Alexey Belyanin
Lecture 3

Semiconductor nanostructures continued:

- Motivation for mid/far-infrared devices;
- Nonlinear dynamics of QC lasers;
- THz physics

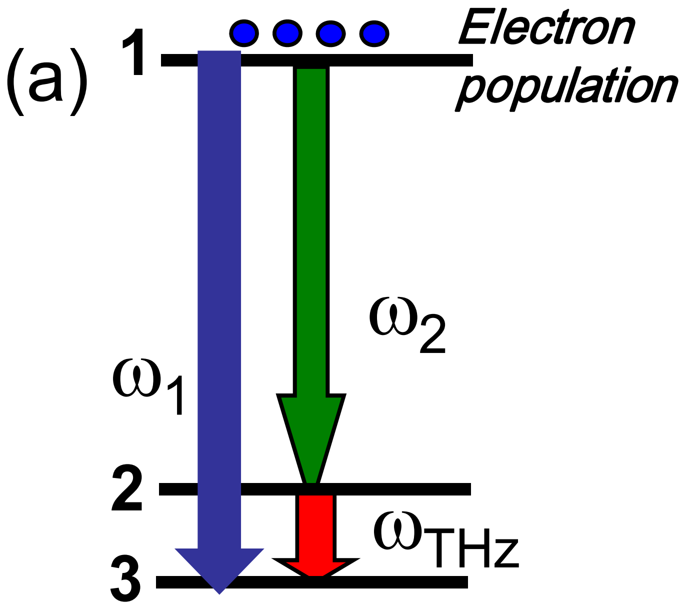
From previous lecture:

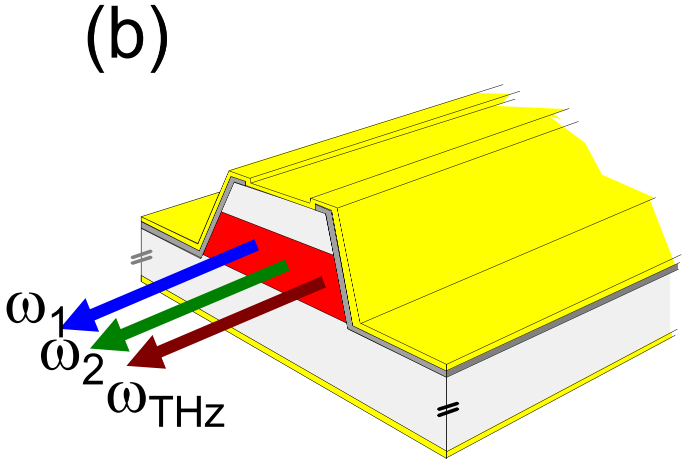
Giant optical nonlinearity of quantum-well nanostructures
+ possibility of electron injection/depopulation

↓

Enables novel optical devices, mostly in the mid/far-IR

First room-temperature THz semiconductor laser



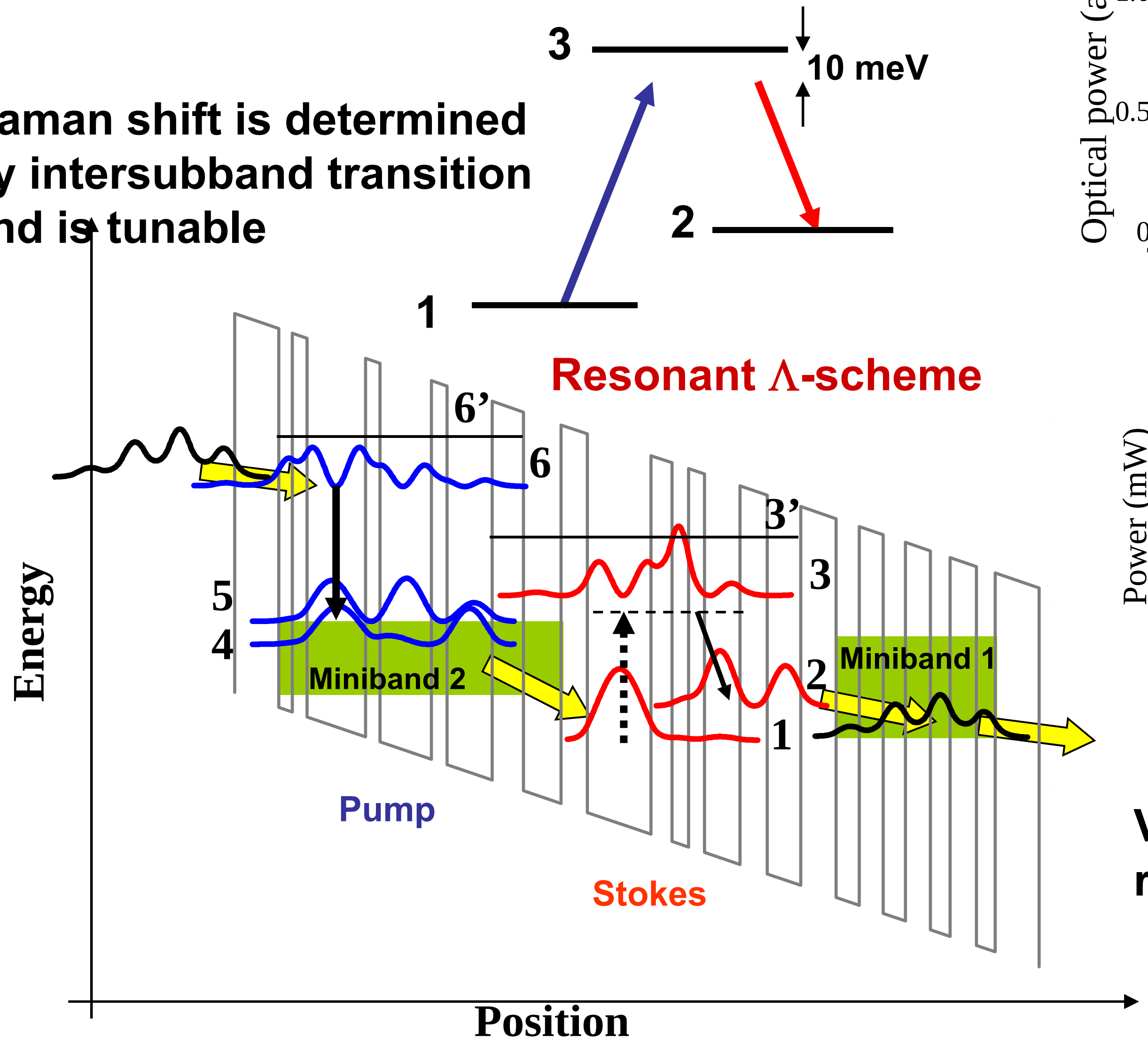


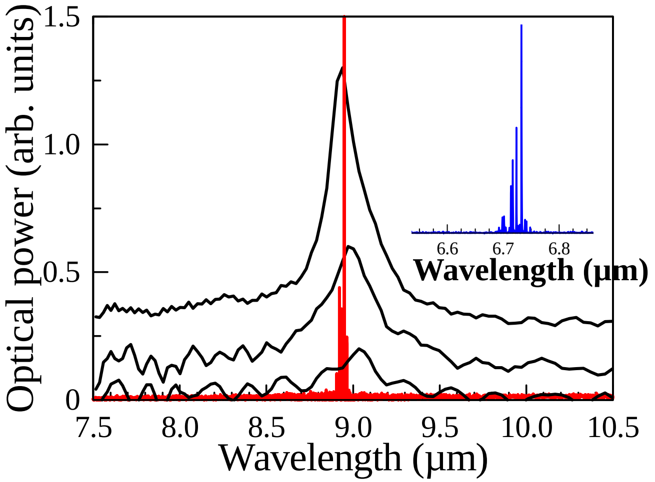
- Make a powerful mid-IR QCL emitting at two modes
- Provide strong nonlinearity for frequency mixing process
- Design a low-loss, phase-matched waveguide for all three modes

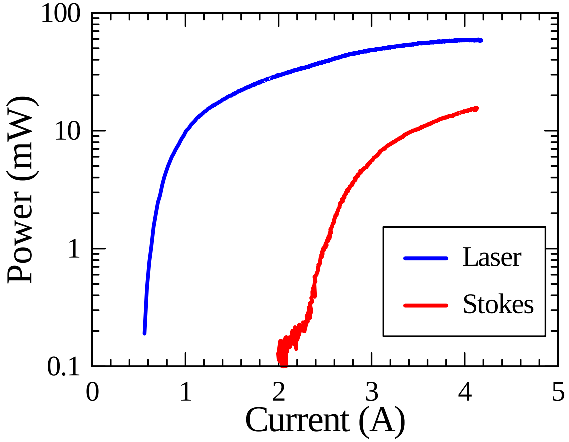
Nature Phot. 2007, APL 2008

Mid-IR Raman injection laser

Raman shift is determined by intersubband transition and is tunable





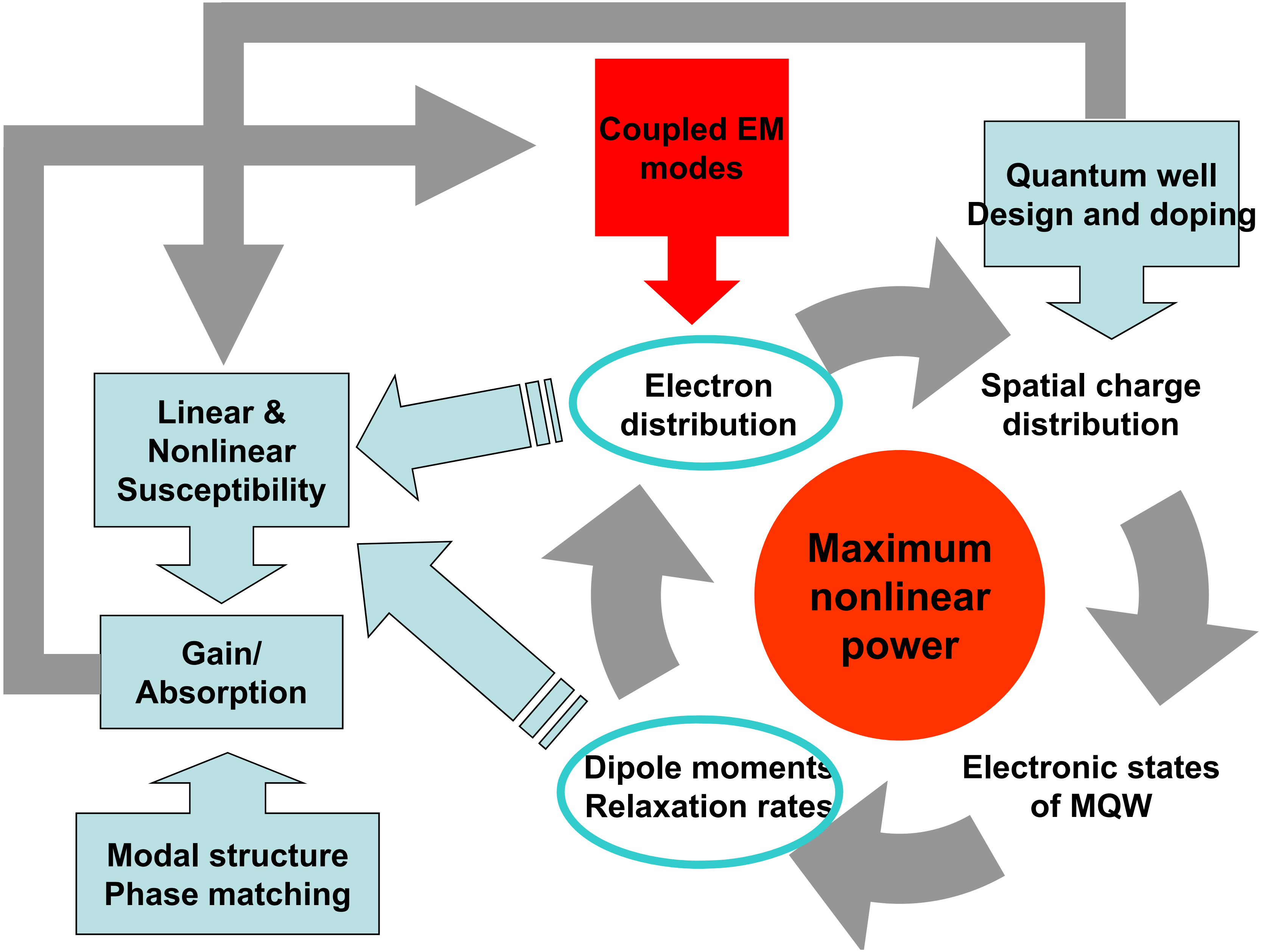


Very large Raman gain at resonance: ~ 10^{-4} cm/W

40 mW Raman threshold
16 mW Stokes power

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

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



Coupled EM modes

Quantum well Design and doping

Linear & Nonlinear Susceptibility

Electron distribution

Spatial charge distribution

Maximum nonlinear power

Gain/ Absorption

Dipole moments Relaxation rates

Electronic states of MQW

Modal structure Phase matching

Why should we suffer through this?

Why should you care?

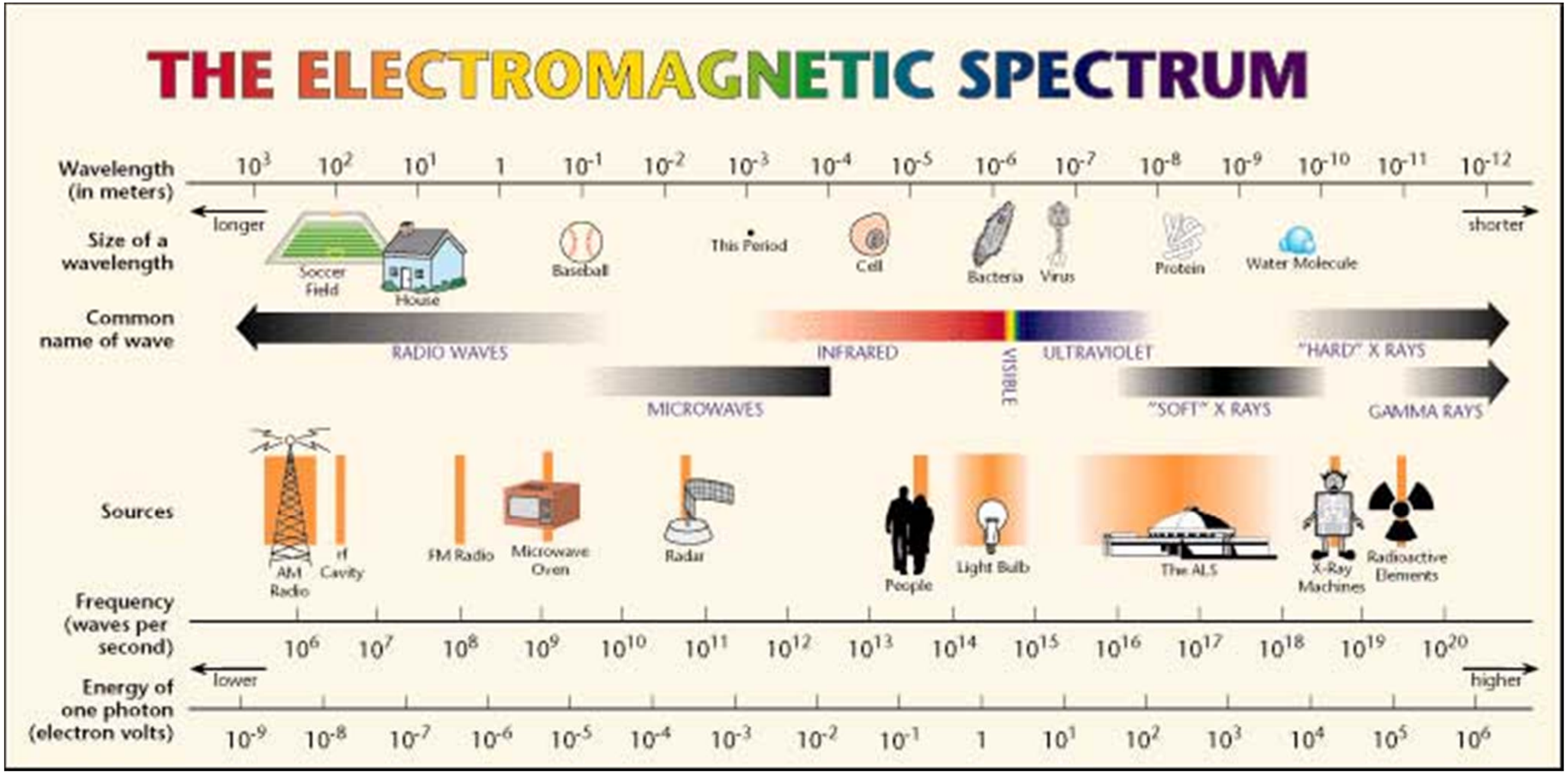
Why nonlinear optics?

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



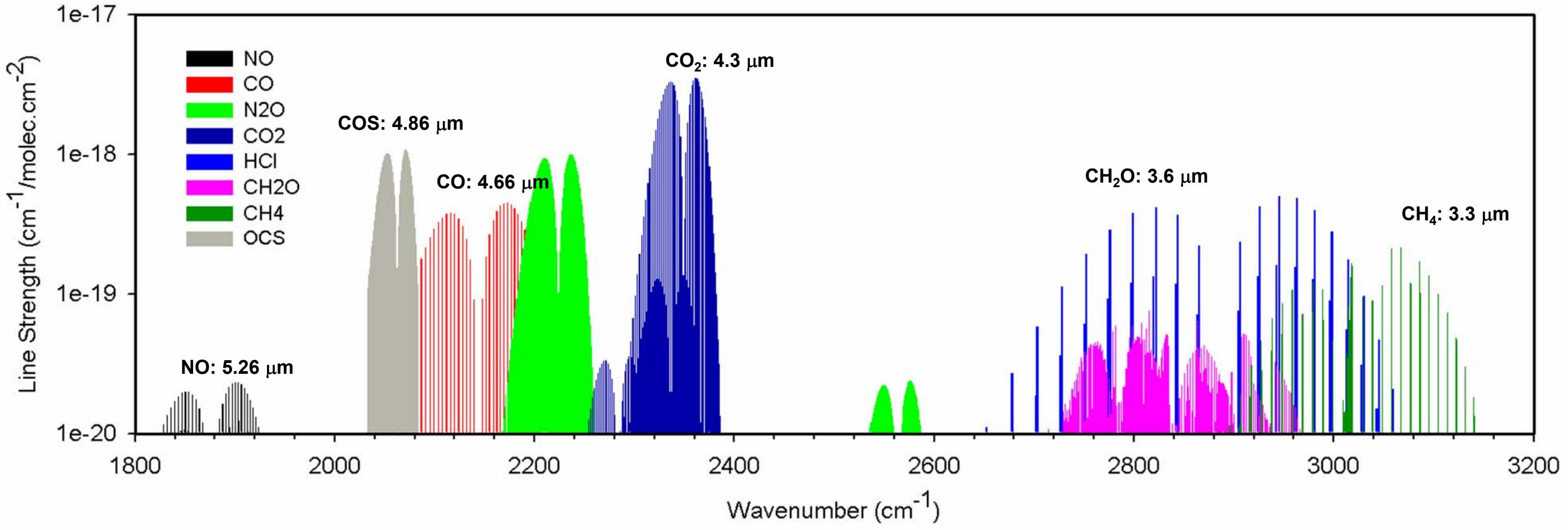
PRIMARY MOTIVATION:

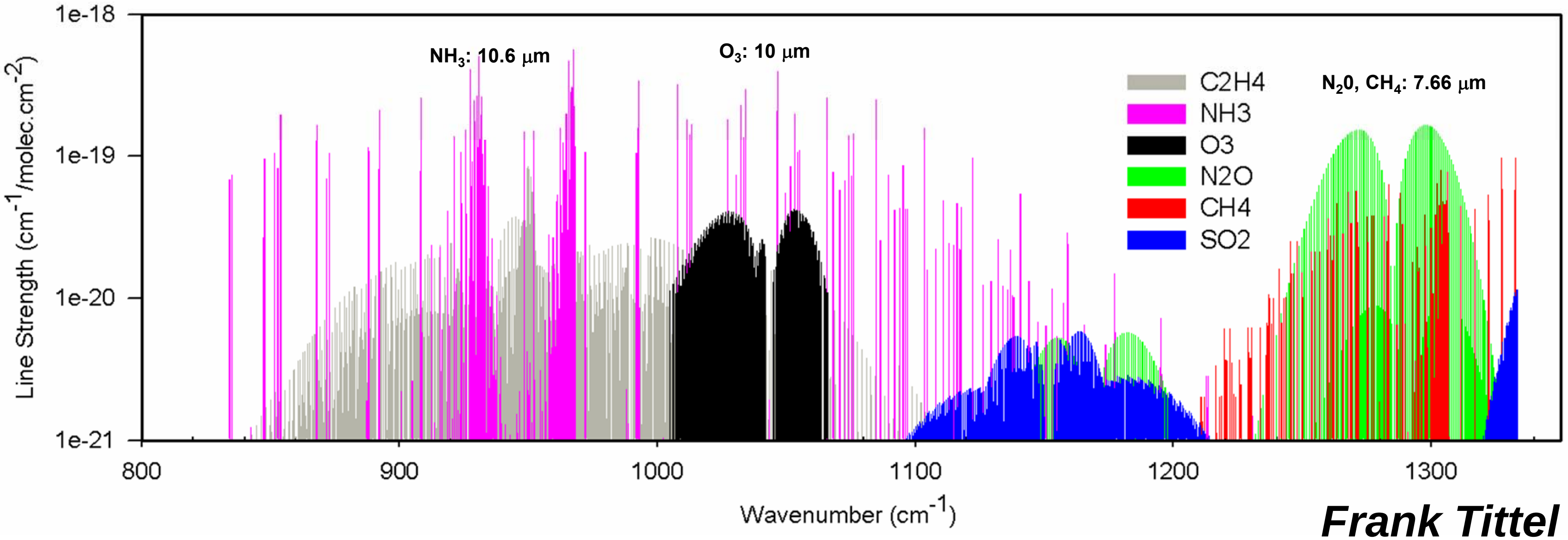
- **Atmosphere has transparency windows in the infrared range**

- **<u>ALL</u> molecules have <u>STRONG</u> spectral fingerprints in the infrared**

Other applications: infrared cameras, target pointers, countermeasures, telecommunications

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





Frank Tittel et al.

Air Pollution: Houston, TX

NASA Atmospheric & Mars Gas Sensor Platforms

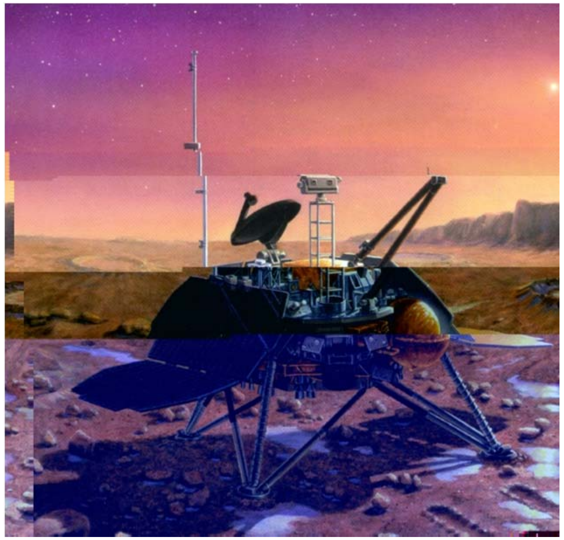
Frank Tittel et al.

Tunable laser sensors for earth’s stratosphere

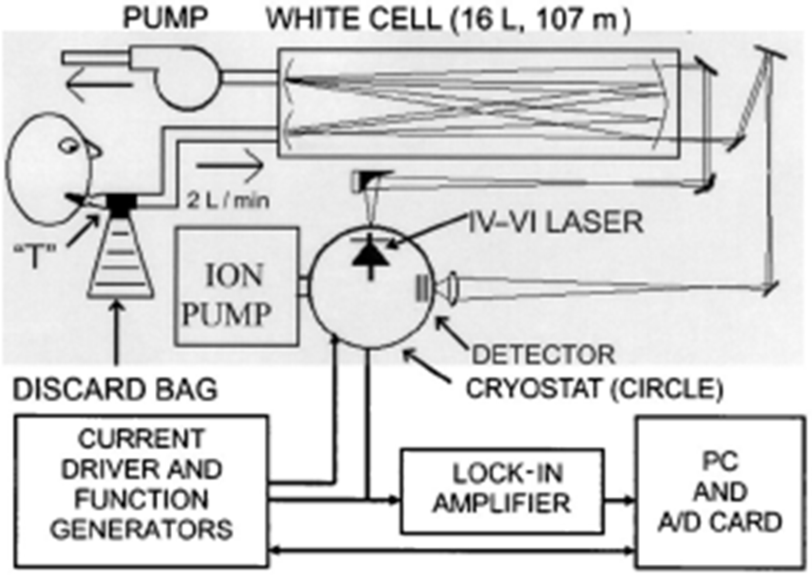
Aircraft laser absorption spectrometers

Dryden Flight Research Center EC97-44358-2 Photographed 29DEC1997
Douglas DC-8 Airborne Laboratory arrival at Dryden (NASA/Tony Landis)

Tunable laser planetary spectrometer

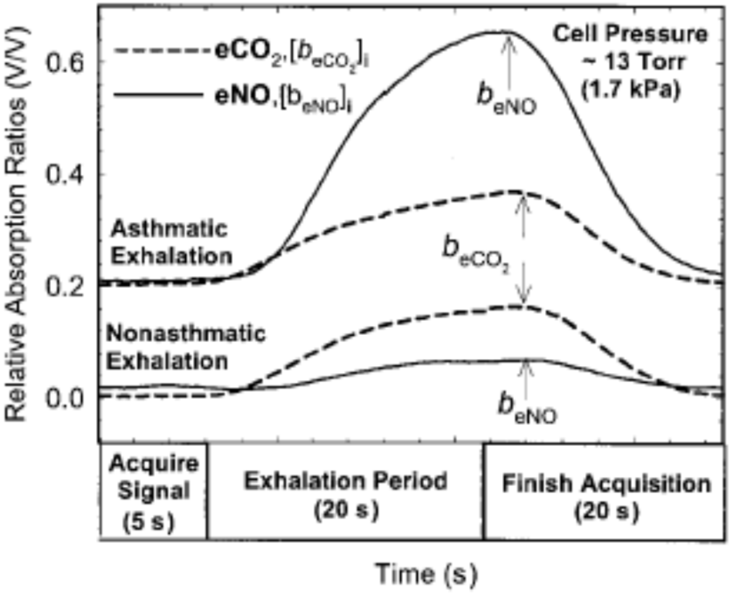


Non-invasive Medical Diagnostics:
Breath analysis



NO: marker of lung diseases

- **Concentration in exhaled breath for a healthy adult: 7-15 ppb**
- **For an asthma patient: 20-100 ppb**



NH_3: marker of kidney and liver diseases

Need fast and compact sensors

Appl. Opt. 41, 6018 (2002)

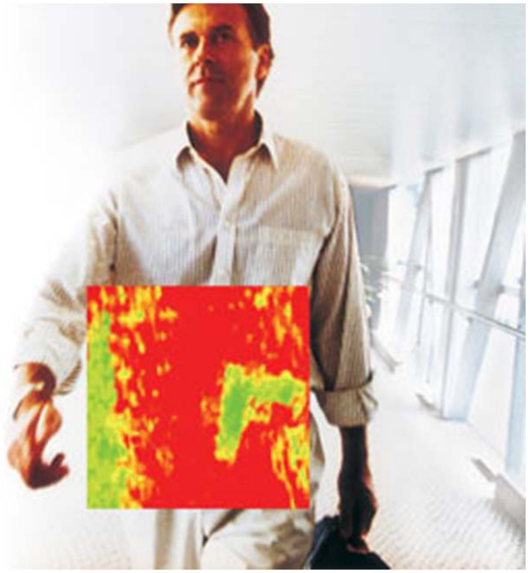
Wide Range of Gas Sensing Applications

- **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

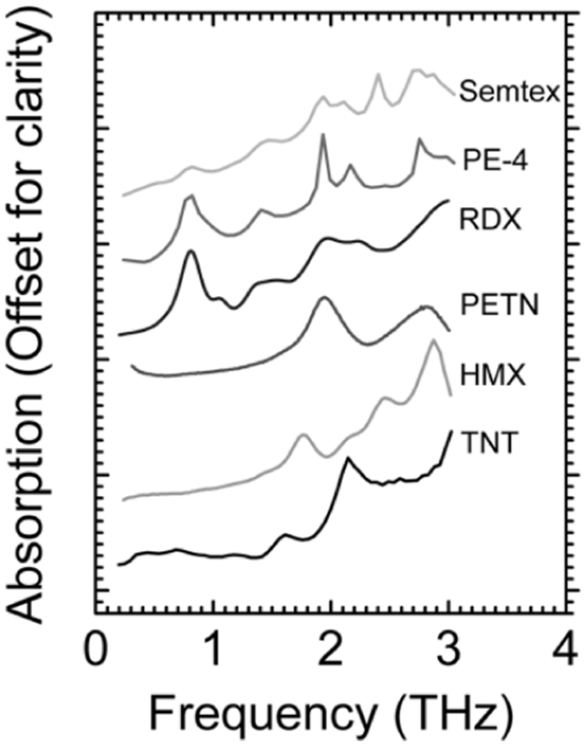
Frank Tittel et al.

World Through Terahertz Glasses

f = 1 THz $\Rightarrow$ E = 4 meV $\Rightarrow$ λ = 300 μm



ThruVision Ltd.



Teraview

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

Many frequency scales in doped semiconductors fall into the THz spectral range

1 THz = 4 meV

- Plasma frequency
- Fermi energy
- Electron scattering rates
- Cyclotron frequency in the magnetic field of ~ 1 Tesla
- Intra-donor transition frequencies
- Phonon frequencies

- Rich information can be extracted from THz spectroscopic studies
- Exotic conditions for atoms and plasma in superstrong magnetic fields

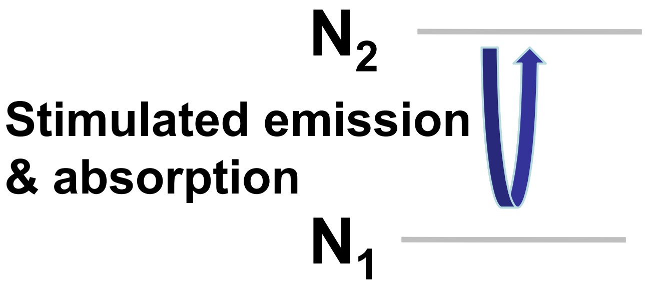
Nonlinear dynamics, phase coherence, and mode locking in quantum cascade lasers

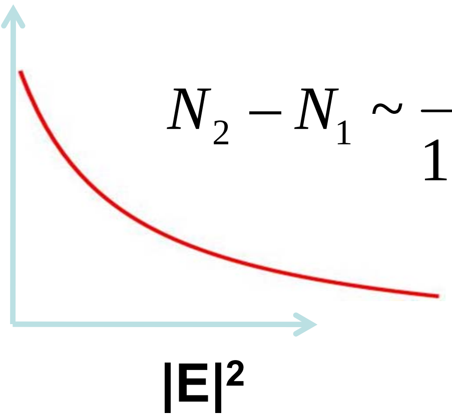
Collaborators:
F. Capasso group, Harvard Univ.
F. Kaertner group, MIT

PRA 2007,2008, OE 2009, PRL 2009, PRL 2011; review: JMO 2011

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

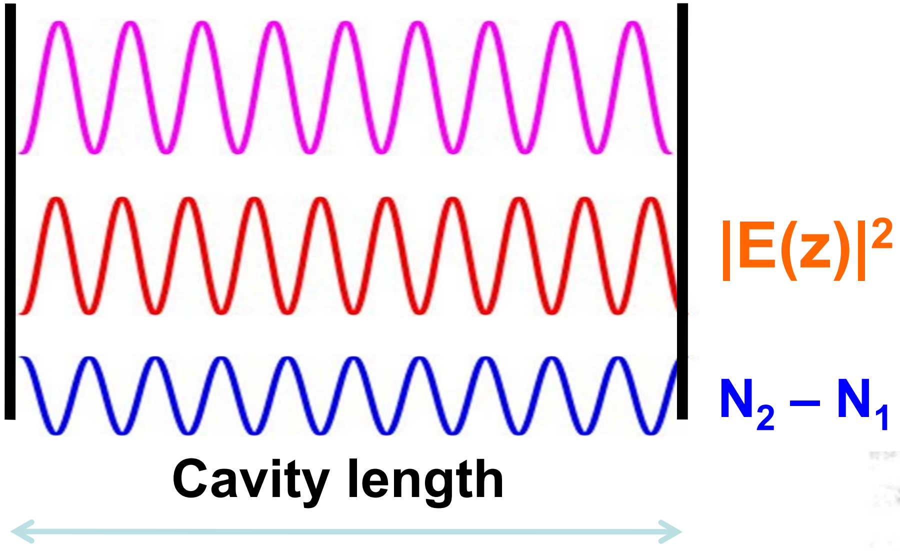




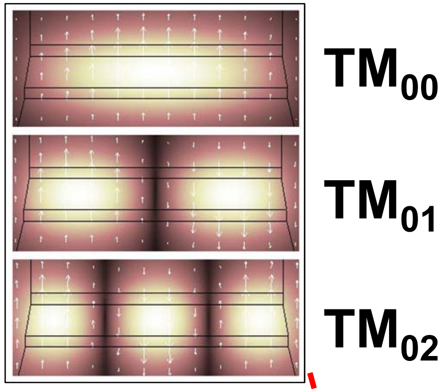
$$N_2 - N_1 \sim \frac{(N_2 - N_1)_{E=0}}{1 + (1/I_s)\sum E_\mu E_\nu}$$

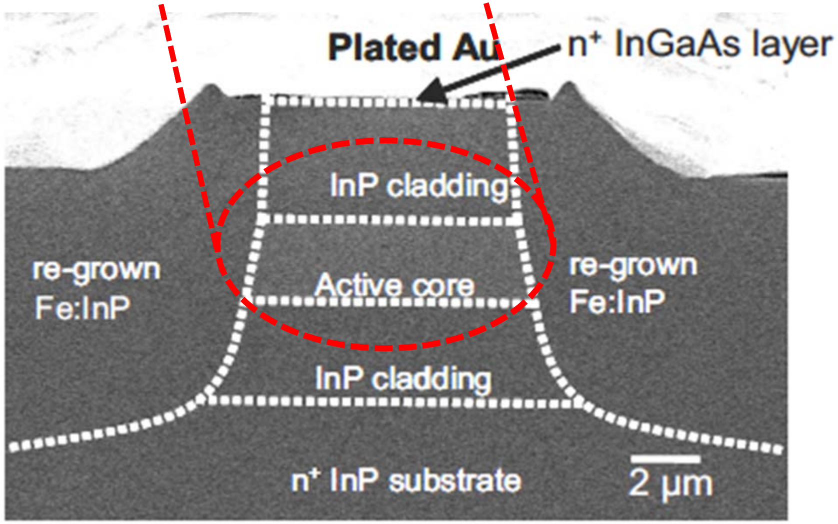
EM modes in QCLs: interaction through saturation nonlinearity

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

Observed signatures:

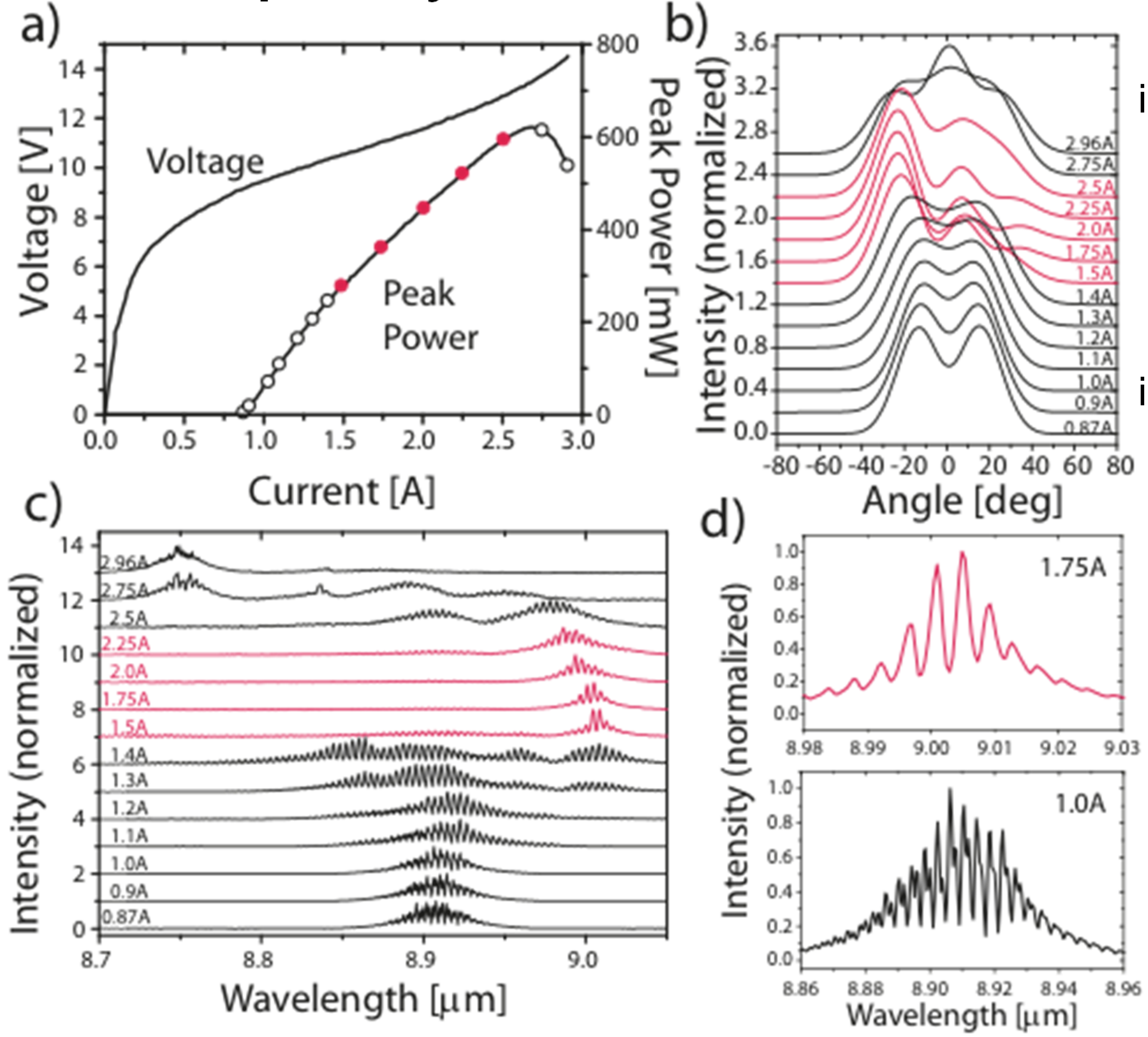
- **Anomalous near-field and far-field beam pattern; beam steering by current**
- **Locking to commensurate frequencies or synchronization of lateral modes to a single comb**
- **These effects appear and disappear as a bifurcation, with a slight change in injection current**

PRL 2009, PRL 2011; review: JMO 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 2011

Complete synchronization of three combs



incoherent

Phase-
locked

incoherent

incoherent

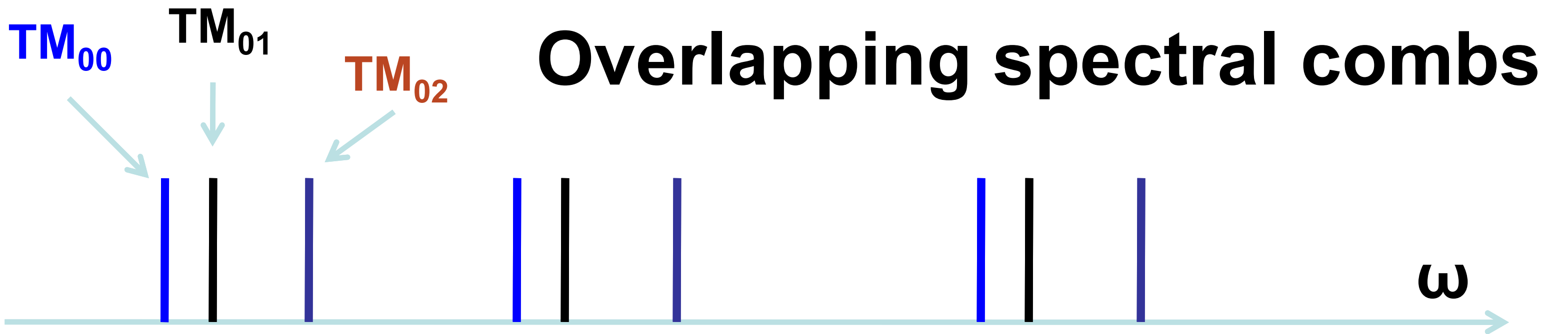
Phase-
locked

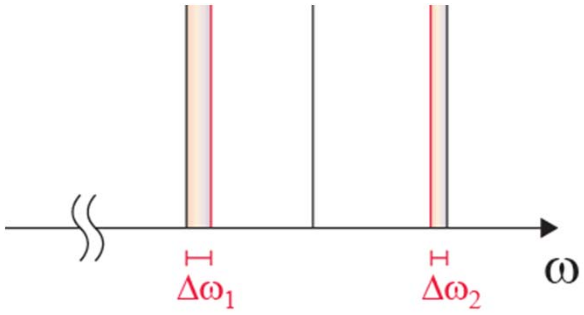
incoherent

Waveguide width 19 μm

PRL 2011

Overlapping spectral combs

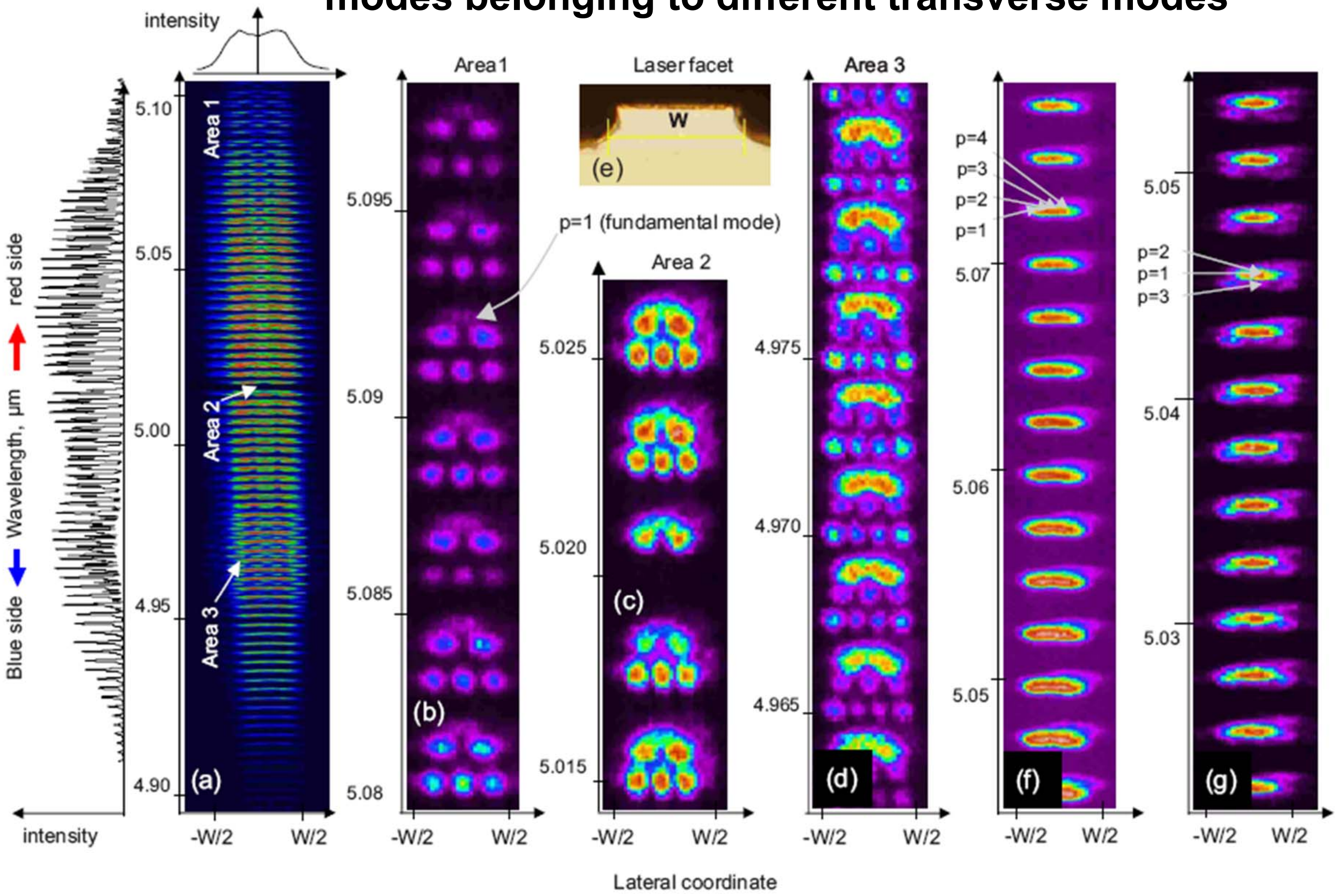




Nonlinear interaction leads to frequency pulling

Three combs can lock into equidistant triplets or even to a single comb (synchronization). No external modulation is needed!

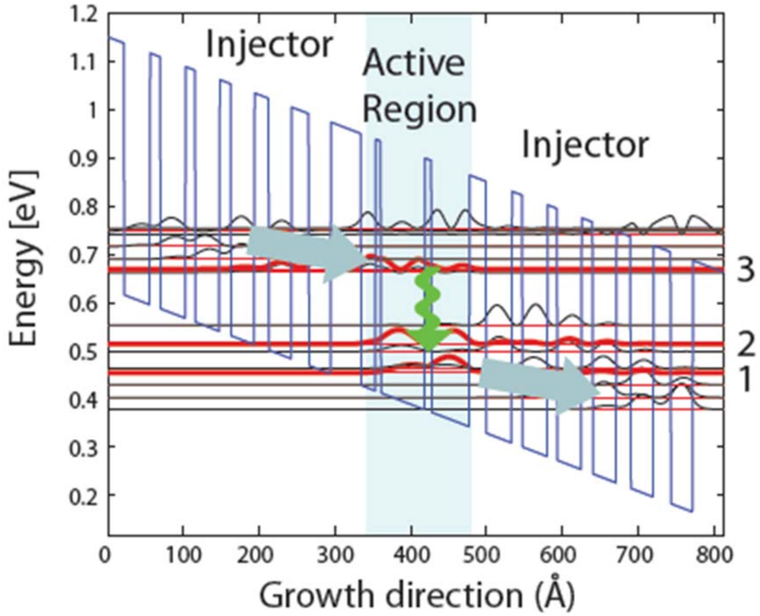
Observations of overlapping combs of longitudinal modes belonging to different transverse modes



Note close grouping of modes with different transverse (and longitudinal) indices

N. Stelmakh et al., *Appl. Phys. Lett.* 94, 013501 (2009)

Why QCLs stand apart in dynamical and multimode behavior



Ultrashort gain recovery: T_1~ 1 ps

Dephasing time T_2 ~ 0.1-0.5 ps

Cavity roundtrip time: T_r ~ 20-80 ps

Photon lifetime: T_c ~ 10 ps

$$T_2 < T_1 << T_{r}, T_c$$

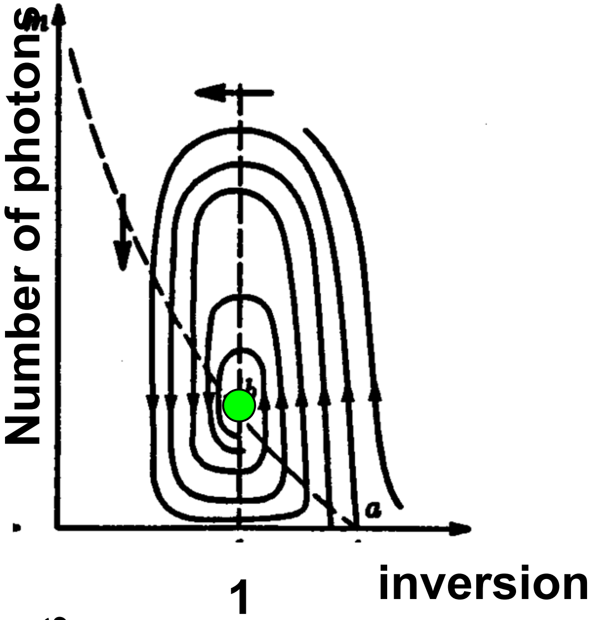
An overdamped Class-A laser:

Polarization and inversion adiabatically follow the field

All other solid-state and diode lasers are Class B:

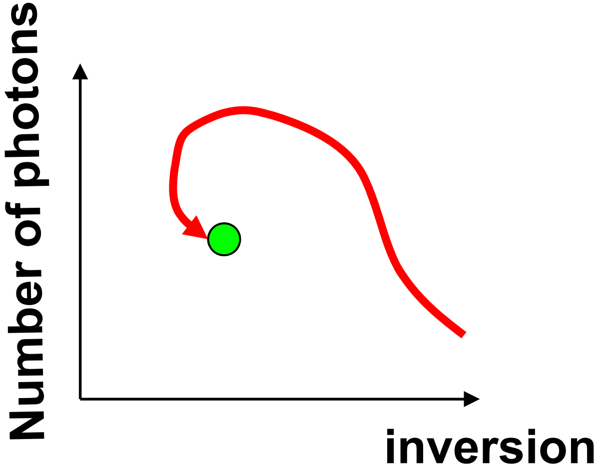
$$T_2 << T_{r,c} << T_1$$

Trivial single-mode dynamics: only aperiodic processes, no relaxation oscillations



Solid-state lasers:
Stable focus

$$\frac{T_1}{T_c} \gg 1$$



QC lasers:
Stable node

$$\frac{T_1}{T_c} \ll 1$$

Overdamped oscillations.
Ultrafast modulation is possible.
How about mode locking?

Maxwell-Bloch Equations

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

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

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

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

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

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:

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

Nonlinear overlap - $G_{jklm}=\int_{AR}\varepsilon E_jE_kE_lE_m dV$

Gain - $g_j=4\pi\omega_0 d^2N_pT_2/(\hbar\mu_j^2)$

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

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

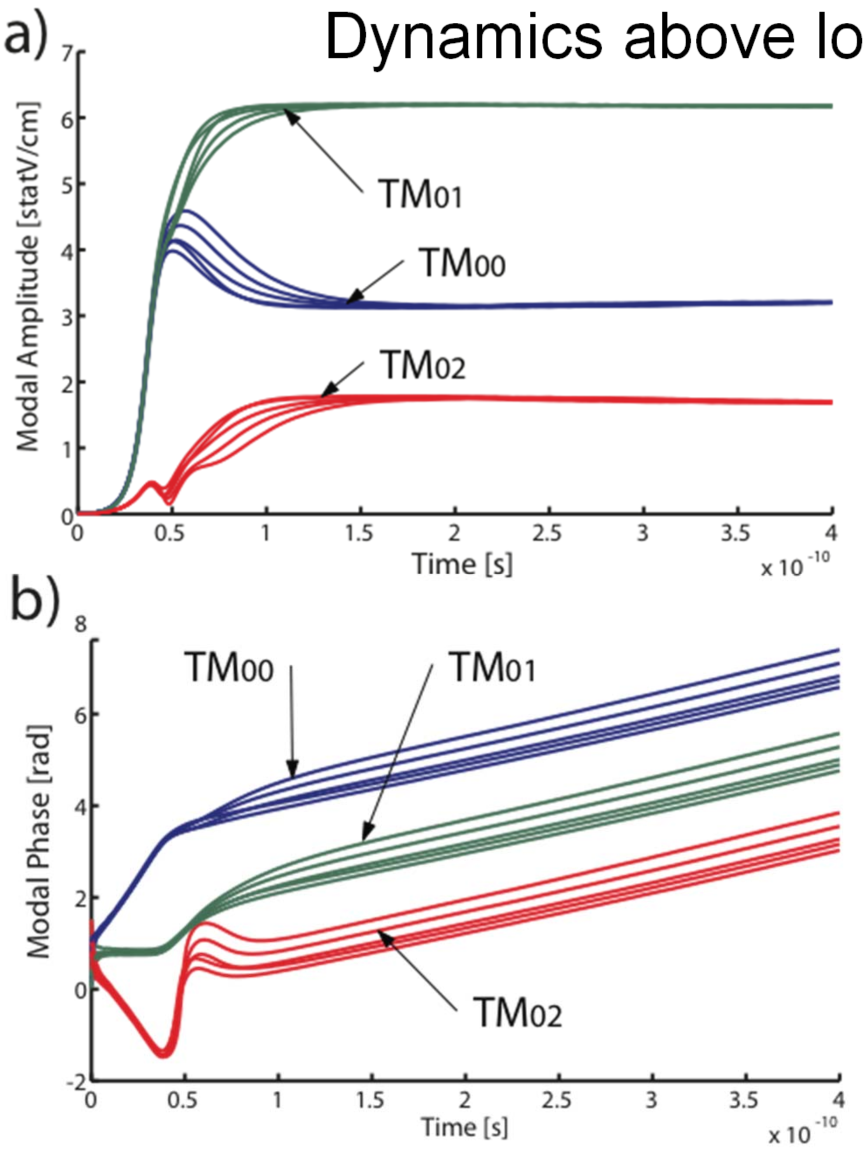
Saturation intensity $I_s=2\hbar^2/(d^2T_1T_2)$

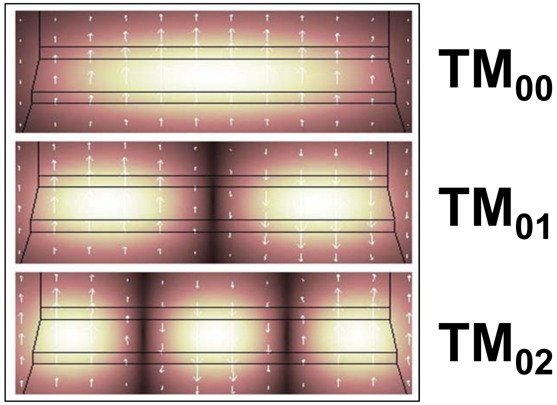
Large dipole moment gives rise to strong nonlinear coupling of laser modes

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)

Dynamics above locking threshold





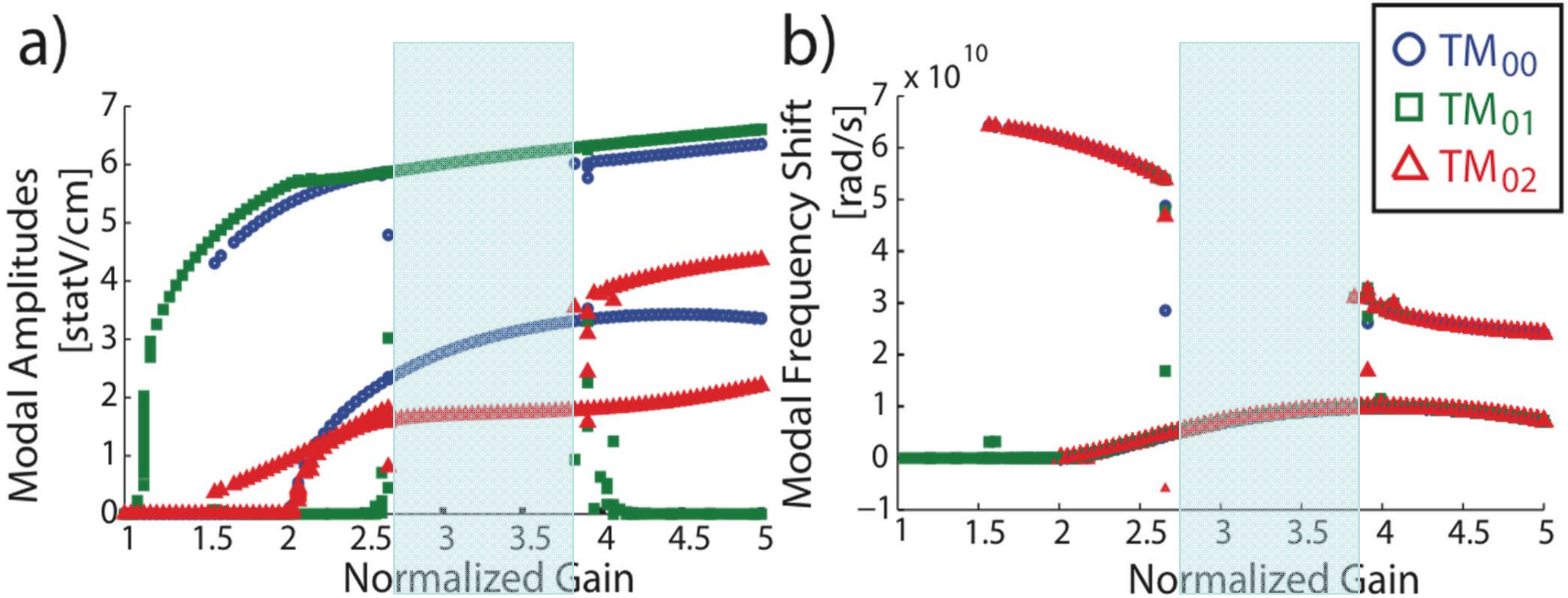
$$a_j(t) = A_j(t)e^{i\Phi(t)}$$

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

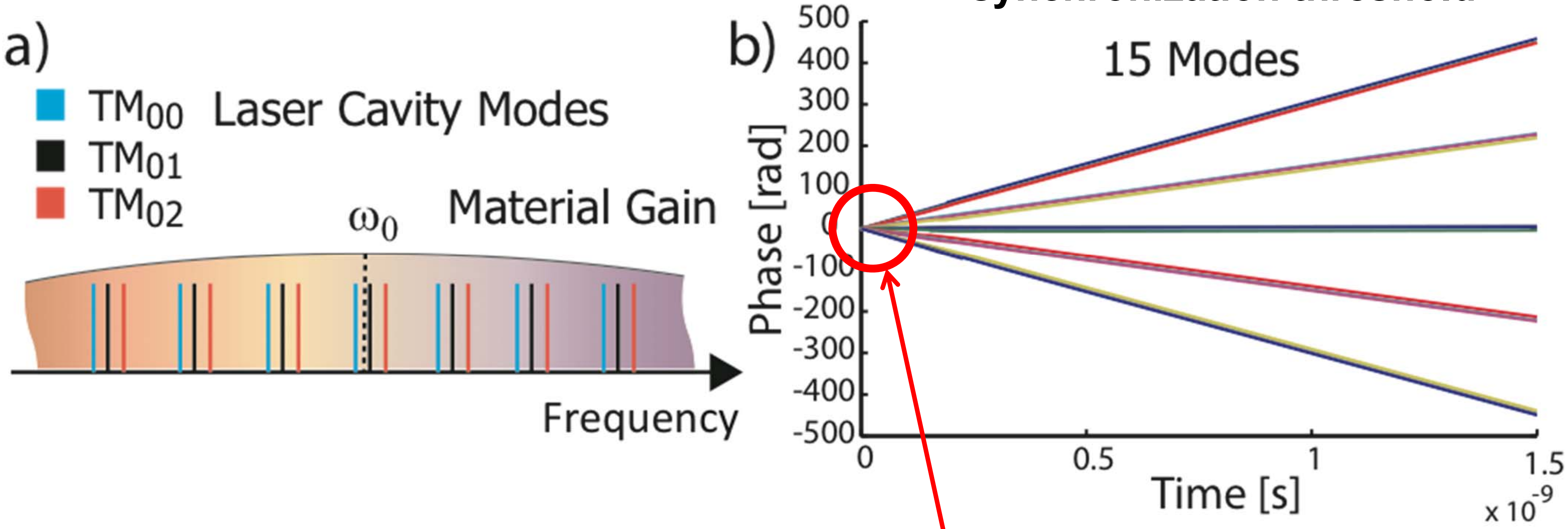


- Single stable multimode state locked to a single frequency at intermediate gains
- Bistable region outside

Longitudinal modes of a linear cavity

$$\propto \exp\left(\pm i\beta_\mu z \pm g_\mu z\right) \quad \beta_\mu \approx N_\mu \pi / L_{cav}, N_\mu \sim 1800$$

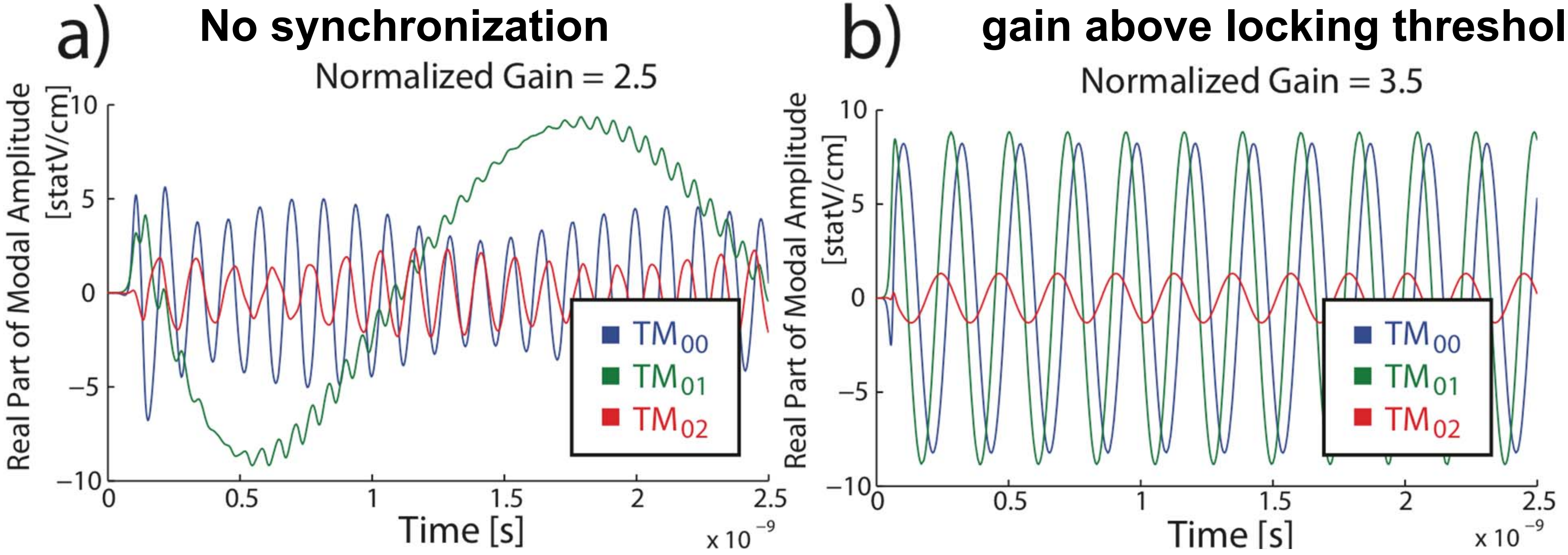
Time evolution above synchronization threshold



Synchronization over ~ 10 ps (not resolved)

- **3 combs of 5 longitudinal modes each**
- **Combs merge into a single comb**
- **Dynamics does not depend on the number of triplets if they are separated**

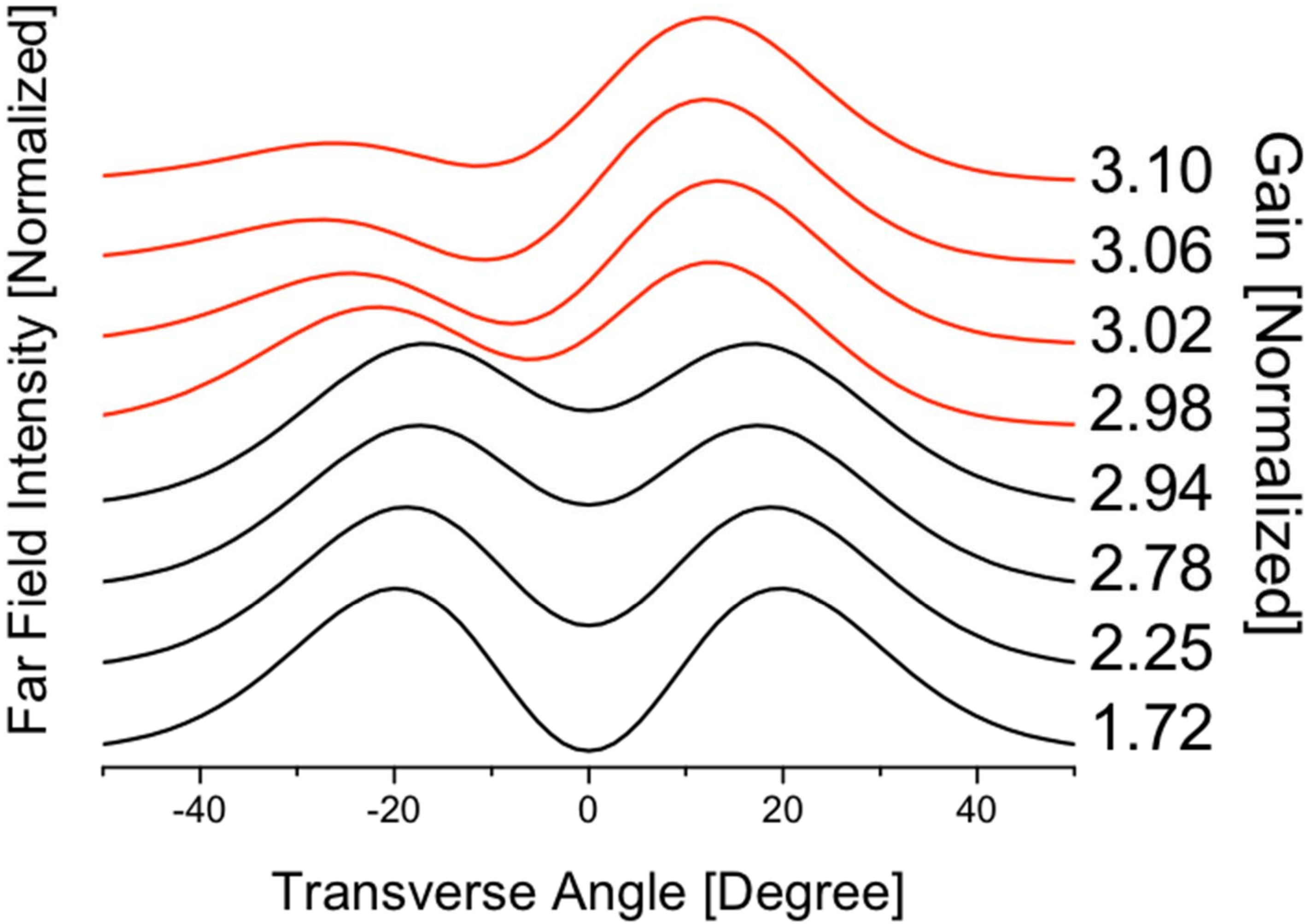
Dynamics of one triplet



No synchronization

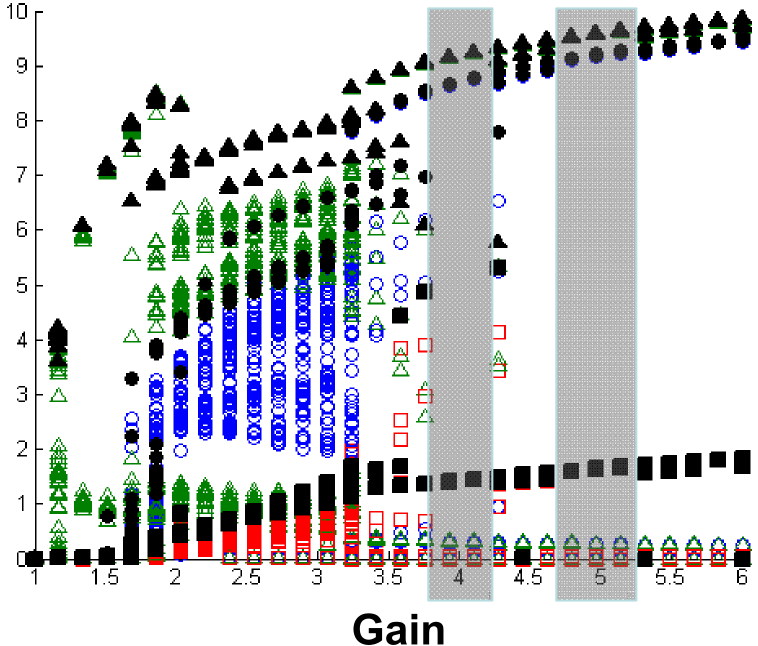
gain above locking threshold

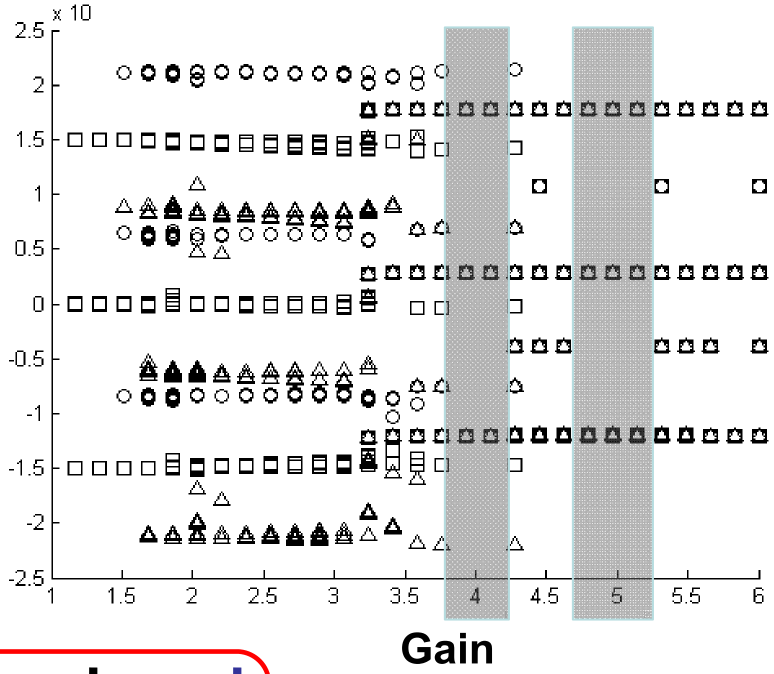
Theoretical far field for different gains above threshold

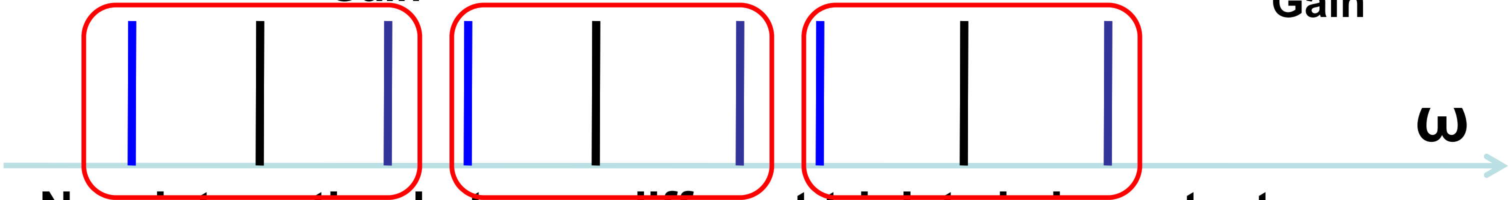


9 Modes - Dynamics of 3 Close Triplets

30 initial conditions for each gain


Amplitudes


Frequencies



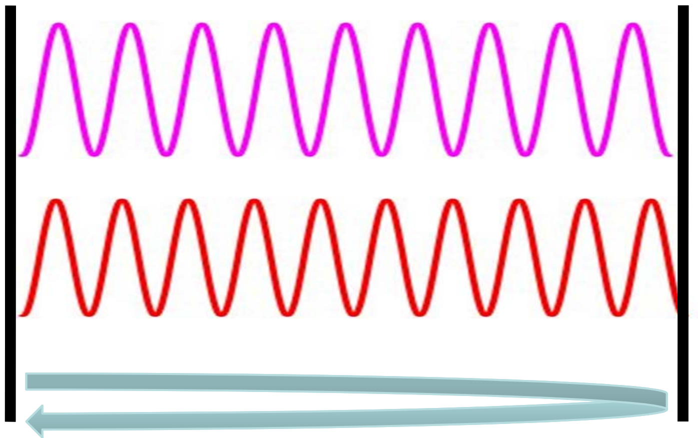
Now interaction between different triplets is important

Dynamics is more complex and less stable

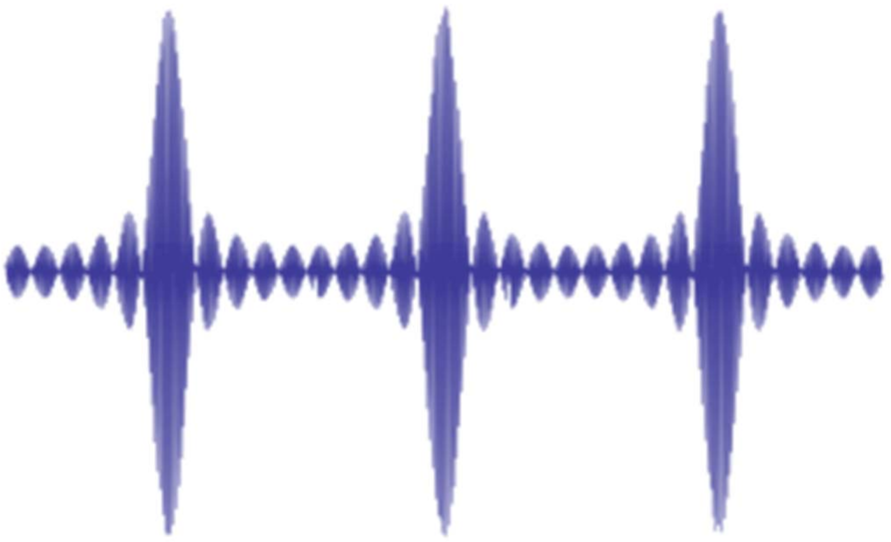
Two regions of synchronization separated by multi-stability or chaotic dynamics

Mode-locked pulses in QCLs

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



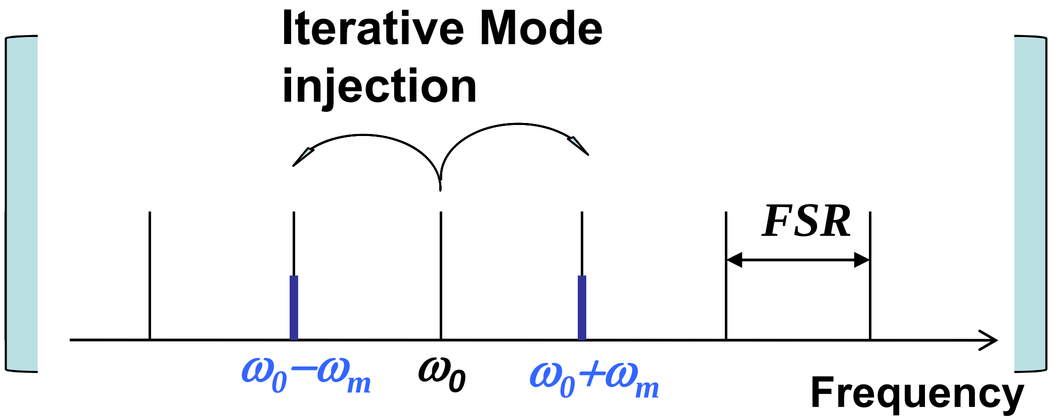
T_{round}



Adding 10 sines

This is very difficult in QCLs where $T_1 << T_{\text{round-trip}}$

ACTIVE MODE LOCKING

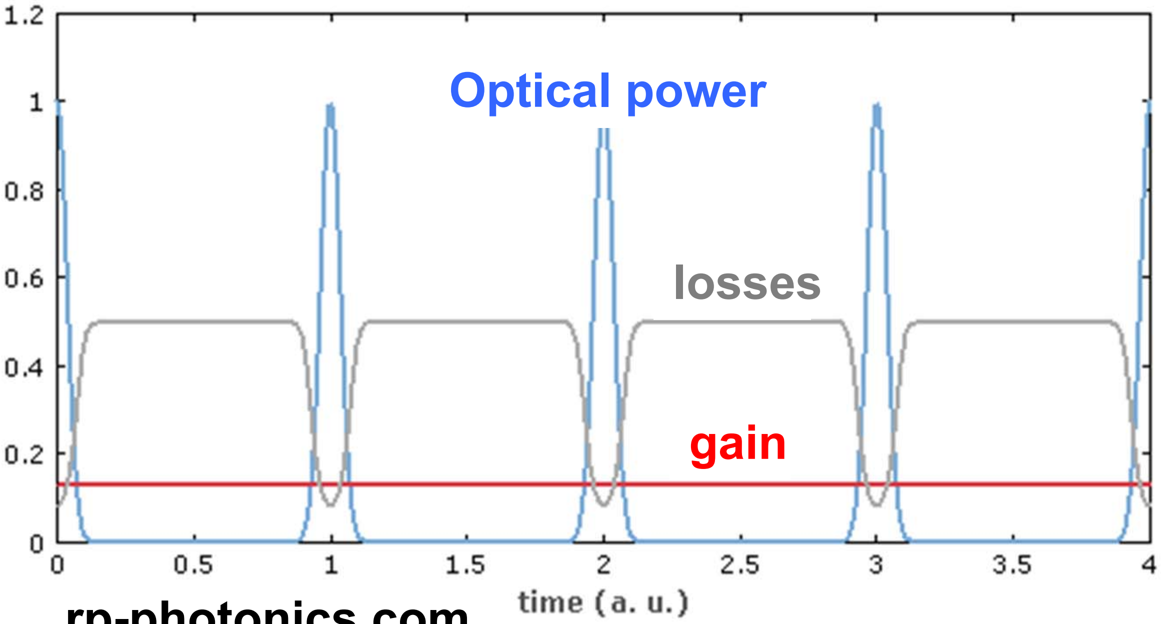


Modulation at the round-trip frequency

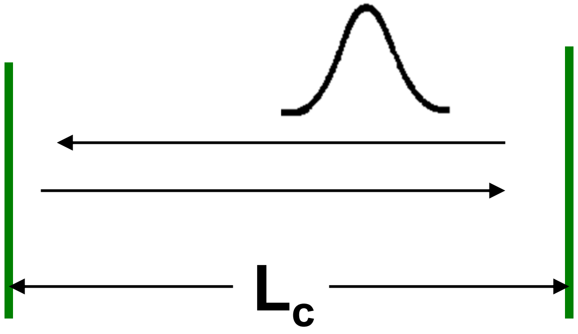
$\cos(\omega_m t)$ active modulation of laser gain creates phase-locked sidebands coincident with the two closest cavity modes

Passive mode locking

Saturable absorption (intensity-dependent losses)

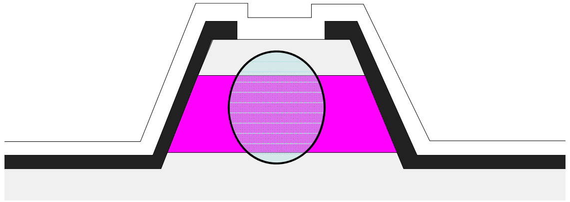


rp-photonics.com



$$\alpha \approx \alpha_0\left(1 - {I(t)}/{I_s}\right)$$

Saturable absorption due to Kerr effect:
$n = n_0 + n_2 I$



Gain should be saturated!

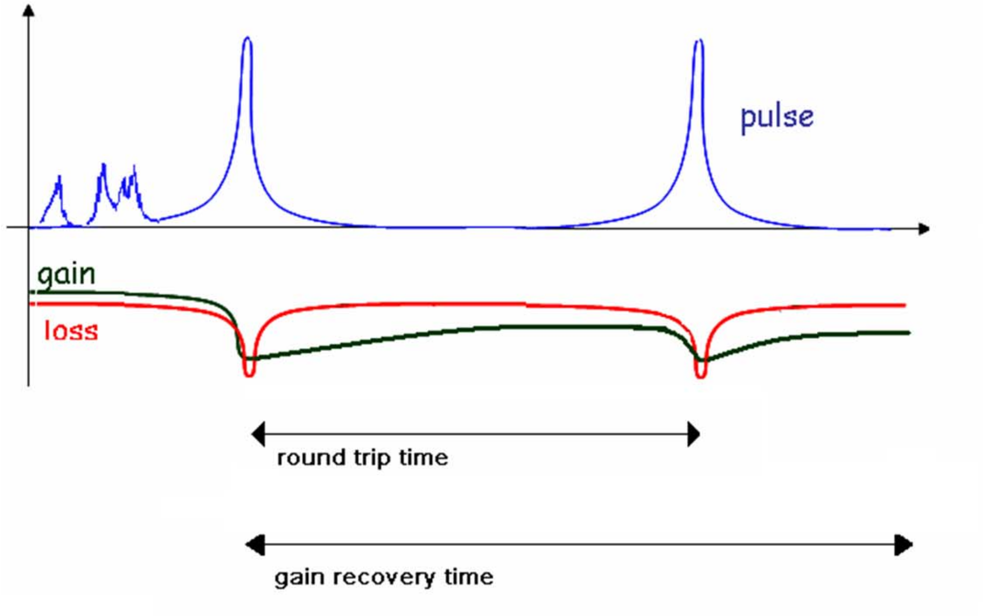
$$g = \frac{g_0}{1 + \frac{I_{average}}{I_s}}$$

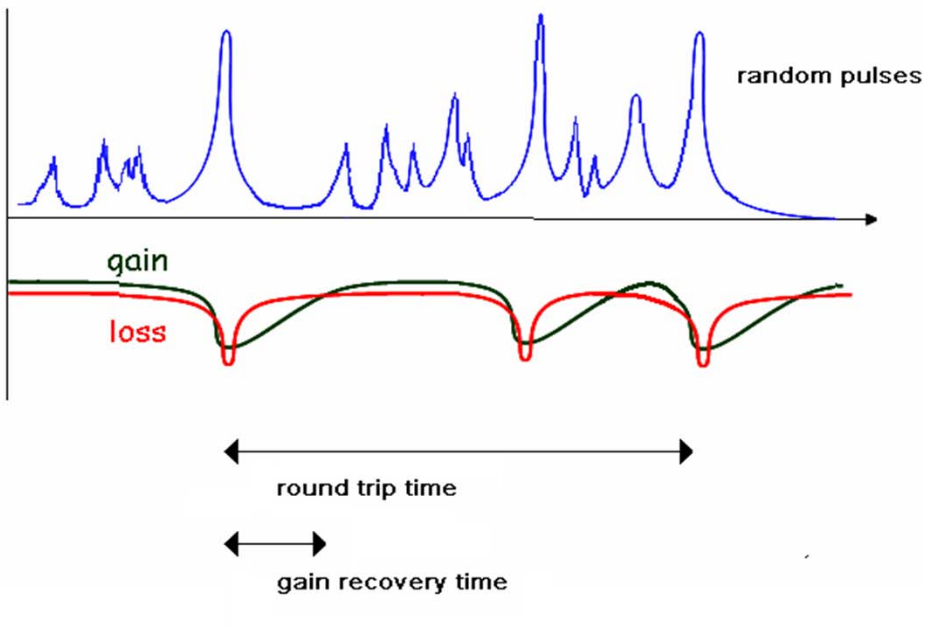
Gain should have long recovery time:

to achieve stable mode locking Haus condition must be met :

gain recovery time > roundtrip time = $2L_c/c$

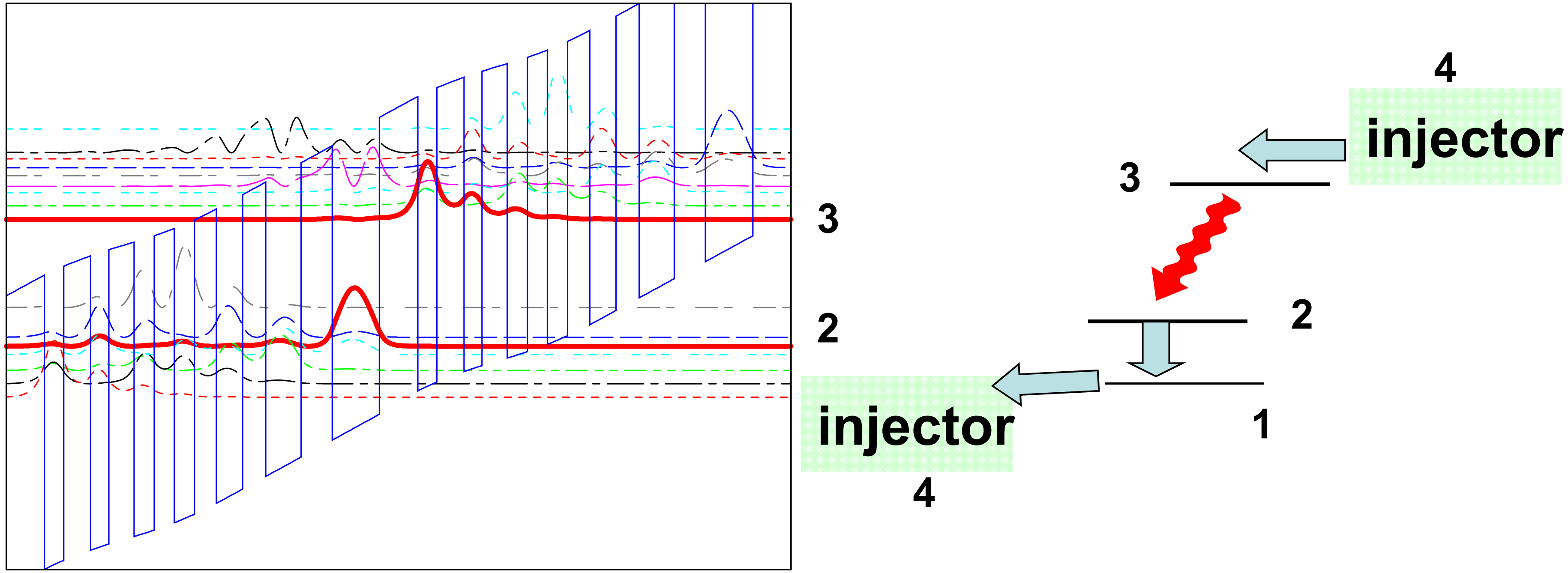
In QCLs this condition is not fulfilled





Attempts to generate mode-locked pulses

Laser structure: superdiagonal

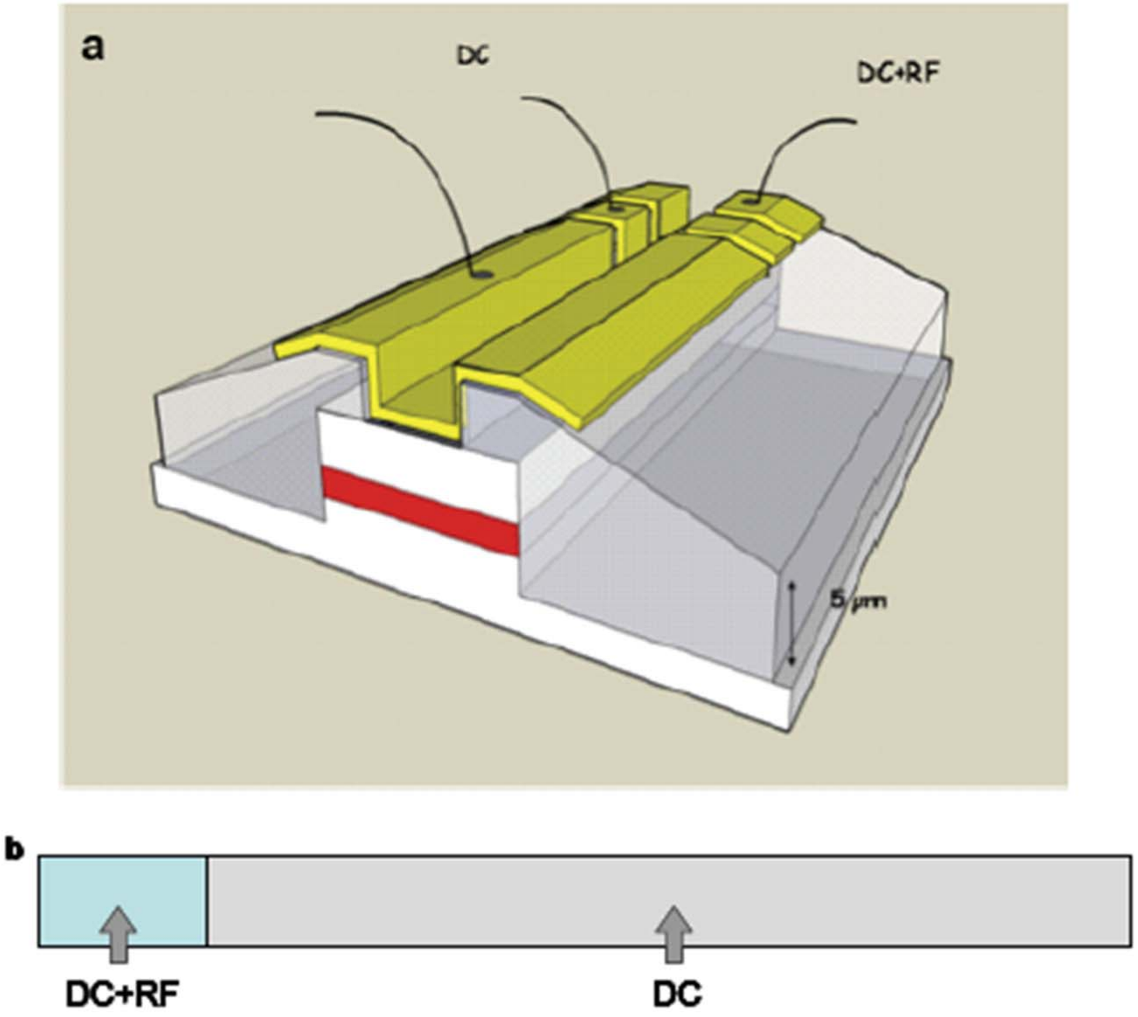


Calculated upper state lifetime ~50 ps
Confirmed by T. Norris measurements

Gain recovery time ??? **Fast components in gain recovery**

Capasso group 2008

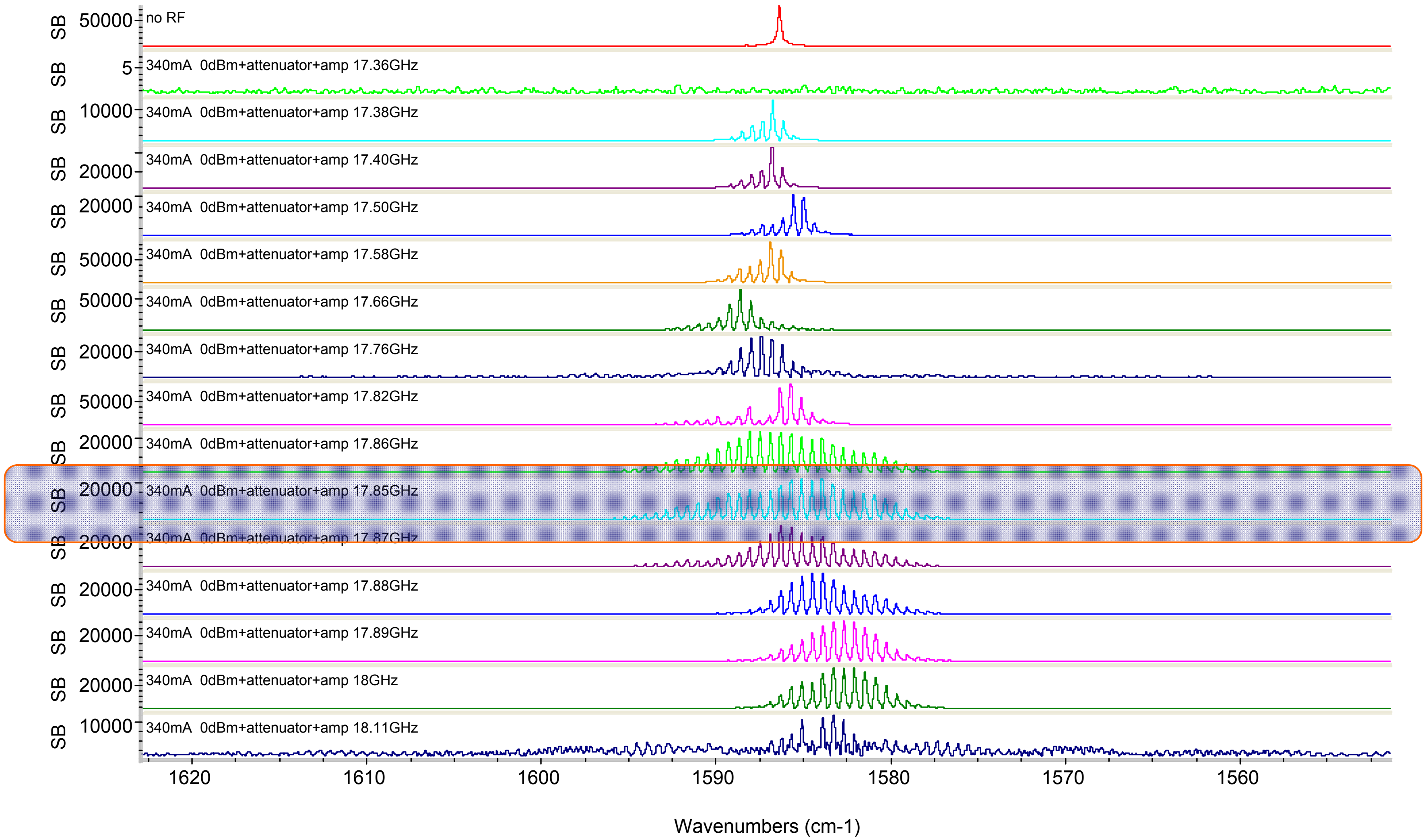
Active mode locking



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

Capasso group, OE 2009, 2010

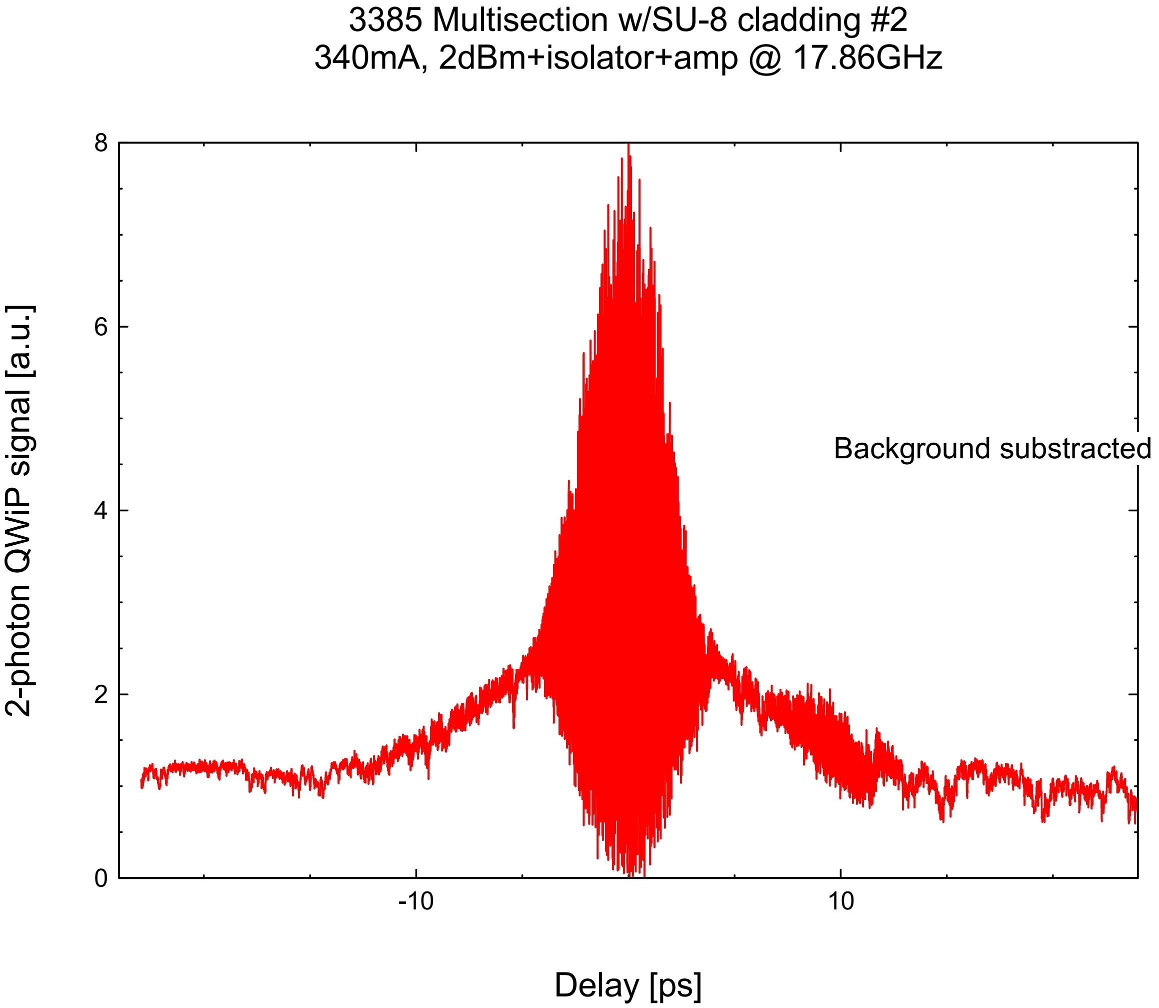
I=340mA, with 35dBm RF power



Resonance @ 17.86 GHz
Power ~ 10 mW

Capasso group

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**

Wang et al. OE 2009

Self mode locking in QCLs?

- Can locking of multiple transverse modes lead to pulsed operation?
- Mode locking in the coherent regime? Faster than dephasing time T_2. RNGH instability, π-solitons, …

Coherent light-matter interaction

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

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

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

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

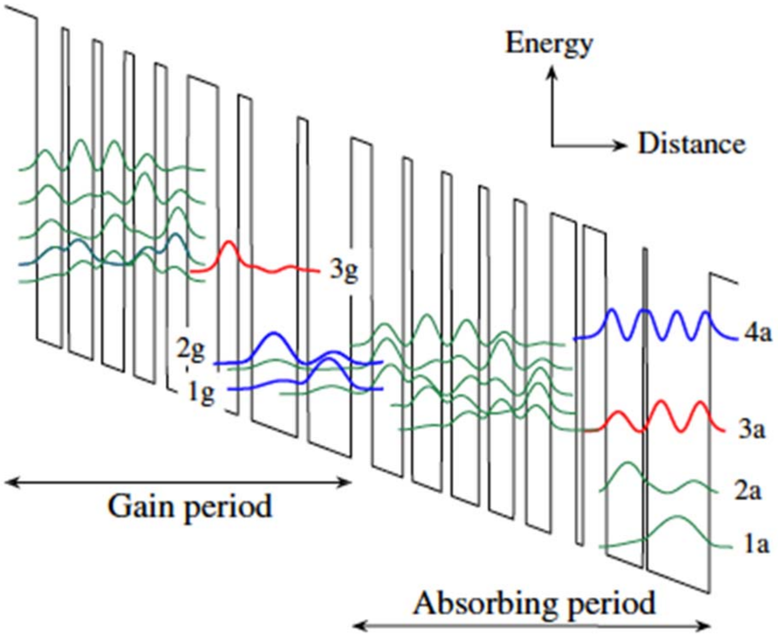
Population inversion $D=\frac{N_{2}-N_{1}}{N}$

Polarization cannot be eliminated if $\frac{dE}{\mathrm{h}}>\gamma_{\perp}$

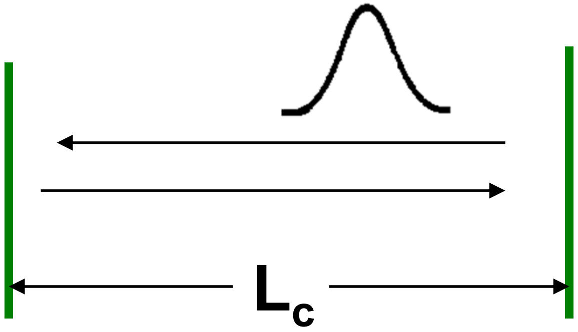
Or for any processes faster than $T_{2}=1/\gamma_{\perp}$

Self-induced transparency mode locking

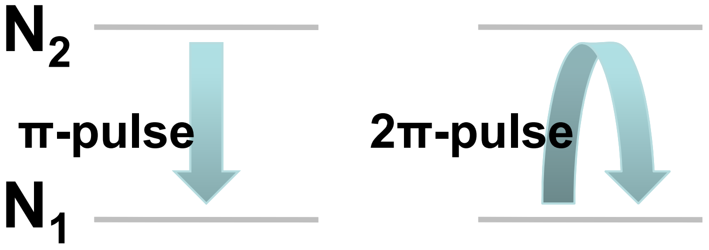
Ultrashort T_1 is an advantage!



Letokhov 1969, Kozlov PRA 1997
Menyuk et al. PRL 2009



Mode-locked pulse is a π-pulse in the gain region and 2π-pulse in the absorbing region



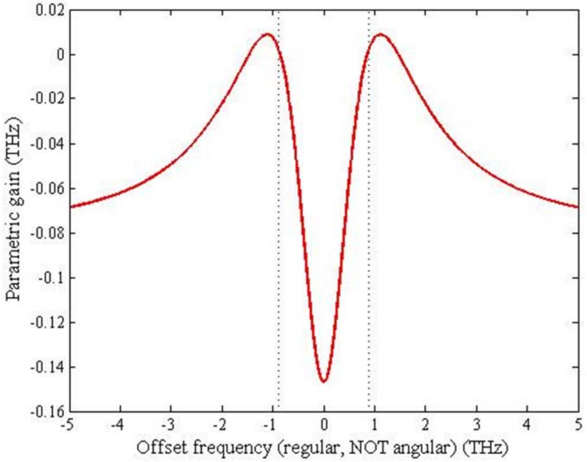
Laser does not self-start; requires injection of ~ 1 ps pulse

“Second threshold”

Regular pulsations and chaos in single-mode lasers

Haken, Oraevskii 1960s

Requires pumping 9 times above threshold and “bad-cavity” laser: cavity line broader than the gain spectrum, or photon lifetime < T_2



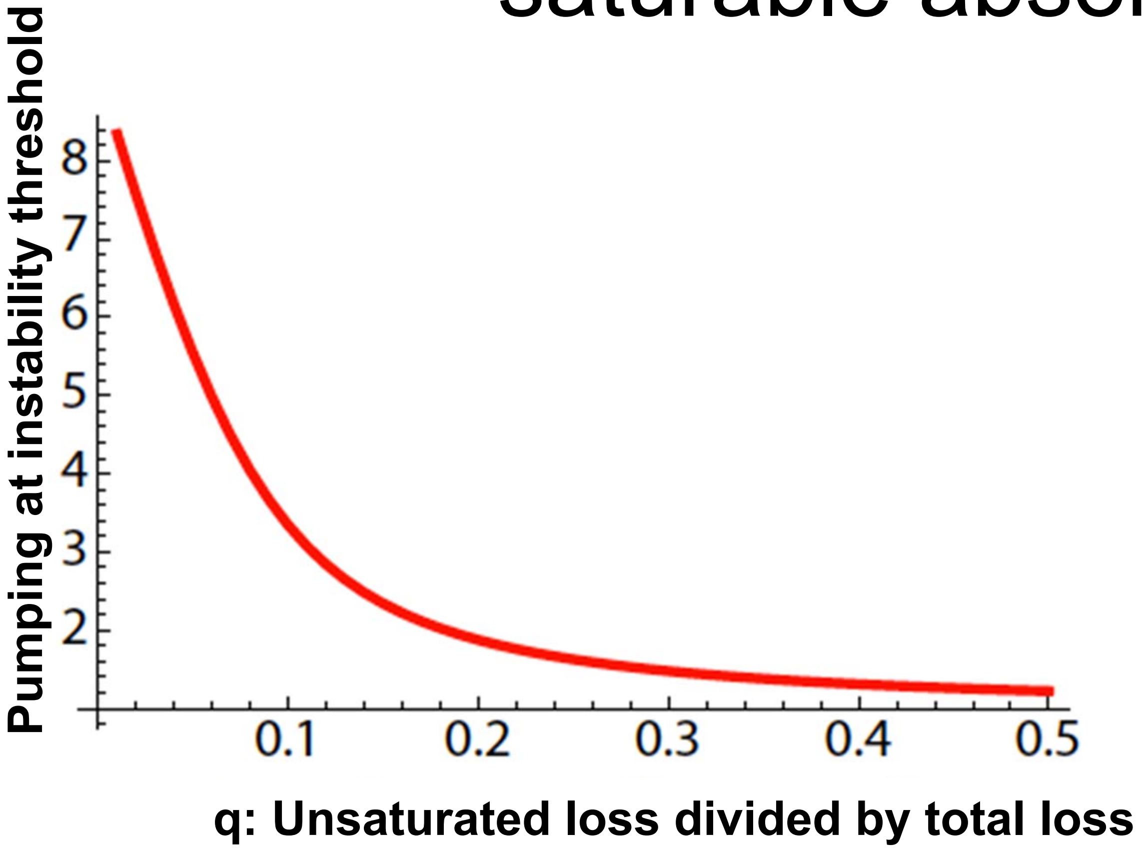
Generation of sidebands in multimode lasers – RNGH instability

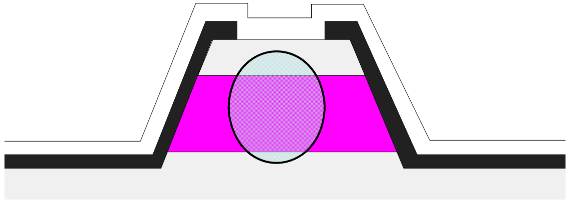
Risken & Nummedal 1968, Graham & Haken 1968

- RNGH instability has been elusive since 1968. No bad cavities, but it still requires pumping 9 times above threshold.
- Claims of observing it in fiber lasers remain controversial.
- Unambiguous observations in QCLs PRA 2007, 2008

Kocharovsky 2001: RNGH instability leads to mode-locked pulses in fiber ring lasers

RNGH threshold is lowered by saturable absorber

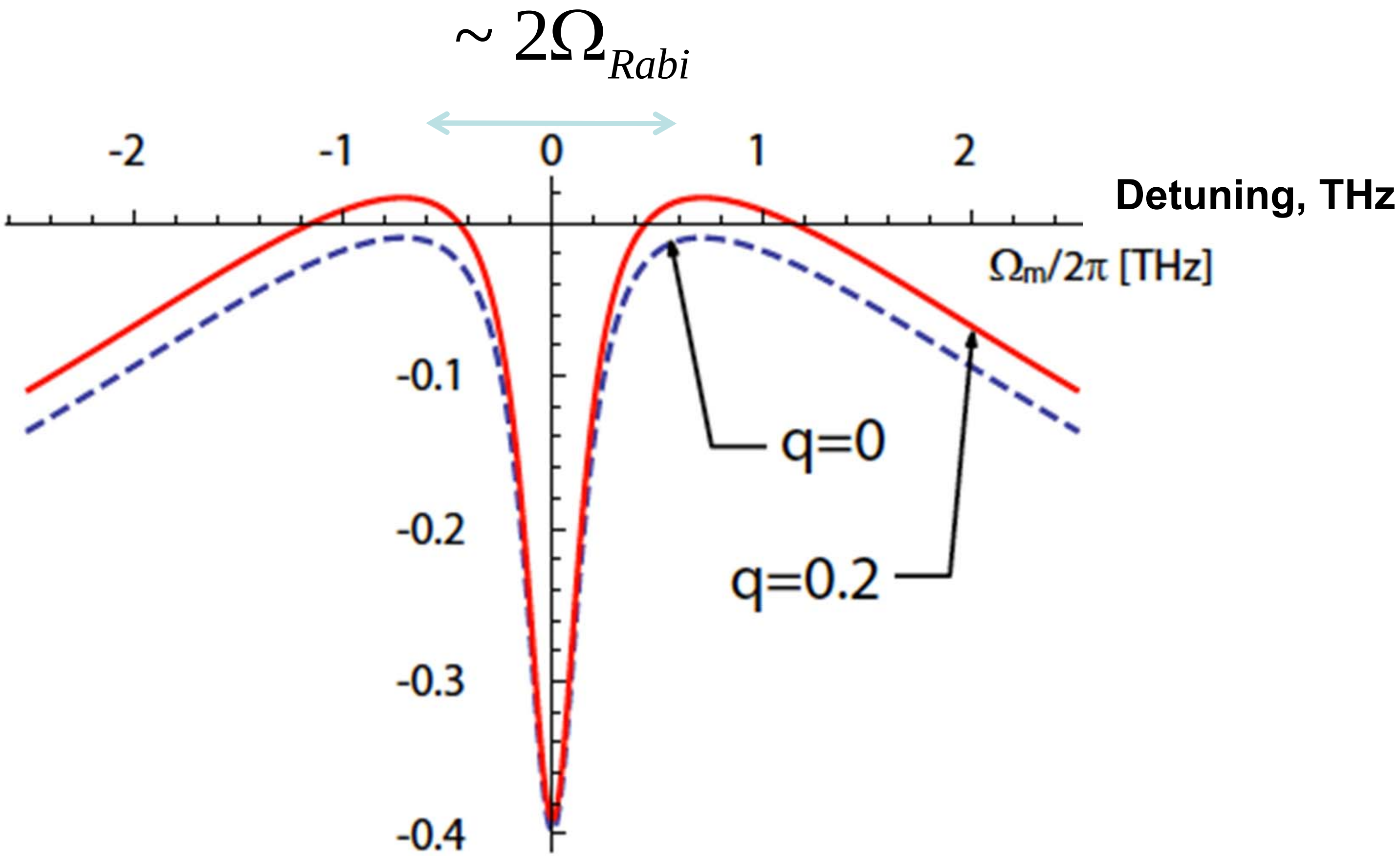




Saturable absorption due to Kerr effect?

PRA 2007,2008; JMO 2011

Nonlinear deformation of the gain spectrum



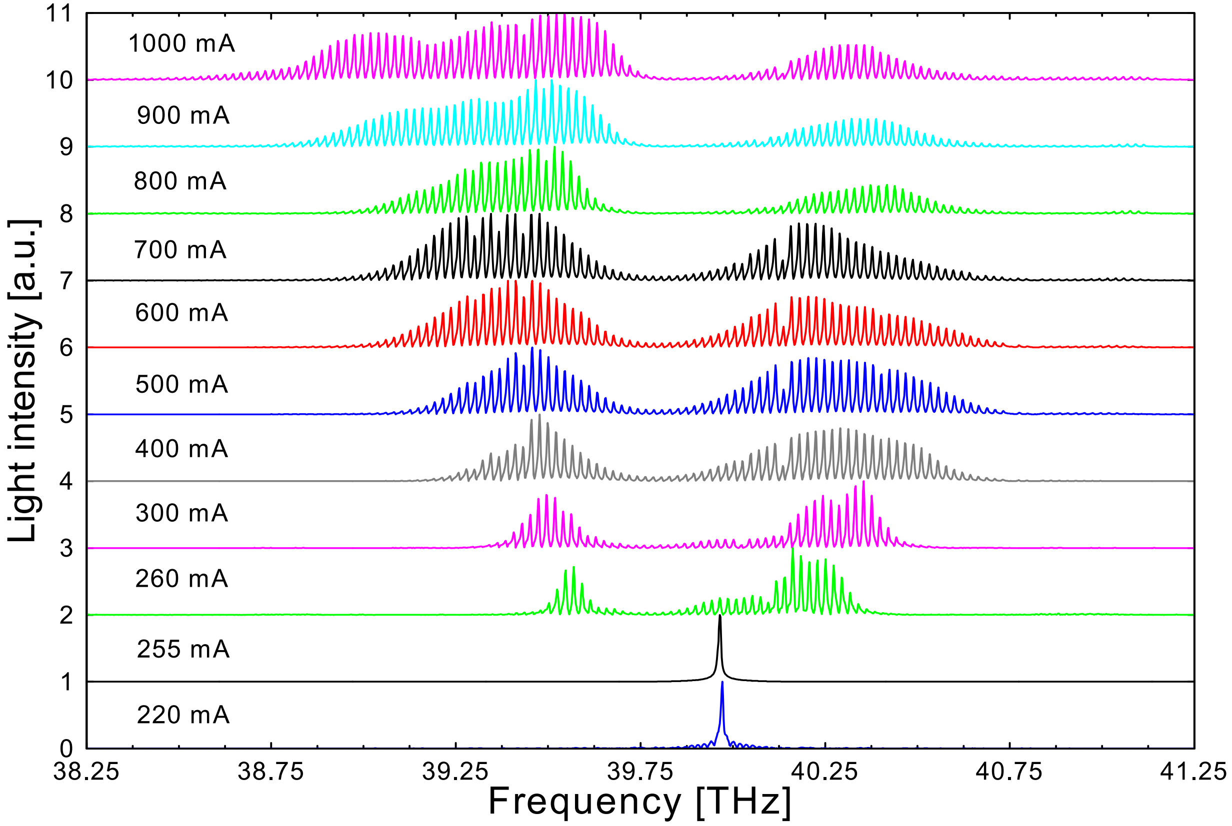
q : normalized loss in a saturable absorber
Pumping is two times above threshold

If q = 0 (no saturable absorber): Instability threshold is: $I_{\mathrm{pump}} > 9\ I_{\mathrm{thr}}$

QCL spectra

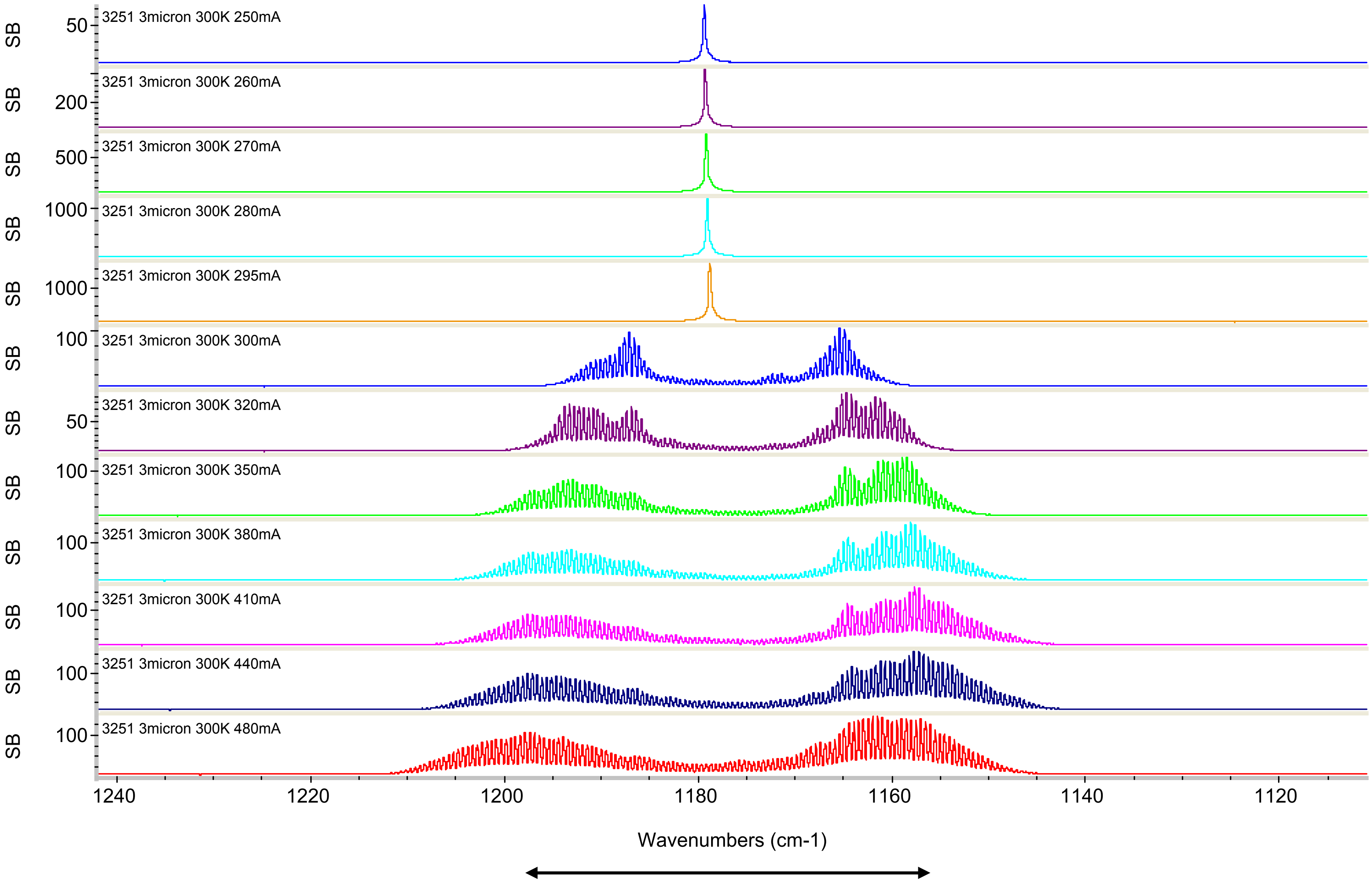
Breaking down of CW single mode into two or multiple spectral humps

Observed in QC lasers of different designs



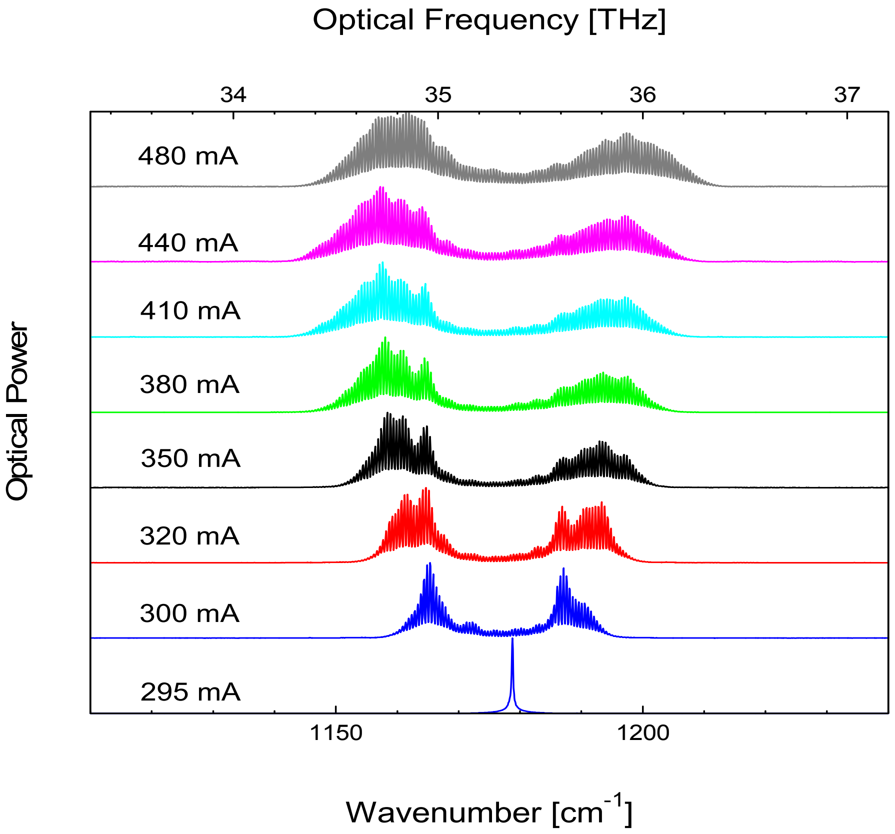
Low threshold ~ 1.2 j_{thr}

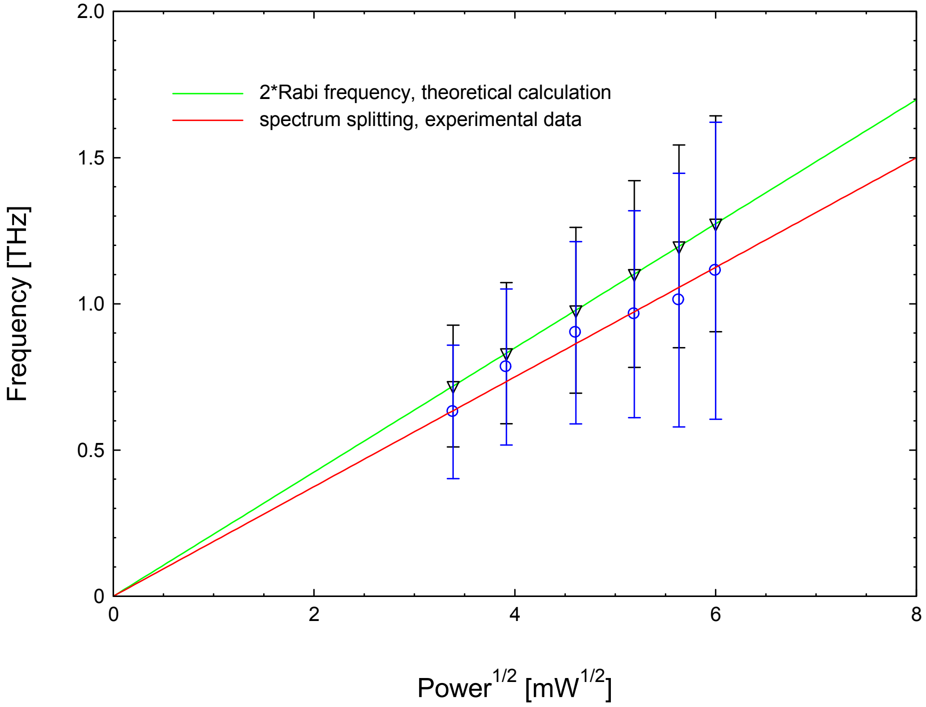
Measurements by Capasso group



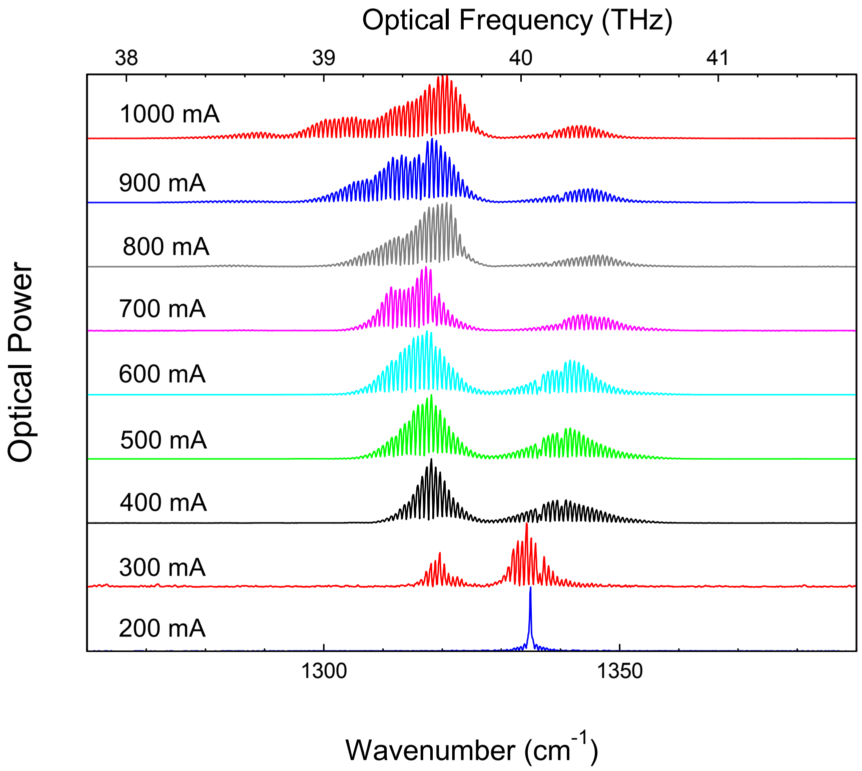
Grows as 2xRabi frequency of the field

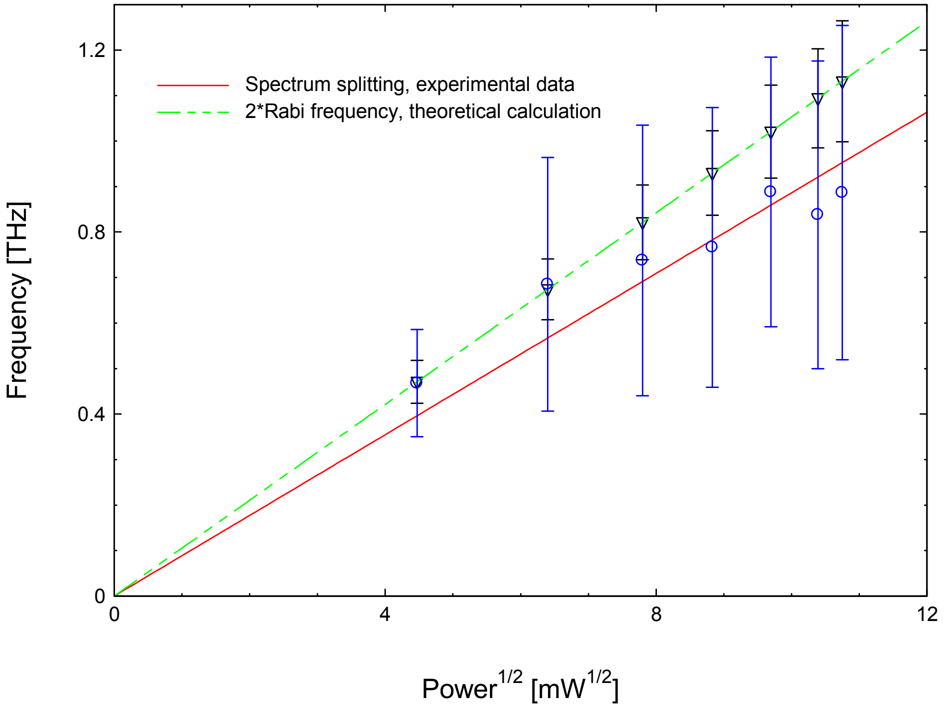
Rabi splitting of the spectra



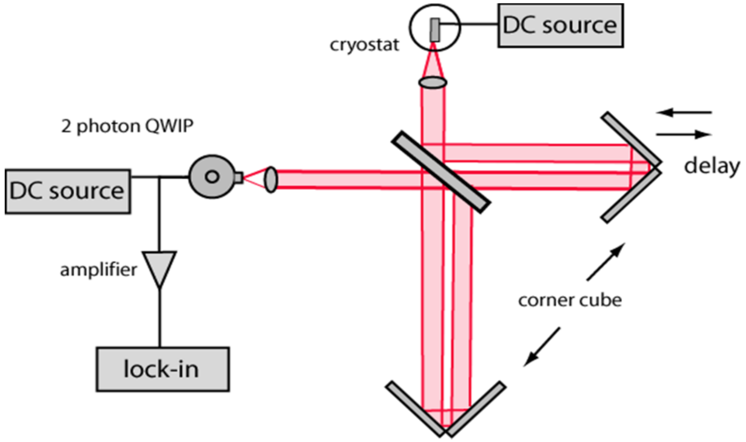


Rabi splitting of the spectra

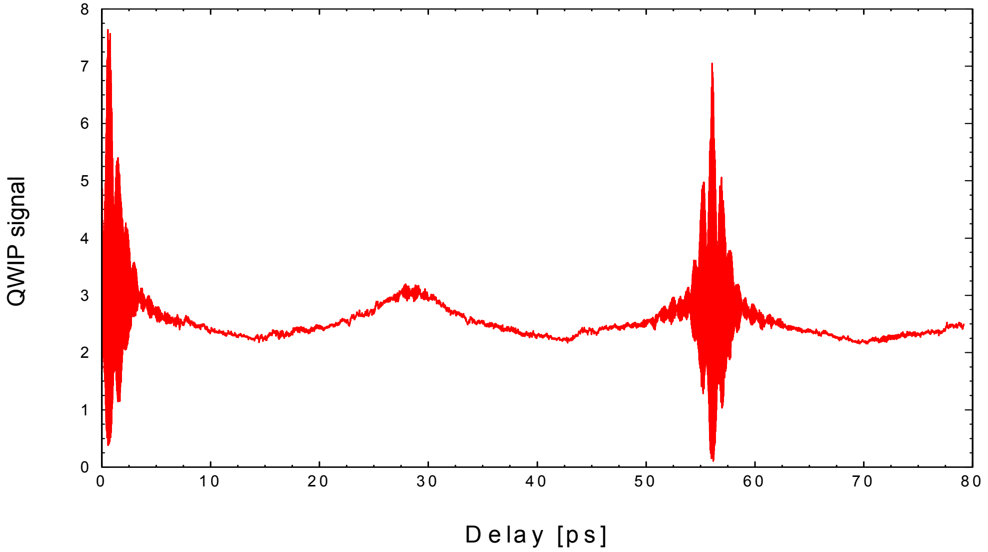




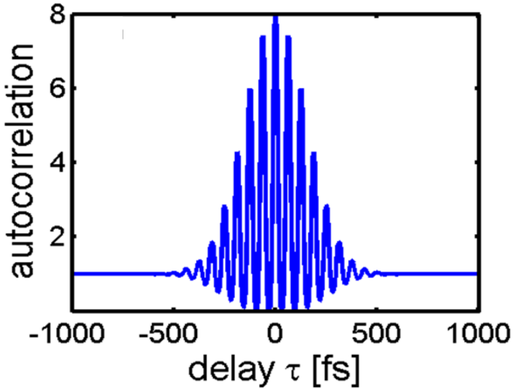
Nonlinear interferometric autocorrelation



Gordon et al. PRA 2008



autocorrelation trace, sample 2743
10 μm wide ridge 77K; current 900mA

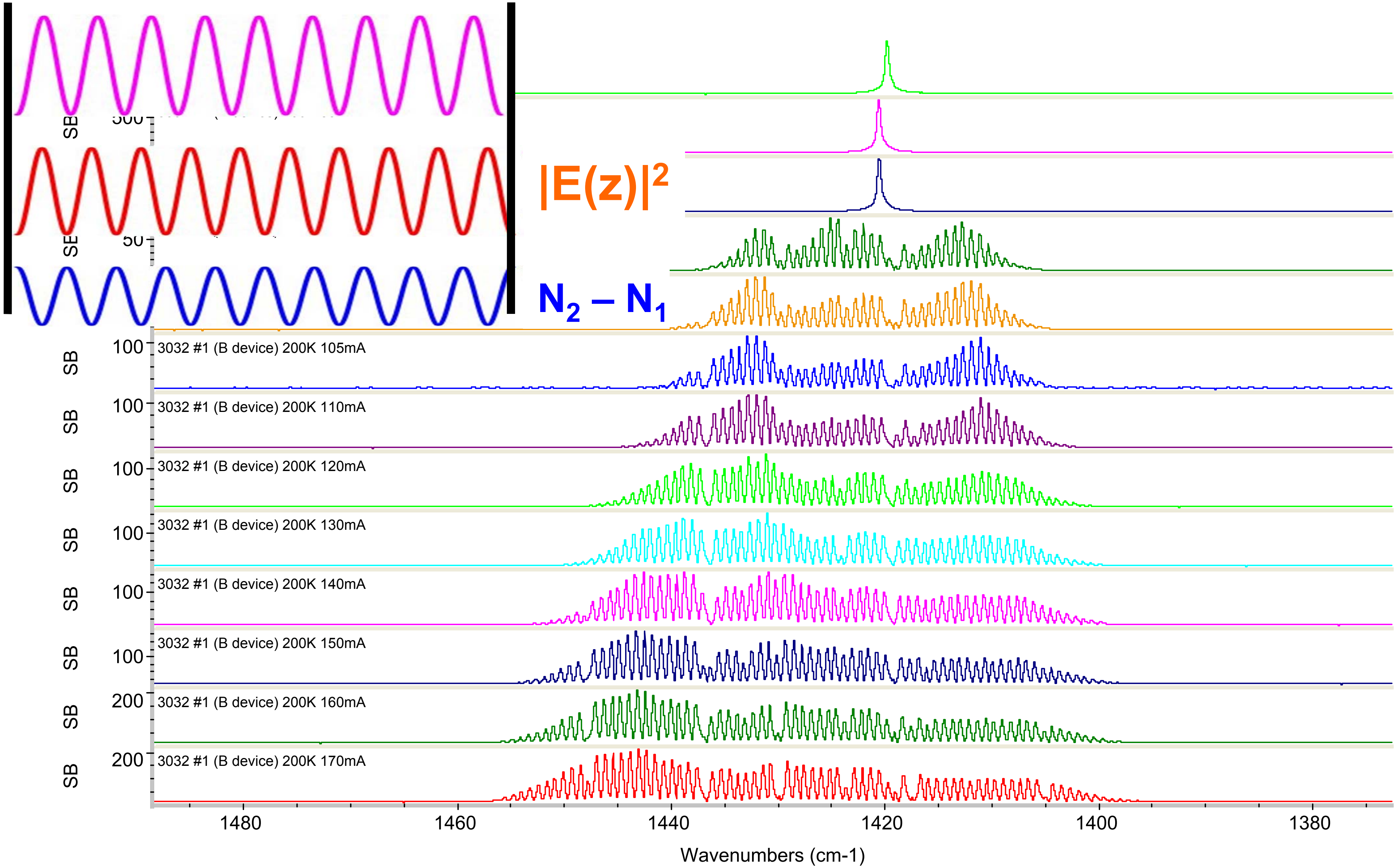


Single pulse, theory

Ratio is not 1:8 – multi-pulse regime or no pulses at all?

Reason: short $T_1 << T_c$? Spatial hole burning?

Spatial hole burning may prevent mode locking



Conclusions

- QCLs show rich nonlinear dynamics and phase-coherent phenomena
- Stable phase locking and synchronization of lateral modes
  - Requires nearly equal thresholds for several lateral modes (buried heterostructure laser);
  - 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?