Nonlinear optical interactions in quantum cascade lasers

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Collaborations:

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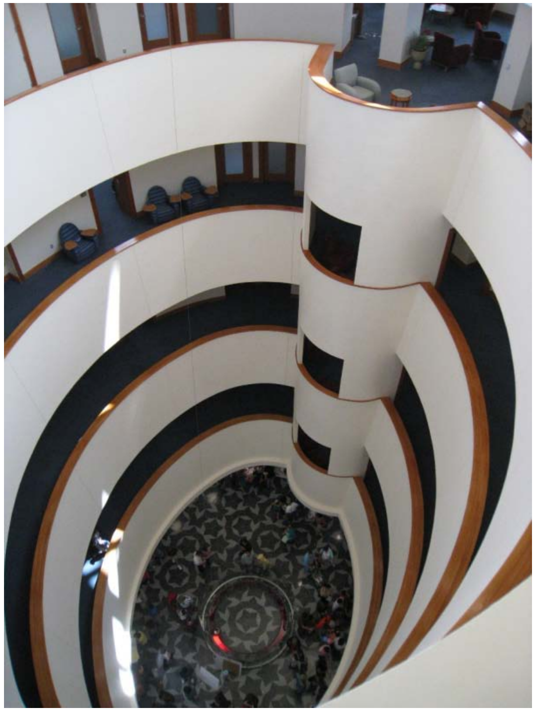
J. Cockburn group, *Sheffield University*

Funding: NSF, NHARP, AFOSR, EU

George Mitchell Physics Buildings

87-ft Foucault pendulum



Roger Penrose standing on Penrose Tiling

Mitchell and Hawking

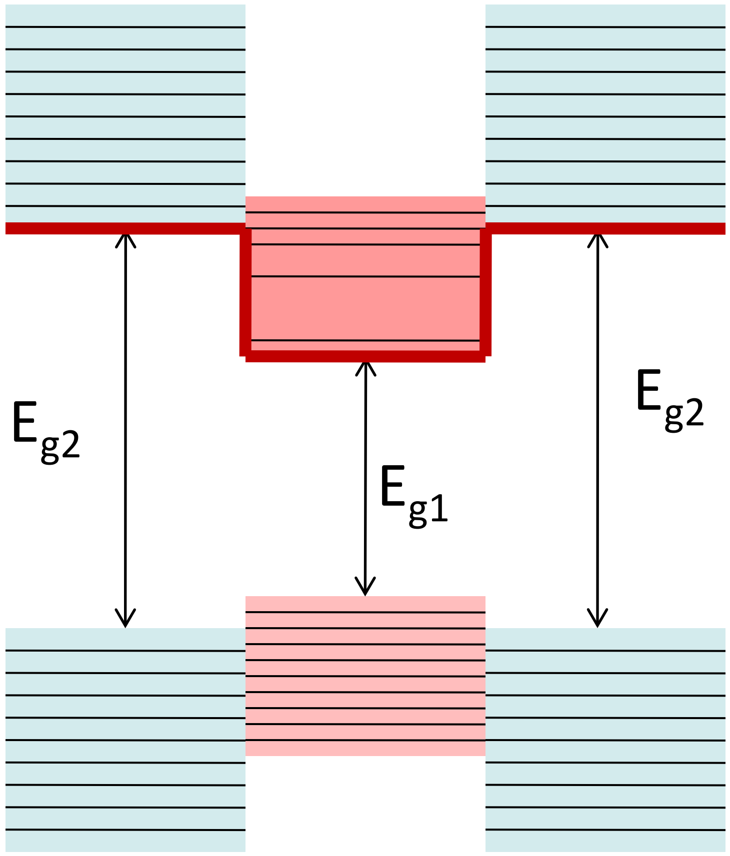
Outline

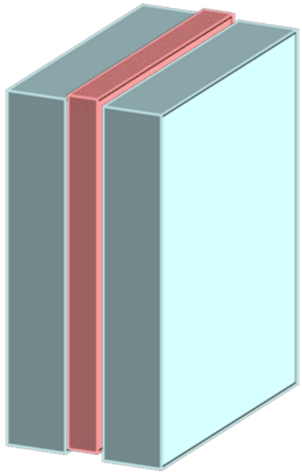
- Introduction: quantum wells and QCLs
- Nonlinear phenomena in QCLs
  - Nonlinear frequency conversion
  - Frequency locking and phase coherence of transverse modes; synchronization
  - Mode locking and ultrashort pulse generation; coherent regimes

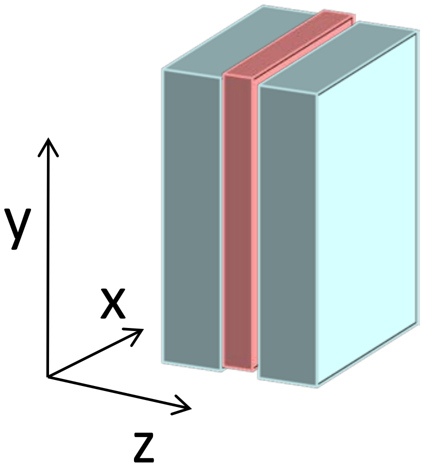
Other stuff

- Ultrafast and collective phenomena in semiconductor magnetoplasmas
  - Nature Phys. 2010, PRLs, PRB 2010
- Magneto-optics in dilute magnetic semiconductors (PRB2011)
- Mid-IR & THz optics in graphene and CNTs
  - TERANO NSF PIRE Center
- Coherent mid/far-IR photodetectors (NSF ERC)
- Cavity QED, Hawking-Unruh radiation

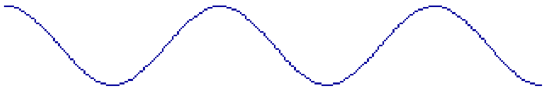
Quantum-confined electron gas







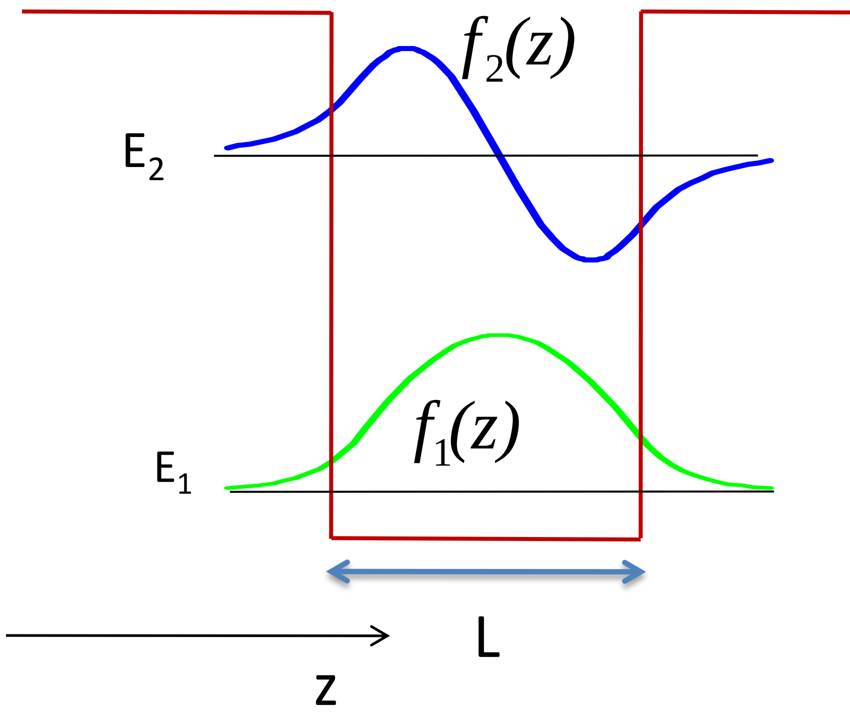
Quantum wells



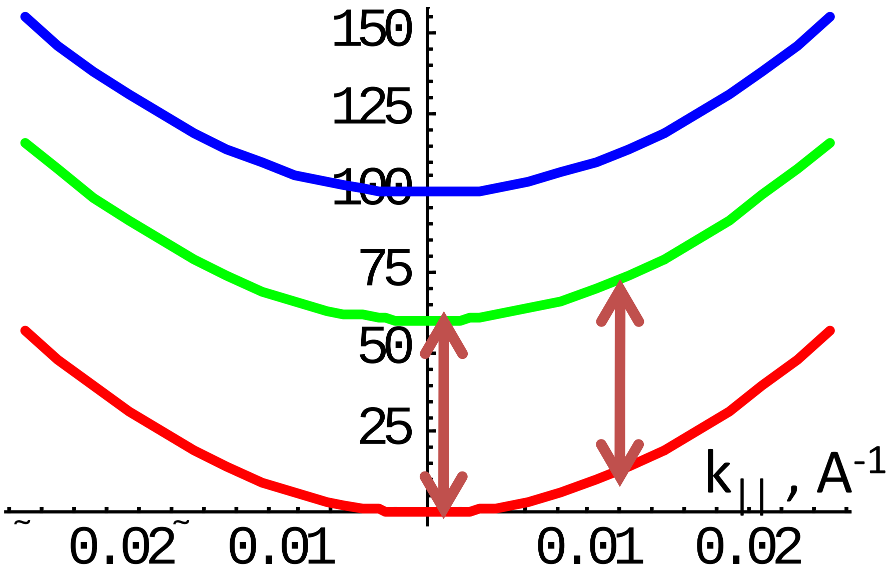
Bulk: $$\psi = e^{i\mathbf{kr}} u_{\mathbf{k}}(\mathbf{r})$$

Quantum well:

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

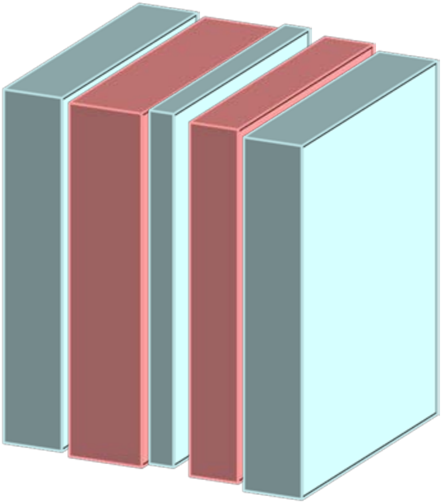


$$E_n \approx E_{n0} + \frac{\hbar^2 k_\parallel^2}{2m_n}; \quad n = 1,2,\ldots$$



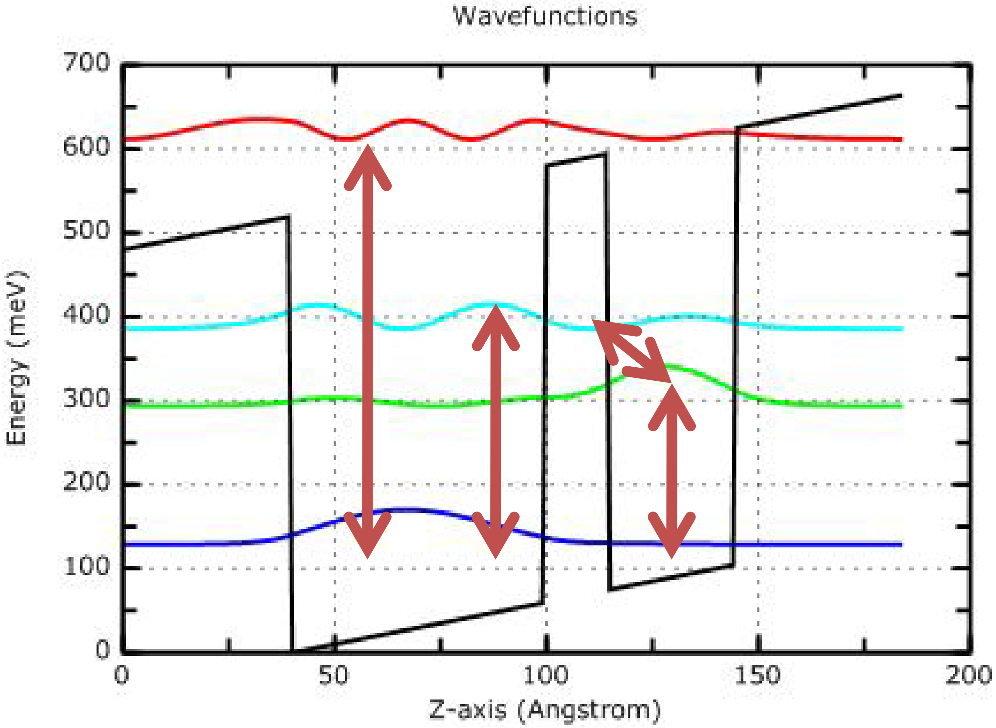
- Large dipole moment ~ L
- Sharp resonances

Build your own nanostructure:



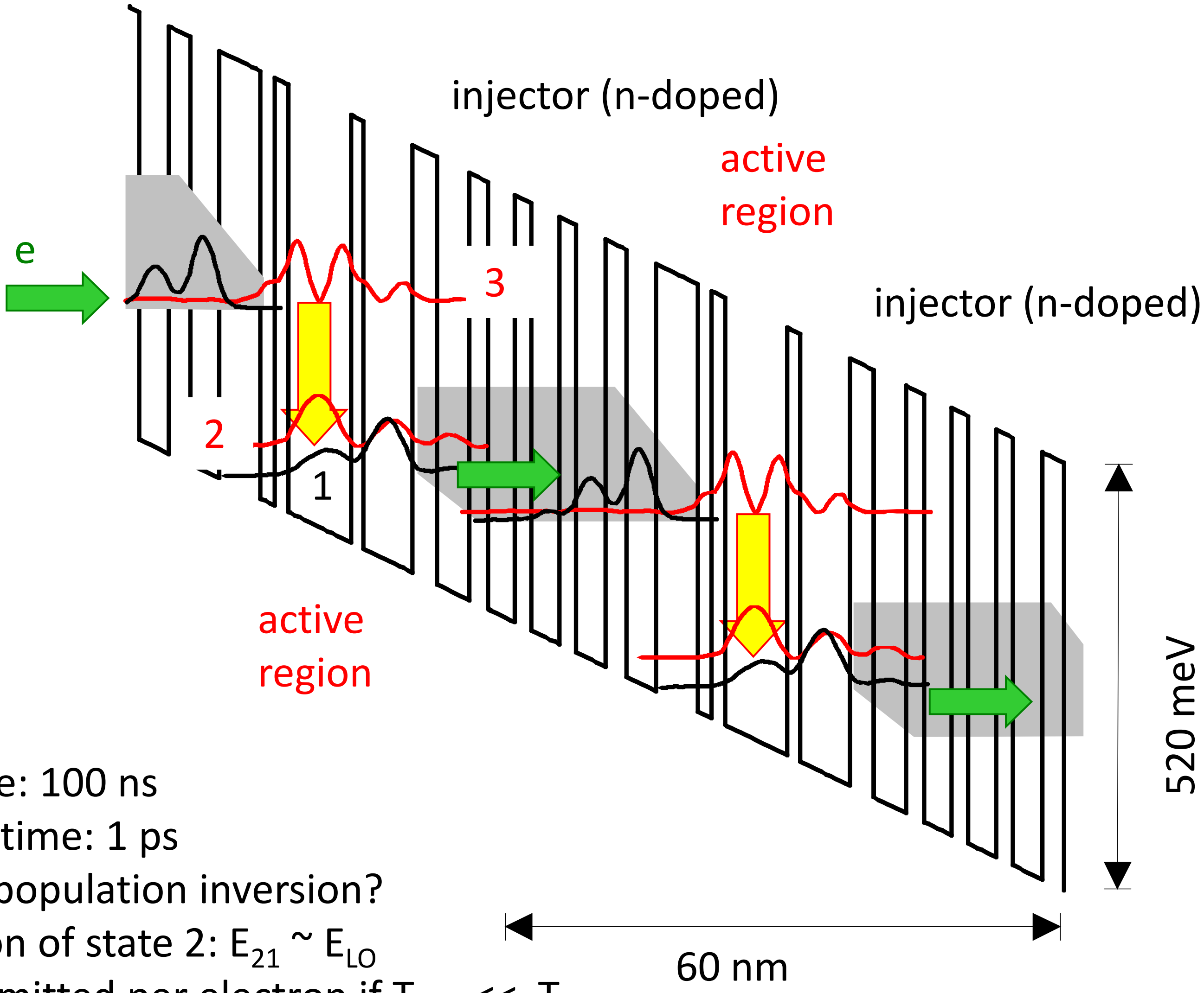
E-field

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



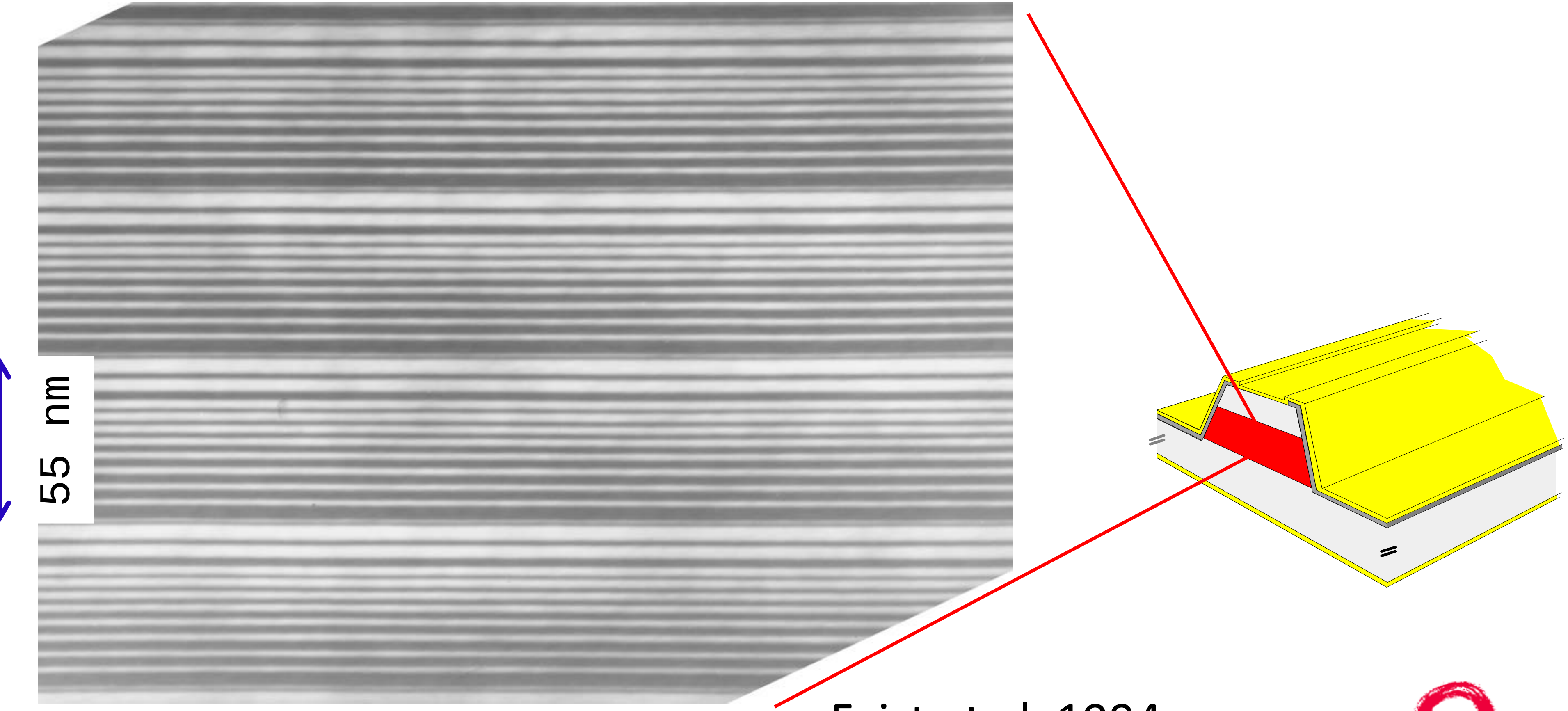
- Sharp resonances
- Tunable frequencies and oscillator strengths
- Ability to control populations by tunneling and coupling to phonons
- Create population inversion
- Enhance optical nonlinearity

One example of QC laser design



Radiative lifetime: 100 ns
Nonradiative lifetime: 1 ps
How to provide population inversion?
Fast depopulation of state 2: $E_{21} \sim E_{LO}$
Many photons emitted per electron if $T_{stim} << T_1$

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



Faist et al. 1994

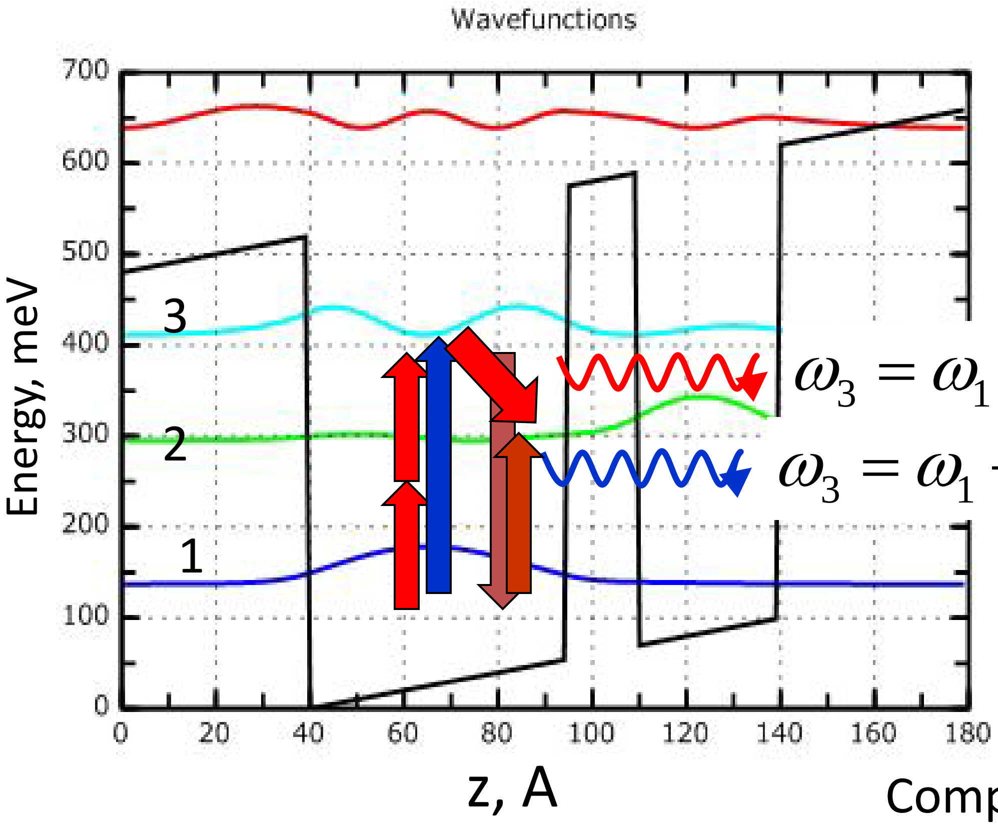
Lucent Technologies
Bell Labs Innovations

How one can enhance optical nonlinearity in QCLs and "engineer" nonlinear optical devices:

- Playing with multiple resonances to generate new frequencies
- Playing with saturation nonlinearity (spectral and spatial hole burning): phase coherence of EM modes and mode locking
- Playing with coherence of the gain medium (intersubband polaritons): Rabi oscillations, Risken-Nummedal-Graham-Haken (RNGH) instability; self-induced transparency; superfluorescence

Resonant nonlinear optics with nanostructures

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



$$\omega_3 = \omega_1 - \omega_2$$

$$\omega_3 = \omega_1 + \omega_2$$

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

Δ_{ij} – detunings

γ_{ij} – linewidths

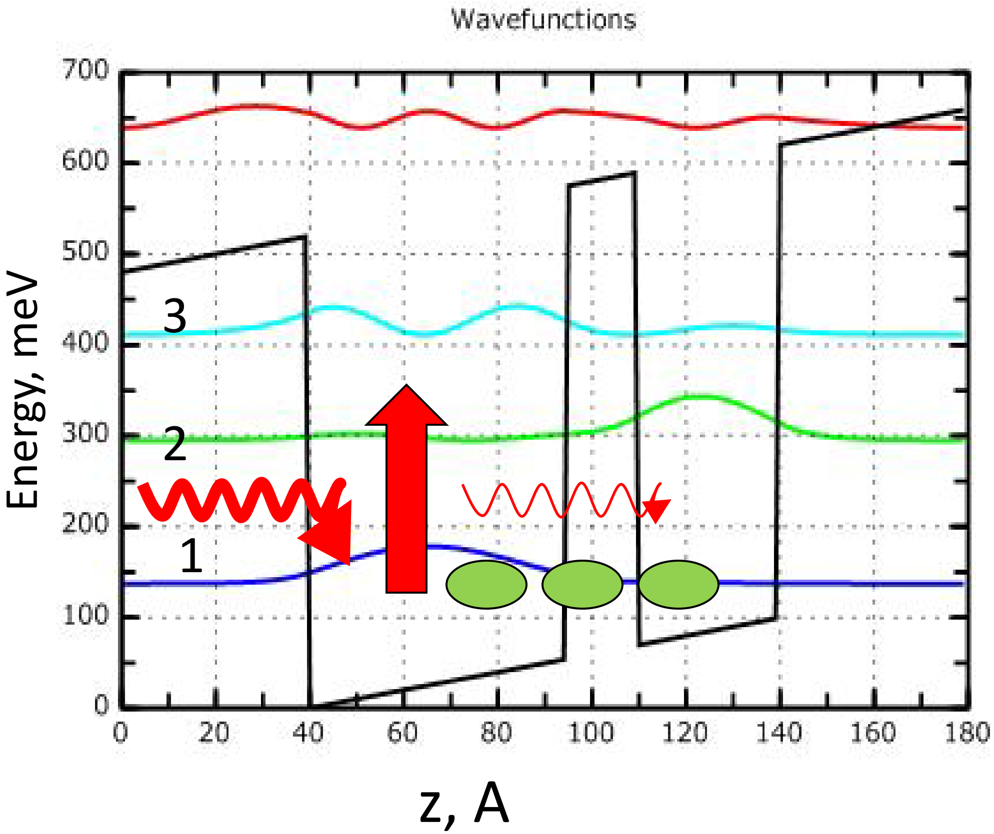
d_{ij} – dipole moments

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

Compare with 1-10 pm/V for bulk crystals

A way to get around resonant absorption

Resonant optical nonlinearity is accompanied by resonant absorption

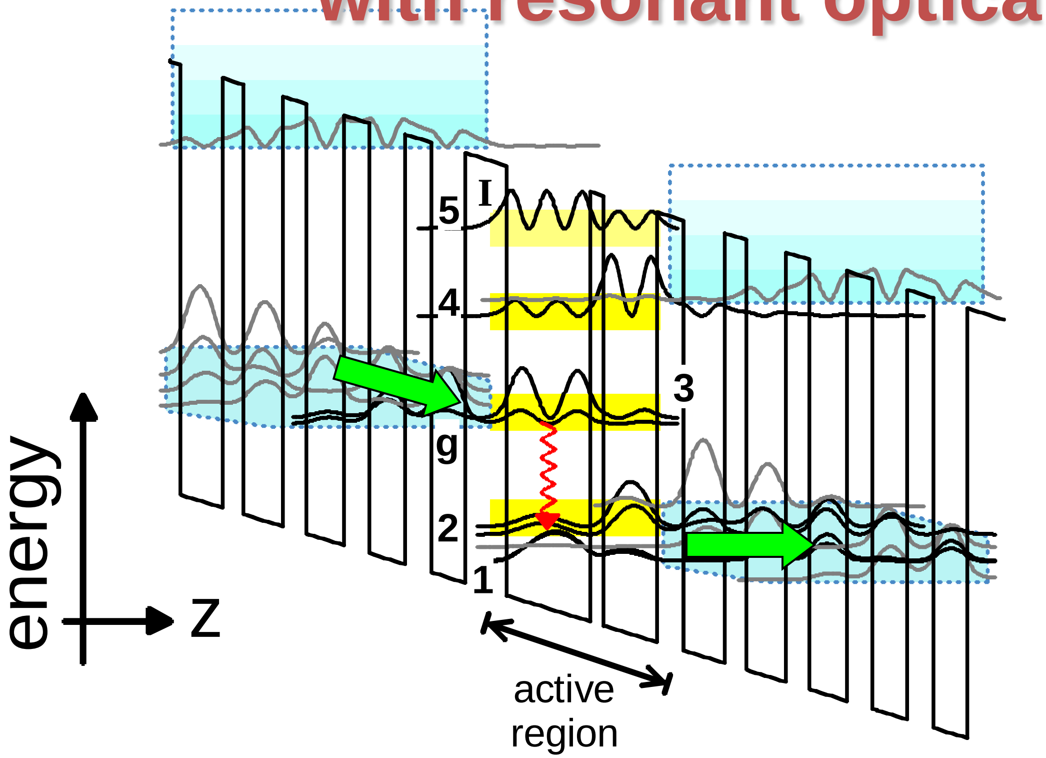


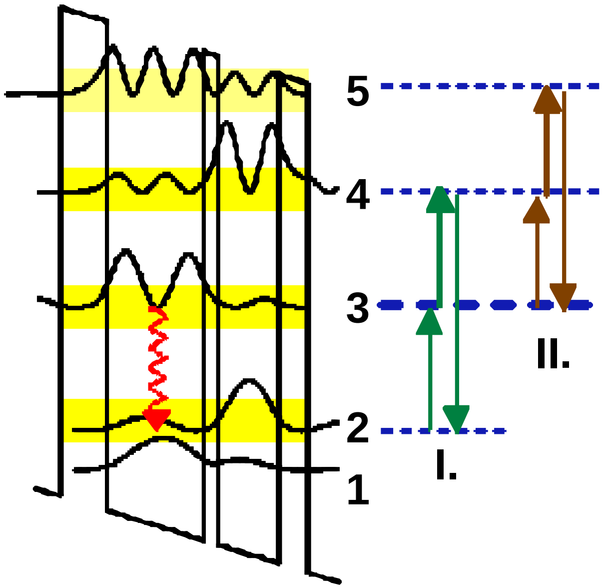
$$\left|\chi^{(2)}\right| \sim \frac{\boxed{N_e} d_{12} d_{13} d_{23}}{\hbar^2 (\gamma_{12}^2 + \Delta_{12}^2)(\gamma_{13}^2 + \Delta_{13}^2)}$$

Solution: create the nonlinear medium with gain

This leads to nonlinear quantum cascade lasers

Monolithic integration of quantum-cascade lasers with resonant optical nonlinearities





Second harmonic generation

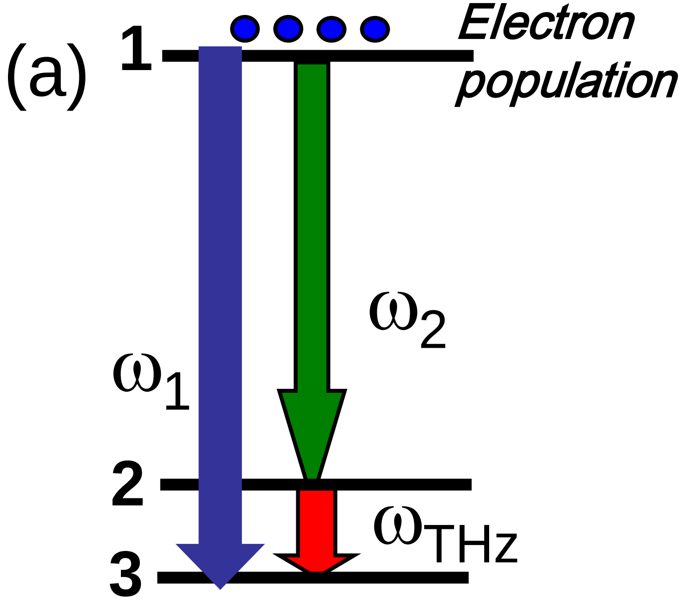
- Maximizing the product of dipoles $d_{23}d_{34}d_{24}$
- Quantum interference between cascades I and II

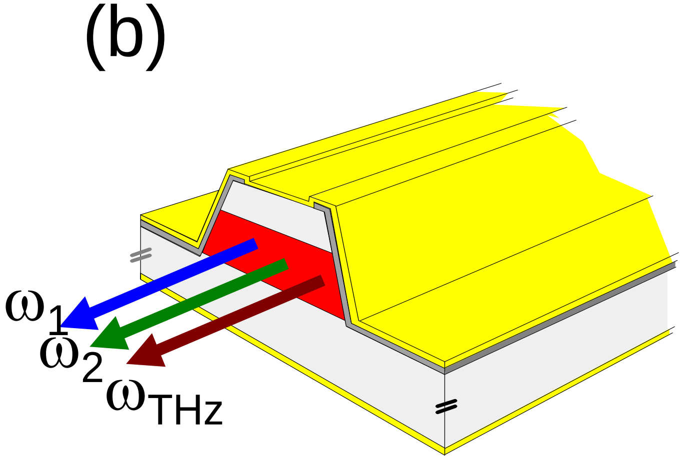
Milliwatt power in SHG:
O. Malis et al. 2004

$\chi^{(2)}$ ~ 10^5 pm/V at ~ 7-9 µm laser wavelength

This is NOT sequential photon absorption/reemission!

Difference frequency generation in two-wavelength QCLs

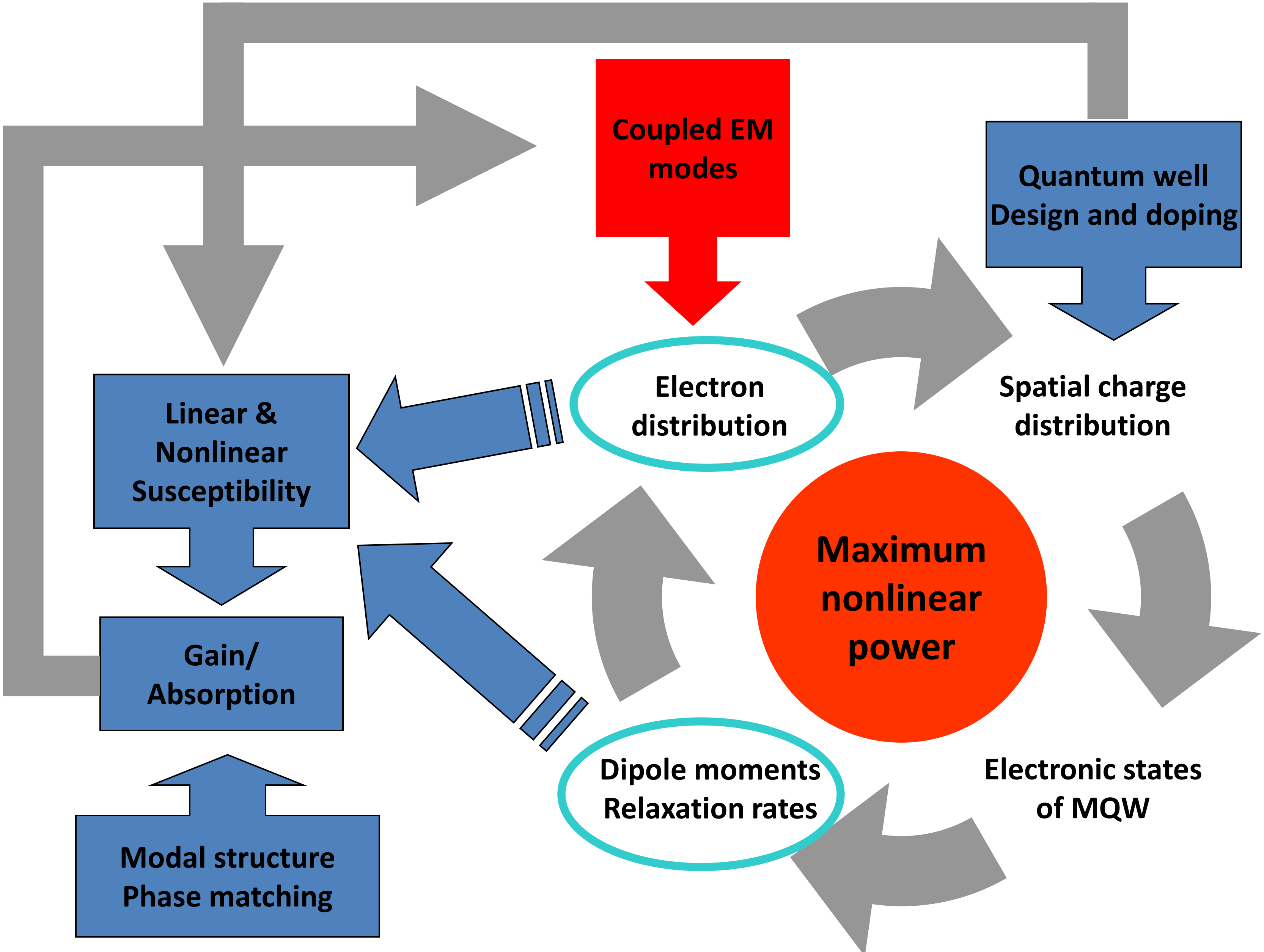




- Make a powerful mid-IR QCL emitting at two modes
- Provide strong nonlinearity for frequency mixing process
- Design a low-loss, phase-matched waveguide for all three modes

$$\omega_{THz} = \omega_1 - \omega_2, \quad k_{THz} = k_1 - k_2$$

Highly nonlinear system of interacting propagating EM fields and electrons in QWs
Requires self-consistent modeling



Why should we suffer through this?

Why should you care?

Why nonlinear optics?

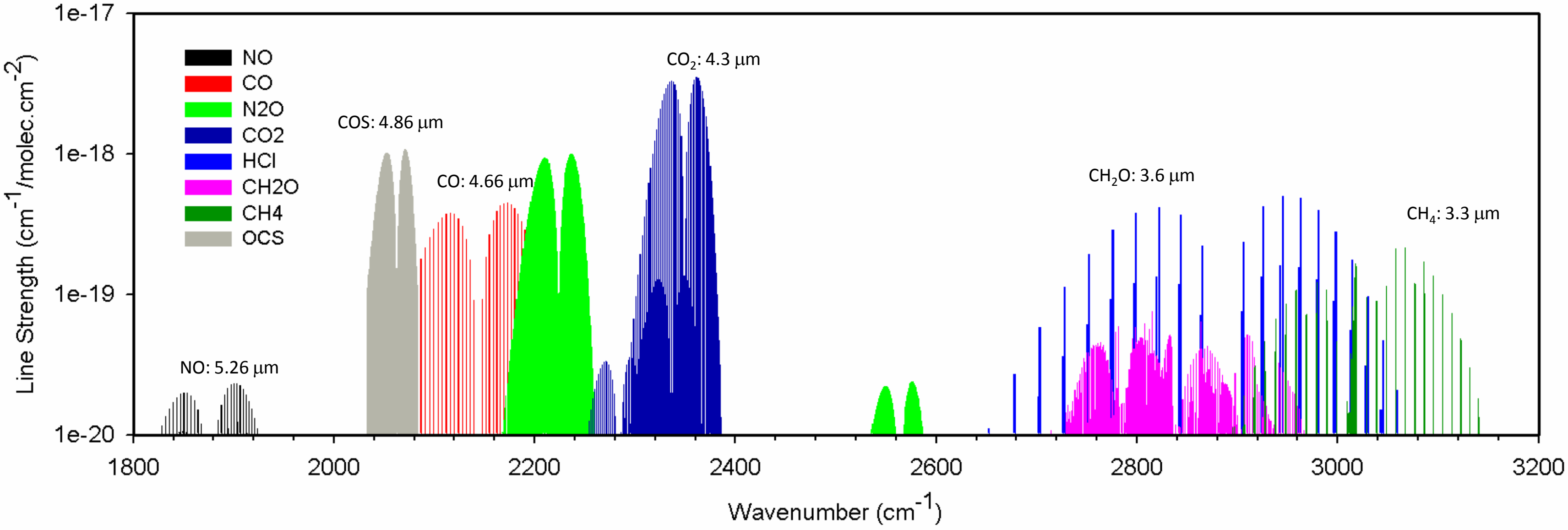
If you need to generate frequencies which you could not reach otherwise

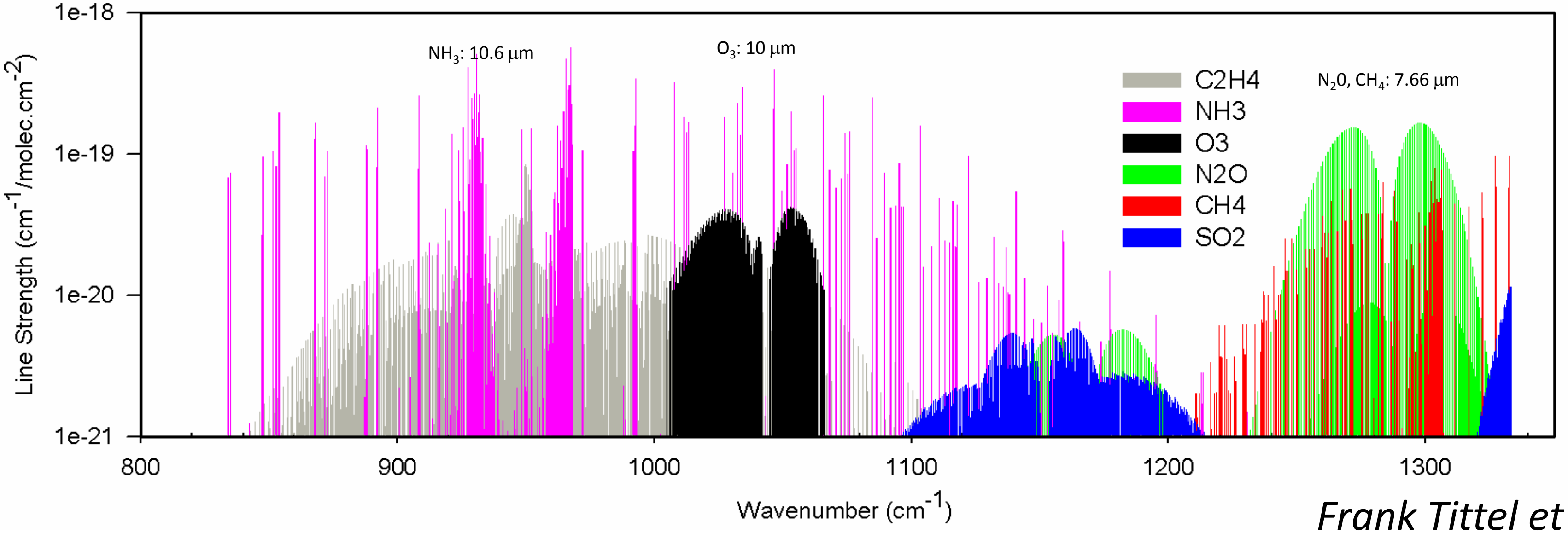
Unique functionalities: broadband tuning, ultrafast modulation, generation of ultrashort pulses, pulse shaping, phase coherence, squeezed and entangled light

Why with intersubband transitions?

- Because it is fun! Freedom of design
- Emerging applications for mid-IR and THz light

HITRAN Simulation of Absorption Spectra (3.1-5.5 & 7.6-12.5 μm)





Frank Tittel et al.

Air Pollution: Houston, TX

8/21/2000

Wide Range of Gas Sensing Applications

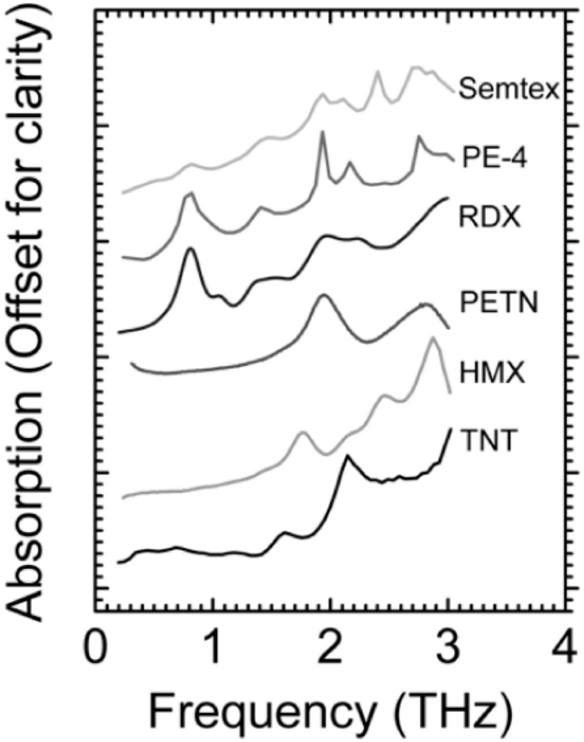
- **Urban and Industrial Emission Measurements**
  - Industrial Plants
  - Combustion Sources and Processes (e.g. early fire detection)
  - Automobile and Aircraft Emissions
- **Rural Emission Measurements**
- **Environmental Gas Monitoring**
- **Spacecraft and Planetary Surface Monitoring**
  - Crew Health Maintenance & Advanced Human Life Support Technology
- **Biomedical and Clinical Diagnostics** (e.g. non-invasive breath analysis)
- **Forensic Science and Security**
- **Fundamental Science and Photochemistry**
  - Life Sciences

Frank Tittel et al.

World Through Terahertz Glasses

$f = 1\ \text{THz} \Rightarrow E = 4\ \text{meV} \Rightarrow \lambda = 300\ \mu\text{m}$

ThruVision Ltd.



Teraview

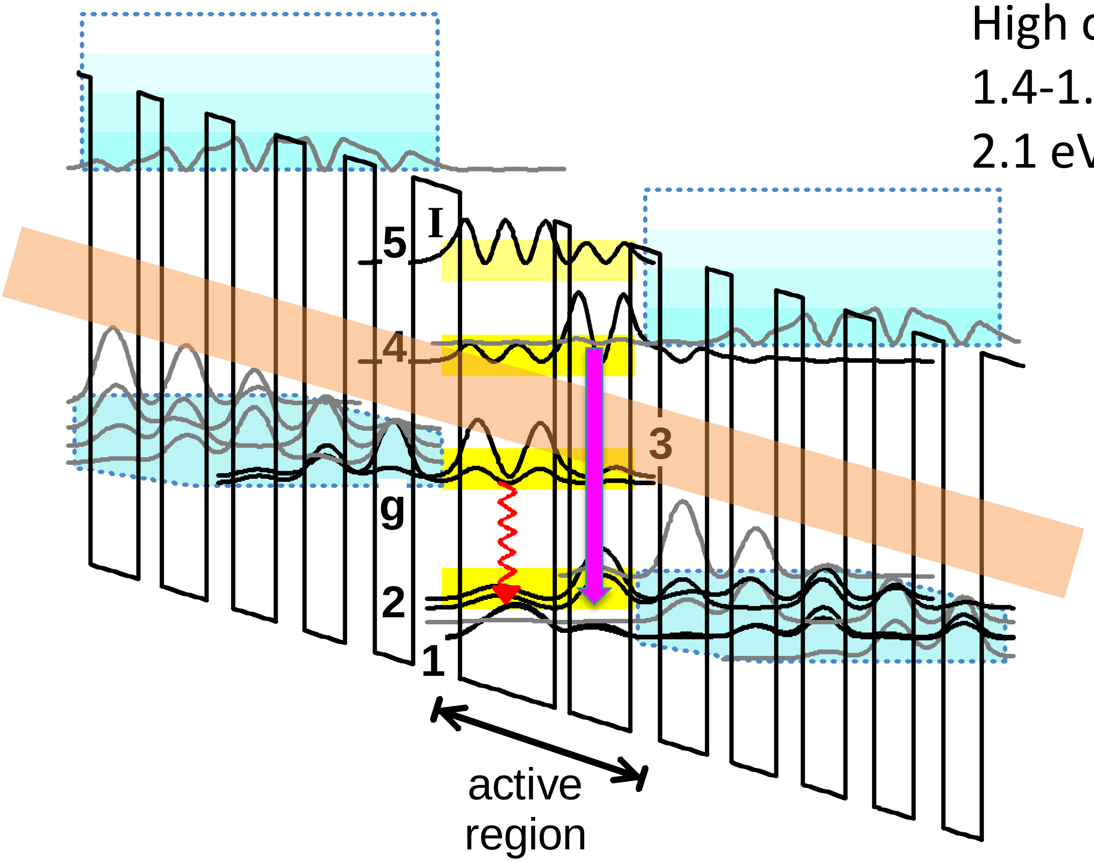
- THz sees through dry opaque cover
- Unique THz spectra of explosives, biomolecules

Extending operation of QCLs into the near-IR range with second harmonic generation

Why bother?

- Direct lasing is prohibited by lateral valleys
- Frequency up-conversion from the “sweet spot” of QCLs at λ ~ 4-6 μm
- Potentially reaching telecom wavelengths at 1.5 μm
- Ultrafast (THz) modulation possible
- Detection by frequency up-conversion: sum-frequency generation $\omega_1 + \omega_2 = \omega_3$

To reach short wavelengths, one needs deep wells



High conduction band offset at Γ-point:
1.4-1.7 eV in InGaAs/AlAsSb/InP
2.1 eV in InAs/AlSb

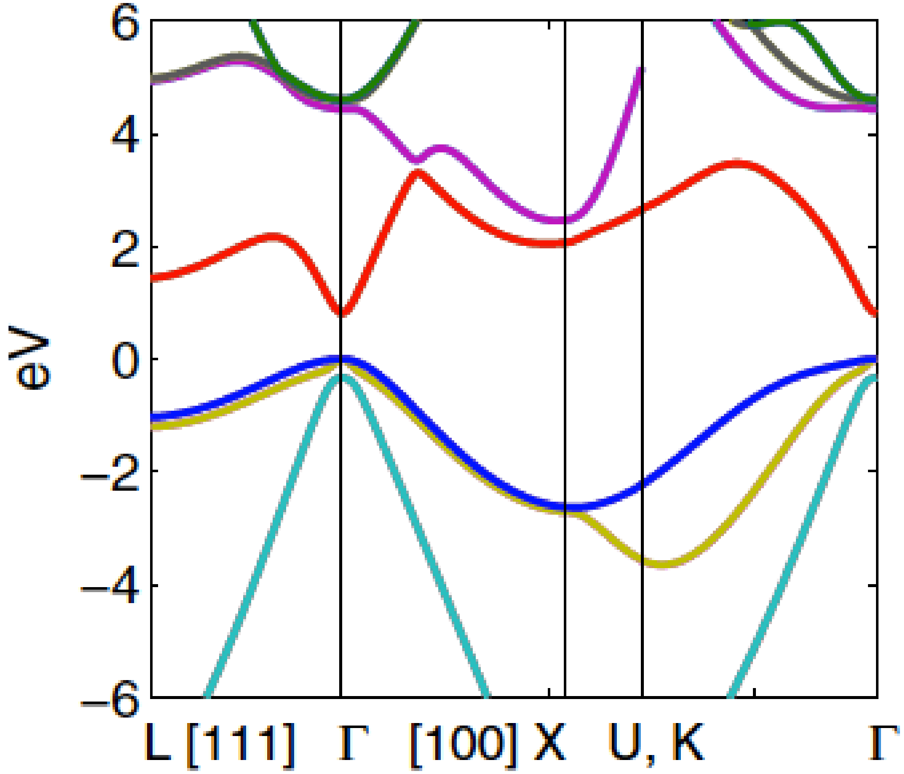
Farewell

$$m_z^*(E) = m^*(1+\alpha E)\ !!$$

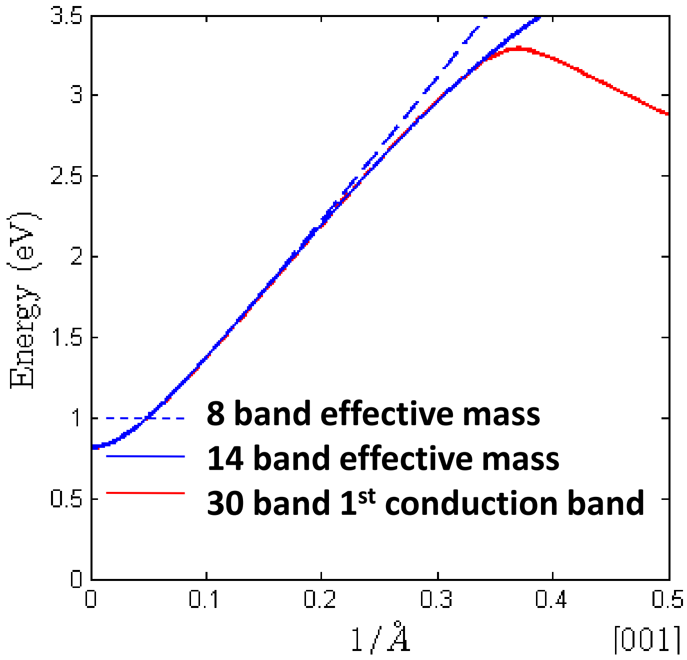
Γ-L,X distance is 0.6-0.7 eV: fatal for lasers
SHG is not sensitive to the position of lateral valleys
Calculation of highly excited subbands (> 1 eV) is a challenge

A 30 band k.p method

$Ga_{0.47}In_{0.53}As$



Comparison of conduction band dispersions



- **The order of conduction band minima:**
Γ – L – X (Y.Cho &, A.B. JAP 2010)
- L valley edge is the limit for laser transition due to scattering.
- **L – Γ : 0.65 eV**
- Vurgaftman et al, JAP 2001 : 0.5 eV

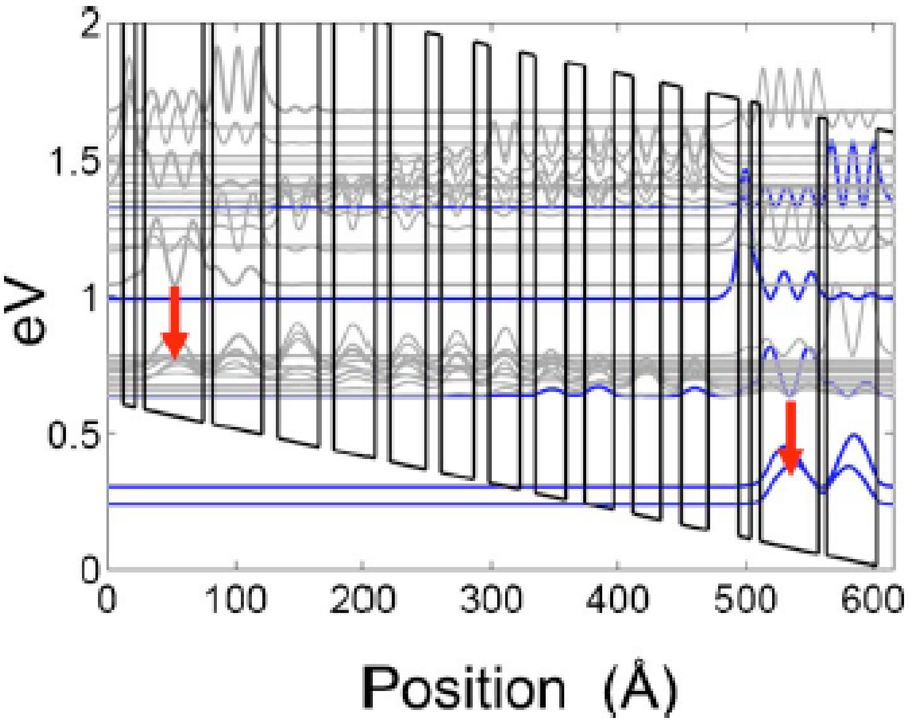
To predict the position of highly excited subbands, one needs to include at least 14 bands

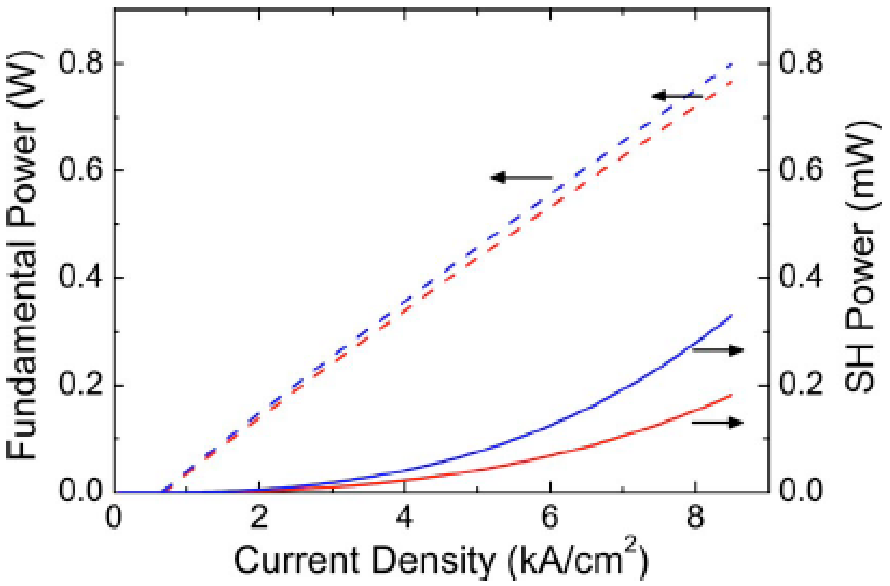
$$\frac{1}{m^*_{14band}(E)} = \frac{E_p}{3}\left(\frac{2}{E_{\Gamma g}+E} + \frac{1}{E_{\Gamma g}+\Delta_0+E}\right) - \frac{E_{P_1}}{3}\left(\frac{1}{E_1-E} + \frac{2}{E_1+\Delta_1-E}\right)$$

Predicted SHG efficiency in a modal phase-matched device: ~ 1 mW/W^2

Solving self-consistent Shroedinger + Poisson + density matrix + Maxwell equations for laser and SH fields

Including saturation, carrier distribution, LO phonon scattering, space charge





Cho & A.B. JAP 2010

Playing with multiple resonances

Second-harmonic, third harmonic, and sum-frequency generation:
PRL ‘03, APL’04, Opt. Lett.’04, JMO 2008, JAP’10; ongoing (with Cockburn)

Raman laser
Mid-infrared: Nature’05, APL’06
Terahertz: ongoing (with Belkin)

DFG Terahertz generation:
Nature Photonics’07, APL’08, JSTQE’09; ongoing (with Belkin)

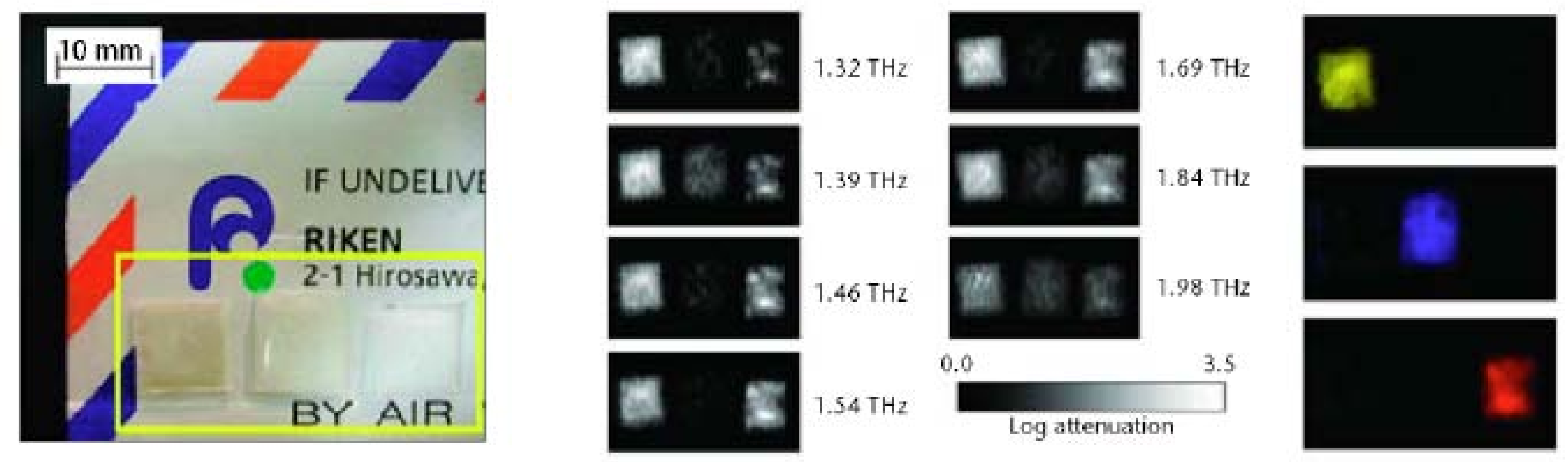
THz in magnetic field, in graphene and CNTs:
Nature Physics 2010; ongoing (with Kono)

Collaboration with M. Belkin (UT-Austin), F. Capasso (Harvard), C. Gmachl (Princeton), J. Kono (Rice), J. Cockburn (Sheffield)

THz spectroscopy and imaging

T-rays allow you to see through any dry optically opaque cover: envelope, clothing, suitcase etc, and locate non-metallic things, even read letters.

T-rays have enough specificity to distinguish “big” molecules; they can be used to detect explosives, drugs, etc.



Three different drugs: MDMA (left), aspirin (center), and methamphetamine (right), have different images in T-rays

K. Kawase, OPN, October 2004

Lack of room-temperature THz semiconductor lasers

$$\hbar\omega = k_B T$$

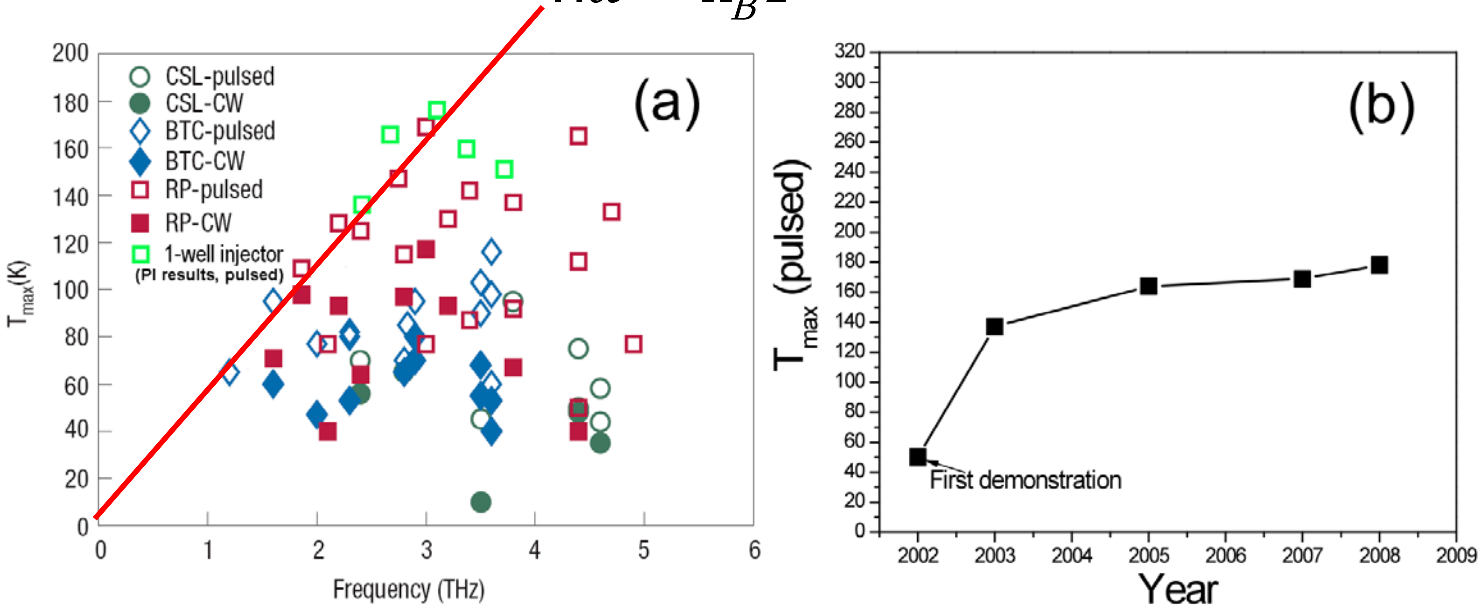
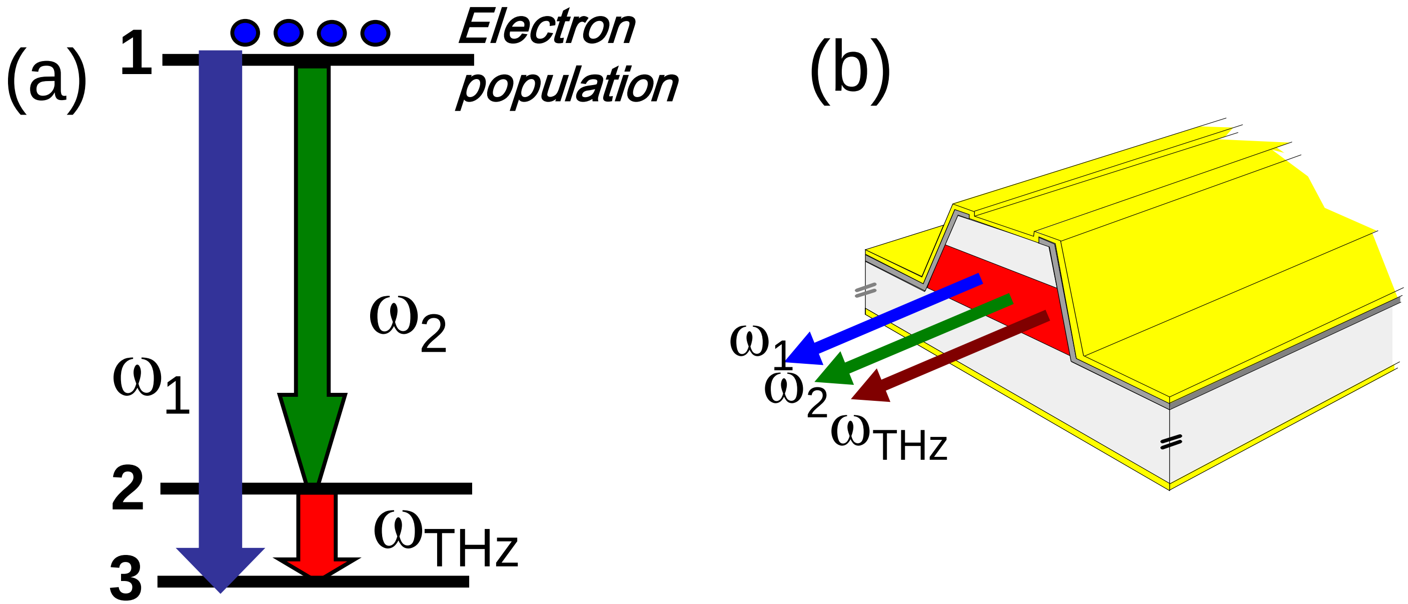


Fig. 1. (a) Maximum operating temperatures of THz QCL designs to date (without magnetic fields). Shown are the data for continuous-wave (CW) and pulsed operations for various THz QCL design concepts: chirped superlattice (CSL), bound-to-continuum (BTC), and resonant-phonon (RP). The data is taken from Ref. [8] and the recent results obtained by the PI working in collaboration with the groups in Harvard, University of Paris-Sud (France) and University of Leeds (U.K.) are added in green [9,48]. (b) The timeline of the maximum operating temperatures achieved with THz QCLs operated in pulsed mode. The points correspond to the data reported in Refs. [7,8,9,16,17]. Note that the maximum operating temperatures for continuous-wave operation of THz QCLs are typically 40-50K below that for pulsed operation.

Data up to 2009

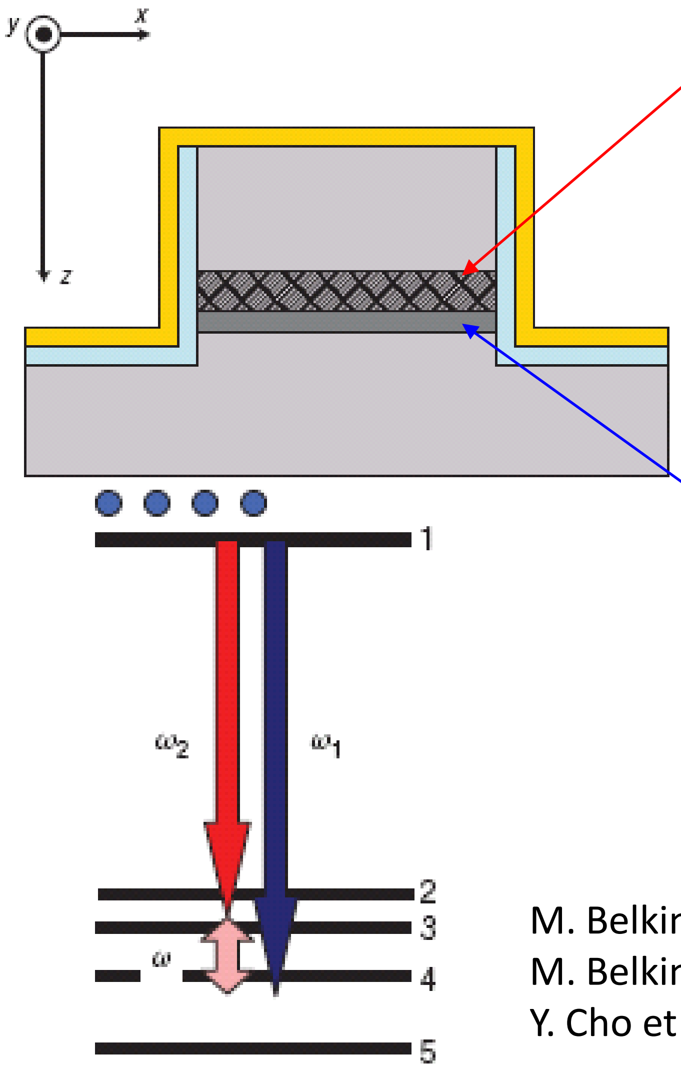
Difference frequency generation in two-wavelength QCLs

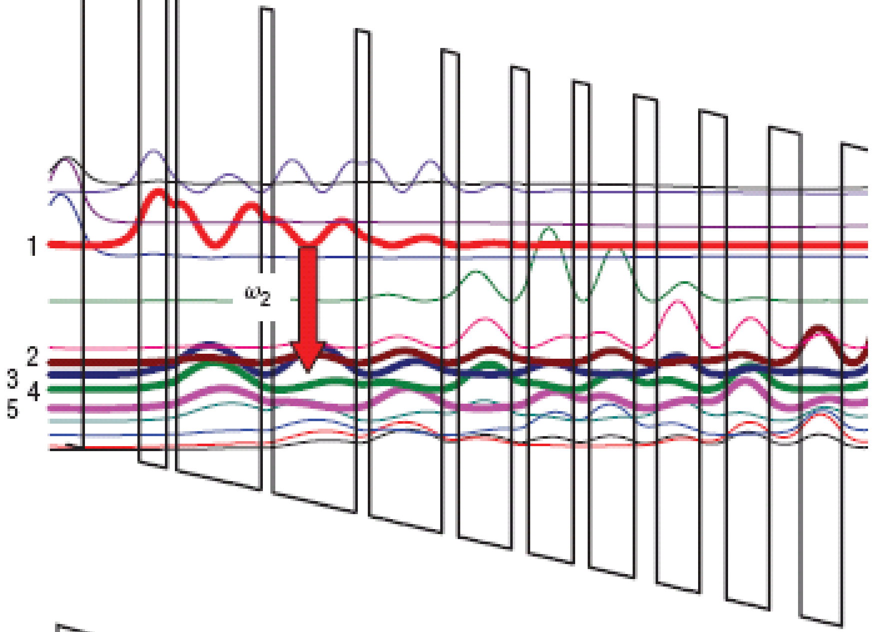


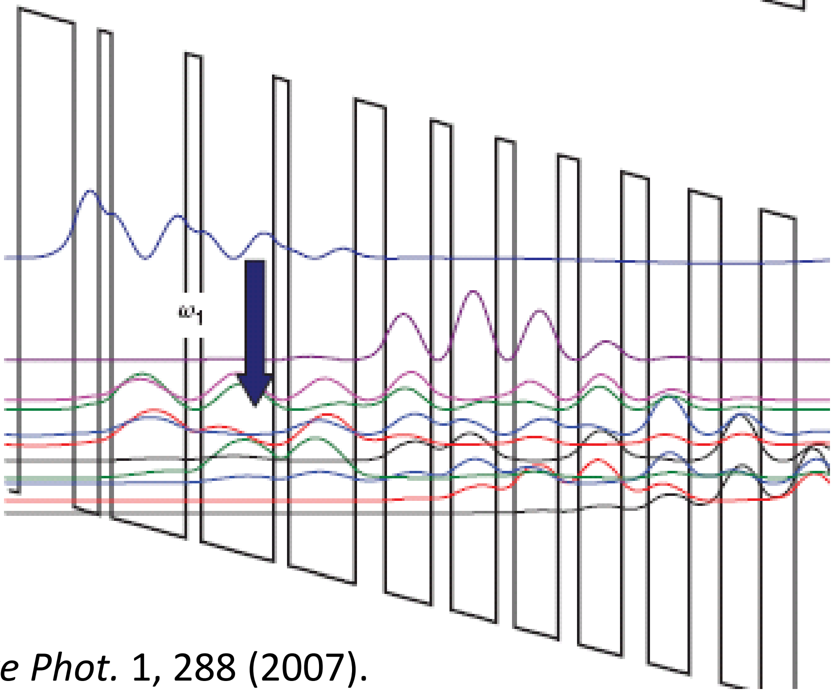
- Make a powerful mid-IR QCL emitting at two modes
- Provide strong nonlinearity for frequency mixing process
- Design a low-loss, phase-matched waveguide for all three modes

$$\omega_{THz} = \omega_1 - \omega_2, \quad k_{THz} = k_1 - k_2$$

THz generation in two-wavelength QCLs

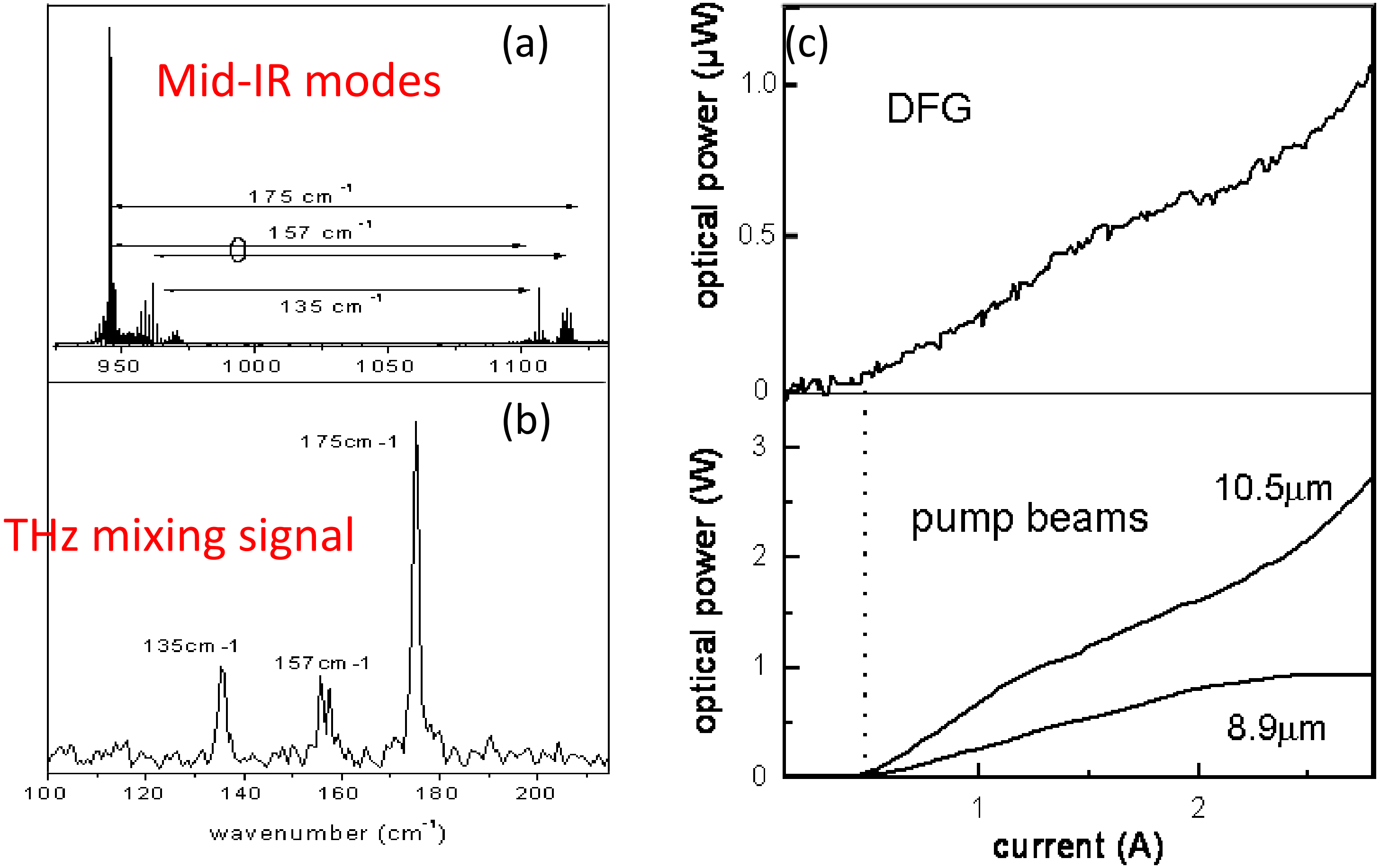






M. Belkin *et al. Nature Phot.* 1, 288 (2007).
M. Belkin et al., APL 96, 201101 (2008)
Y. Cho et al. submitted

First room-temperature THz semiconductor laser

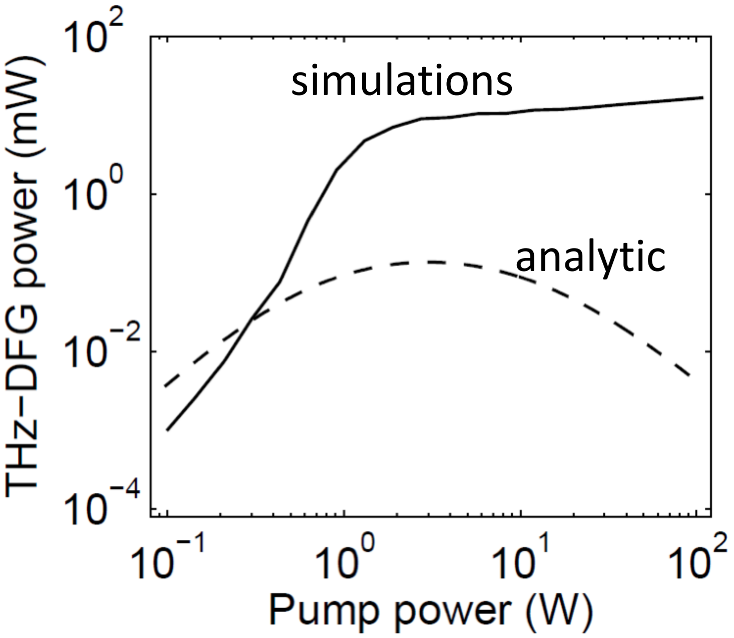


Mid-IR modes

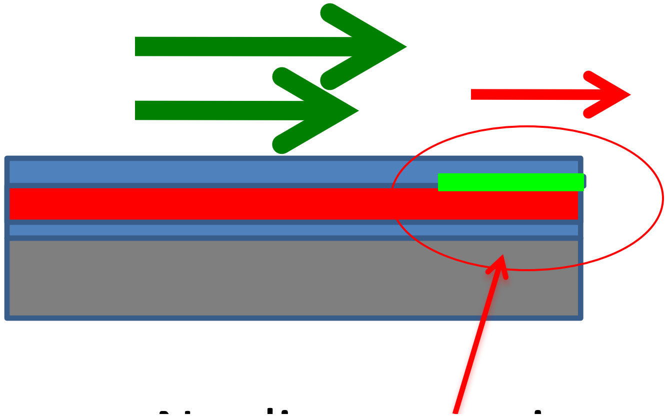
THz mixing signal

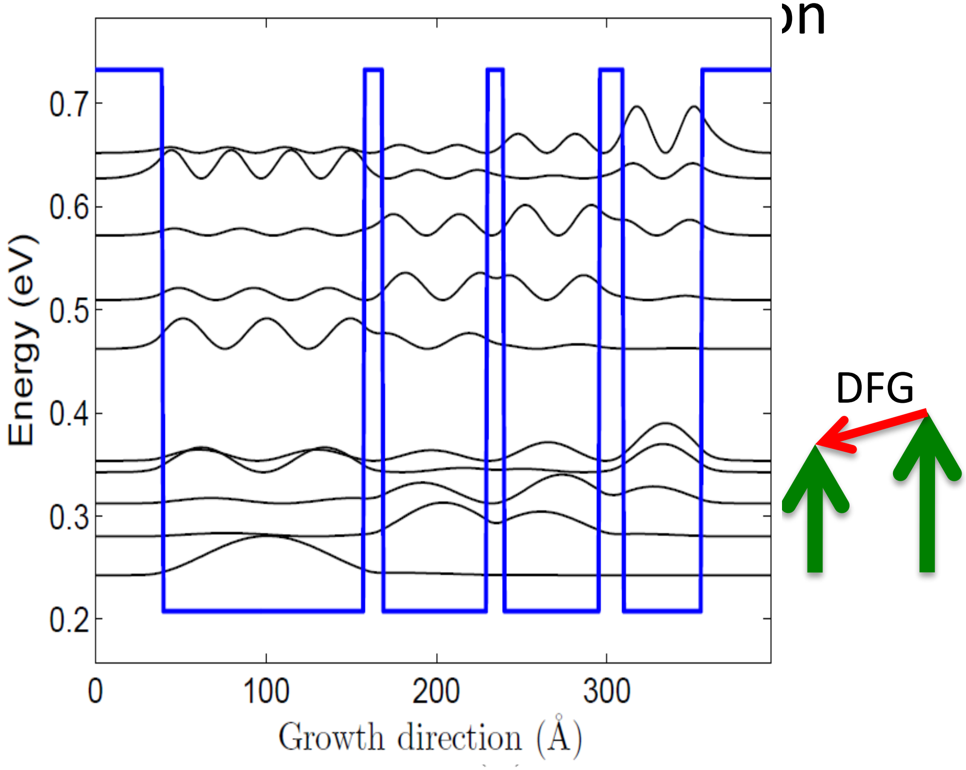
Belkin et al. APL'08

In-plane integration of laser and nonlinear section



Self-consistent simulations vs. analytic theory which neglects Raman coherences
Cho et al. 2011





THz Raman lasers

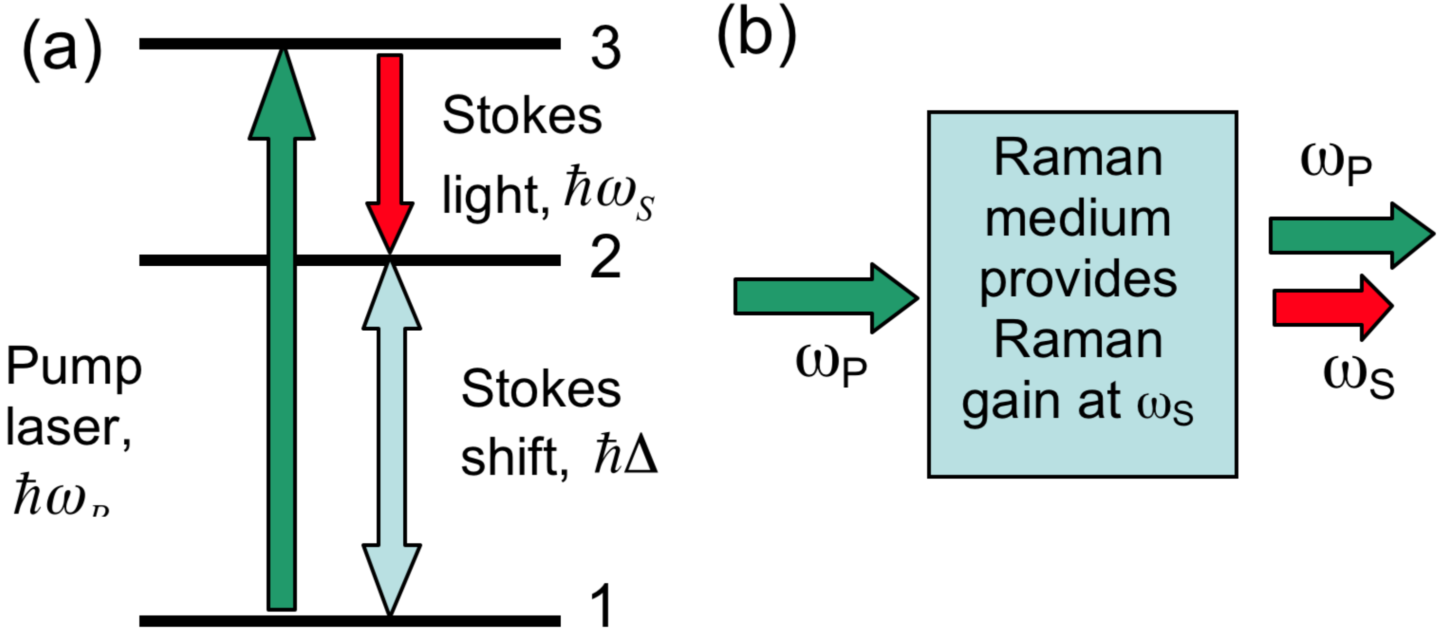
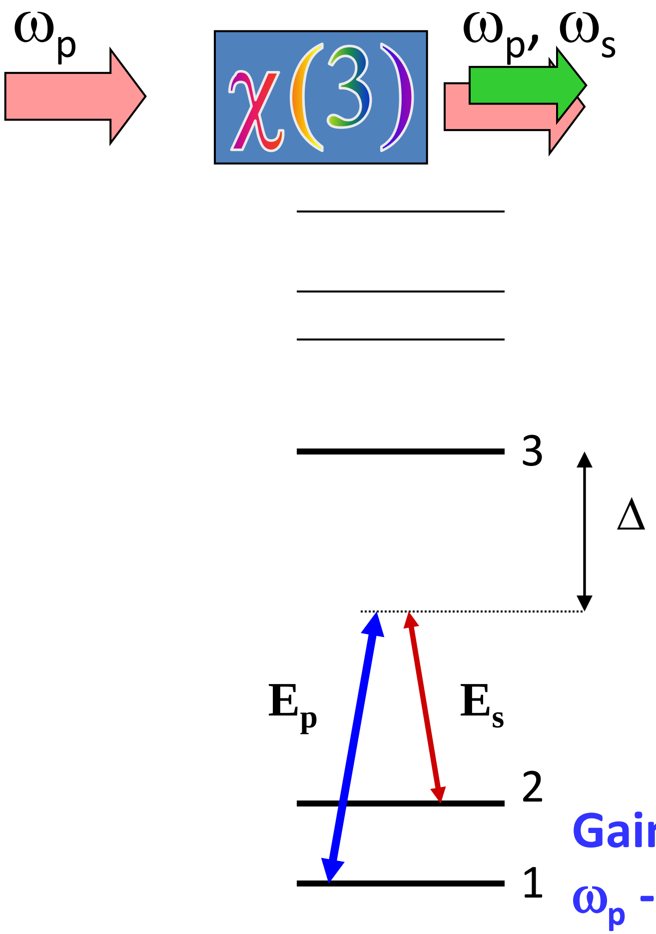


Fig. 2. Schematics of the Raman process (resonant case). (a) Band diagram showing electron transitions and relevant energy levels; (b) schematics of the Stokes wave generation: the pump wave at frequency ω_P produces Raman gain in the medium with Raman nonlinearity. The gain is maximal at frequency $\omega_S = \omega_P - \Delta$. The gain results in the generation of the Stokes wave ω_S via the laser action

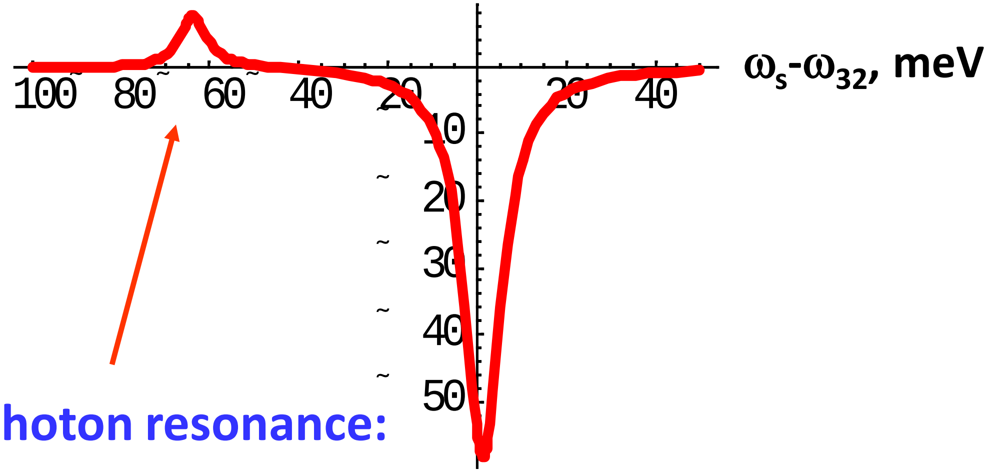
Nature 2005 (mid-IR); THz in progress

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



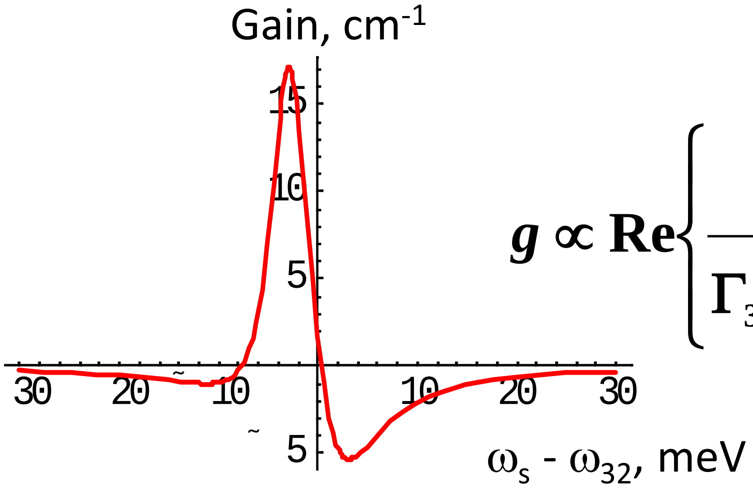
- Very large detuning Δ to avoid absorption
- No real transitions to upper state 3
- Raman shift ω_{21} is fixed to be the phonon frequency

Signal gain



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

Stokes gain at arbitrary detuning



One-photon absorption

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

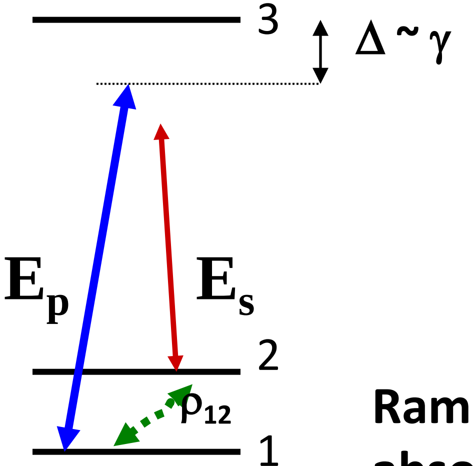
“Two-photon” gain

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

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

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

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



Rabi frequency

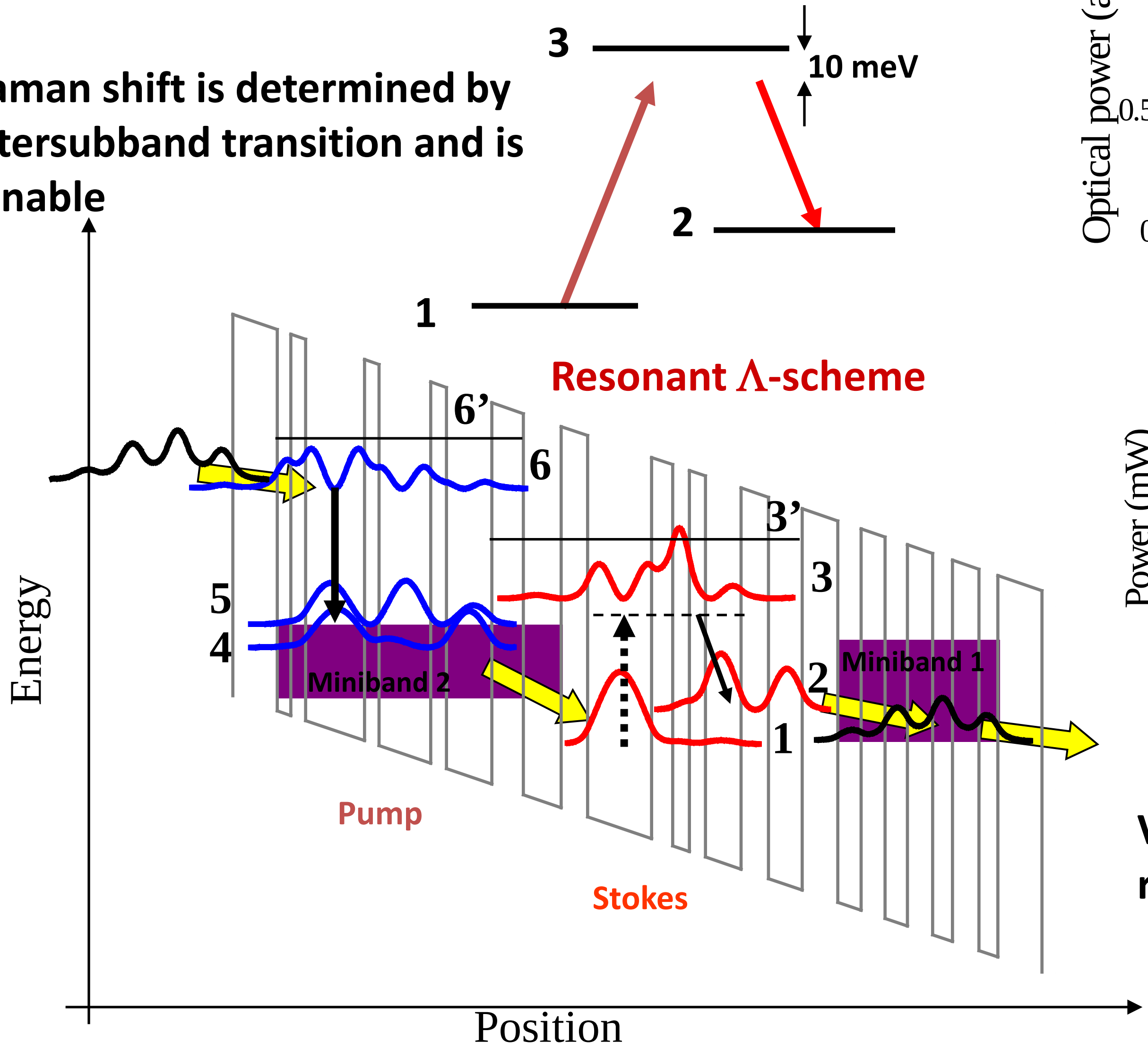
$$\Omega_p = \frac{d_{31} E_p}{\hbar}$$

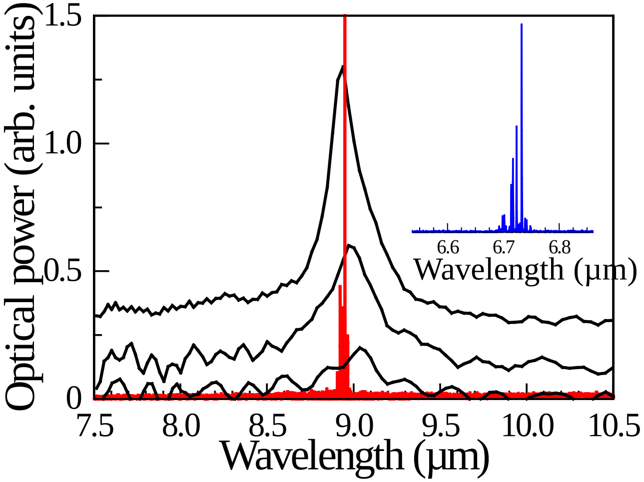
$$\Omega_s = \frac{d_{32} E_s}{\hbar}$$

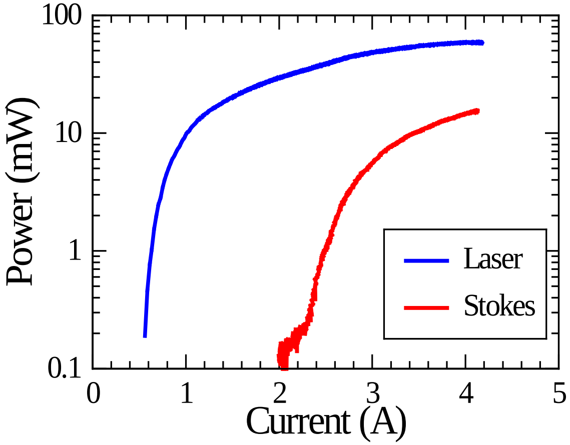
Raman gain is enhanced, but resonant absorption of the pump limits the interaction length

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

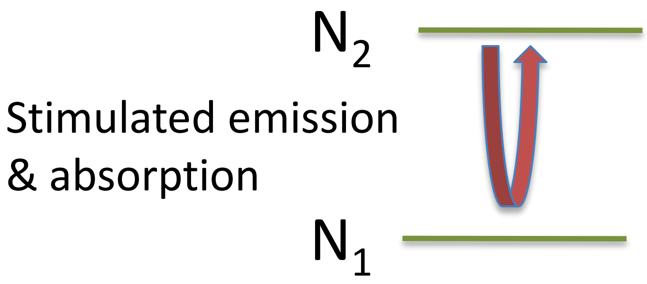
Nature 433, 845 (2005)

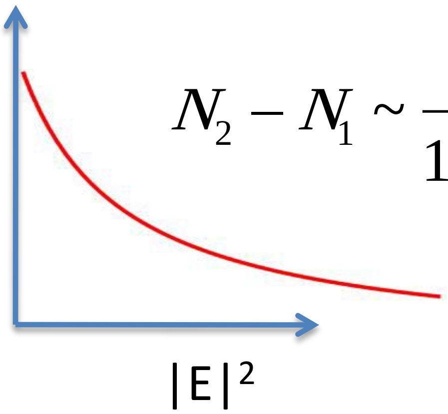
Conclusions for this part

- “Extreme” frequency conversion to near-infrared or THz seems feasible
- Nonlinear signal power is lower than for direct lasing, but there are unique benefits:
  - Room-temperature THz operation
  - Reaching over the lateral valley cutoff wavelength
  - Ultrafast modulation
  - Broader tunability (in Raman lasers)

Nonlinear interactions and phase coherence of laser modes

- QCL as a “two-level” but multimode laser
- Saturation nonlinearity and its many faces:
  - – Limits growth of laser field
  - – couples different modes, leading to mode competition, phase coupling, and mode locking

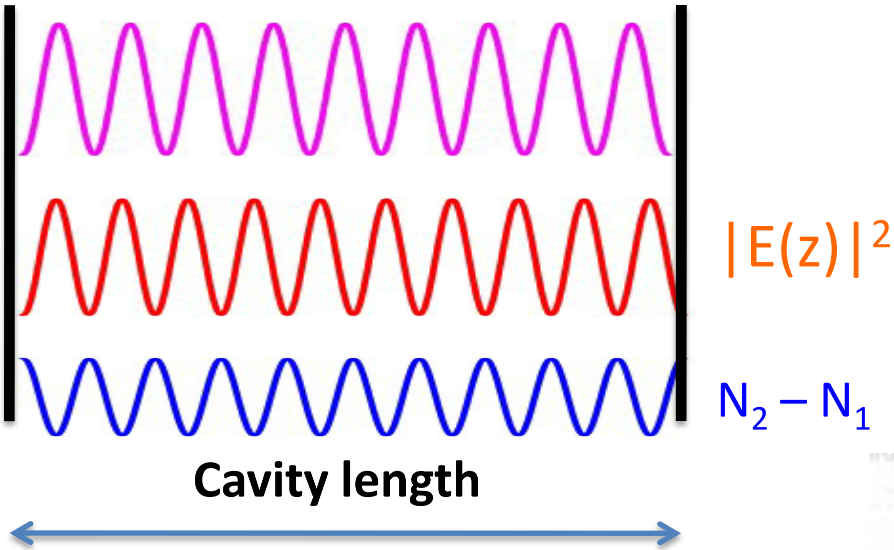




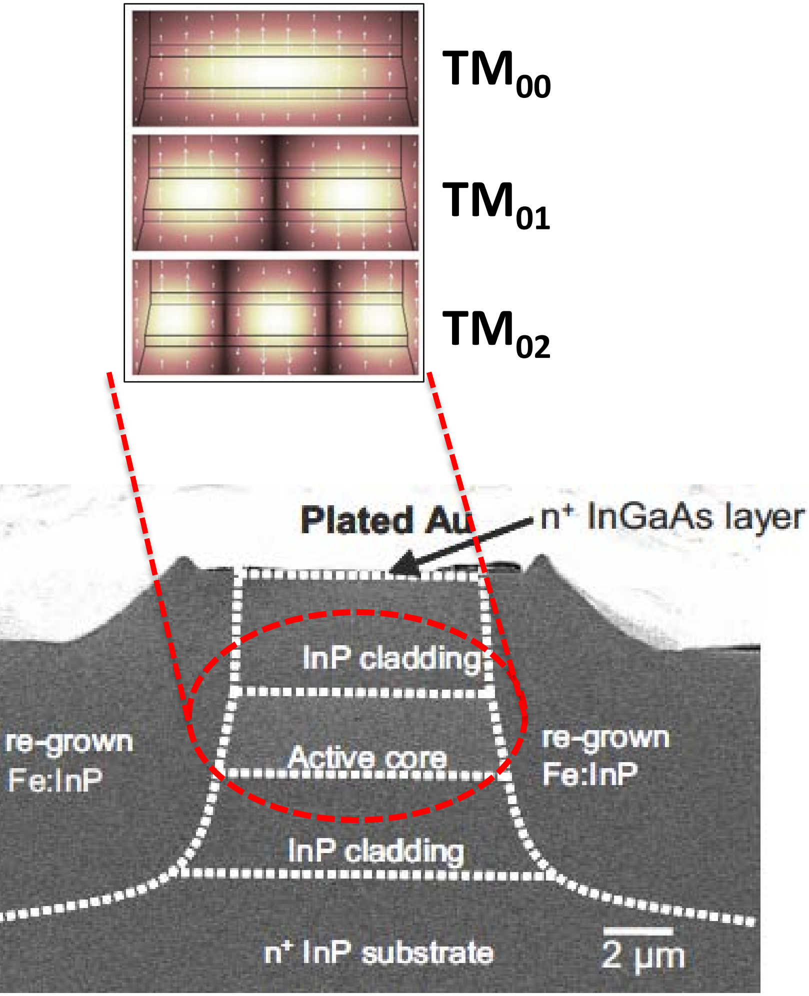
$$N_2 - N_1 \sim \frac{(N_2 - N_1)_{E=0}}{1+(1/I_s)\sum E_\mu E_\nu}$$

EM modes in QCLs: interaction through saturation nonlinearity

Longitudinal modes



Transverse Modes



Cavity cross-section

Saturation is inhomogeneous in space and in frequency (hole burning)

$$N_2 - N_1 \sim \frac{(N_2 - N_1)_{E=0}}{1+(1/I_s)\sum E_\mu E_\nu}$$

Frequency and phase locking of transverse modes

Experimental signatures:

- Anomalous near-field and far-field beam pattern; beam steering; multistability
- Locking to commensurate frequencies or synchronization of lateral modes to a single comb

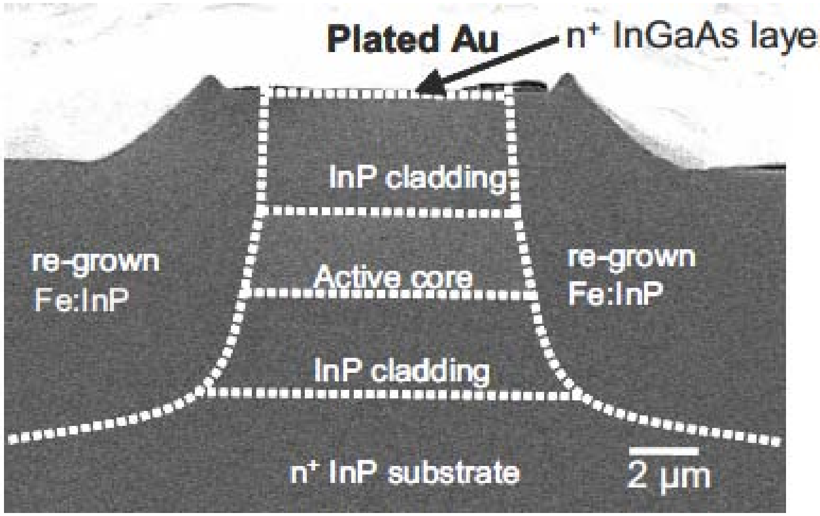
PRL 2009, OE review 2010, PRL 2011, JMO review 2011

Huge amount of research on transverse mode coherence, stationary or non-stationary pattern formation, coupled laser arrays etc.

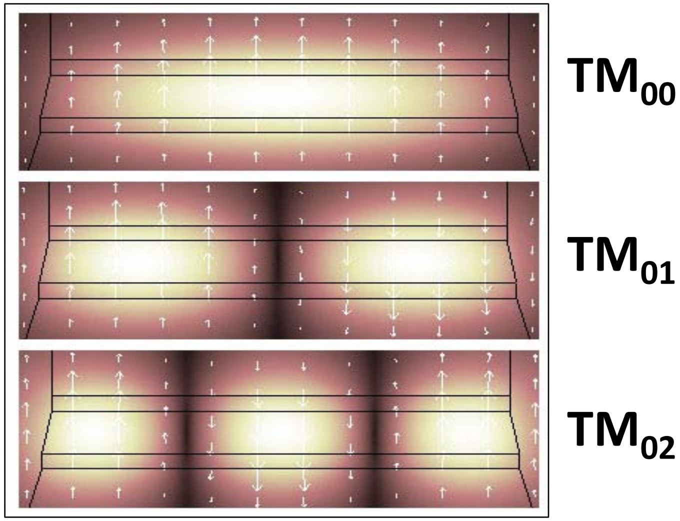
- Numerous studies in diode lasers but they have different nature of nonlinearity, different dynamical behavior
- Synchronization is achieved by periodic modulation, external optical injection or feedback
- applications in communications and optical information processing (chaos synchronization, control of pattern formation, spatial and polarization entanglement)
- Recent studies of lateral mode structure in QCLs: Gellie et al. JAP 2009 (THz), Stelmakh et al. APL 2009 and Bewley et al. JQE 2005 (mid-IR)
- Lateral mode coherence and synchronization in QCLs: Yu et al. PRL 2009, Wojcik et al. OE 2010, PRL submitted

Experiment (Capasso group, Harvard):
Multi-lateral mode regimes in buried heterostructure QCLs

7 µm wavelength, 12-24 µm active region

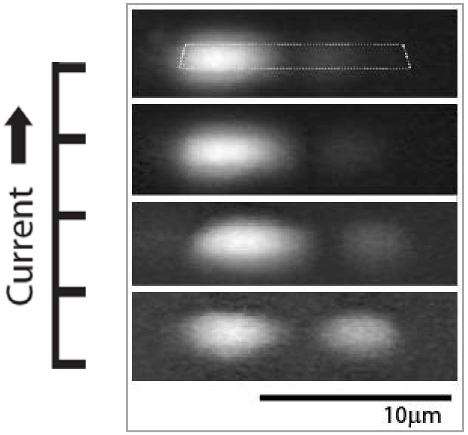


calculated modes



BH laser -> close thresholds for several modes

NSOM measurements:
Beam steering



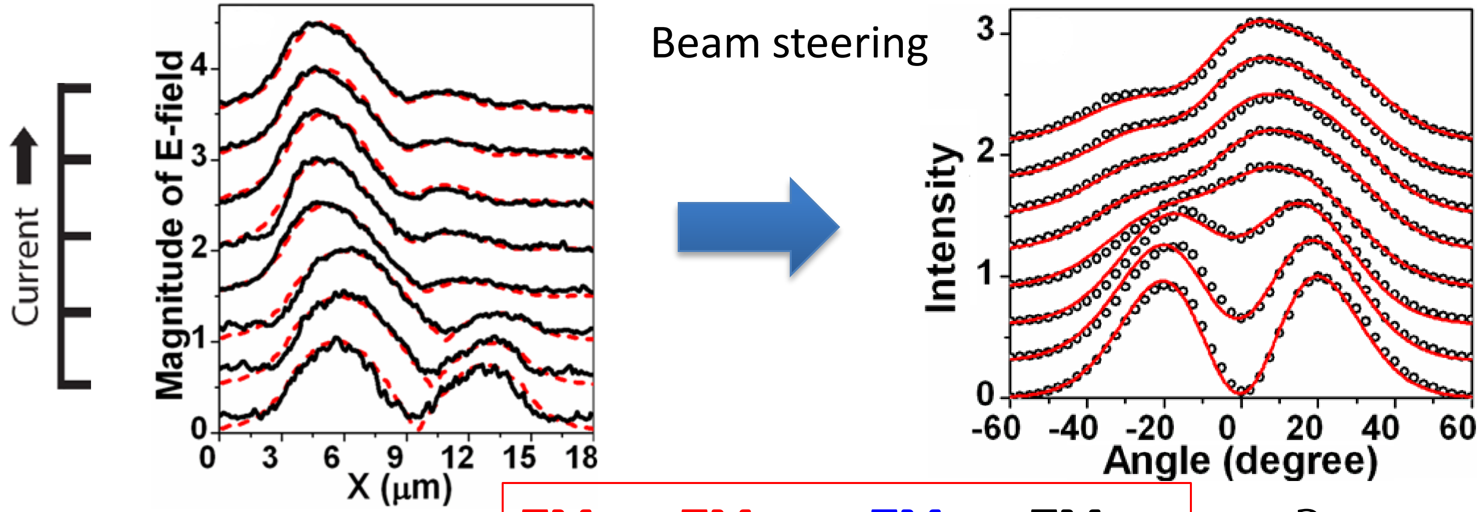
N. Yu et al., *Phys. Rev. Lett.* 102, 013901 (2009)

Evidence for phase locking between lateral modes

Near field

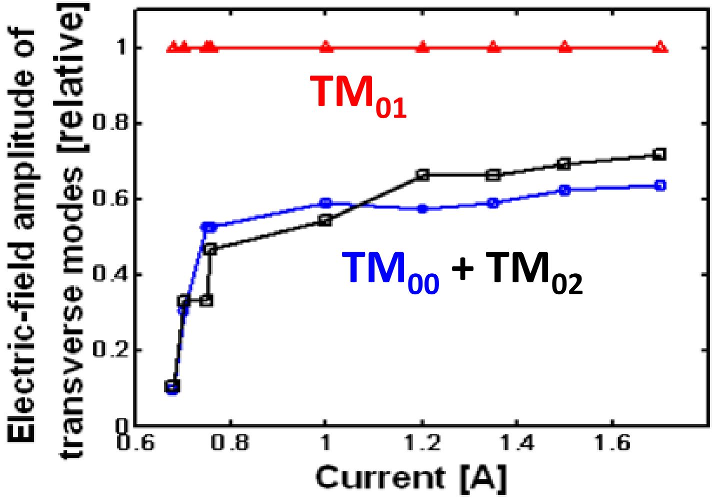
Beam steering

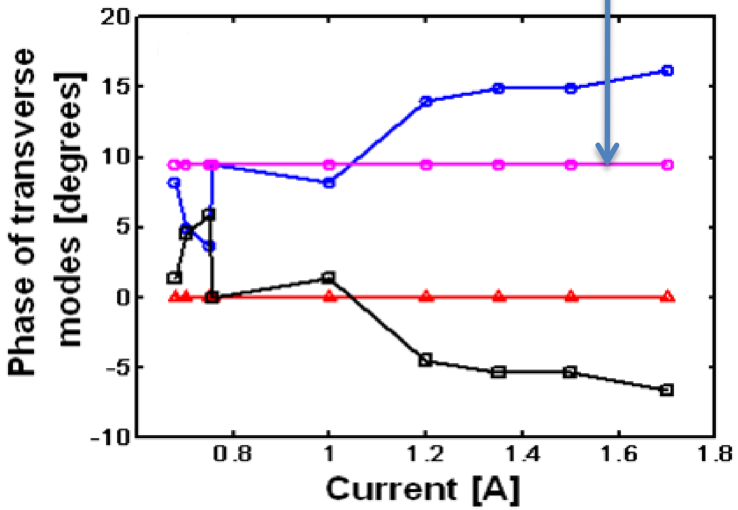
Far field



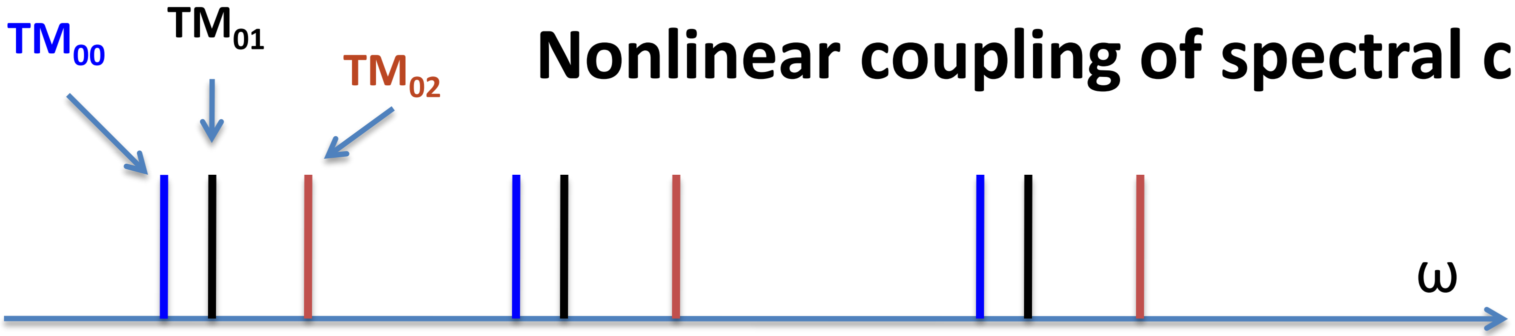
$\mathrm{TM}_{01} + \mathrm{TM}_{01} \rightarrow \mathrm{TM}_{00} + \mathrm{TM}_{02}$

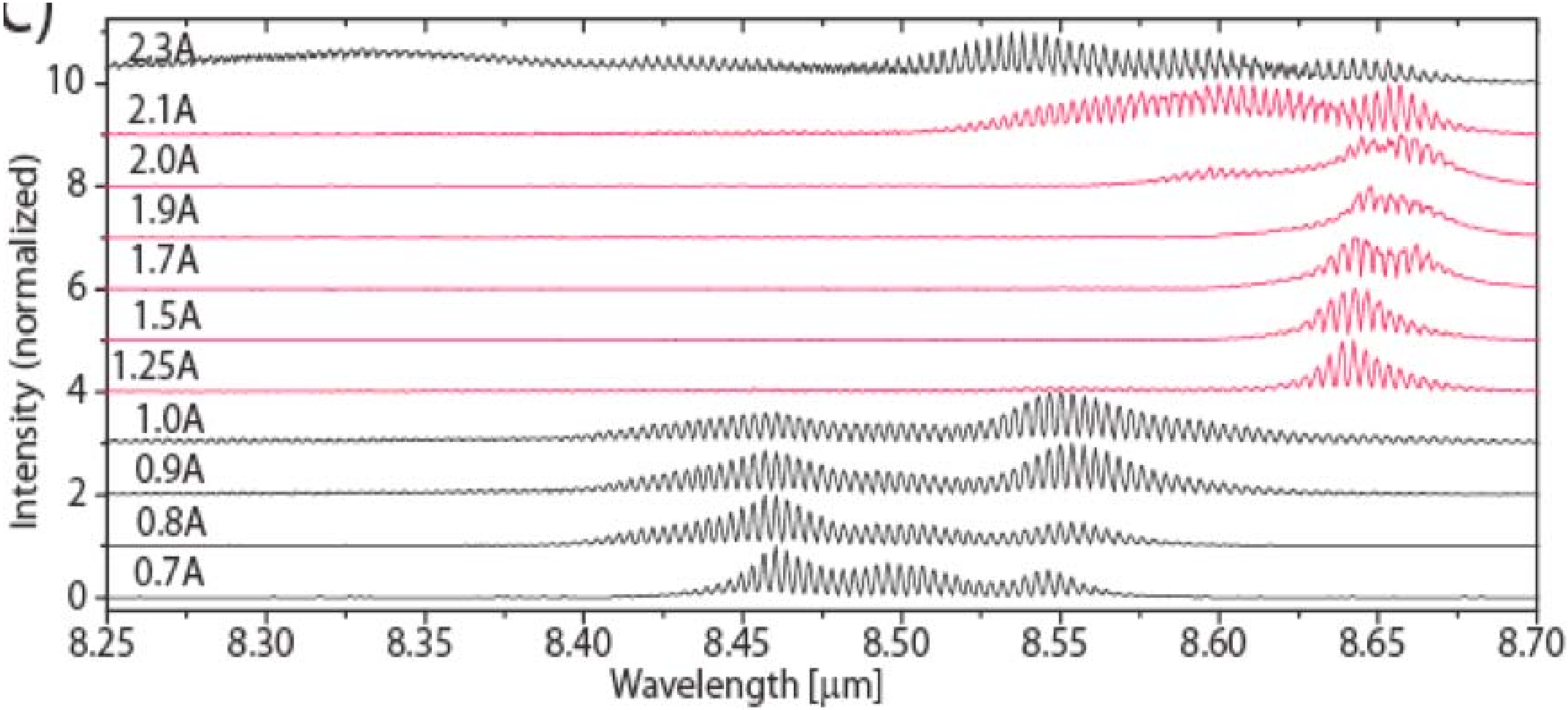
$2\varphi_{01} - \varphi_{00} - \varphi_{02}$





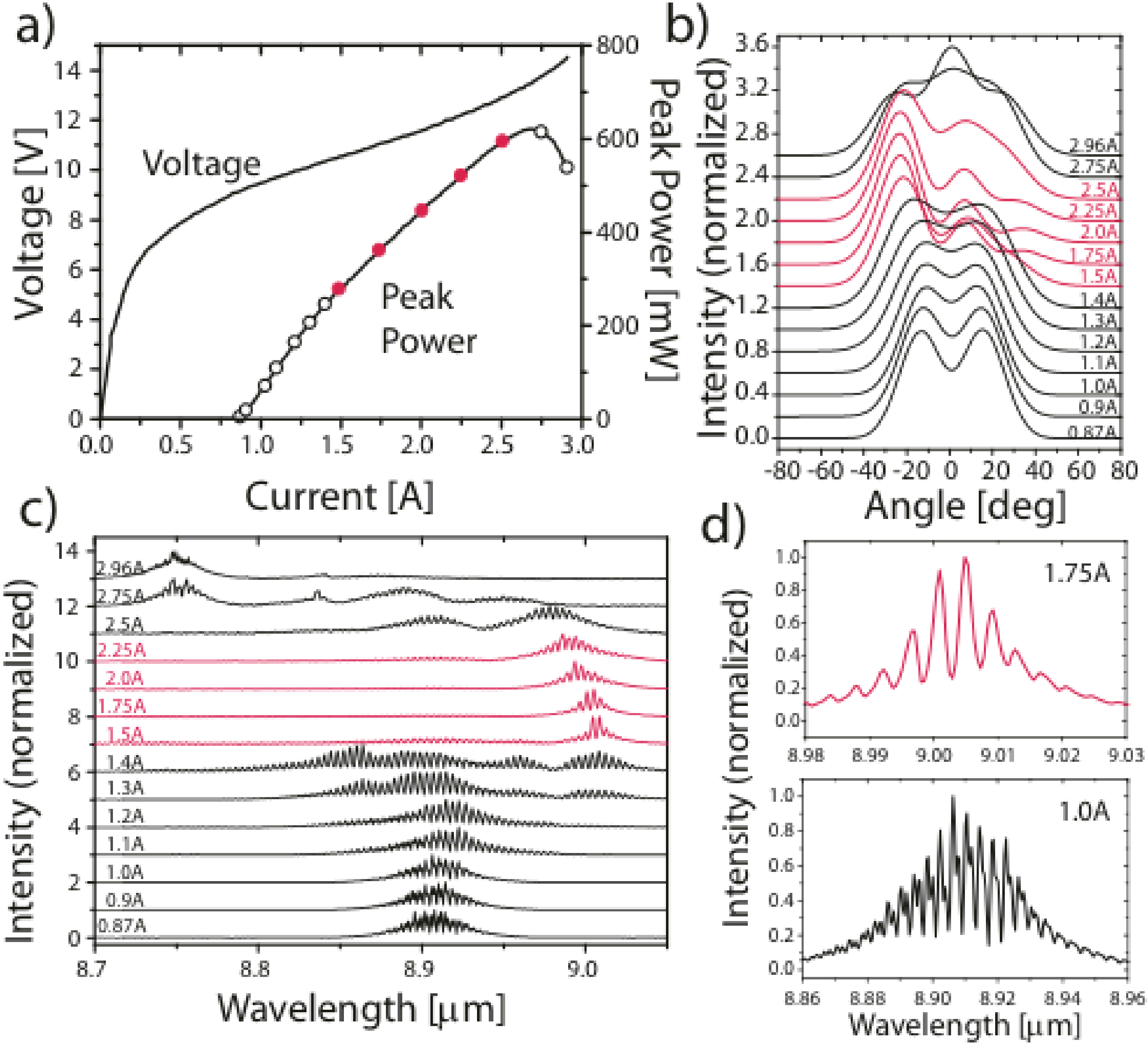
Nonlinear coupling of spectral combs





Three combs can lock into equidistant triplets or even to a single comb (synchronization)

Synchronization of modal combs



Waveguide width 19 µm

PRL 2011

Maxwell-Bloch Equations

$$\frac{d\sigma}{dt}+\gamma_{\perp}\sigma=\frac{-id}{2\hbar}D\sum_{\lambda}a_{\lambda}\boxed{E_{\lambda}(\mathbf{r})}$$

$$\frac{dD}{dt}+\gamma_{\parallel}(D-D_{p})=\frac{-id}{\hbar}\sum_{\lambda}\boxed{E_{\lambda}(\mathbf{r})}(a_{\lambda}^{*}\sigma-a_{\lambda}\sigma^{*})$$

$$\frac{da_{\lambda}}{dt}+(\kappa_{\lambda}+i\Delta_{c\lambda})a_{\lambda}=4\pi i\omega_{0}Nd\,\frac{1}{Vc}\int\sigma\boxed{E_{\lambda}(\mathbf{r})}dV$$

Field $$E(\mathbf{r},t)=\sum_{\lambda}(1/2)a_{\lambda}(t)\exp(-i\omega_{0}t)\boxed{E_{\lambda}(\mathbf{r})}+\text{c.c.}$$

Polarization $$P=Nd\sigma e^{-i\omega_{0}t}+\text{c.c.}$$

Population inversion $$D=\frac{N_{2}-N_{1}}{N}$$

"Linear" cavity modes

- Adiabatic elimination of inversion and polarization
- $X^{(3)}$ approximation

Coupled equations for modal amplitudes:

$$\frac{da_j}{dt}+\underbrace{(\alpha_j+i(\omega_{cj}-\omega_0))a_j}_{\text{Cavity dispersion/loss}}=\frac{g_j}{2}\left(\underbrace{\sum_k a_k\int_{AR}\varepsilon E_j E_k\,dV}_{\text{Modal gain}}\underbrace{-\frac{2}{I_s}\sum_{k,l,m}G_{jklm}a_k a_l^* a_m}_{\text{Nonlinear mixing}}\right)$$

Nonlinear overlap - $G_{jklm}=\int_{AR}\varepsilon E_j E_k E_l E_m\,dV$

$T_1=1/\gamma_{\parallel}$

$T_2=1/\gamma_{\perp}$

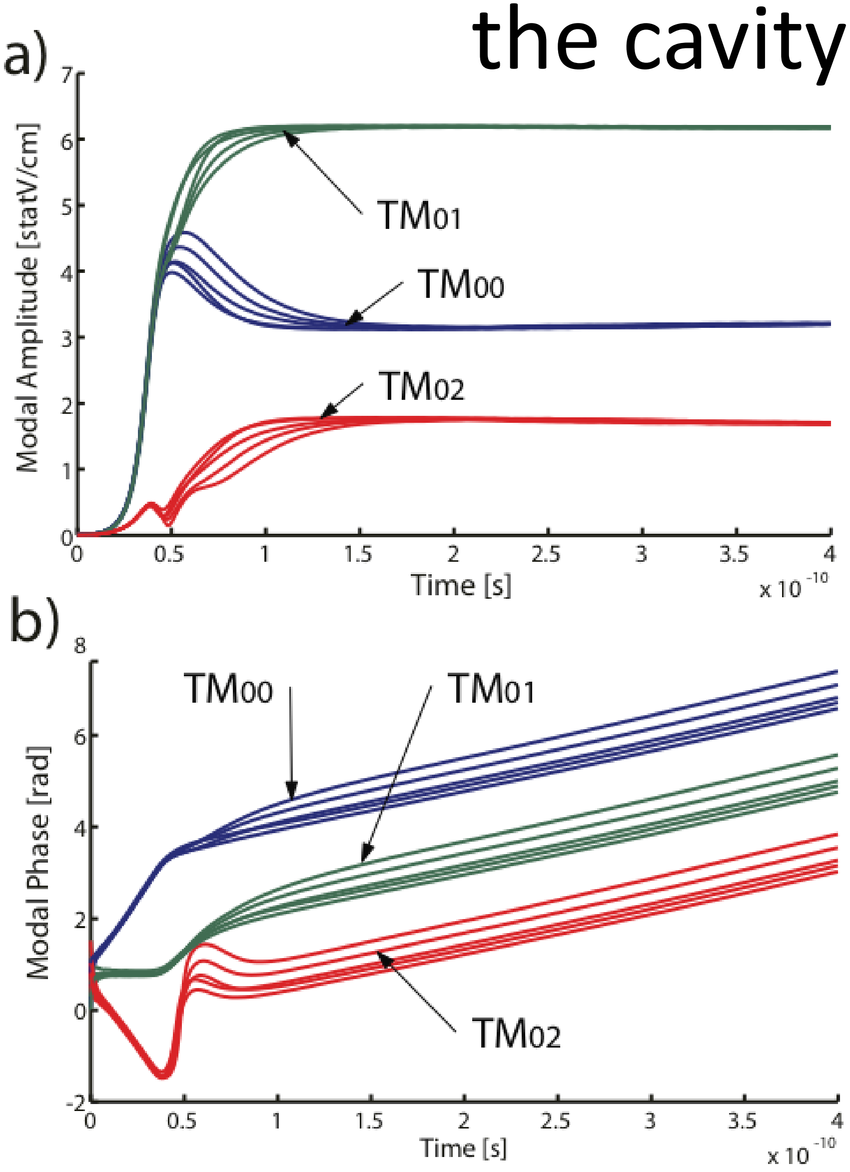
Gain - $g_j=4\pi\omega_0 d^2 N_p T_2/(\hbar\mu_j^2)$

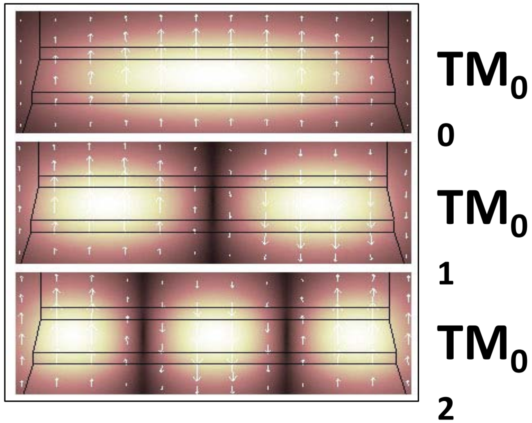
Saturation intensity - $I_s=2\hbar^2/(d^2 T_1 T_2)$

Large dipole moment gives rise to strong nonlinear coupling of laser modes

Fast gain relaxation T_1 ~ 1 ps (Type A laser) overdamps relaxation oscillations and leads to stable phase locking. No saturable absorber or external modulation!

Mean field approximation (averaging over the cavity length)





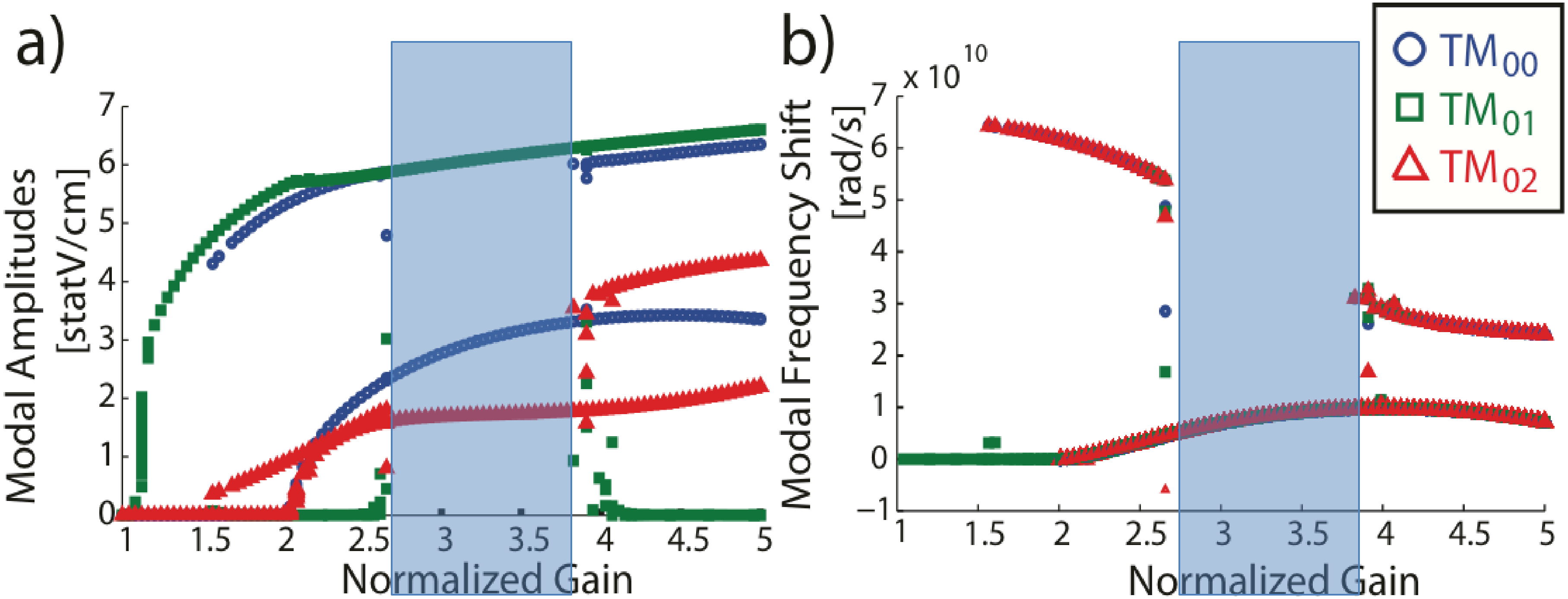
$$a_j(t) = A_j(t)\, e^{i\Phi(t)}$$

Modal amplitudes A(t) and phases Φ(t) for five different initial conditions

Locking to a single frequency

$$\Omega = \frac{d\Phi(t)}{dt}$$

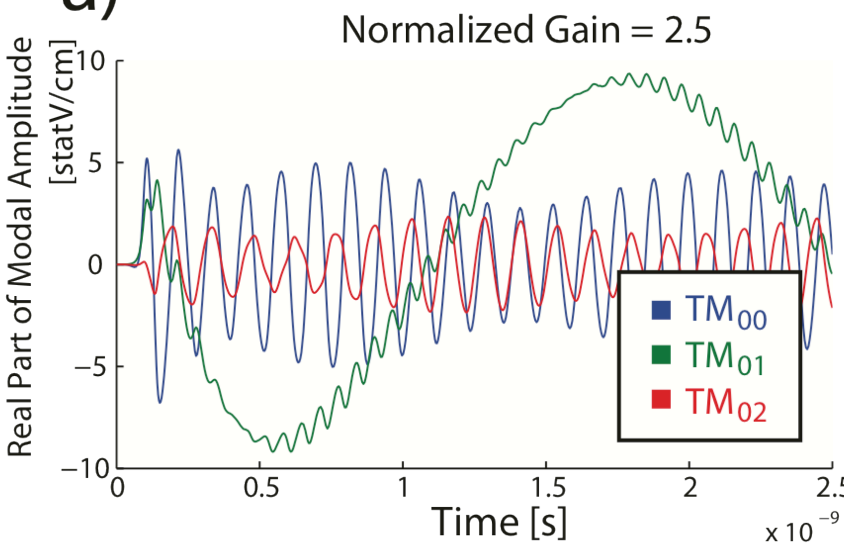
For each gain: determine all stable solutions starting from a large set of random initial phases and amplitudes



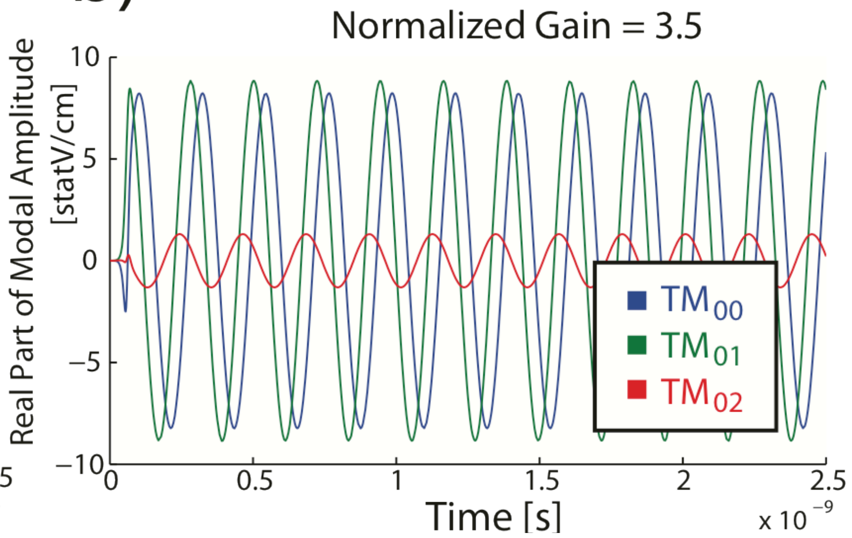
- Single stable multimode state locked to a single frequency at intermediate gains
- Bistable region outside

Dynamics of one triplet

a) **No synchronization**

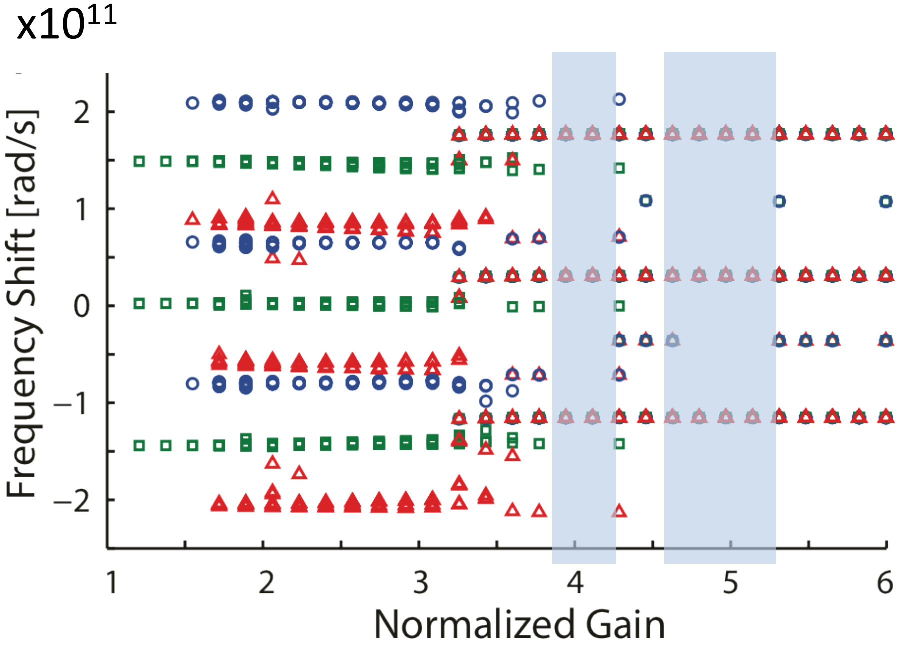


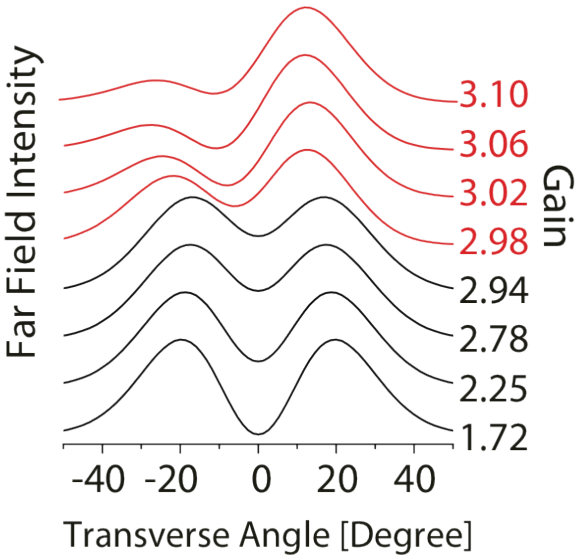
b) **gain above locking threshold**



Simulations for 60 random initial conditions per each value of gain

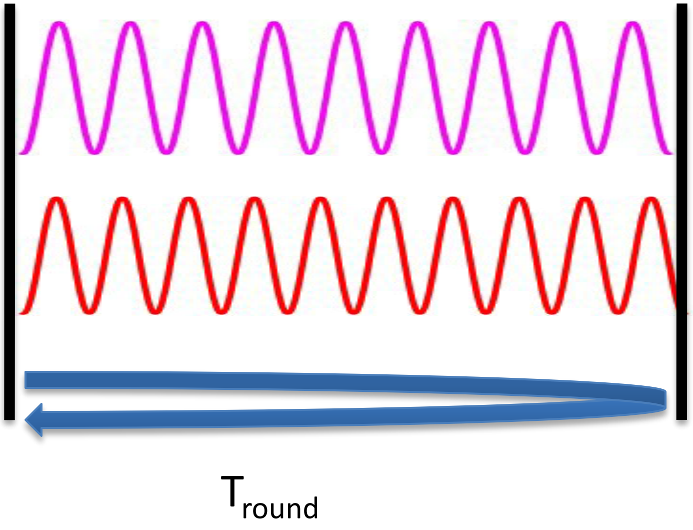
- Three lateral modes, each has three longitudinal modes
- Note islands of synchronization

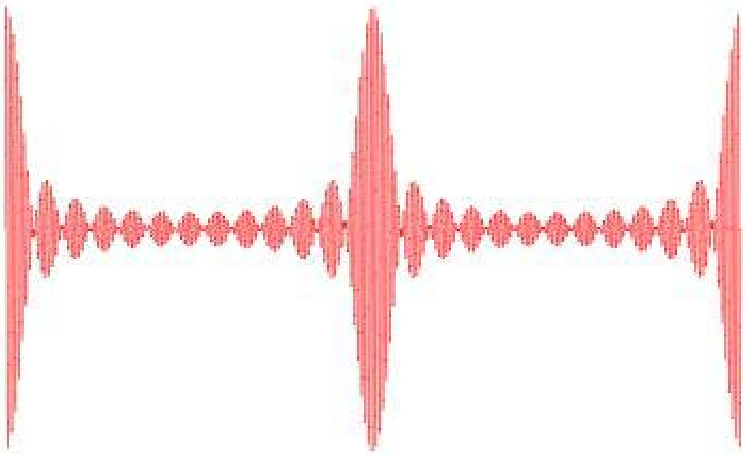



Theoretical far field

Locking longitudinal modes in QCLs

- Can we generate ultrashort pulses in the mid/far-infrared?

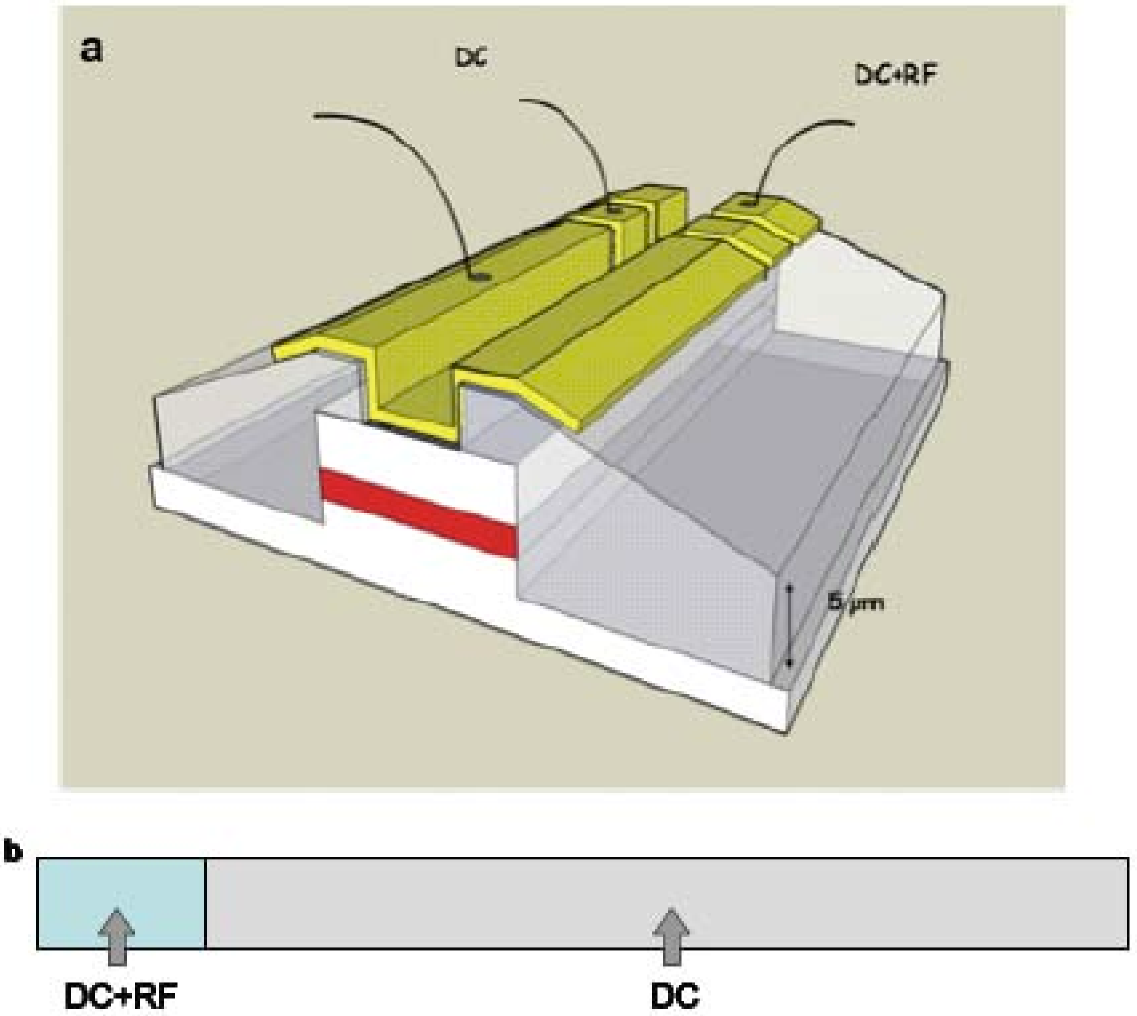




Adding 10 sines with zero initial phases

This is very difficult in QCLs where $T_1 \ll T_{round}$

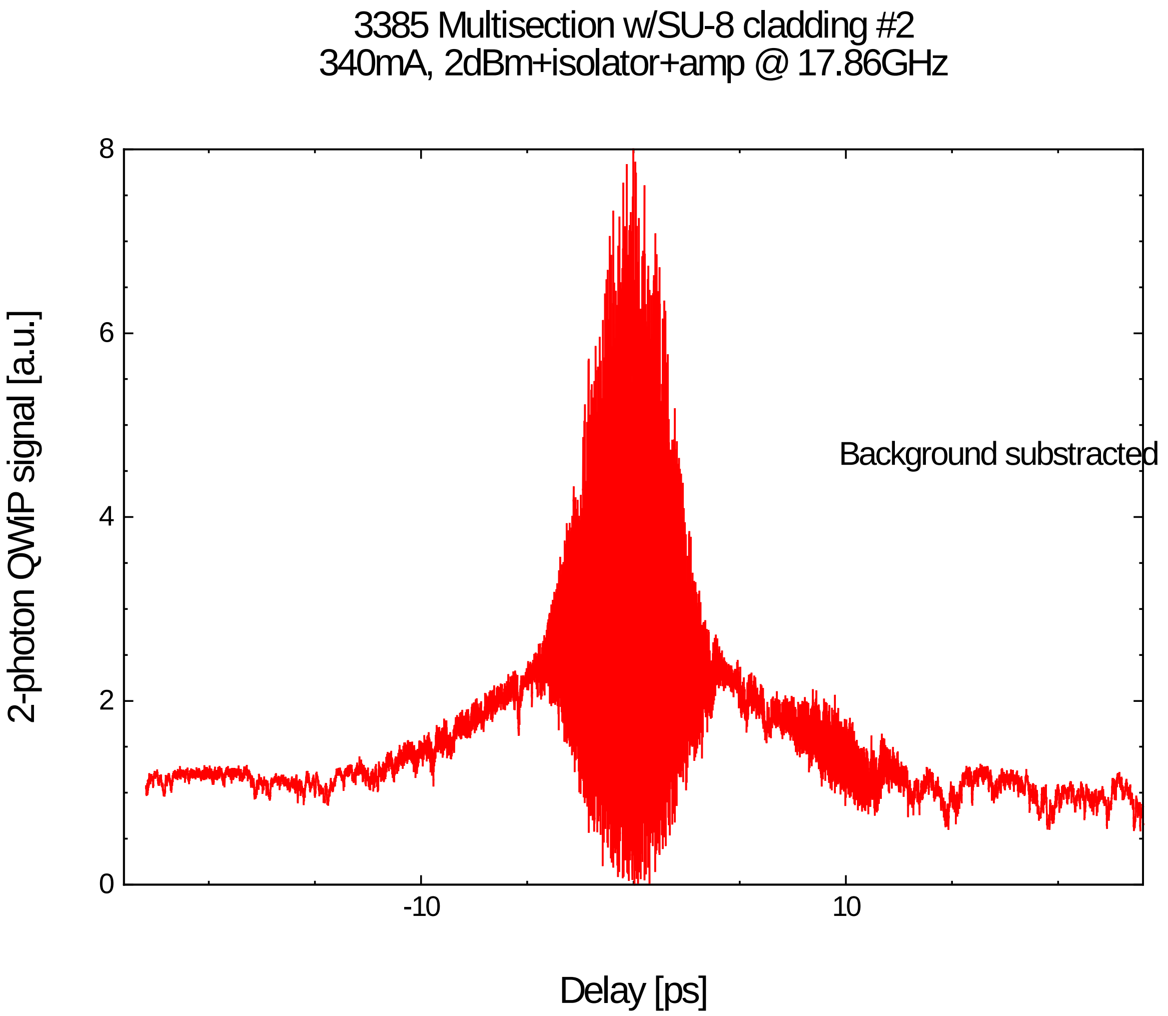
Active mode locking (Capasso group, 2009)



Gain is modulated in a short section at the round-trip frequency f = $1/T_r$

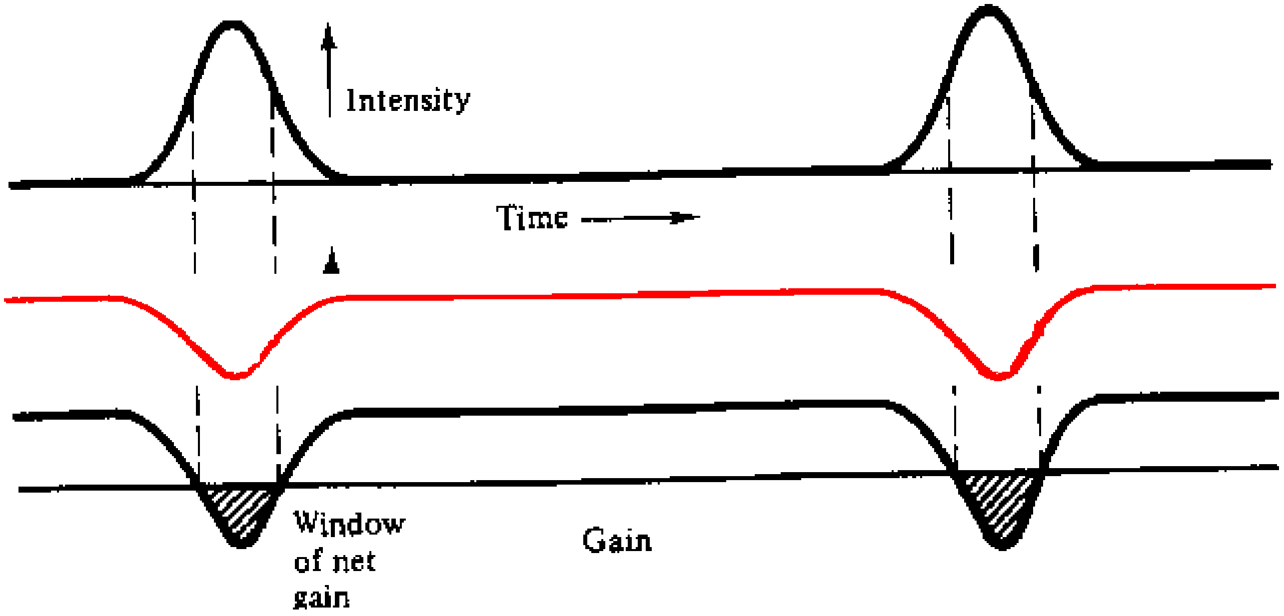
Optics Express 2009, 2010

2-Photon Autocorrelation shows 3-ps pulses



Pulse width
Estimated from
The interference
Part ~ 3ps

High-amplitude modulation is required;
Mode locking exists only very close to laser threshold



Gain = g_0

$T_1 << T_r$

In QCLs gain has a fast recovery time:

$T_1 < T_r = 2L_c/c$

$$g = \frac{g_0}{1 + \frac{I(t)}{I_s}}$$

It responds to instantaneous intensity

Stays unsaturated

Only continuous lasing possible ??

Possible solutions

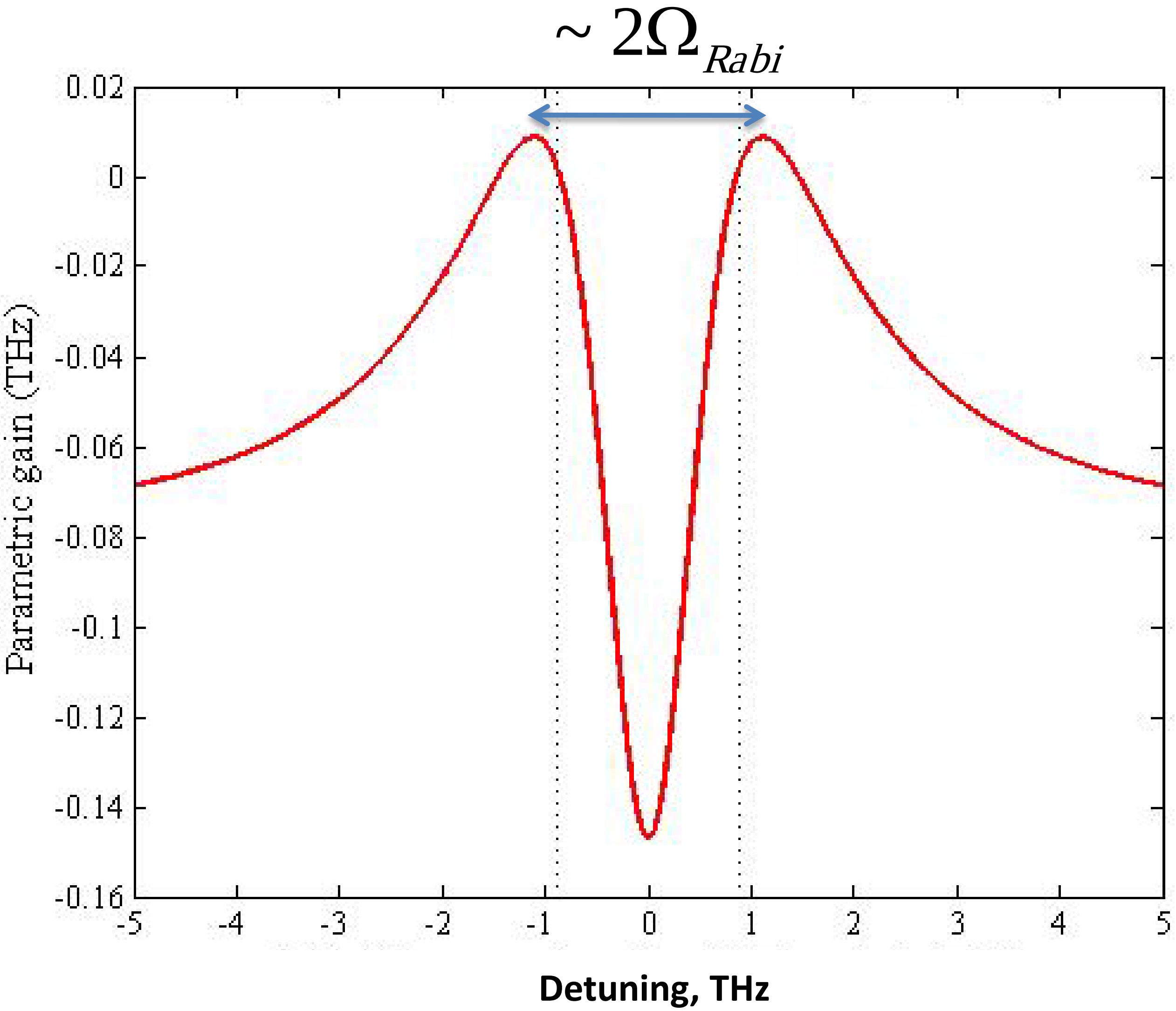
- Can locking of multiple transverse modes lead to pulsed operation?
- Mode locking in the coherent regime? Parametric RNGH instability, π-solitons

Conclusions

- QCLs show unique nonlinear dynamical behavior
- Stable phase locking and synchronization of lateral modes
  - Requires nearly equal thresholds for several lateral modes (buried heterostructure laser);
  - Requires near-degeneracy of the spectrum: closely spaced modes with different transverse order
  - Note that stable locking of longitudinal modes belonging to ONE transverse mode is impossible without a saturable absorber

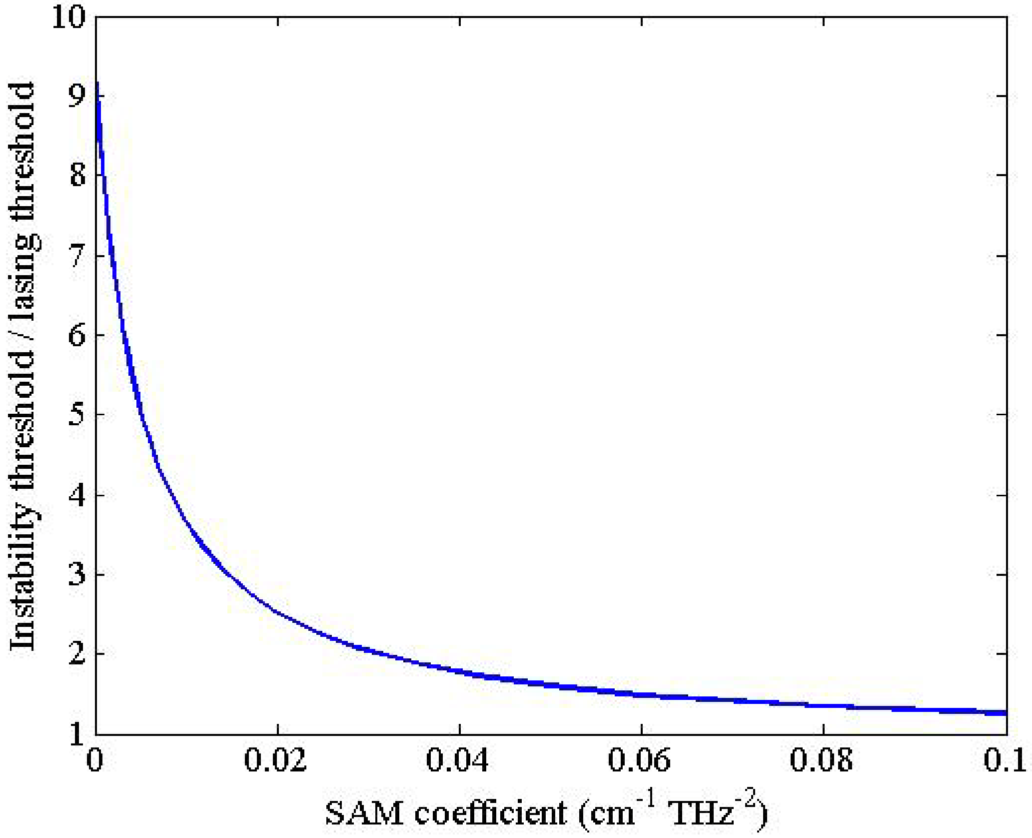
- CONTROL over the phases of locked modes?
- Can we make stable pulses out of locked lateral modes?
- Only active mode locking was achieved so far. Is single-pulse operation via passive mode locking possible?

Nonlinear deformation of the gain spectrum

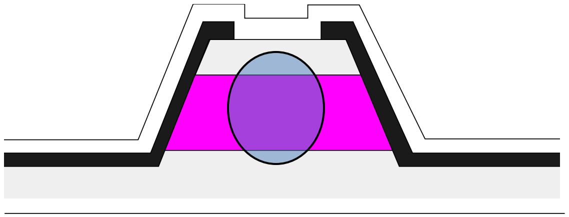

$\sim 2\Omega_{Rabi}$

However, instability threshold is: $I_{\text{pump}} > 9\ I_{\text{thr}}$

Threshold is lowered by saturable absorber



(Unsaturated loss divided by saturation intensity)



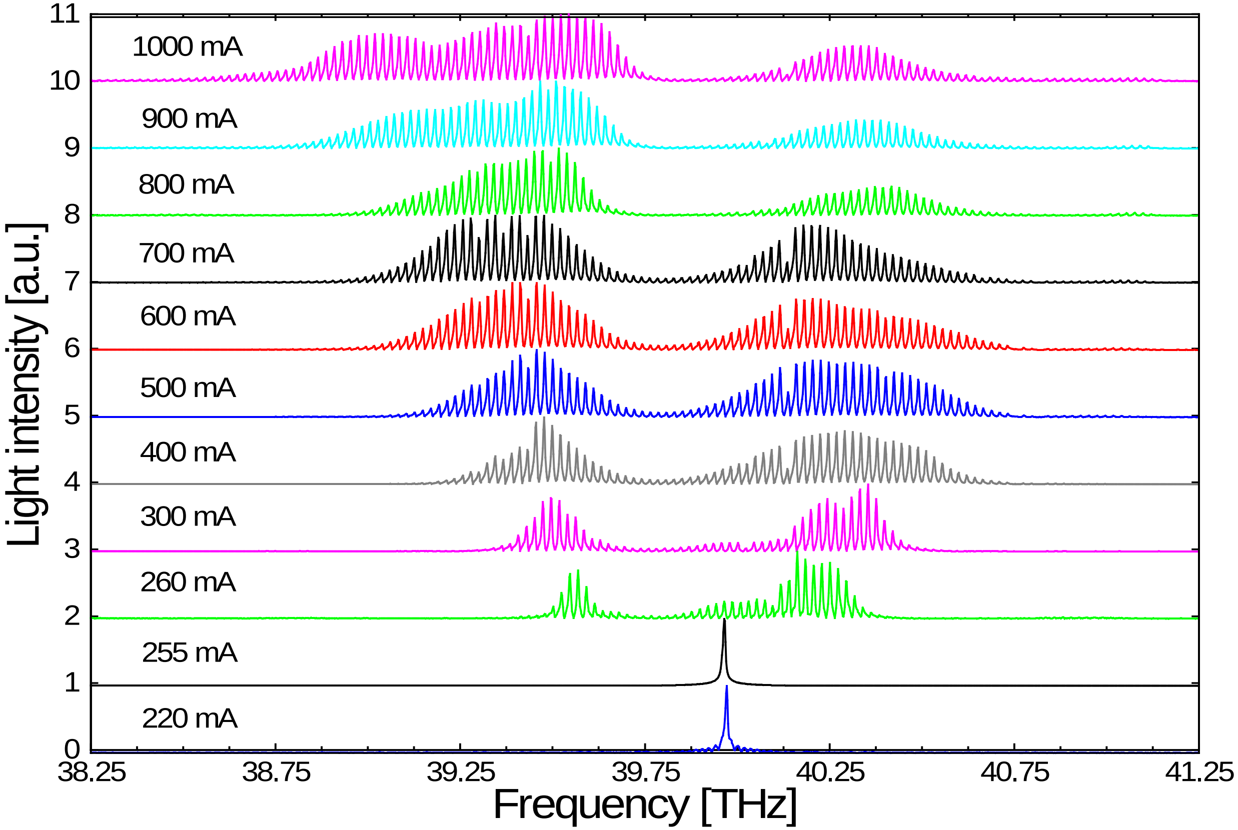
Saturable absorption due to Kerr effect

PRA 2007,2008; JMO review 2011

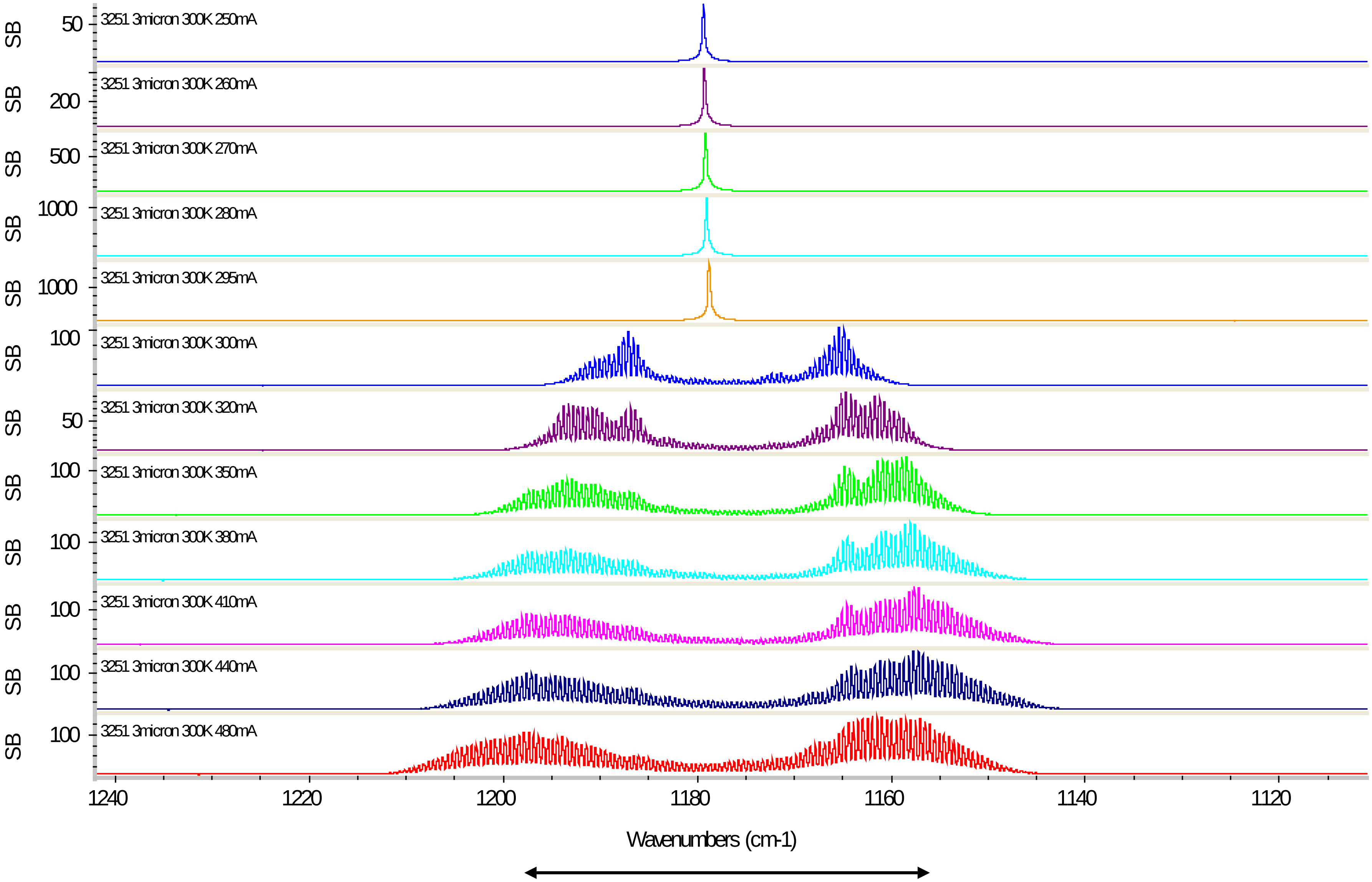
QCL spectra

Breaking down of CW single mode into two or multiple spectral humps

Observed in QC lasers of different designs



Low threshold ~ 1.2 j_{thr}



Grows as 2xRabi frequency of the field

Rabi splitting of the spectra