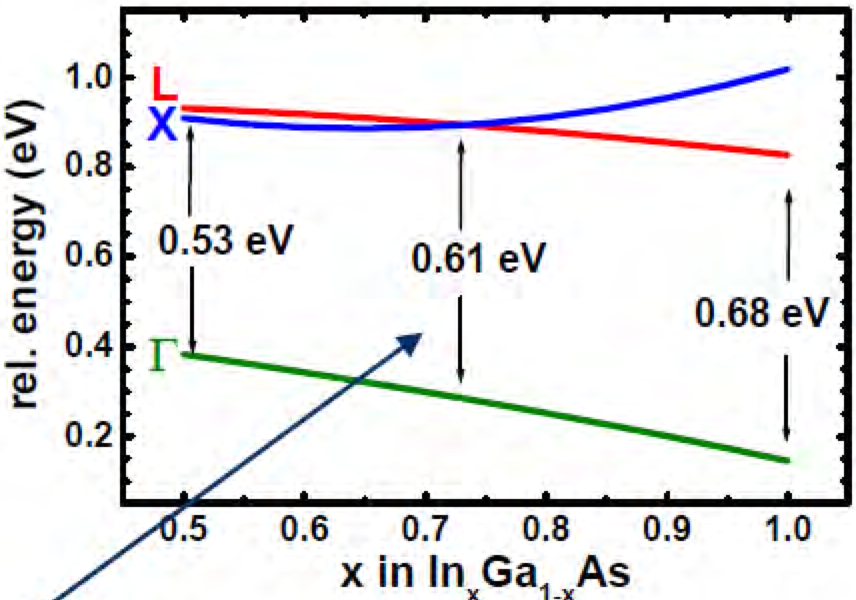
Tmax=400 K and high peak power (λ=3.3 μm)
Devenson et al. (Montpellier), APL 91, 141106, (2007)

Record short λ (λ=2.6 μm)
Cathabard et al., (Montpellier), APL 96, 141110, (2010)

5 - CONCLUSIONS

Lateral valleys set the limit wavelength

The indirect-Γ separation increases with In content. The maximum transition energy increases correspondingly.



$0.53\text{eV} \Rightarrow \lambda_{min} \approx 3.8\ \mu m$

$0.61\text{eV} \Rightarrow \lambda_{min} \approx 3.3\ \mu m$

$0.68\text{eV} \Rightarrow \lambda_{min} \approx 2.9\ \mu m$

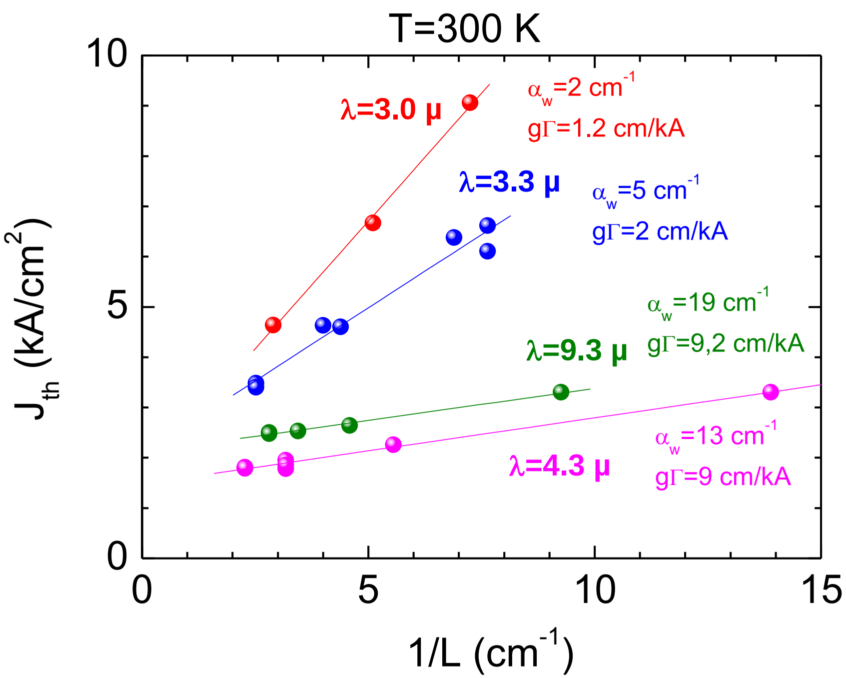
For relaxed InAs,
$\lambda_{min} \approx 2.7\ \mu m.$

$In_xGa_{1-x}As$ with $x \approx 0.72$

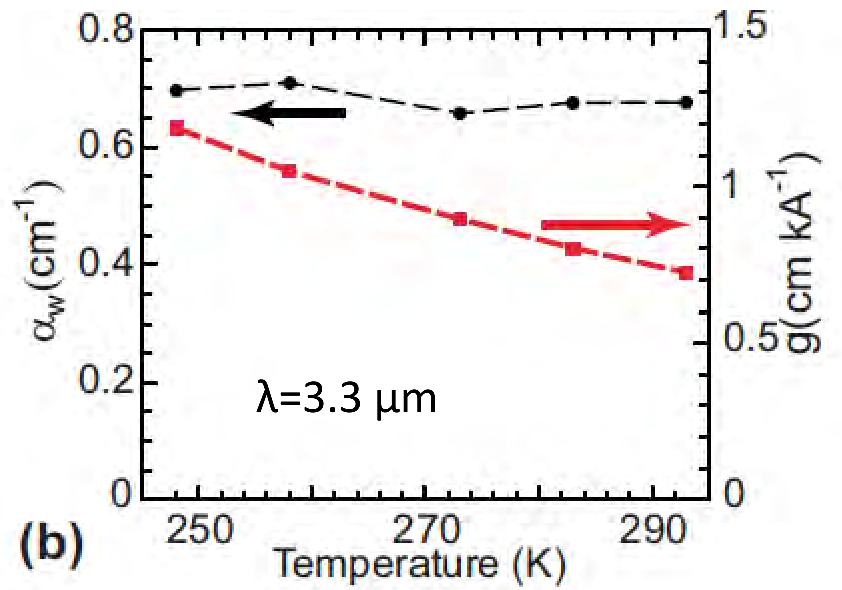
(courtesy of T. Masselink)

Intersubband gain is low (loss too)

InAs / AlSb



InGaAs / Al(In)As



Low gain due to interface roughness

- Reduced lifetimes
- Broader transitions

InAs vs. InP :
higher gain,
but less efficient waveguide

State of the art

- RT CW and Watt above 3.3 µm (InP)
- RT, CW above 3.02 µm (InP)
- RT above 2.9 µm (InAs)
- QC Lasers above 2.6 µm (InAs)