ETH zürich

Quantum cascade lasers frequency combs in the mid-infrared and terahertz

Jerome Faist

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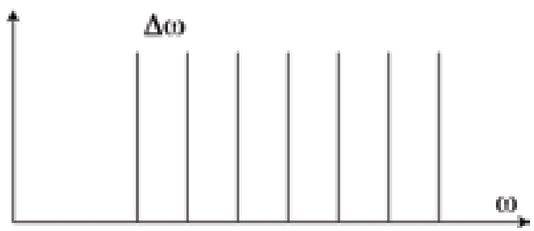
Optical frequency comb

- Source with equidistant optical modes



- Incredibly useful!
  - Links the accuracy of an optical transition (10^{-18}) to the one of a microwave signal

LETTER
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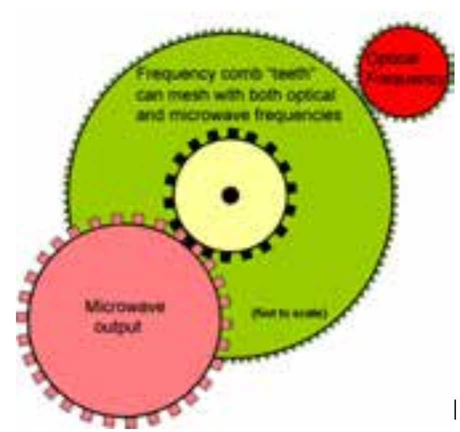
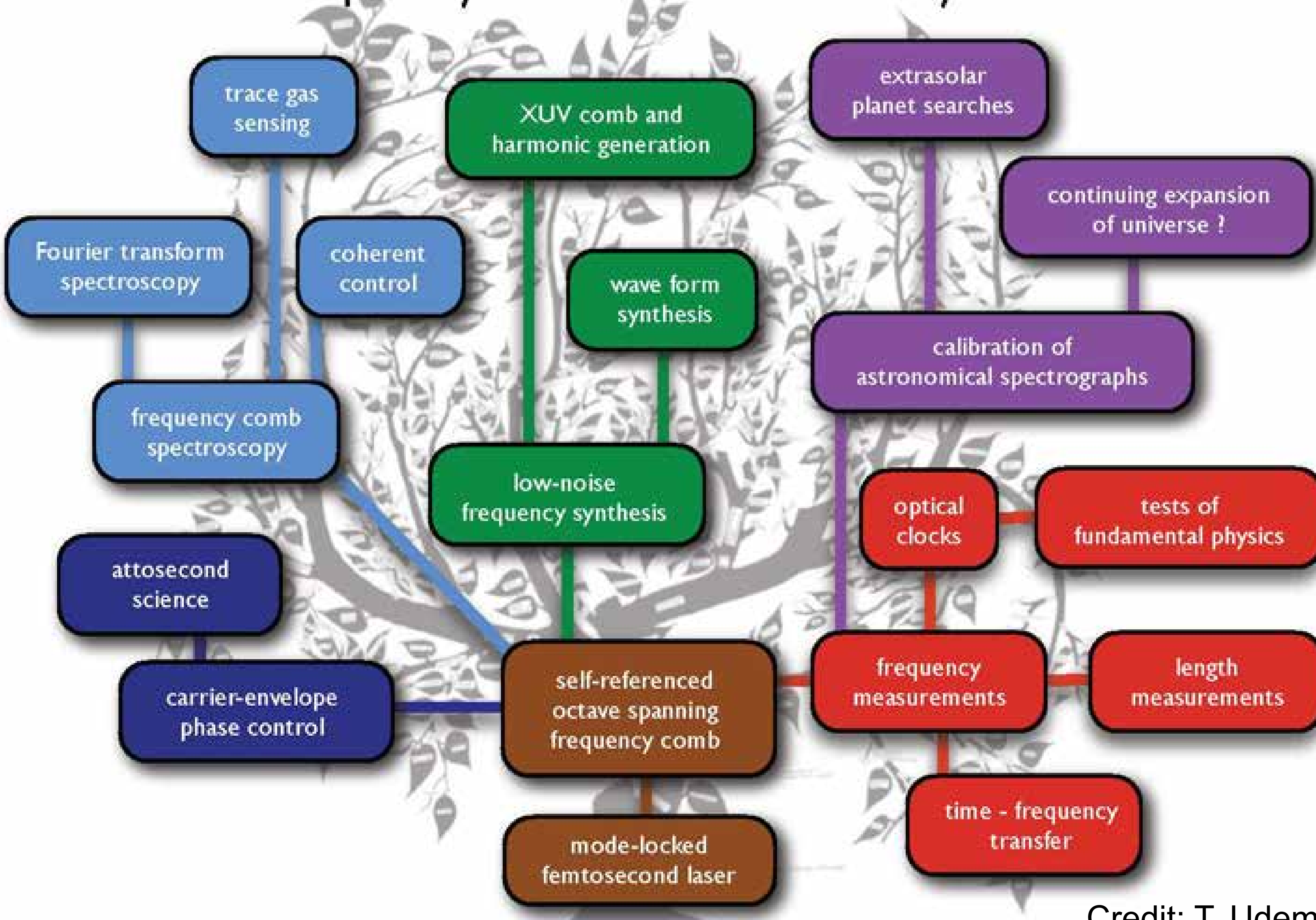
An optical lattice clock with accuracy and stability at the 10^{-18} level

B. J. Bloom[1,2]*, T. L. Nicholson[1,2]*, J. R. Williams[1,2]†, S. L. Campbell[1,2], M. Bishof[1,2], X. Zhang[1,2], W. Zhang[1,2], S. L. Bromley[1,2] & J. Ye[1,2]

Both clocks stabilize their lattice laser frequencies to a Cs clock via a self-referenced Yb fibre comb, and their trapping light intensities were stabilized after being delivered to the atoms. The lattice vector shift was

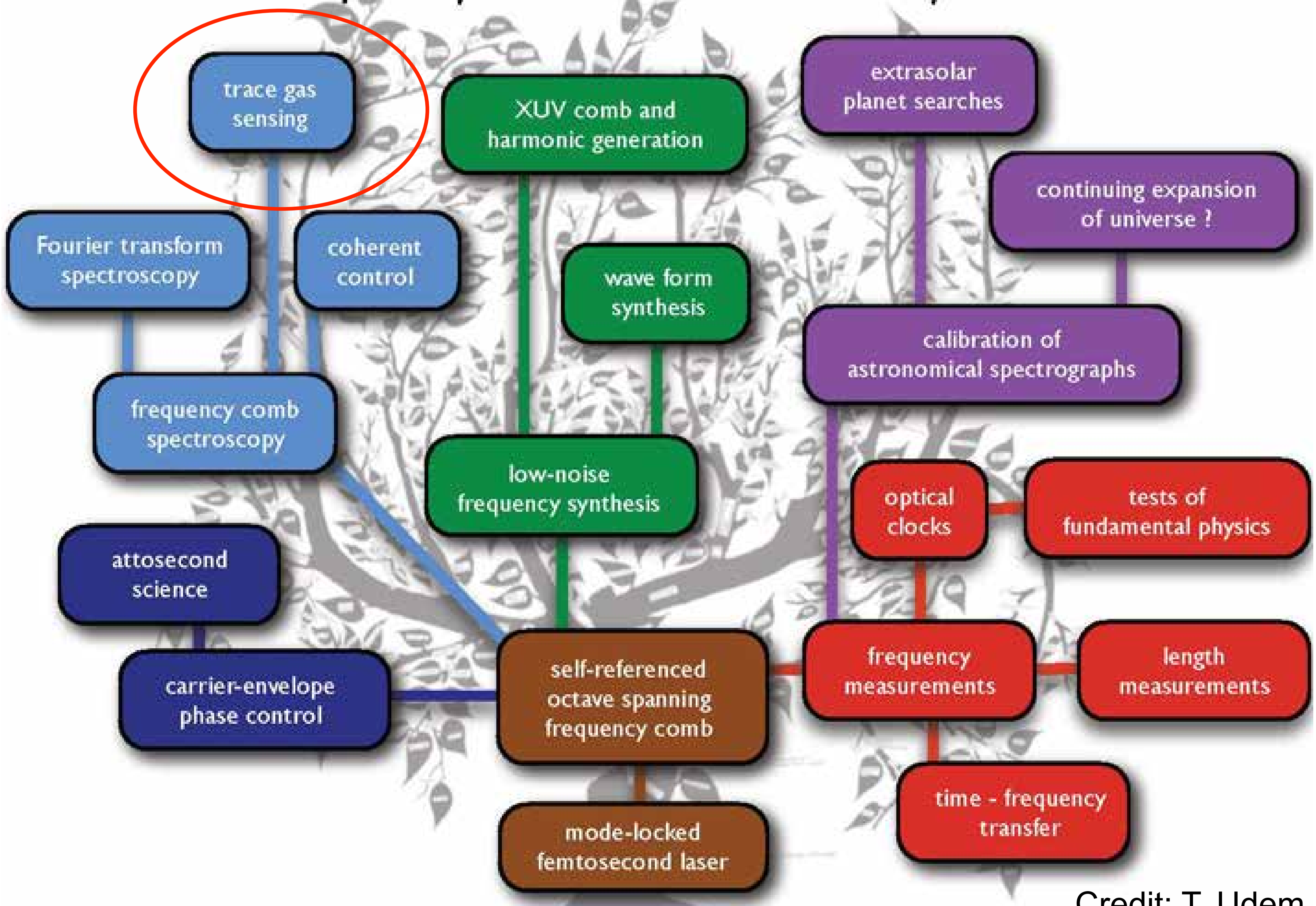


Frequency combs - evolutionary tree



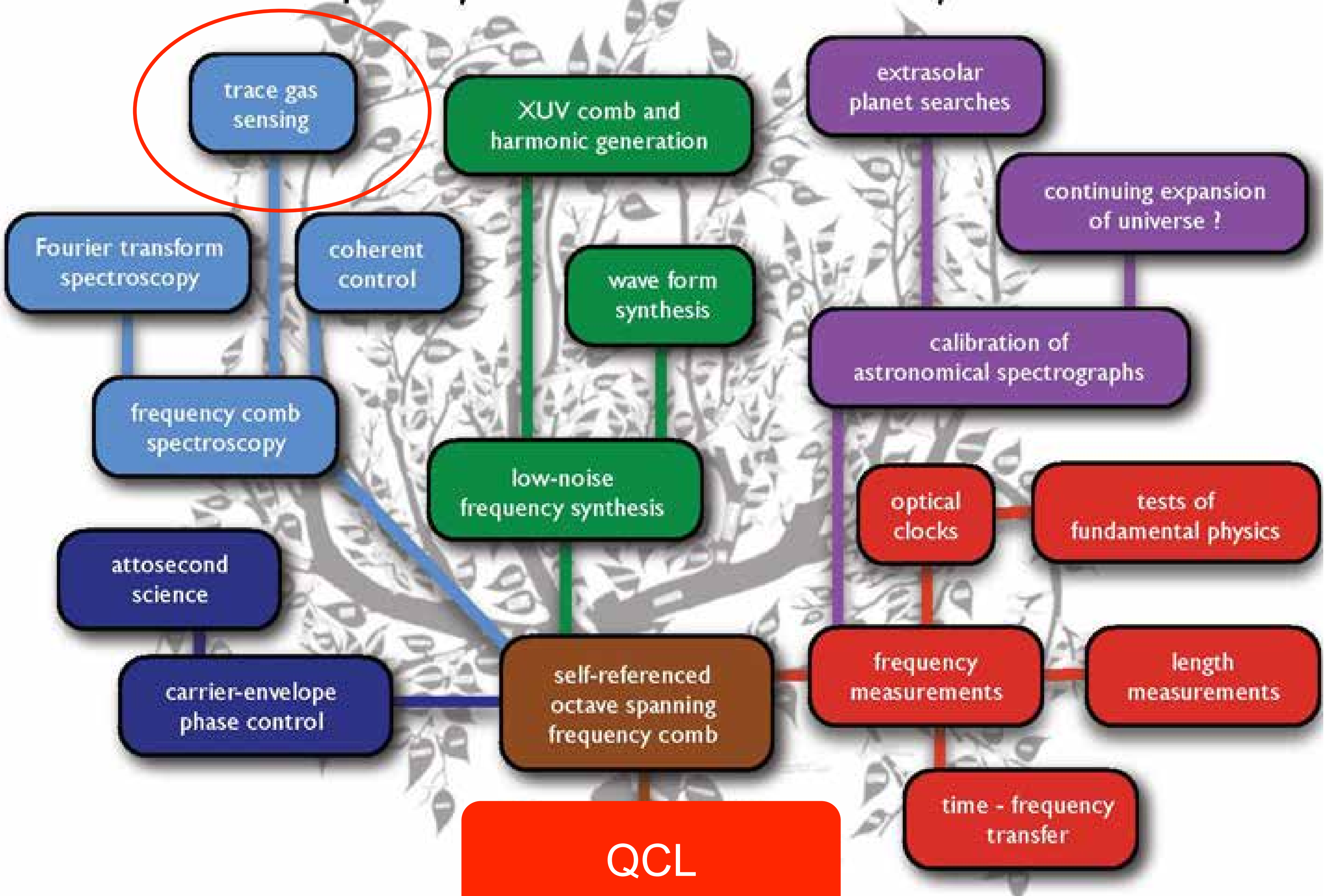
Credit: T. Udem

Frequency combs - evolutionary tree



Credit: T. Udem

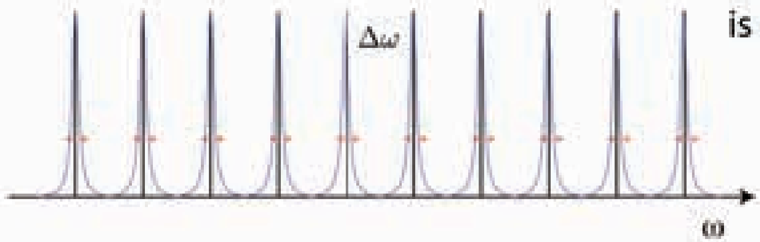
Frequency combs - evolutionary tree



Credit: T. Udem

What is a comb?

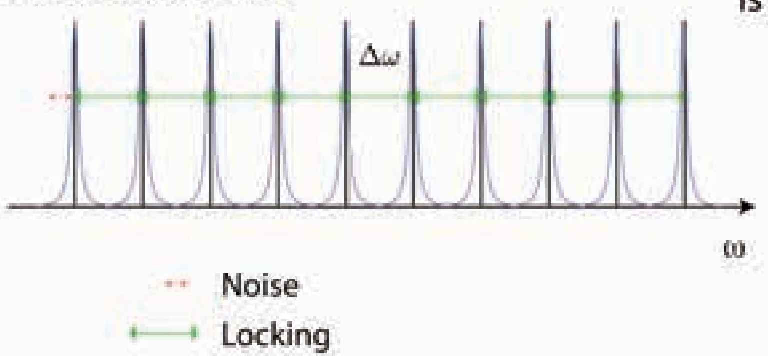
Array of N independent DFBs combined:



line-to-line frequency noise is uncorrelated

$$\delta(\omega_i - \omega_j) \approx \sqrt{2}\delta(\omega_i)$$

Optical frequency comb:



line-to-line frequency noise is correlated

$$\delta(\omega_i - \omega_j) << \delta(\omega_i)$$

The beat tone has a very narrow linewidth in a comb

Still, the array source may still be very interesting for some applications!!!

Need to lock the modes!

First mode locking

- Christiaan Huygens noticed that two pendulum clocks would synchronized themselves if hanged at the same mantelpiece
- Due to a small mechanical coupling between the pendulum

Christiaan Huygens (1629-1695)

Can be generalized to many pendulum!

- Here for 32 metronomes are synchronised

Works also for laser

- Injection locked lasers

Locking range:

Adler's formula

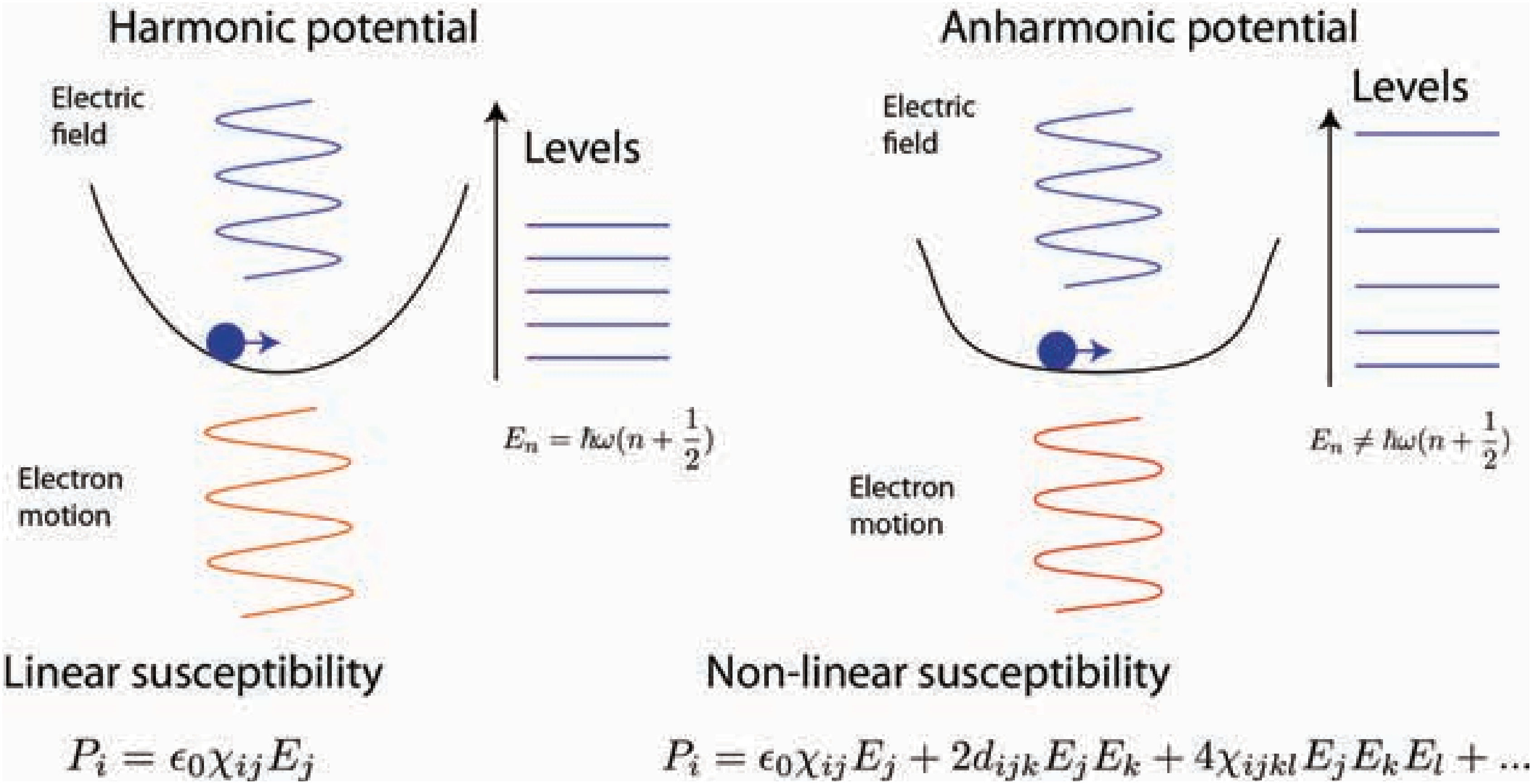
$$\omega' = \omega_0 \quad \text{if} \quad \omega' - \omega_0 < \Delta\omega_{lock} \quad \text{with} \quad \Delta\omega_{lock} = \frac{2\omega_0}{Q}\sqrt{\frac{P_{inj}}{P_0}}$$

- How do you get to lock different frequencies?

R. Adler, "A study of locking phenomena in oscillators," Proc. IEEE **61**(10), 1380–1385 (1973)

Use non-linearities that couple different frequencies

Non-linear polarization



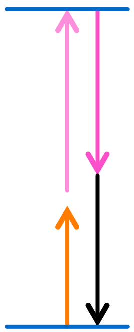
Two – level systems are non-linear!

- Optical gain in an harmonic potential is not possible
  - Active material must be also non-linear!
- Two-level systems have (at least) third order non-linearities
- The latter will generate sidebands

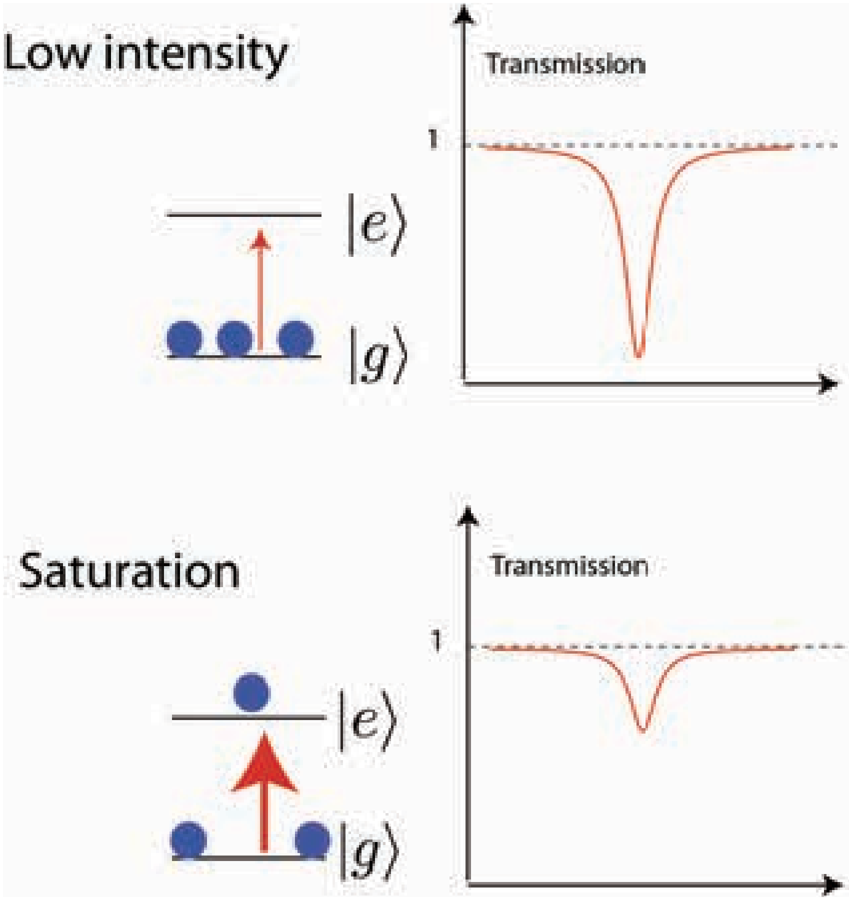
$$P_{NL}(\omega;\omega_1,\omega_2,\omega_3) = 4\chi_{ijkl}E_jE_kE_le^{i(\omega_1\pm\omega_2\pm\omega_3)t}$$

- Energy conservation for single frequency fields

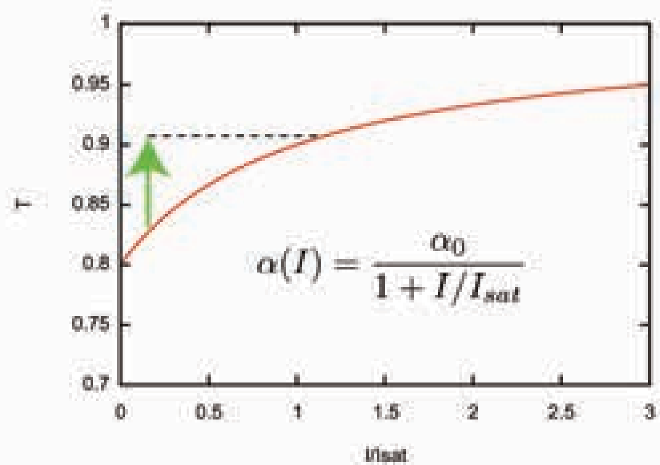
$$\omega = \omega_1 \pm \omega_2 \pm \omega_3$$



Another non-linearity: saturable absorption



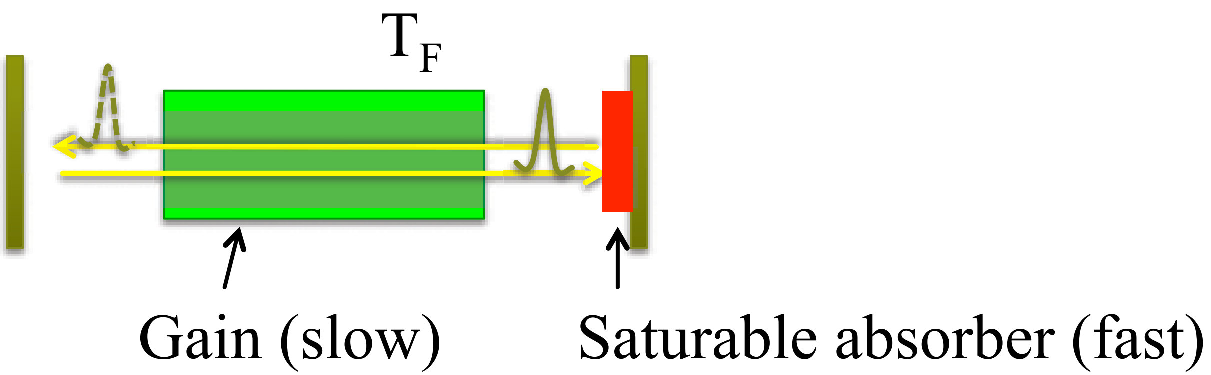
Saturation of the absorption



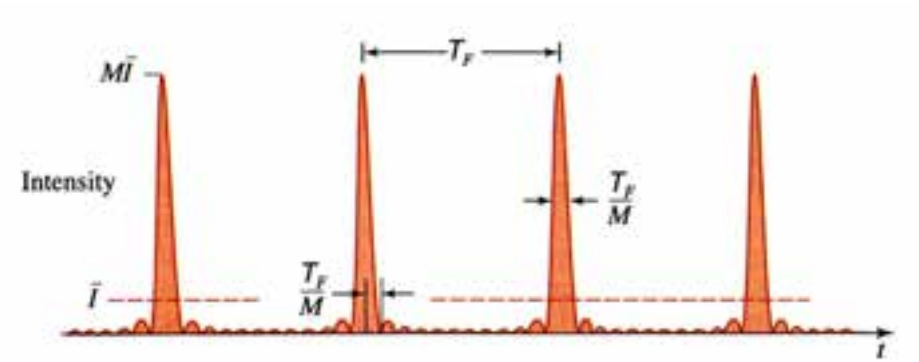
The transmission of high-intensity pulses is enhanced

Mode-locking

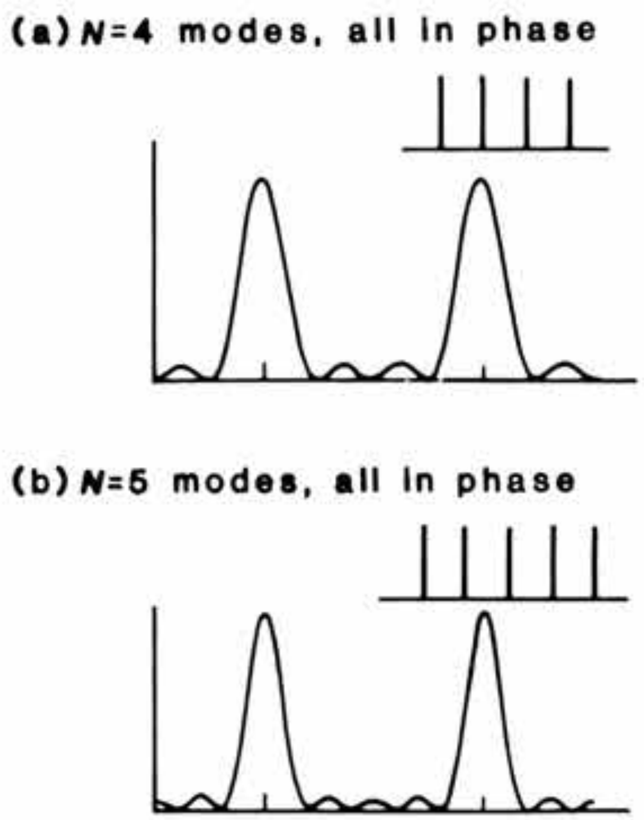
- Using a saturable absorber to mode-lock
  - Round trip gain for pulse is larger than for c.w. operation

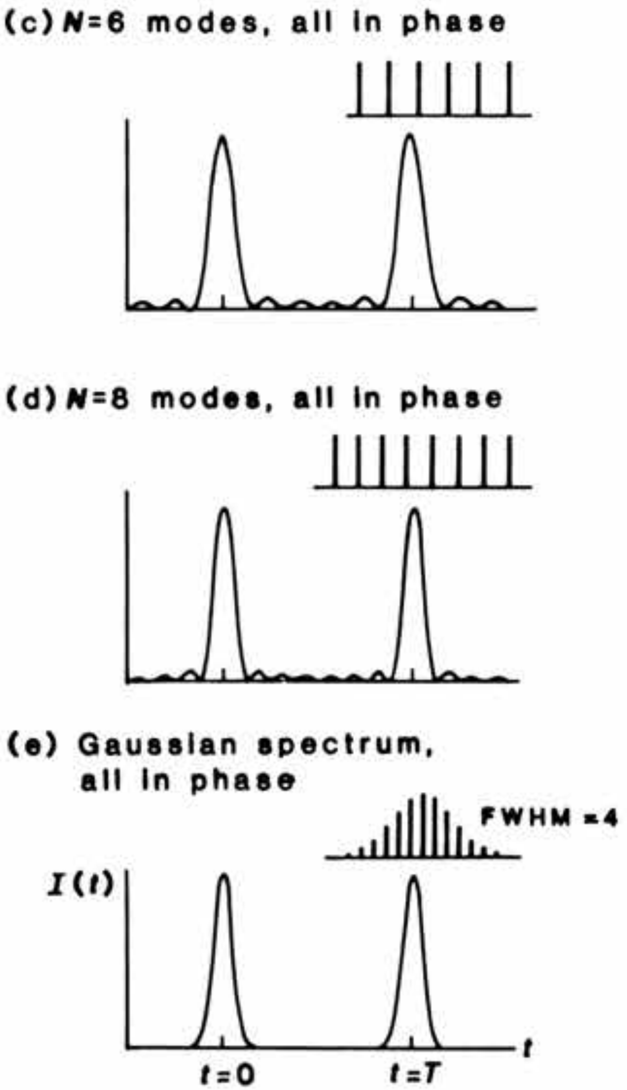


- Result: train of pulses



Locking of N modes





Time domain picture

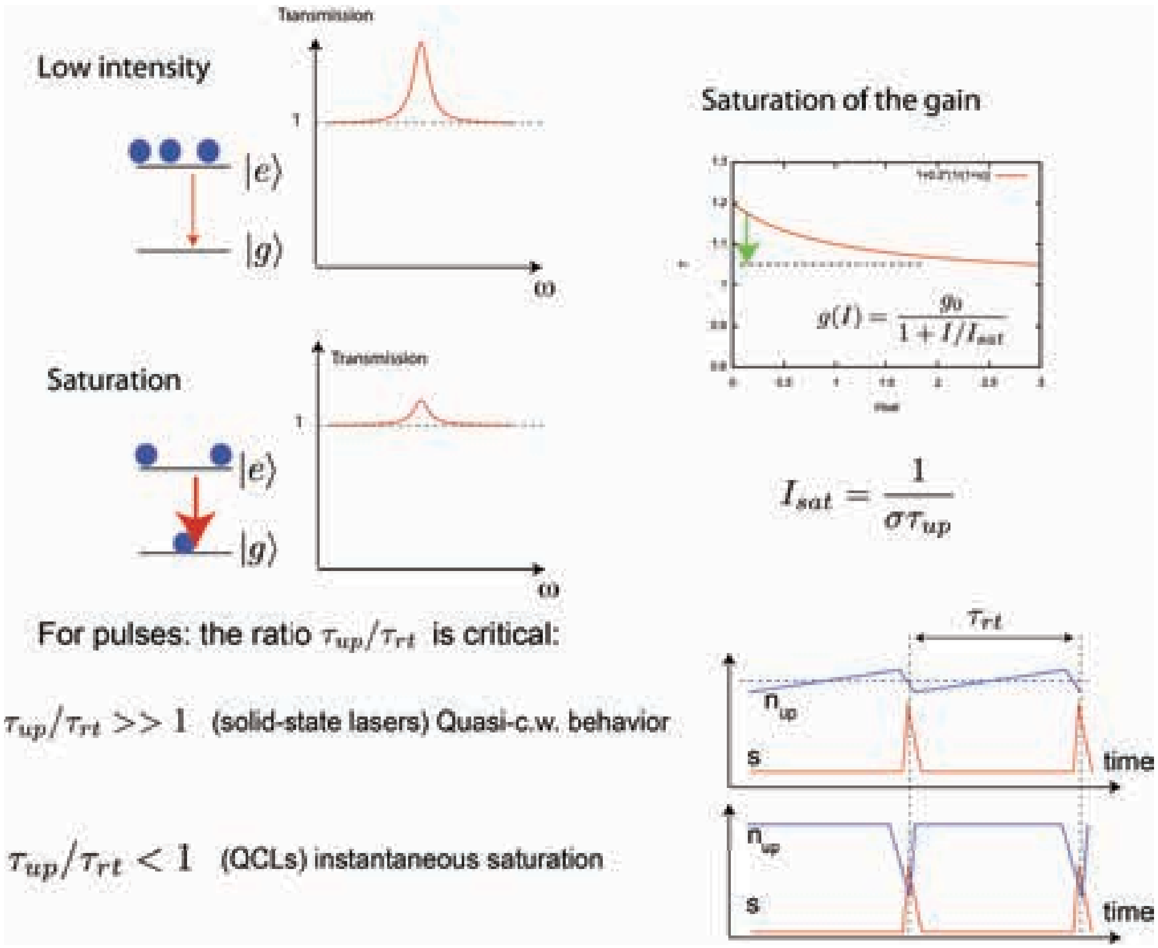
- Time domain approach (fast saturable absorber)
  - Equation:

$$\frac{1}{T_R}\frac{\partial}{\partial T}a = (g-\ell)a + \frac{g}{\Omega_g^2}\frac{\partial^2}{\partial t^2}a + \gamma|a|^2 a$$

  - Solution $a_o(t) = A_o\,\mathrm{sech}(t/\tau)$
  - Pulse duration: $\tau = \sqrt{\frac{2g}{\gamma A_0^2}}\frac{1}{\Omega_g}$ ~ inverse gain bandwidth
    - For broadband QCL, would be 50-100fs
  - Equation similar for Kerr combs

H. Haus, "Mode-locking of lasers," *IEEE J Sel Top Quant*, vol. 6, no. 6, pp. 1173–1185, 2000.

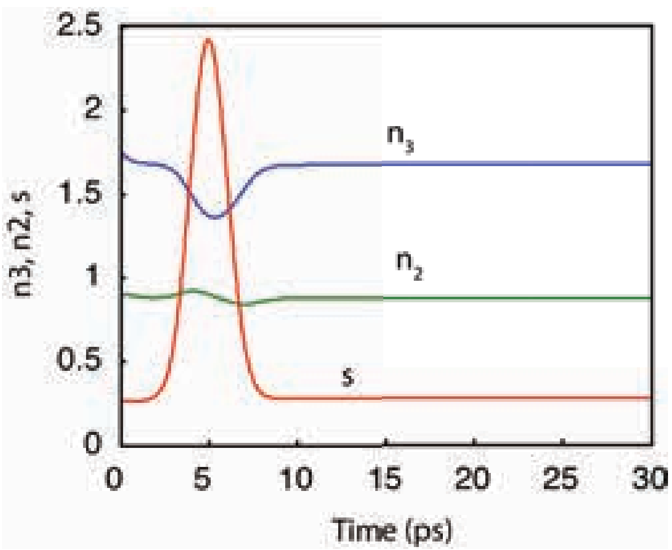
Saturation also valid for the gain!

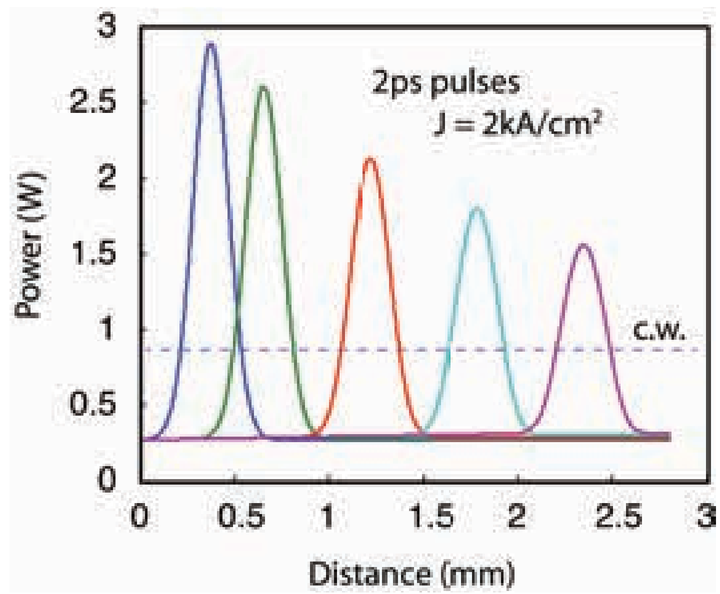


In QCL the gain saturates fast

- Very short upper state lifetime -> cannot store energy

Simulation of a pulse propagating in a QCL (rate equation)





- Passive mode-locking very difficult..

QCLs can be modulated fast (active ML)

- No resonance, low pass filter transfer function

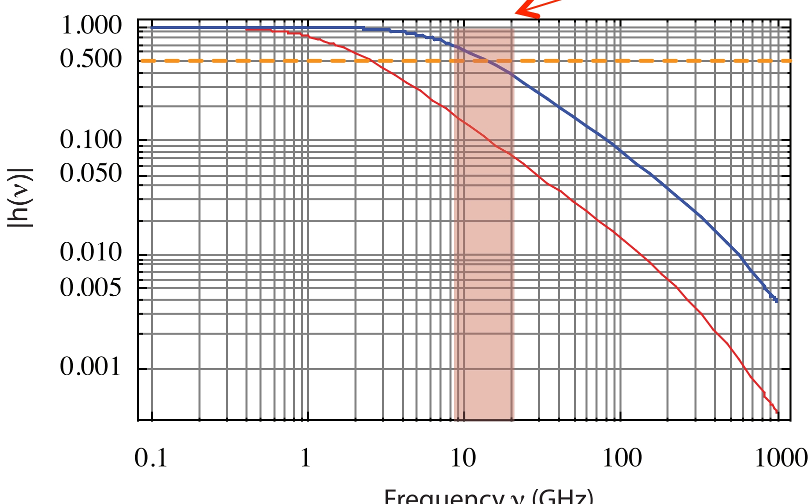
$$|h(\omega')|^2 = \frac{1}{1+\omega'^4+\omega'^2\left(\frac{\tau_p}{\tau_{stim}}+2\frac{\tau_p}{\tau_{up}}+\frac{\tau_p\tau_{stim}}{\tau_{up}^2}-2\right)}$$

- However very short τ_{stim} at high powers

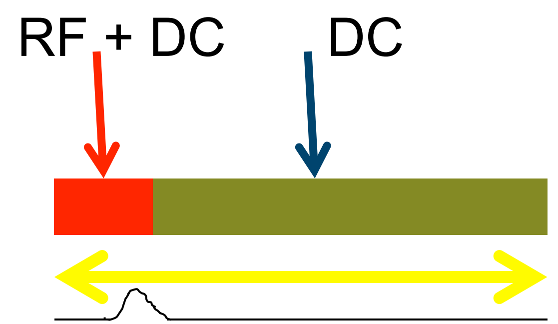
R. Paiella, (2000-2002)
S. Barbieri, C. Sirtori (2007-)
B. Hinkov, IQCSW poster

$$\omega'^2 = \omega^2\tau_{stim}\tau_p,$$

Round trip frequency (3-6mm L_{cav})

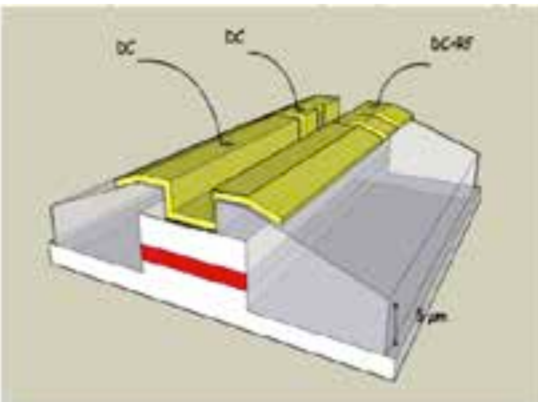


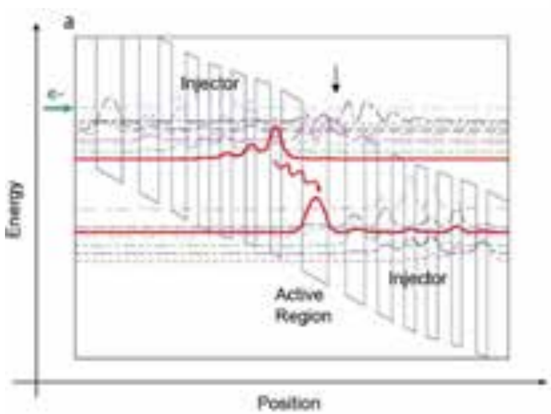
Two-section device

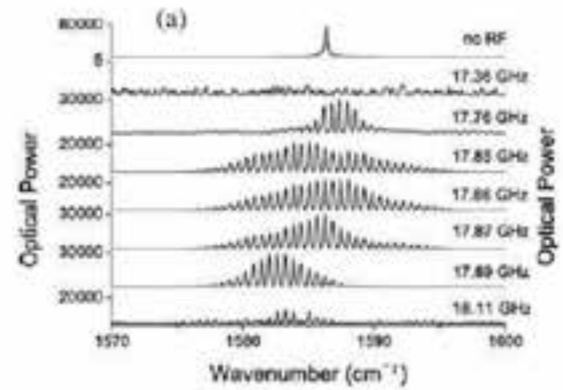


Active mode locking in QCLs:

- Designs with long upper state lifetime

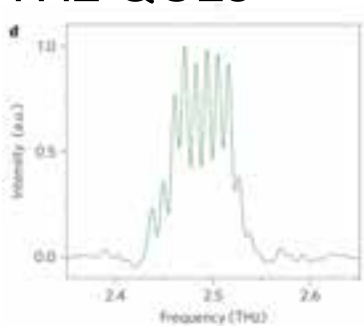


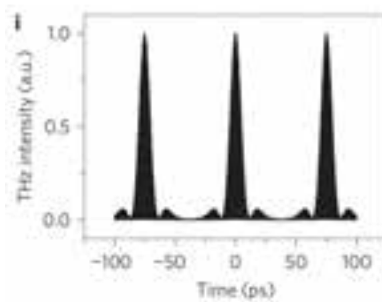




C. Y. Wang,et al., ” *Opt Express*, vol. 17, no. 15, pp. 12929–12943, 2009.

- THz QCLs





S. Barbieri et al., *Nature Photon*, vol. 5, no. 5, pp. 306–313, 2011.

Limitations of active mode-locking:

- The minimum pulse width τ now depends also on the round-trip frequency Ω_m
  - Neglect the gain saturation -> will make things worst

$$\tau = \sqrt[4]{\frac{2g}{M\Omega_m^2\Omega_g^2}}$$

- As a result, one gets 1-2ps pulses for typical values
  - 3mm device, modulation depth M=20%, Ω_g = 200cm^{-1} FWHM gain

A. Siegman
H. Haus, "Mode-locking of lasers," *IEEE J Sel Top Quant*, vol. 6, no. 6, pp. 1173–1185, 2000.

Another possible picture

- Time domain approach (fast saturable absorber)
  - Equation:

$$\frac{1}{T_R}\frac{\partial}{\partial T}a = (g-\ell)a + \frac{g}{\Omega_g^2}\frac{\partial^2}{\partial t^2}a + \gamma|a|^2 a$$

Pulse duration

$$\tau = \sqrt{\frac{2g}{\gamma A_0^2}}\frac{1}{\Omega_g}$$

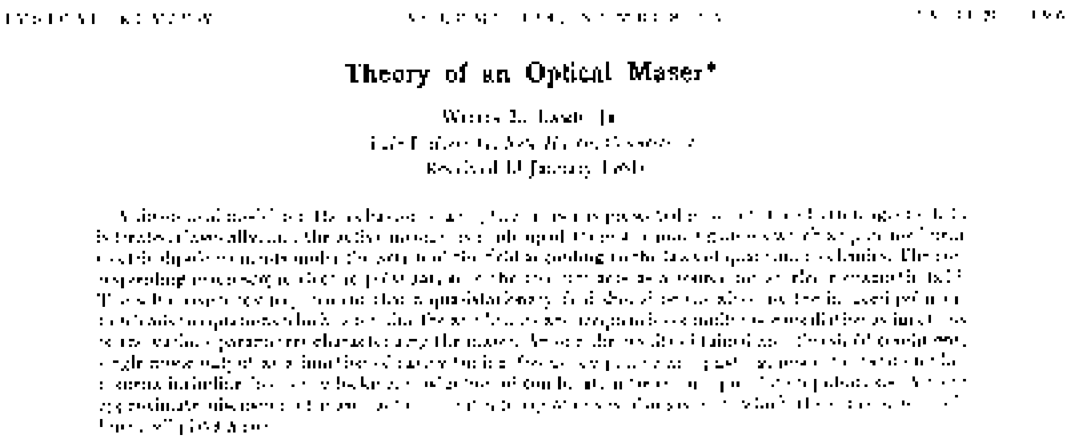
  - Solution $a_a(t) = A_a\,\mathrm{sech}(t/\tau)$

$$\frac{1}{\tau^2} = \frac{\gamma A_a^2 \Omega_g^2}{2g}$$

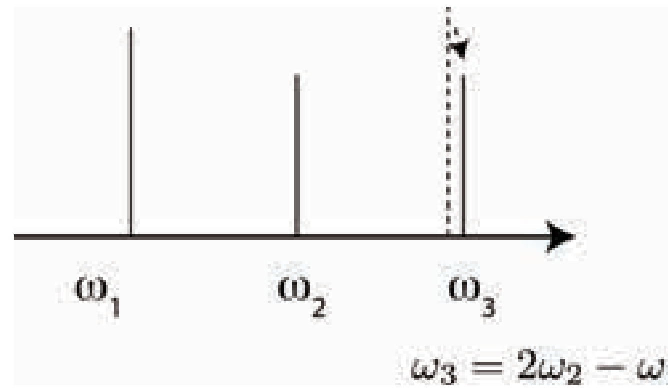
  - Similar for Kerr combs
- Frequency domain approach (modal decomposition)
  - Coupling of discrete modes

Three mode laser

- Taken from a classic paper:

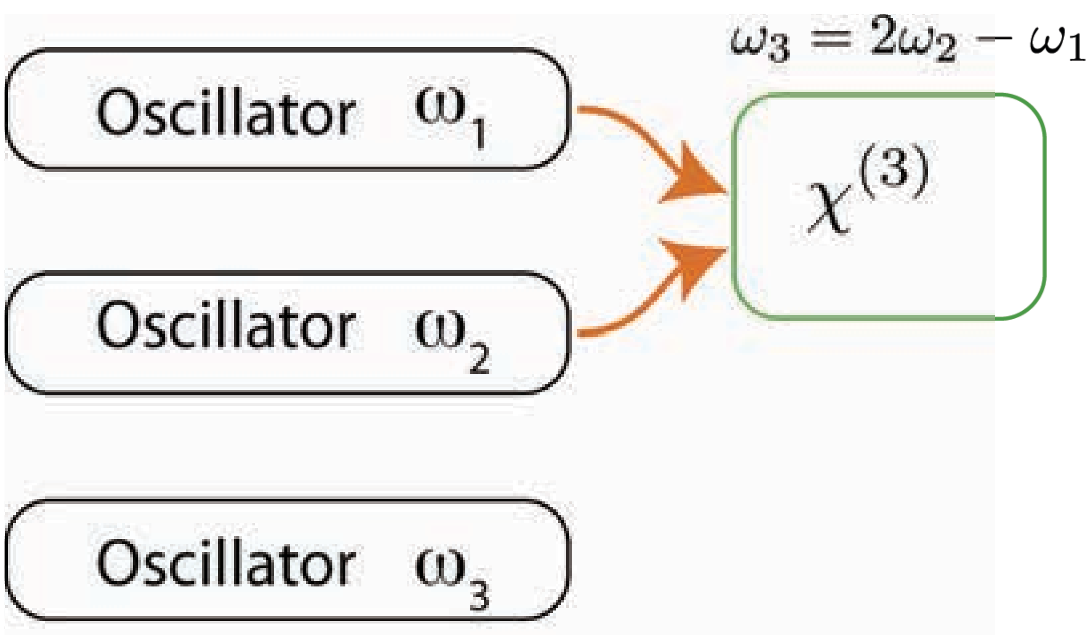


Theory of an Optical Maser*

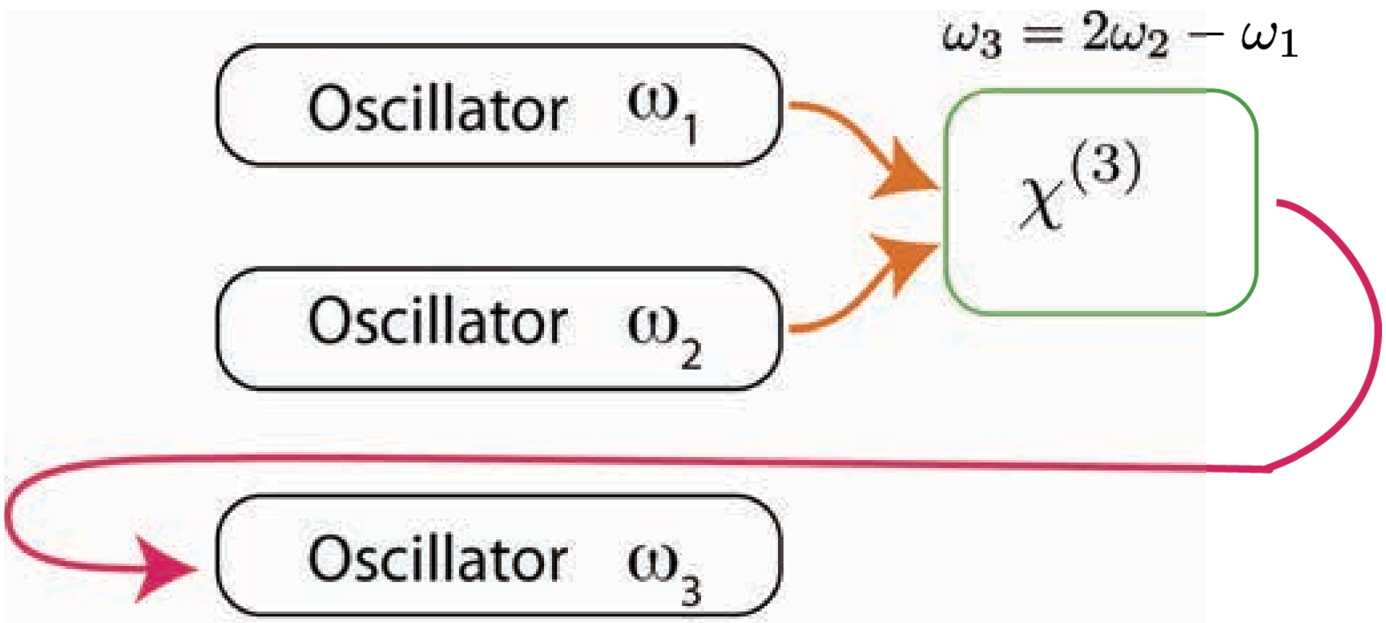


- Injection at ω_3 will yield three mode operation (with equidistant frequencies)

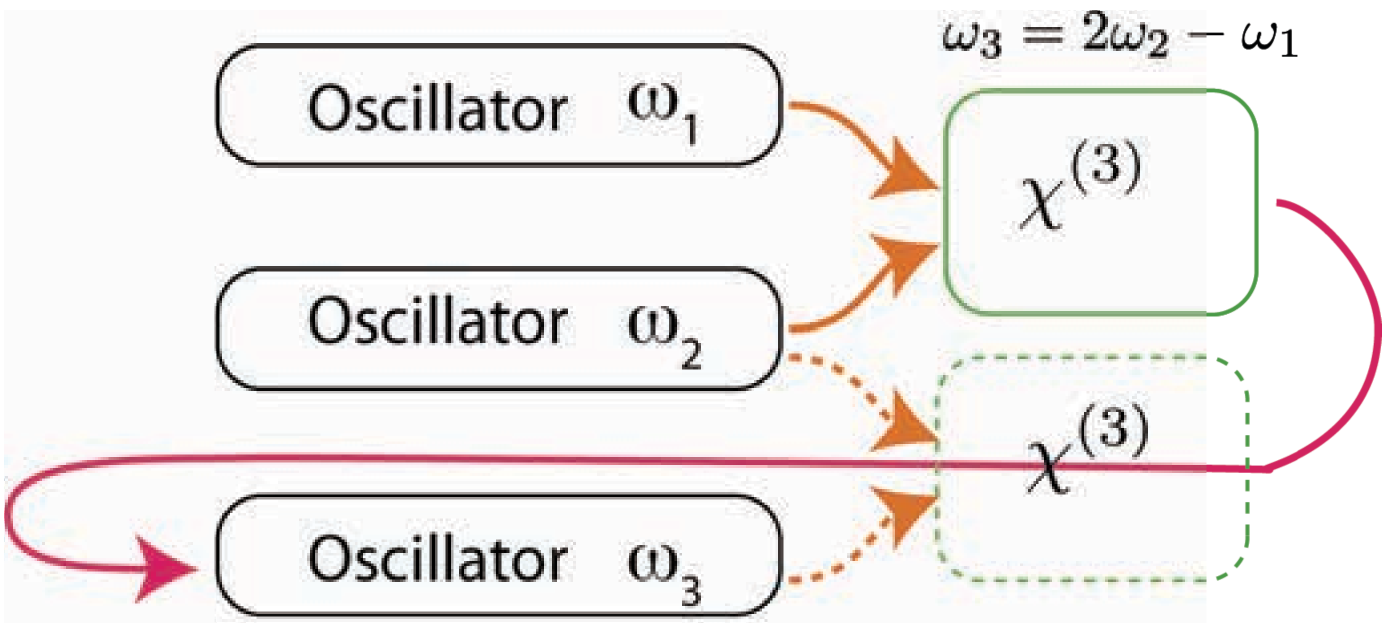
Mode-locking as a injection locking



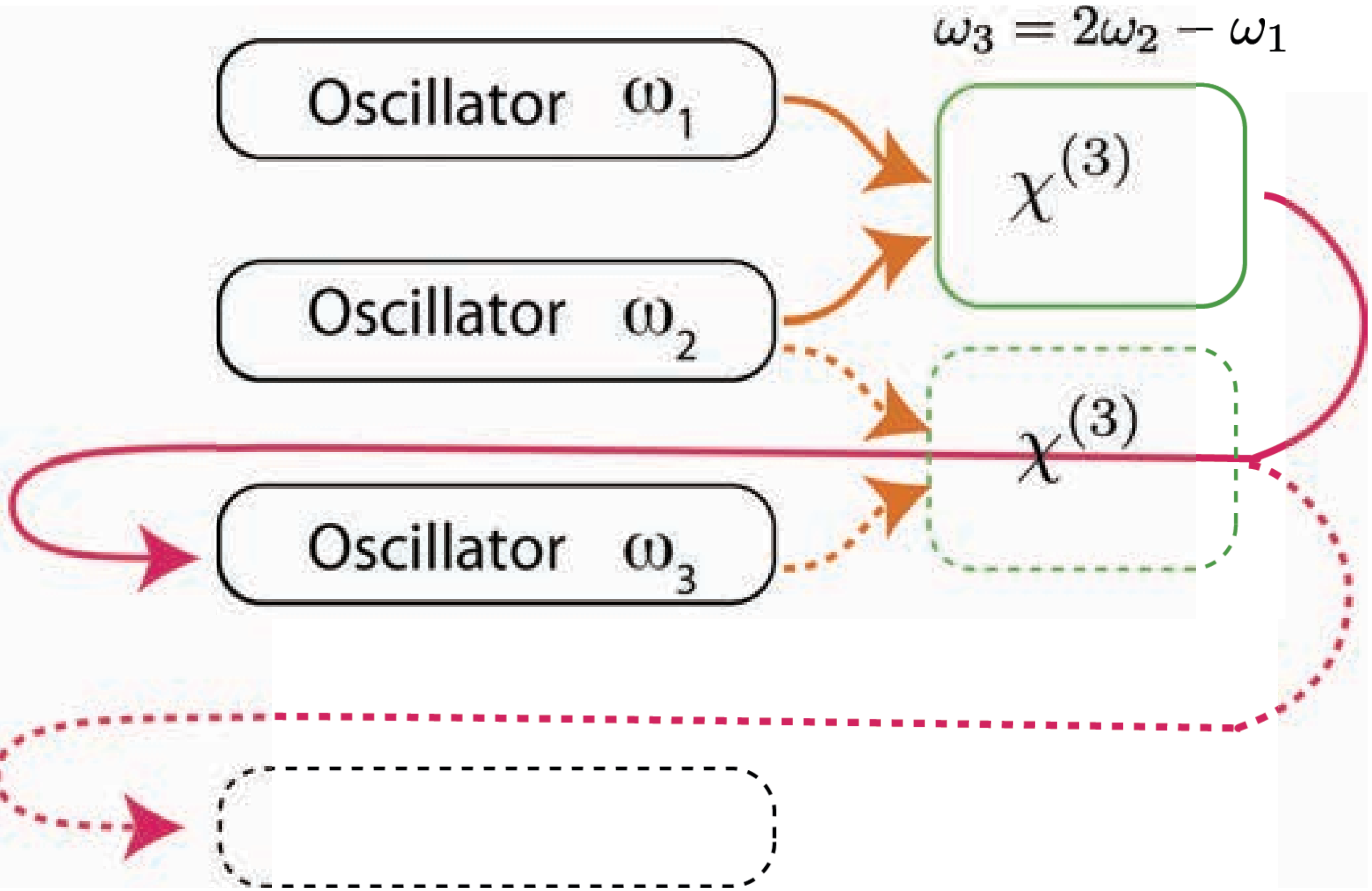
Mode-locking as a injection locking



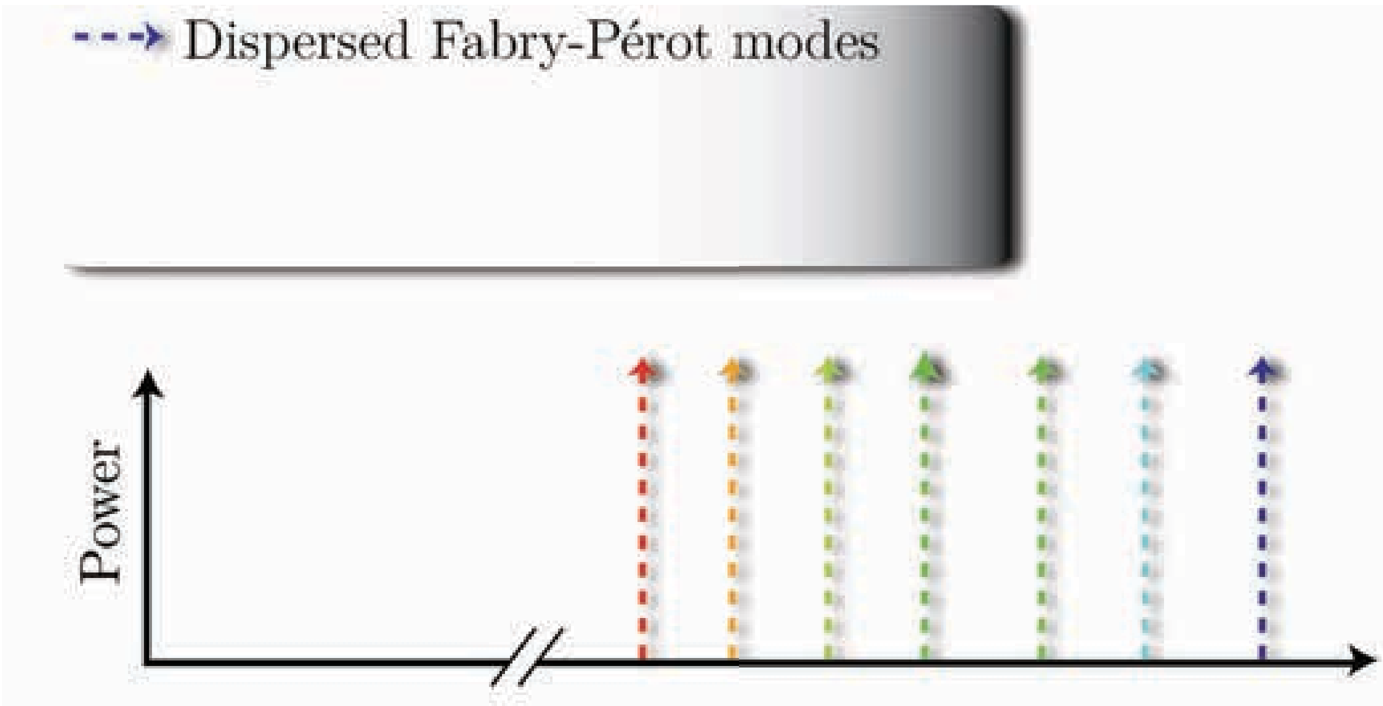
Mode-locking as a injection locking



Mode-locking as a injection locking



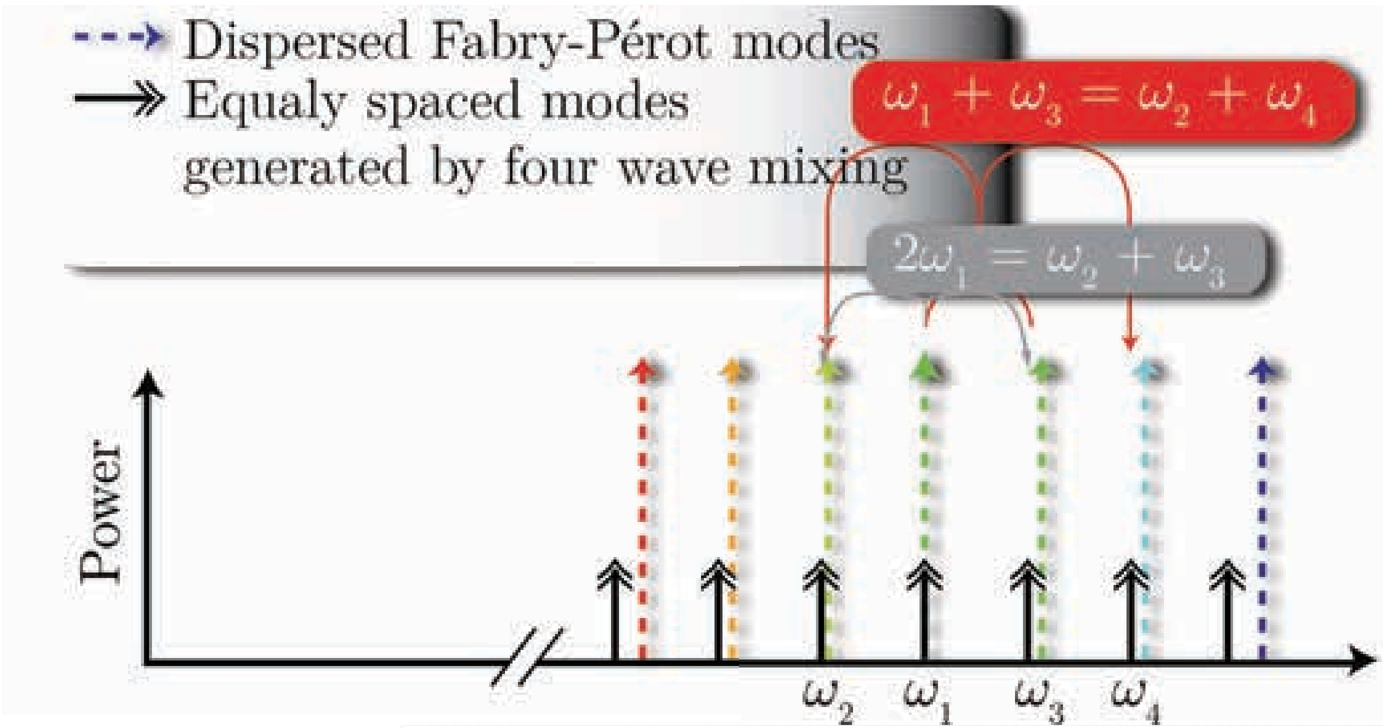
Four wave mixing in QCLs



$$\omega_1 + \omega_2 = \omega_3 + \omega_4$$ frequency matching

$$\vec{\beta}_1 + \vec{\beta}_2 = \vec{\beta}_3 + \vec{\beta}_4$$ phase matching

Four wave mixing in QCLs

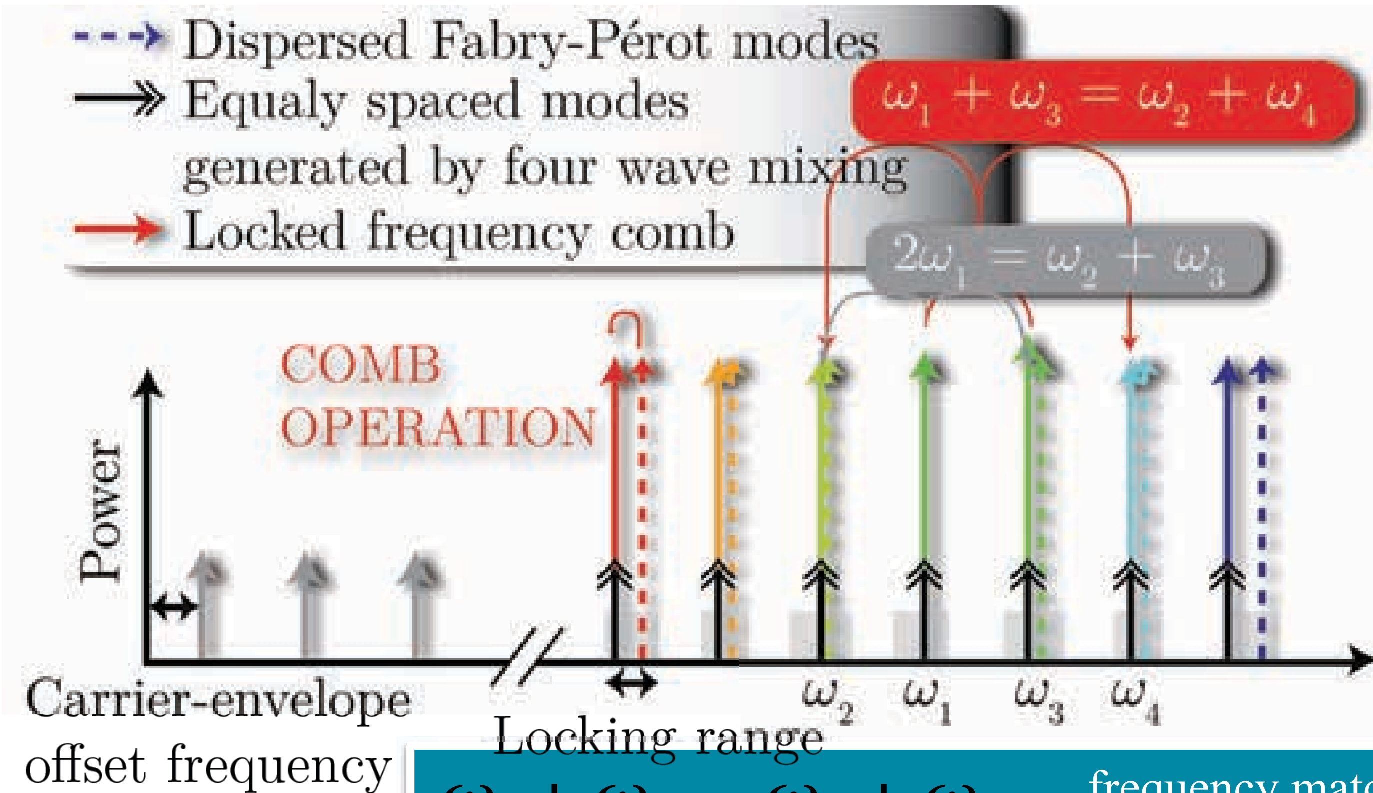


$$P_{NL} \propto \chi^{(3)} E^3$$

$$\omega_1 + \omega_2 = \omega_3 + \omega_4$$ frequency matching

$$\vec{\beta}_1 + \vec{\beta}_2 = \vec{\beta}_3 + \vec{\beta}_4$$ phase matching

Four wave mixing in QCLs



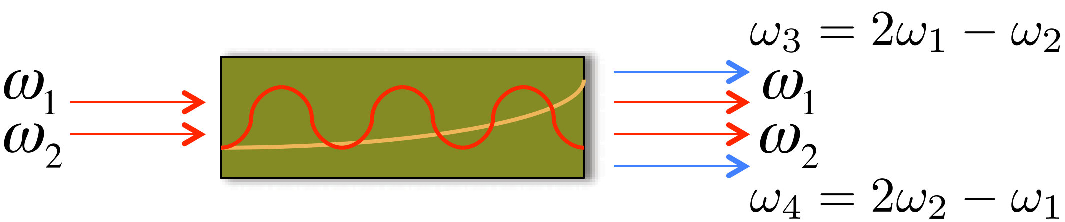
$$P_{NL} \propto \chi^{(3)} E^3$$

$$\omega_1 + \omega_2 = \omega_3 + \omega_4$$ frequency matching

$$\vec{\beta}_1 + \vec{\beta}_2 = \vec{\beta}_3 + \vec{\beta}_4$$ phase matching

Four wave mixing in QCLs

- Measure directly the four wave mixing product



Rosencher, E. *et al*. Science 271, 168 –173 (1996).
Walrod, D. *et al.*, APL, 59 (23), p. 2932, 1991.

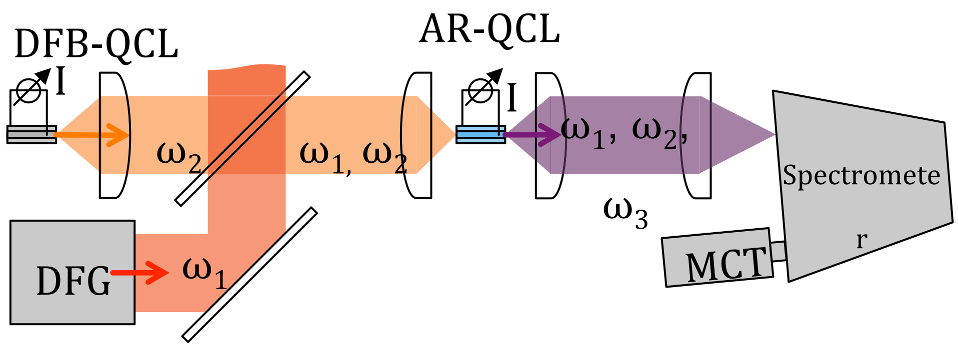
- Need to prevent lasing to have a quantitative measurement
  - Use double anti-reflection coated devices

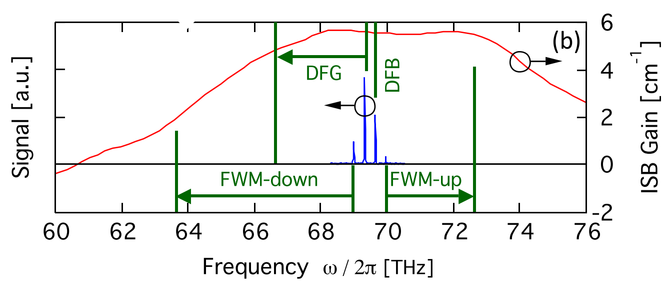
P. Friedli, et al., ," *Appl Phys Lett*, vol. 102, no. 22, p. 222104, 2013.

P. Friedli, H. Sigg

Experimental setup

- Mix a DFB QCL with a source operating by DFG (widely tunable)



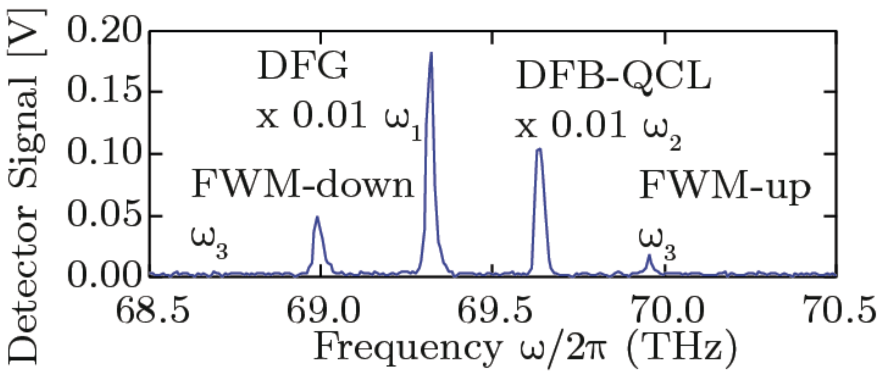


P. Friedli, et al., ,” *Appl Phys Lett*, vol. 102, no. 22, p. 222104, 2013.

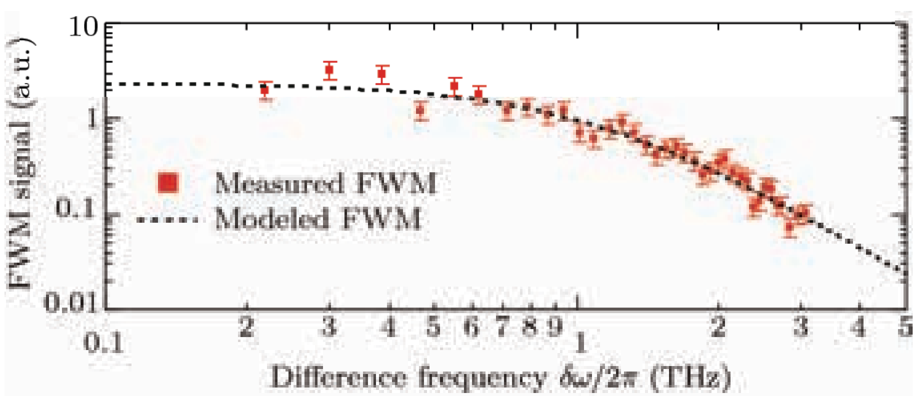
P. Friedli, H. Sigg

Result: four wave mixing product

- Quantitative agreement with the model



$\chi^{(3)} = (0.9 \pm 0.2)\times 10^{-15}\,\mathrm{m}^2\mathrm{V}^{-2}$



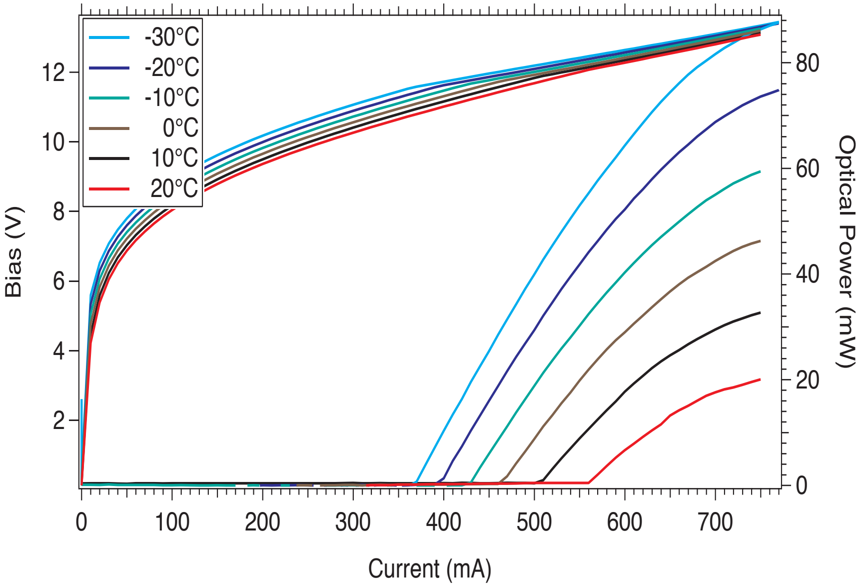
P. Friedli, et al., ," *Appl Phys Lett*, vol. 102, no. 22, p. 222104, 2013.

Interpretation

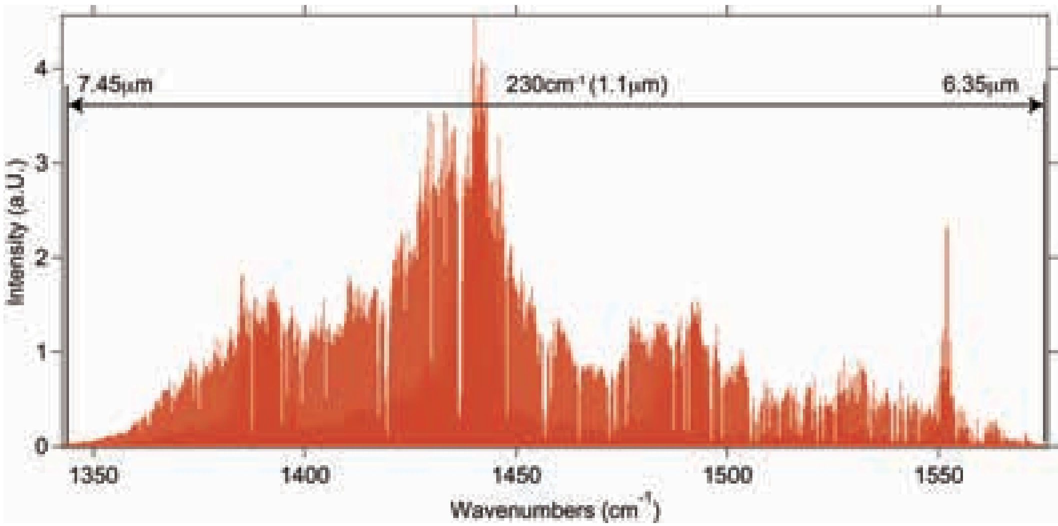
- The beating of ω_1 and ω_2 in the active does modulate the population inversion in time (and space)
- The resulting modulation in (complex) refractive index creates the additional sidebands
- In contrast to normal semiconductor lasers
  - It is fast (<1ps)
  - Therefore broadband!

Broadband multimode lasers

LIV



High Resolution Laser Spectra at -20°C



EPI-UP mounted

6 mm long device

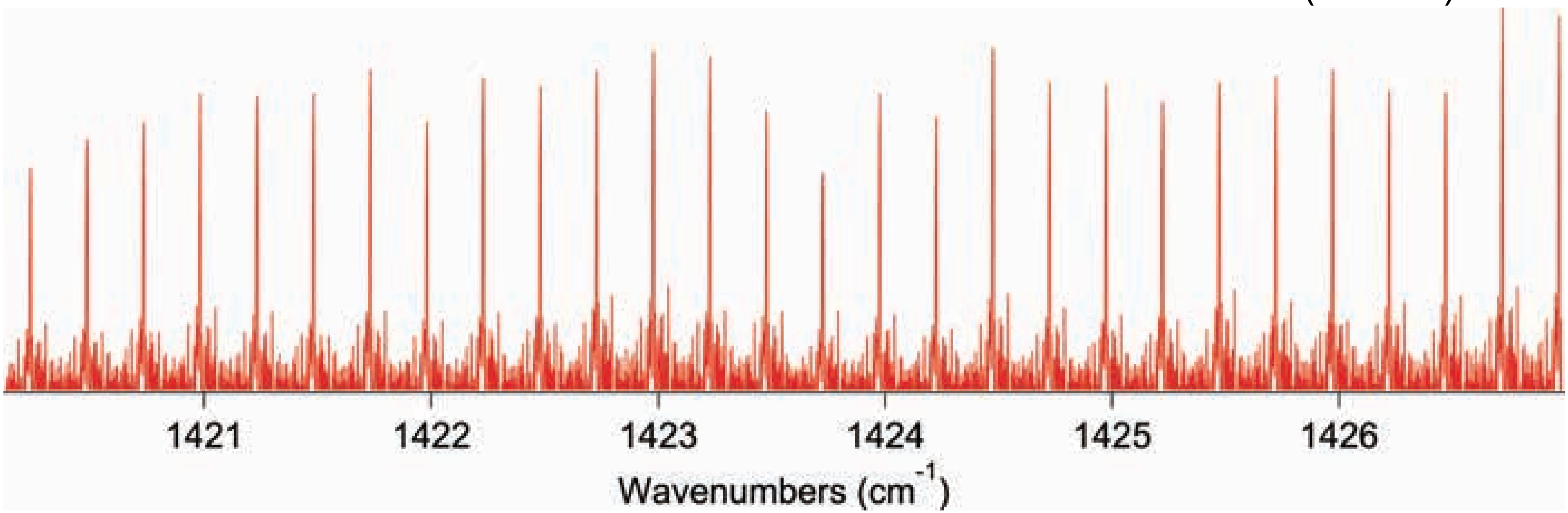
Center wavelength 1440cm^{-1} (6.94μm)

$\Delta\lambda/\lambda$ = 16%

Is it a comb?

- Measure the (high resolution) spectrum

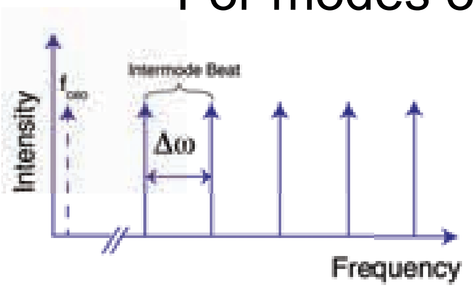
$\Delta\nu =$ 0.0026cm^{-1} (80MHz)



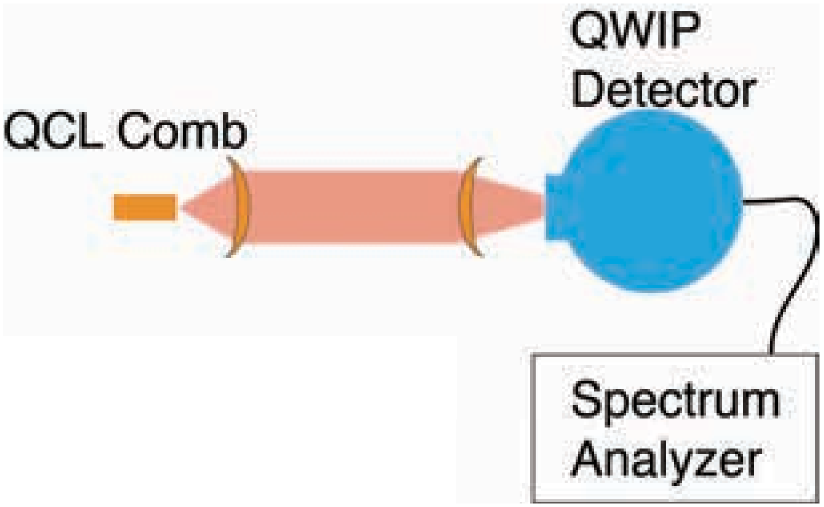
- Does not tell anything about the correlations!!
- The measurement of $g^{(1)}$ does not yield the phases either

Is it a comb?

- Beat note measurement of the photocurrent (at 7.5GHz)
  - For modes of amplitude E_k, the photocurrent at $\Delta\omega$

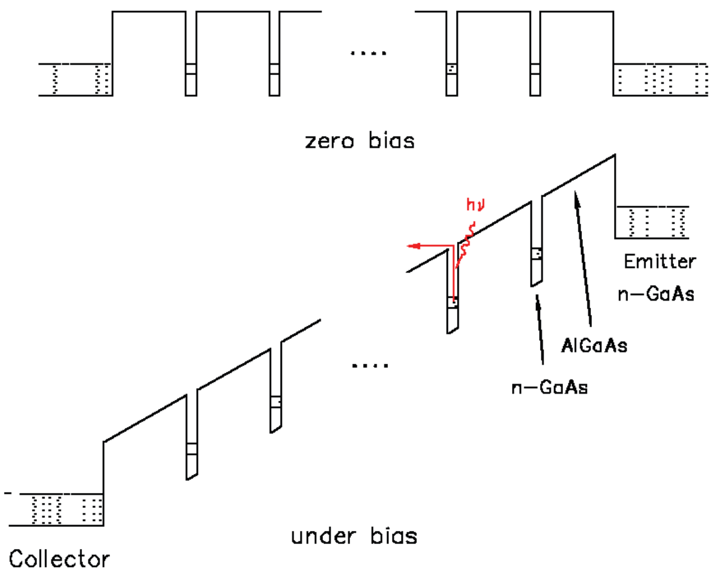


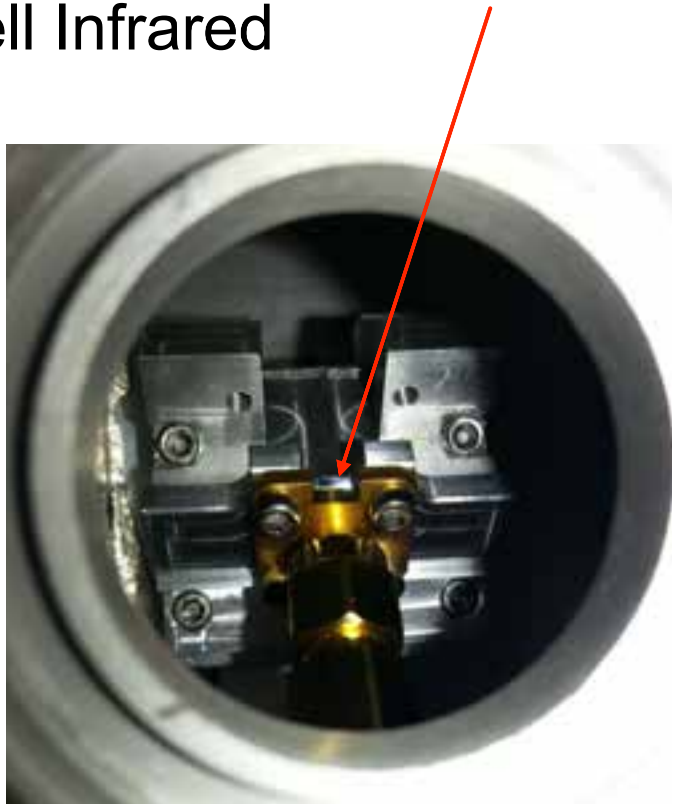
$$I^{\Delta\omega} = \sum_k E_k E_{k+1} \cos(\phi_{k+1} - \phi_k)$$



Fast detector

- Use an intersubband Quantum Well Infrared Photoconductor (QWIP) detector

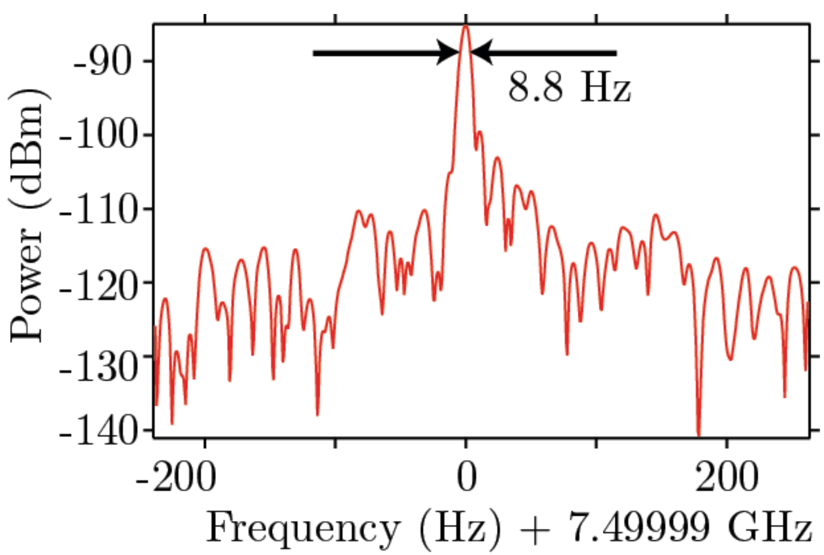


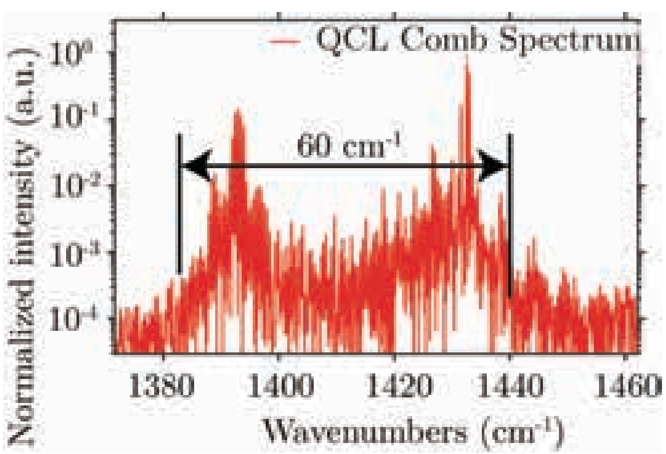


Detector: H.C. Liu
Ref: H. Schneider and H.C. Liu, Springer book

Beatnote spectrum

- The very narrow width confirms the correlations between modes
  - Uncorrelated lines could not be narrower than Schawlow-Townes (100's Hz)
  - However the signal is only about 2% of the c.w. photocurrent

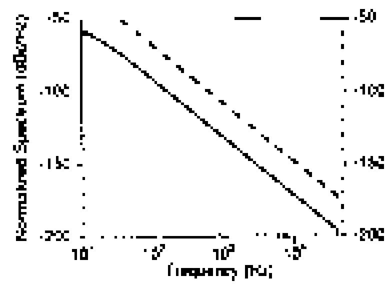




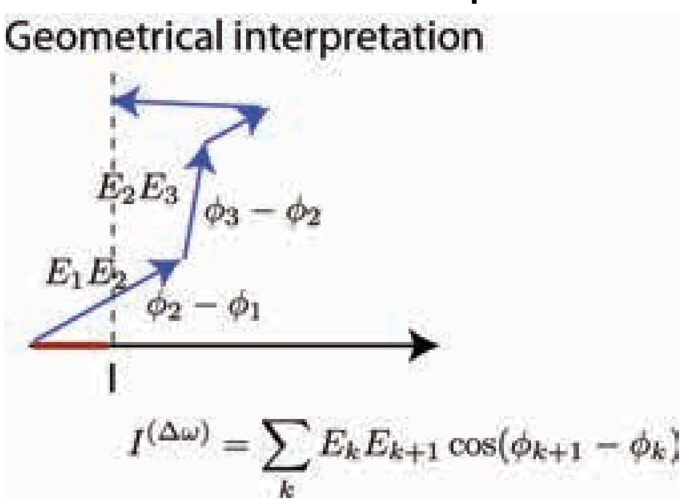
"Time jitter" measurement

- Photocurrent is a "classical way" of measuring noise

Noise of Mode-Locked Lasers

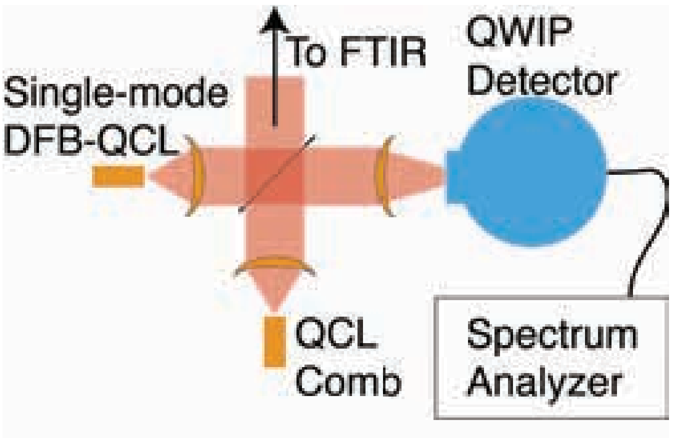


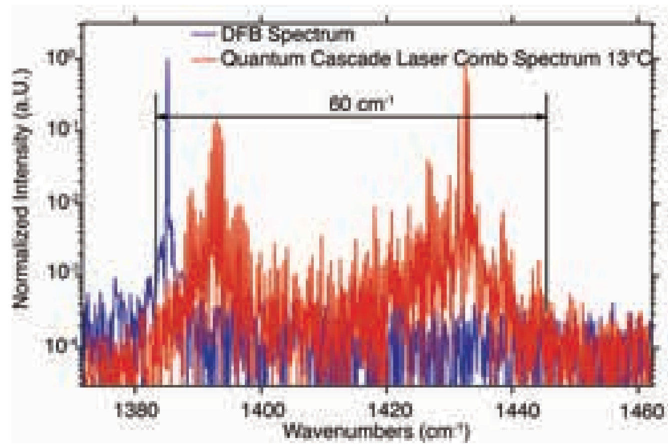
  - Combines the relative phases of the modes

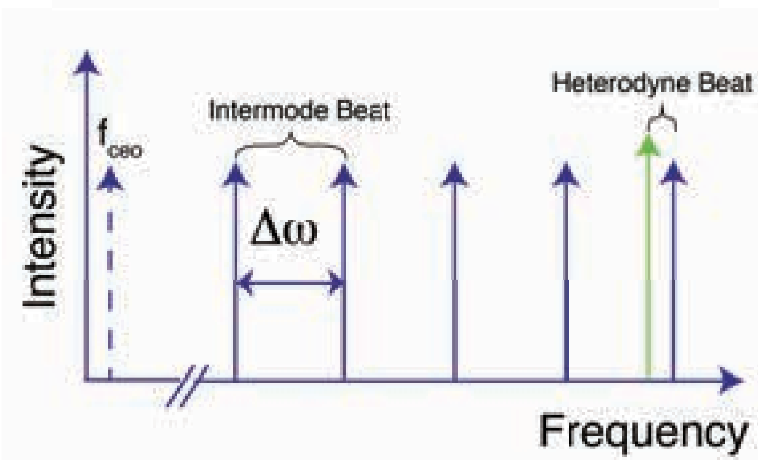


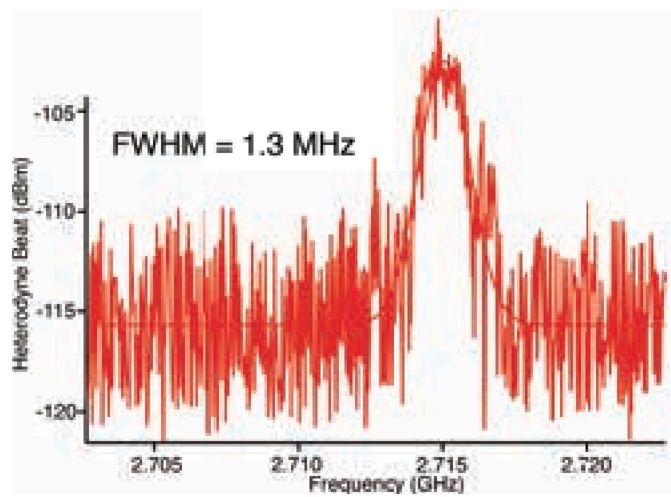
However, the quantitative analysis assumes a mode-locked output with single pulses

Compared to the linewidth of a single line



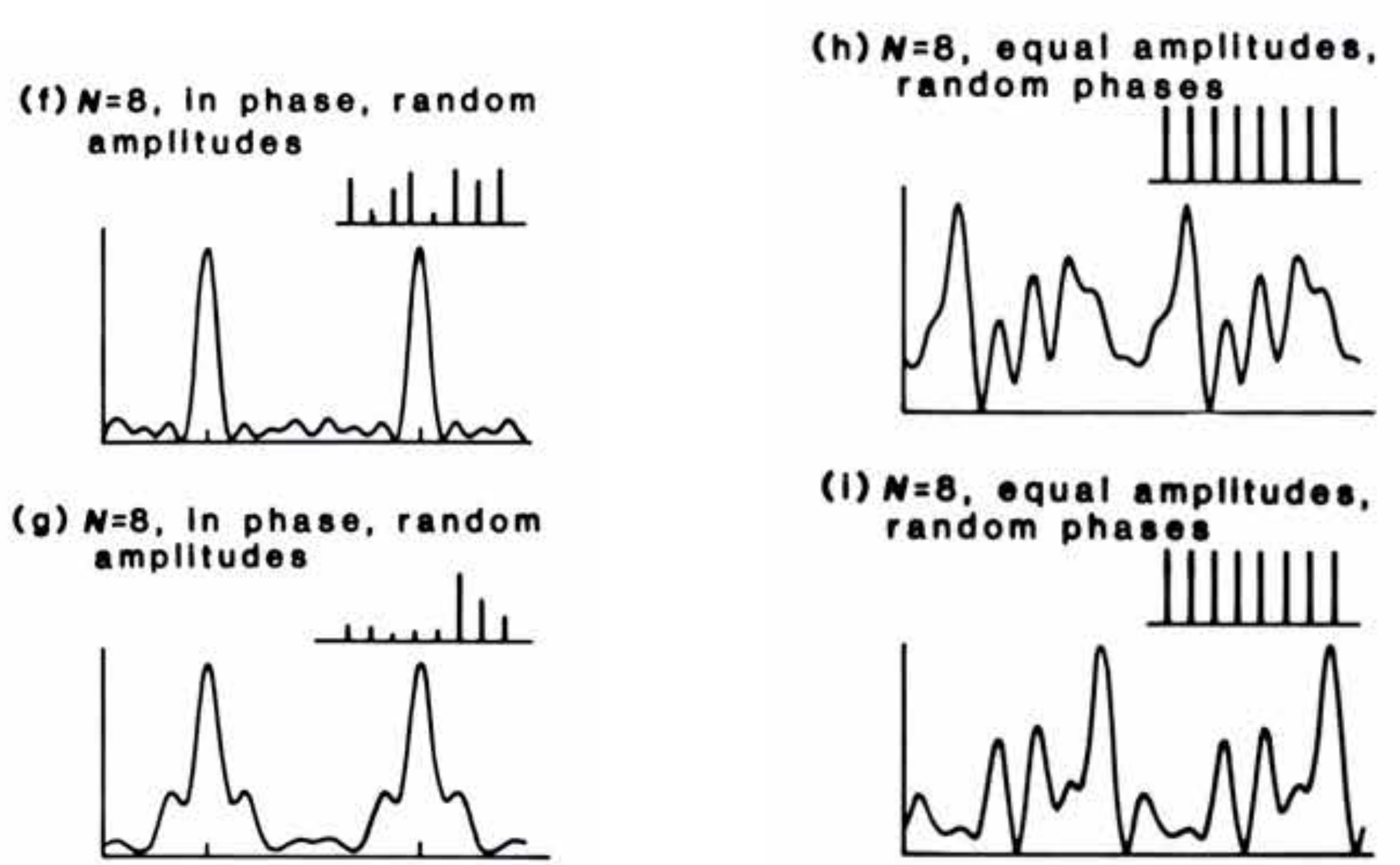




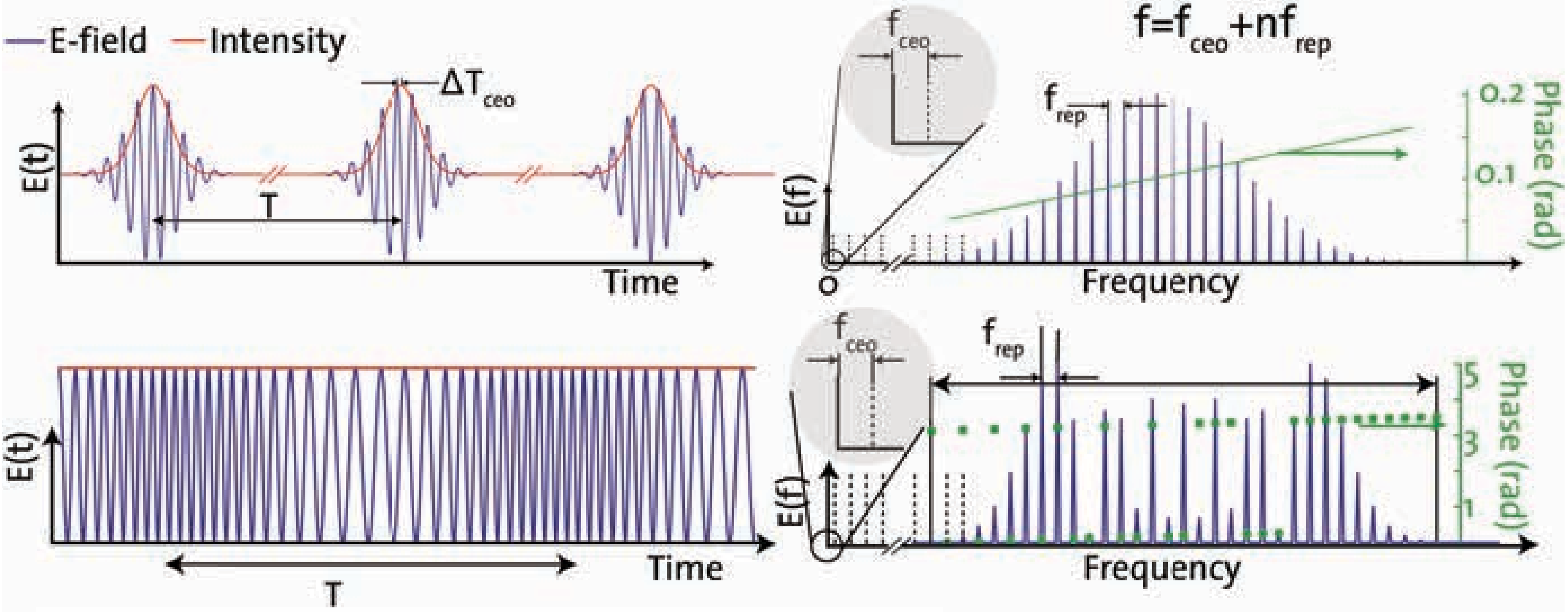


What about the phases?

Effect of random phases..



Optical frequency comb

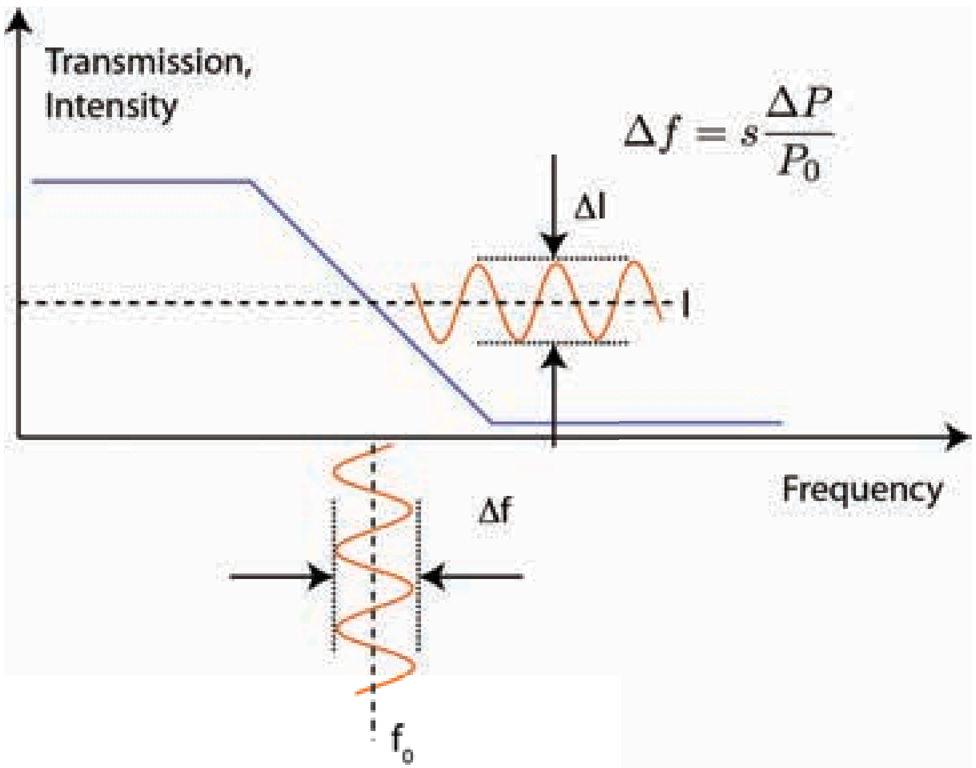


$$E(t) = E_0 \cos\left(2\pi f_c t + m \cos\left(2\pi\ f_{rep} t\right)\right)$$

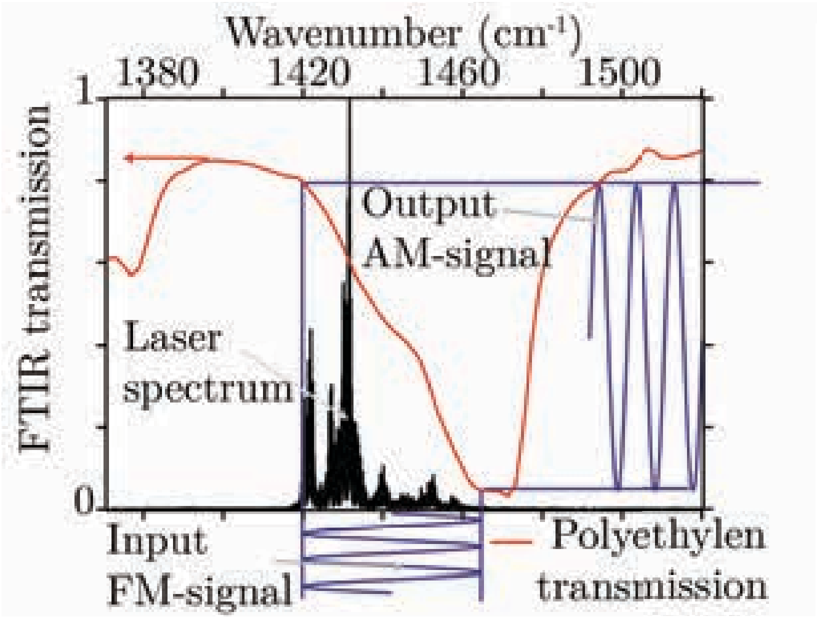
$$m = \frac{f_\Delta}{f_{rep}}$$

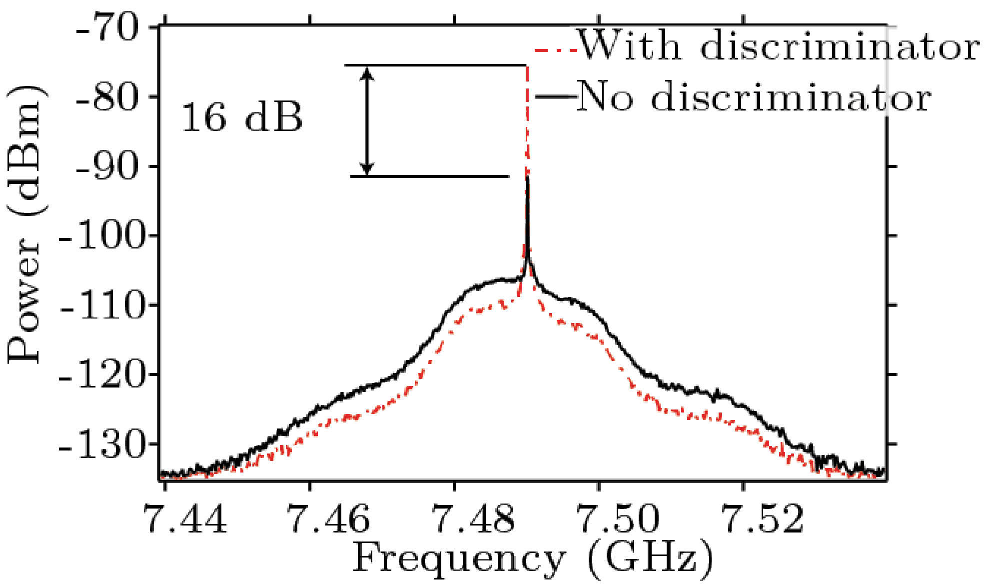
Frequency to amplitude conversion

- Optical discriminator: frequency dependent absorption
  - FM -> AM conversion



Optical discriminator - FM nature mode locking!





Sheet of polyethylene acts as an optical discriminator

A. Hugi, et al., *Nature*, vol. 492, 229–233 (2012)

Is it a surprise?

- QCLs are "type A" lasers
  - The medium follows the optical field dynamics

$$\kappa << \gamma_{\perp}, \gamma_{21}$$

- Similar to gas lasers such as HeNe (red alignment laser)

Volume 5, Number 10 APPLIED PHYSICS LETTERS 15 November 1964

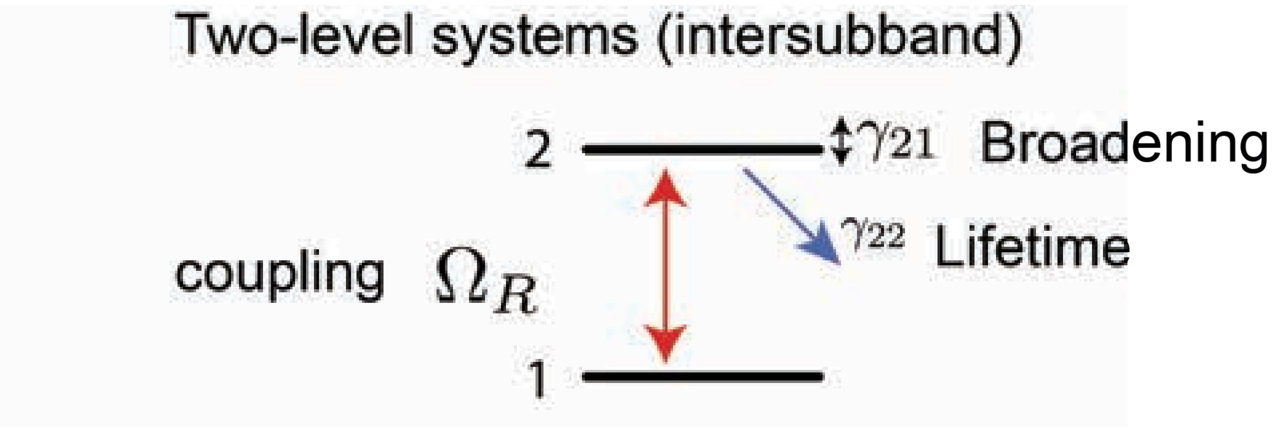
FM OSCILLATION OF THE He-Ne LASER[1]

S. E. Harris
Consultant to Sylvania Electronic Systems
Mountain View, California and
Department of Electrical Engineering
Stanford University, California

Russell Targ
Electronic Defense Laboratories
Sylvania Electronic Systems
Mountain View, California
(Received 25 September 1964)

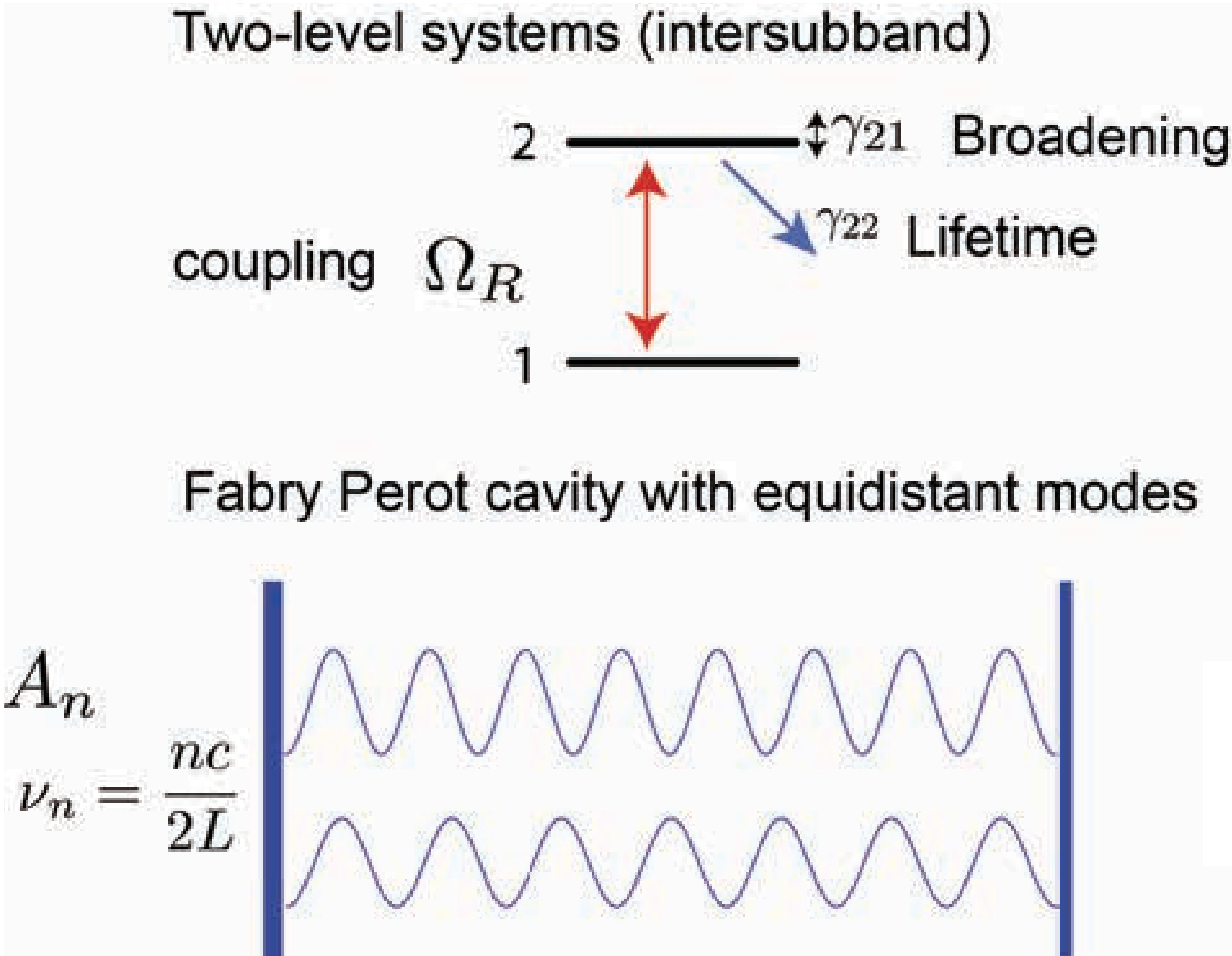
(internal phase perturbation; interferometry; KDP crystal; E)

Modal decomposition approach



J. B. Khurgin, et al *Appl Phys Lett*, vol. 104, no. 8, p. 081118, Feb. 2014.

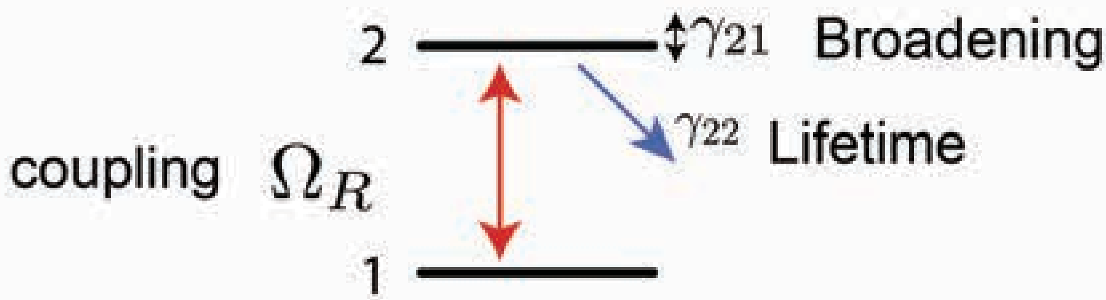
Modal decomposition approach



J. B. Khurgin, et al *Appl Phys Lett*, vol. 104, no. 8, p. 081118, Feb. 2014.

Modal decomposition approach

Two-level systems (intersubband)



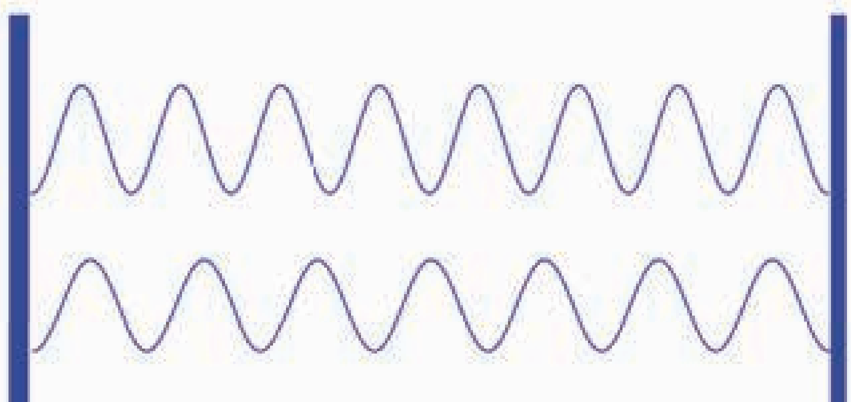
Motion of ρ

$$\frac{d\rho}{dt} = -\frac{i}{\hbar}[H,\rho]$$

Fabry Perot cavity with equidistant modes

A_n

$$\nu_n = \frac{nc}{2L}$$



Motion of mode A_n

$$\left(\nabla^2 - \frac{\epsilon_r}{c^2}\frac{\partial^2}{\partial t^2}\right) A_n = \mu_0 \frac{\partial^2}{\partial t^2} P_n$$

J. B. Khurgin, et al *Appl Phys Lett*, vol. 104, no. 8, p. 081118, Feb. 2014.

Time dependence of the mode amplitude An

- Modal expansion formalism

$$\frac{dA_n}{dt} = (G_n - 1)A_n - iD_nA_n - G_n \sum_{k,l=-N/2}^{N/2} A_m A_k A_l^* B_{kl} C_{kl} \kappa_{klmn}$$

Gain

Cavity Dispersion

Four wave mixing driven by population pulsation:

- Spatial hole burning
- Self Gain Saturation
- Cross-gain saturation

FWM terms are resonant!

$$\frac{dA_n}{dt} = (G_n - 1)A_n - iD_nA_n - G_n \sum_{k,l=-N/2}^{N/2} A_m A_k A_l^* \boxed{B_{kl} C_{kl} \kappa_{klmn}}$$

- Overlap between modes

$$\kappa_{n,l,k,m} = \frac{1}{l_c} \int_0^{l_c} \sin(k_n z) \sin(k_l z) \sin(k_k z) \sin(k_m z) dz$$

- Width of the gain curve (and FWM process)

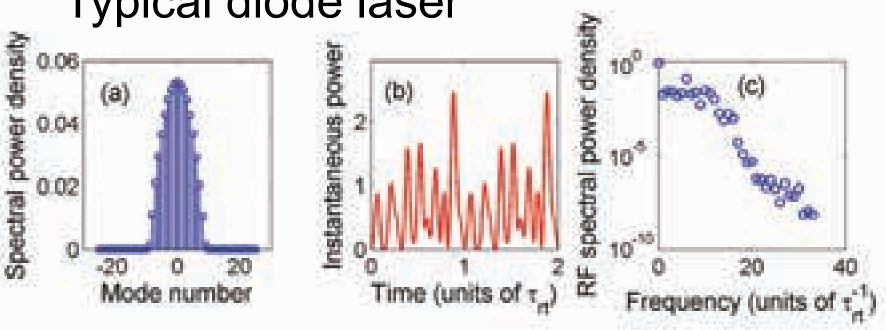
$$B_{kl} = \frac{\gamma_{12}}{2i} \left(\frac{1}{-i\gamma_{12} - l\omega} - \frac{1}{i\gamma_{12} - k\omega} \right) \qquad \omega = \frac{2\pi}{t_{rt}}$$

- Population population beating

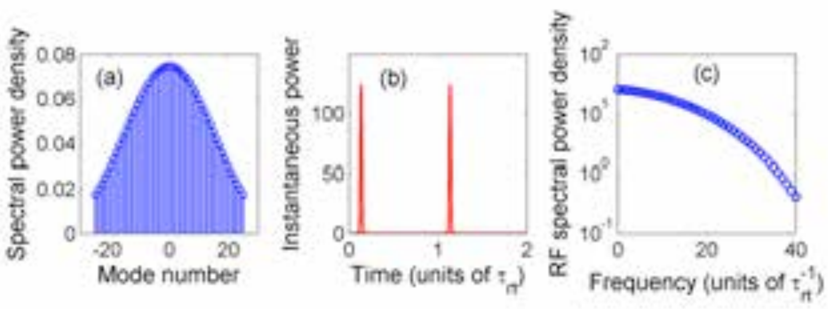
$$C_{kl} = \frac{\gamma_{22}}{\gamma_{22} - i(l-k)\omega}$$

Simulation results

Typical diode laser



with saturable absorber

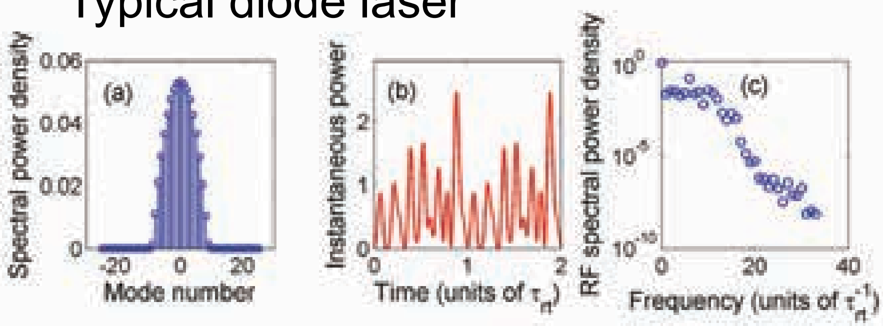


t_{21} ~ 1ns
$t_{21(absorber)}$=1.0 ps, $t_{coh(absorber)}$=0.1ps

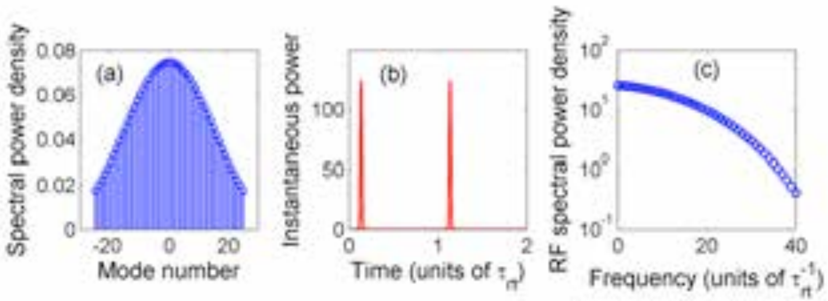
Simulation results

Self-FM modulation in QCLs

Typical diode laser



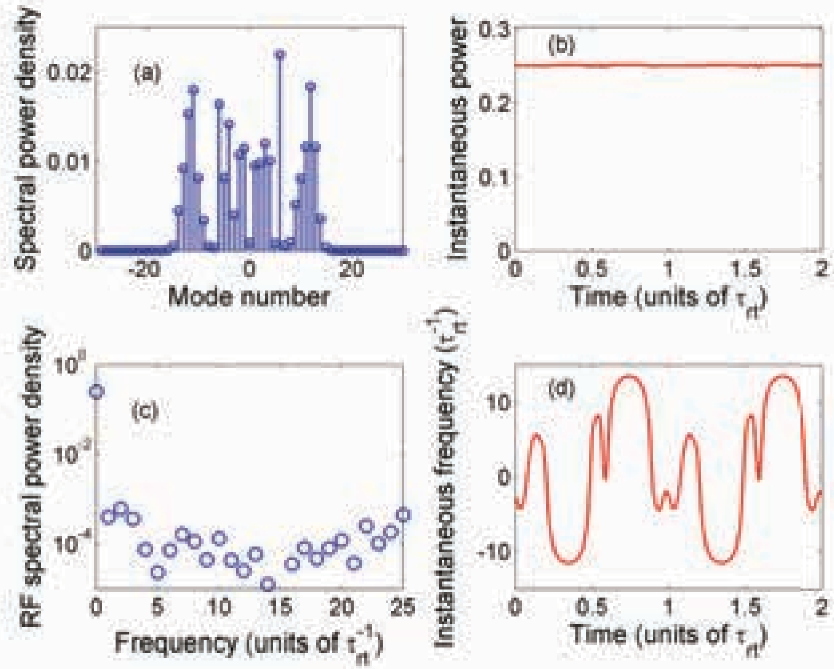
with saturable absorber



t_{21} ~ 1ns

$t_{21(absorber)}$=1.0 ps, $t_{coh(absorber)}$=0.1ps

Typical QCL without dispersion



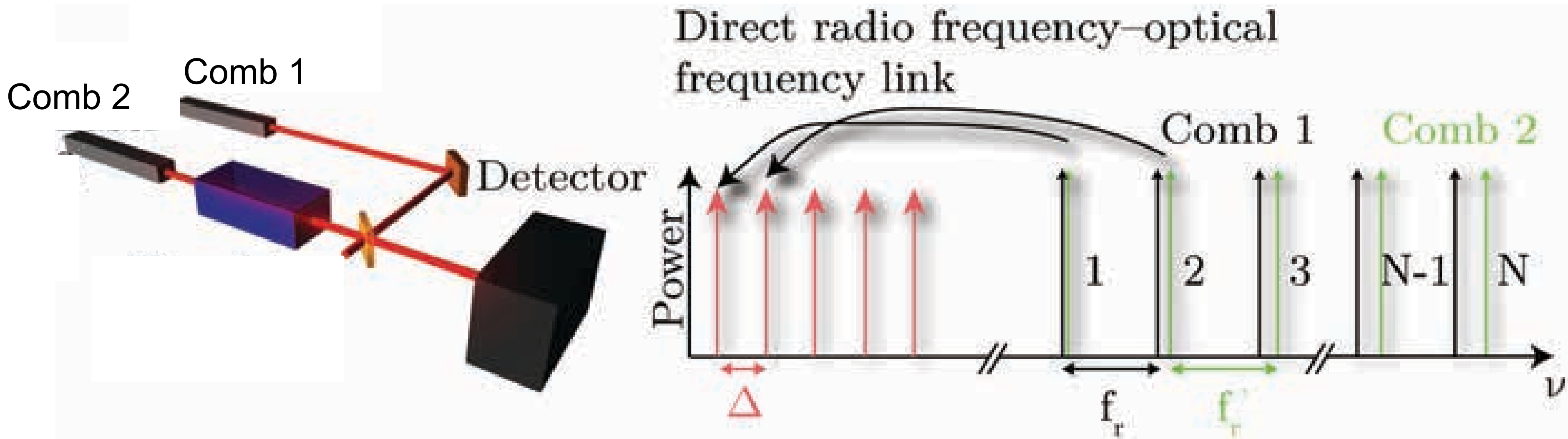
t_{21}~1ps, no saturable absorber

J. B. Khurgin, et al *Appl Phys Lett*, vol. 104, no. 8, p. 081118, Feb. 2014.

Use a dual comb technique

- Mix two combs optically: benchmark one comb with another one

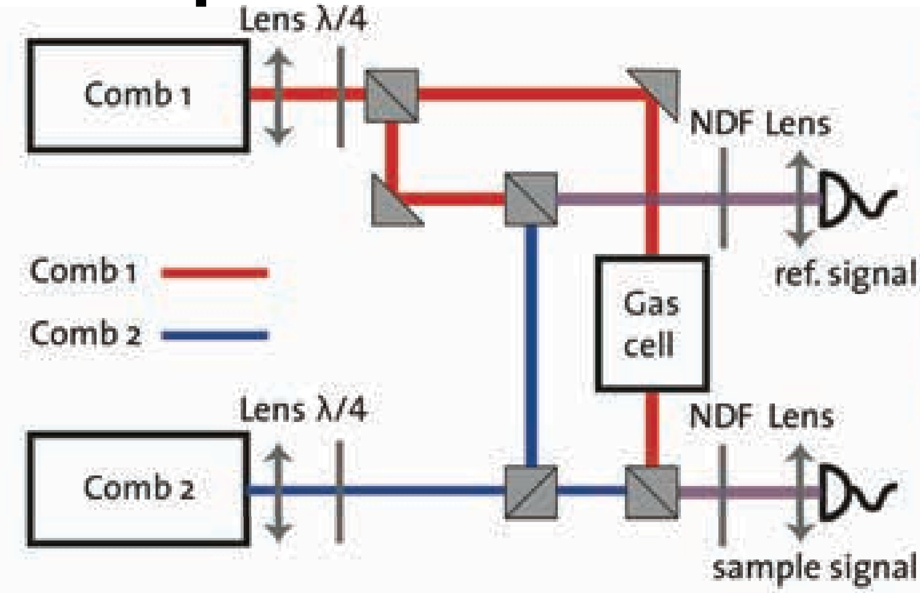
A. Schlatter, et al, *Appl. Phys. B*, vol. 88, pp. 385–391, (2007)

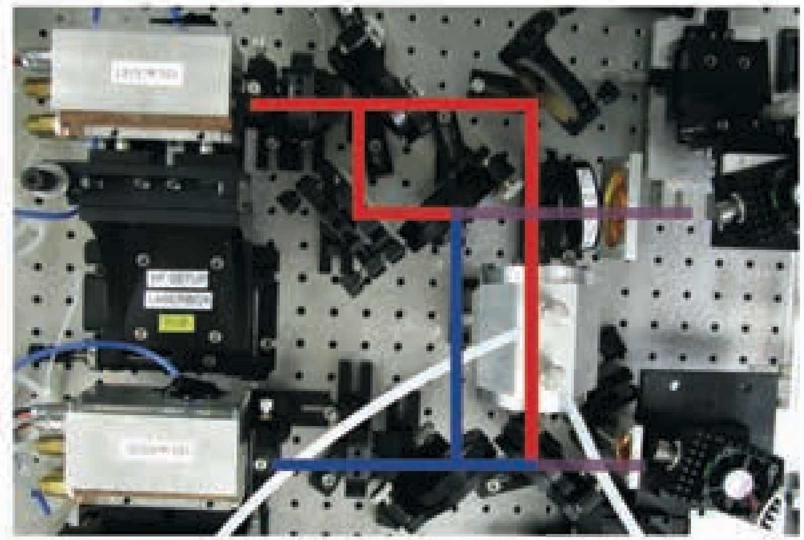


- Used also for spectroscopy (see G. Villares' talk)

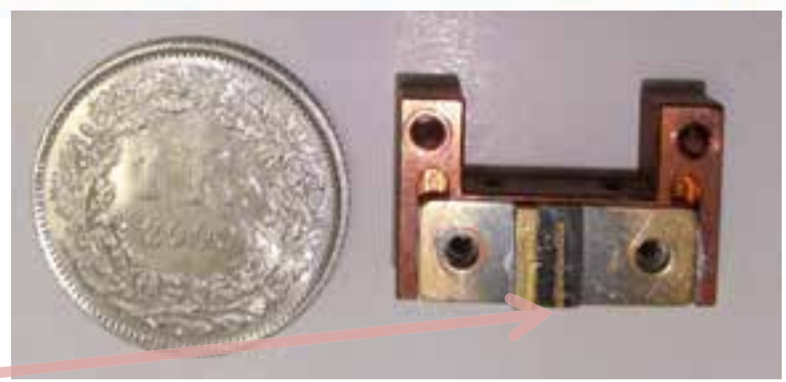
S. Schiller, "Spectrometry with frequency combs," Opt Lett, vol. 27, no. 9, pp. 766–768, 2002.
Keilmann, F., et al. Time-domain mid-infrared frequency-comb spectrometer. Opt. Lett. 29, 1542–1544 (2004)

Setup: mid-IR dual comb spectroscopy



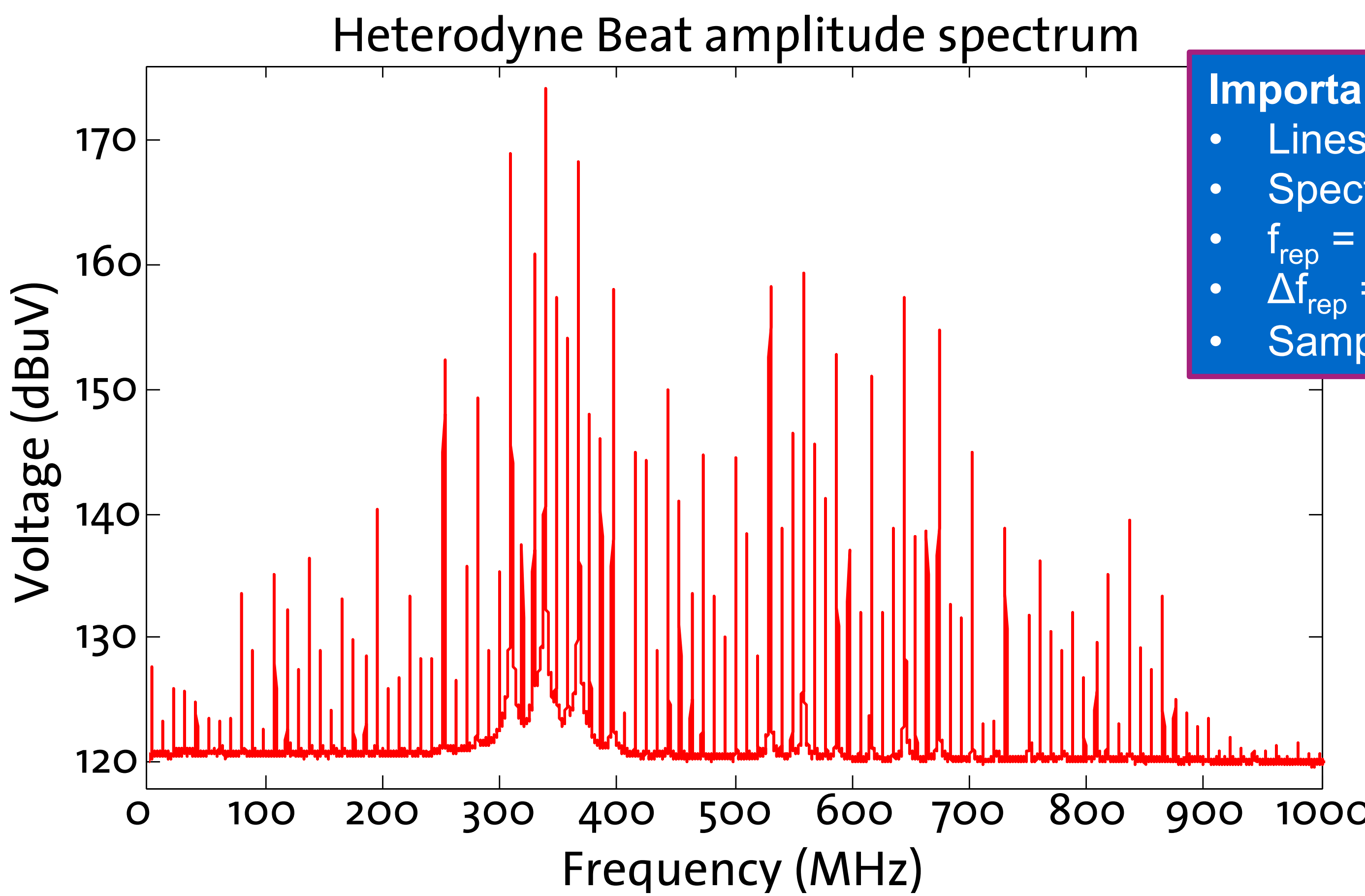


Characteristics
- Compact setup (65cm x 65cm x 25cm)
- No cryogenic cooling needed



6 mm long

Free running heterodyne beat measurements

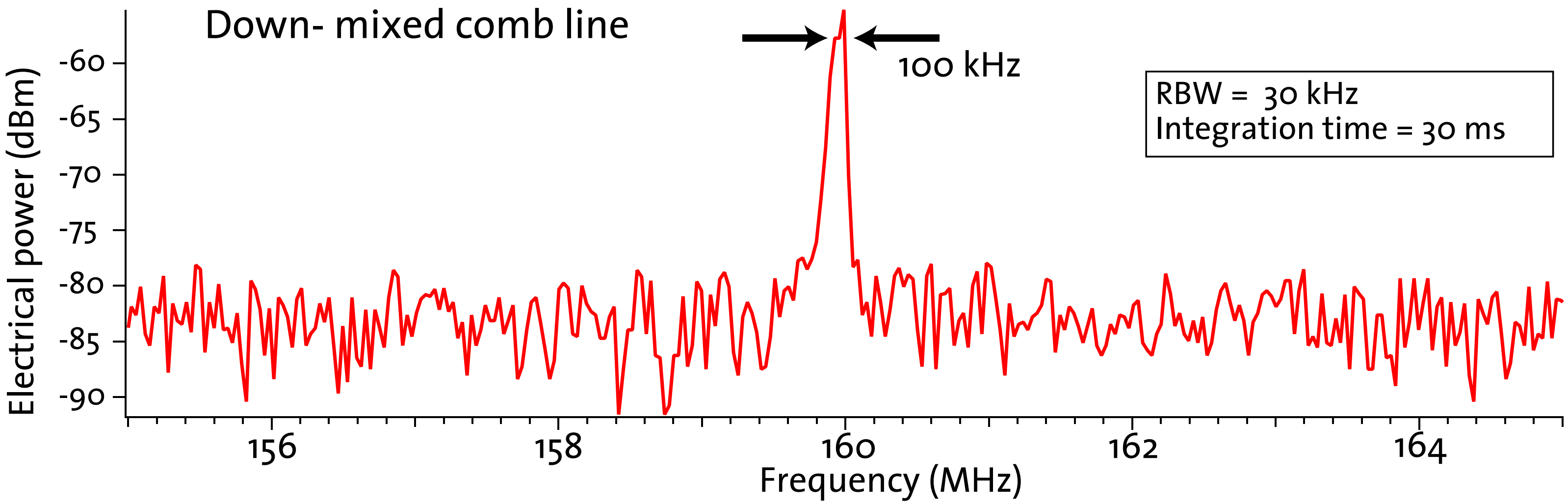


Important numbers:

- Lines = 100;
- Spectral coverage = 25 cm^{-1}
- f_{rep} = 7.5 GHz
- Δf_{rep} = 10 MHz
- Sampling = 0.25 cm^{-1}

G. Villares et al., Nat. Comm (in press)

Free running heterodyne beat measurements



Comb tooth linewidth (free running)

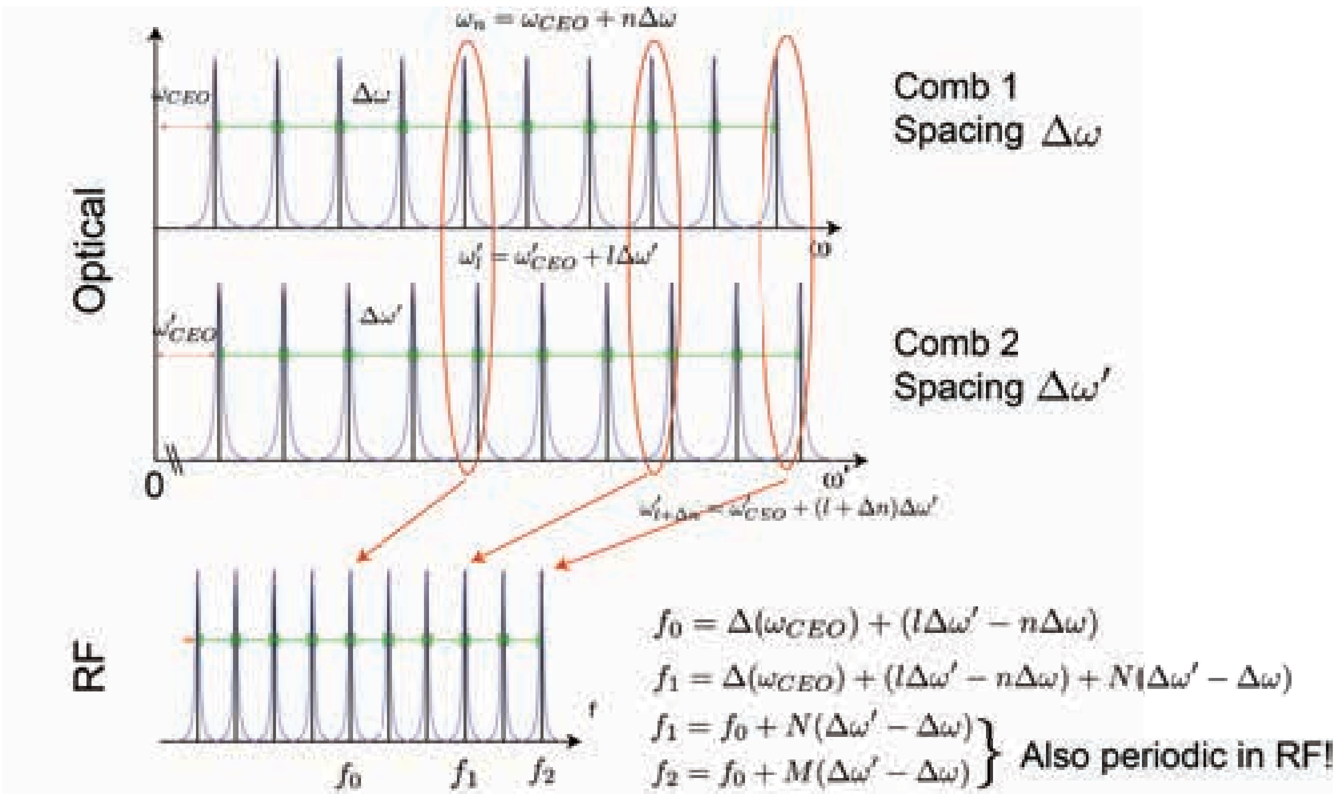
- FWHM = 100 kHz

Limited by the relative drift of the combs

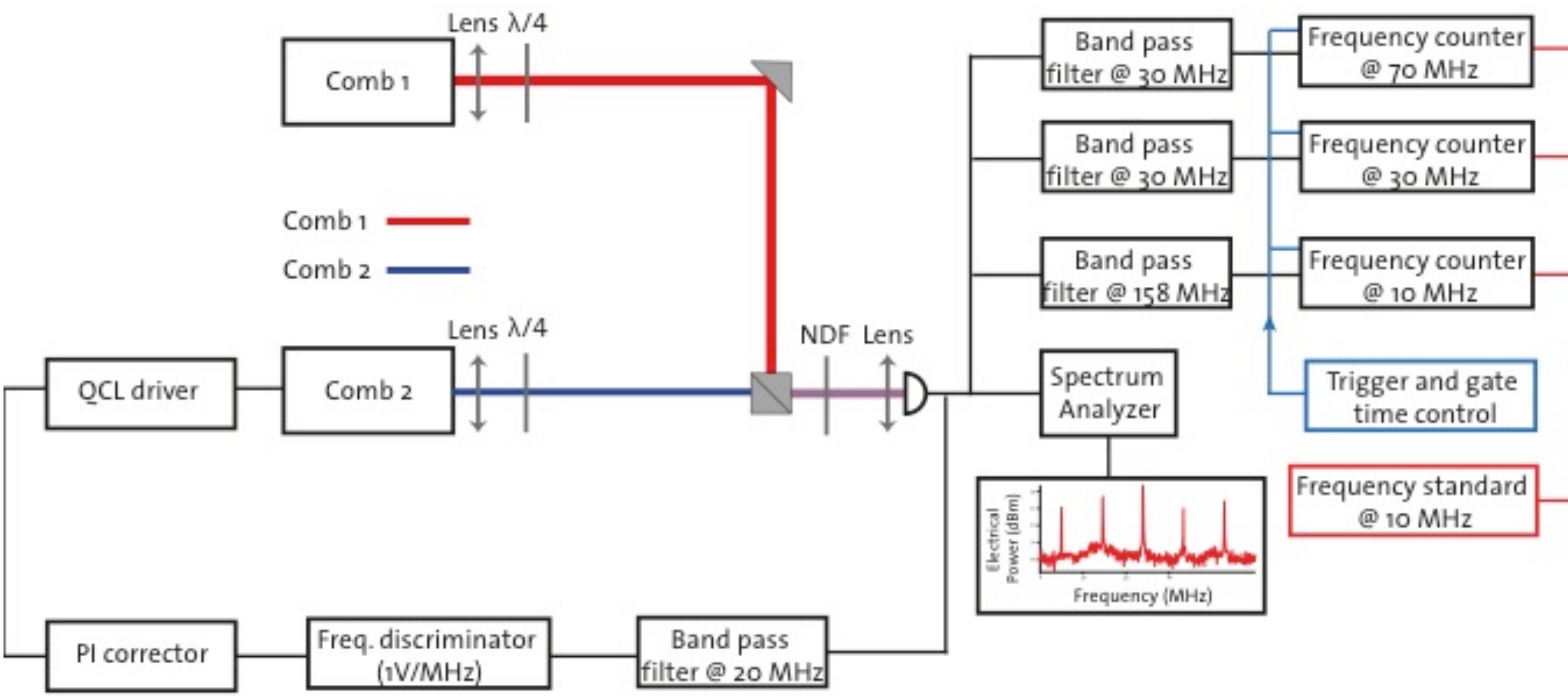
G. Villares et al., Nat. Comm (in press)

Comb equidistance measurement

- Measuring frequency differences removes the CEO fluctuations
  - The equidistance in the RF domain mirrors the one in the optical domain



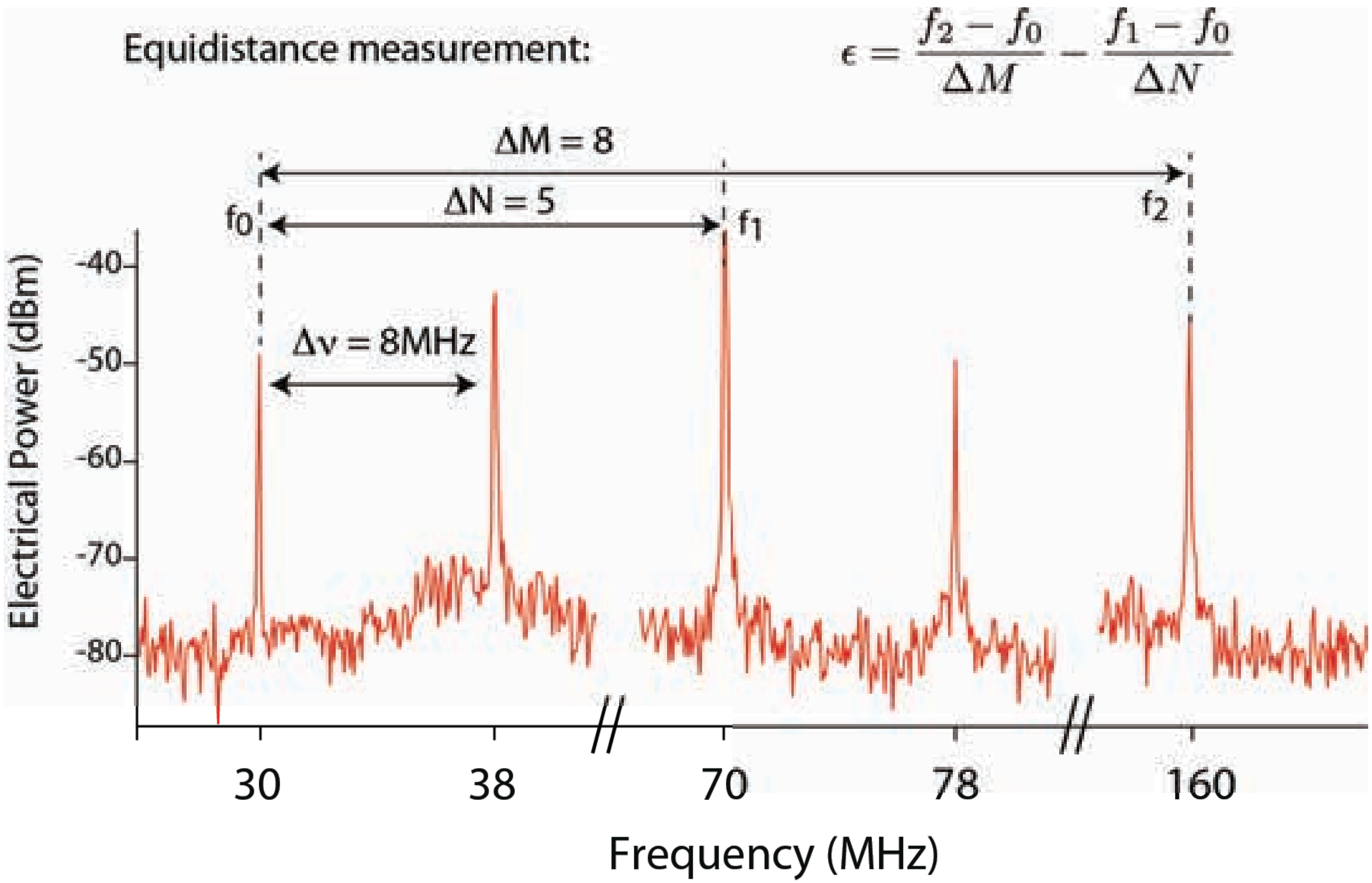
Stabilization and comb equidistance measurement



P. Del'haye et al, *Nature*, vol. 450, pp. 1214–1217, 2007

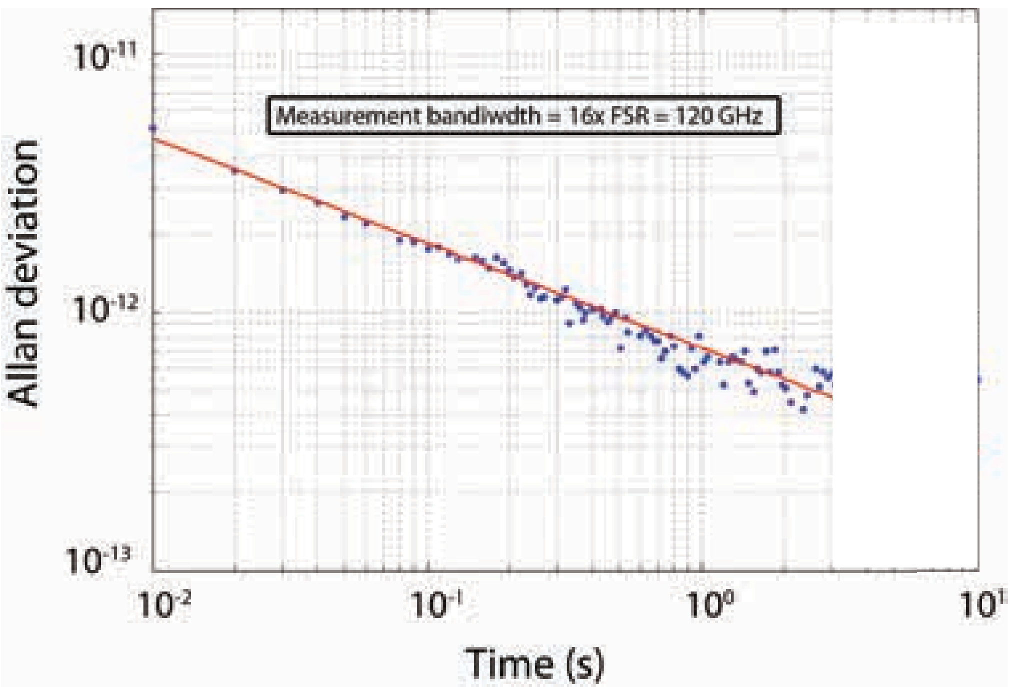
G. Villares et al., Nat. Comm (in press)

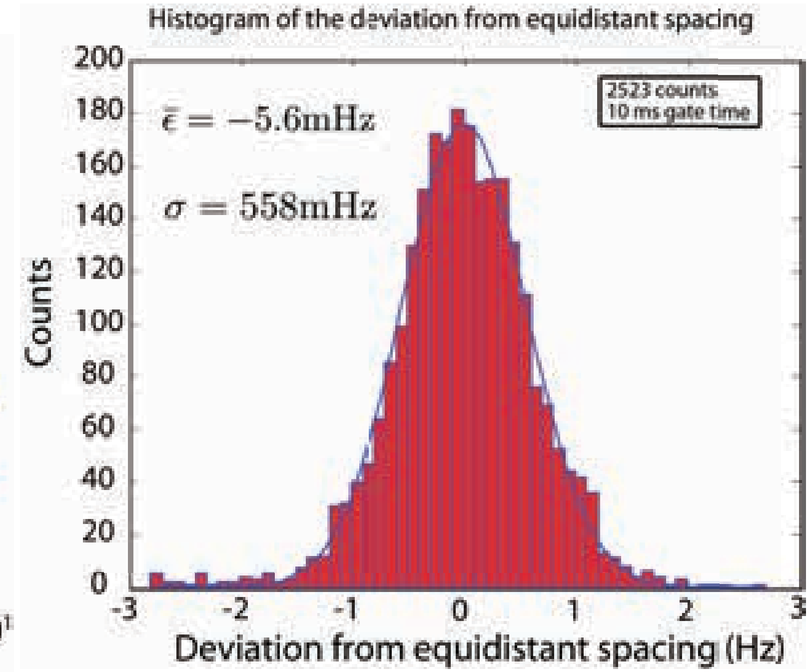
Equidistance between peaks



G. Villares et al., Nat. Comm (in press)

Results: mHz level accuracy





- The round trip frequency is 7.5GHz
- The individual comb is equidistant to a level of 5×10^{-13}

G. Villares et al., Nat. Comm (in press)

Noise limits of combs

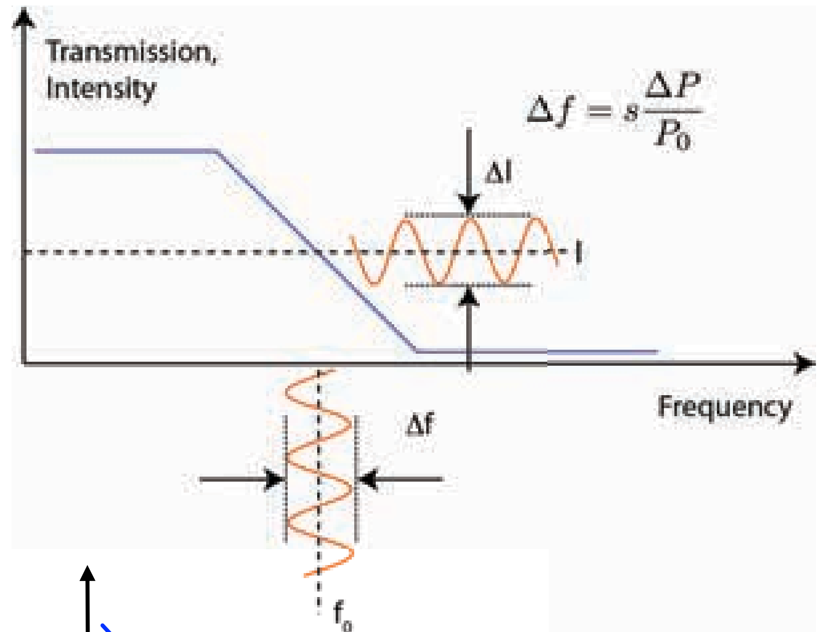
- What is the ultimate linewidth of a comb?
- For c.w. lasers, we have the Schawlow-Townes limit:

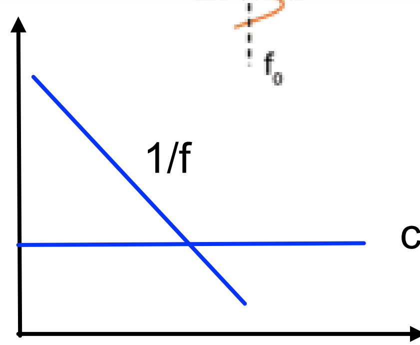
$$\Delta\nu = \frac{\pi h\nu\, \Delta{\nu_c}^2}{P} \frac{\alpha_m}{\alpha_{tot}} \frac{n_3}{n_{3t} - n_{2t}} (1 + \alpha_{LEF}^2)$$

- Very narrow, because
  - QCLs are powerful (10-100mW c.w.)
  - Operate at long wavelength (5-10μm)
  - Build with low loss cavities (~6mm cavity)
- However, for QCL this linewidth (~100Hz) is usually much narrower than the one due to technical noise (100kHz)

Use again an optical discriminator

- Measure the frequency noise power spectral density (PSD)
  - Is the amount of frequency noise at each frequency
- Technical noise decreases with f $(1/f)^n$
- Schawlow-Townes is flat





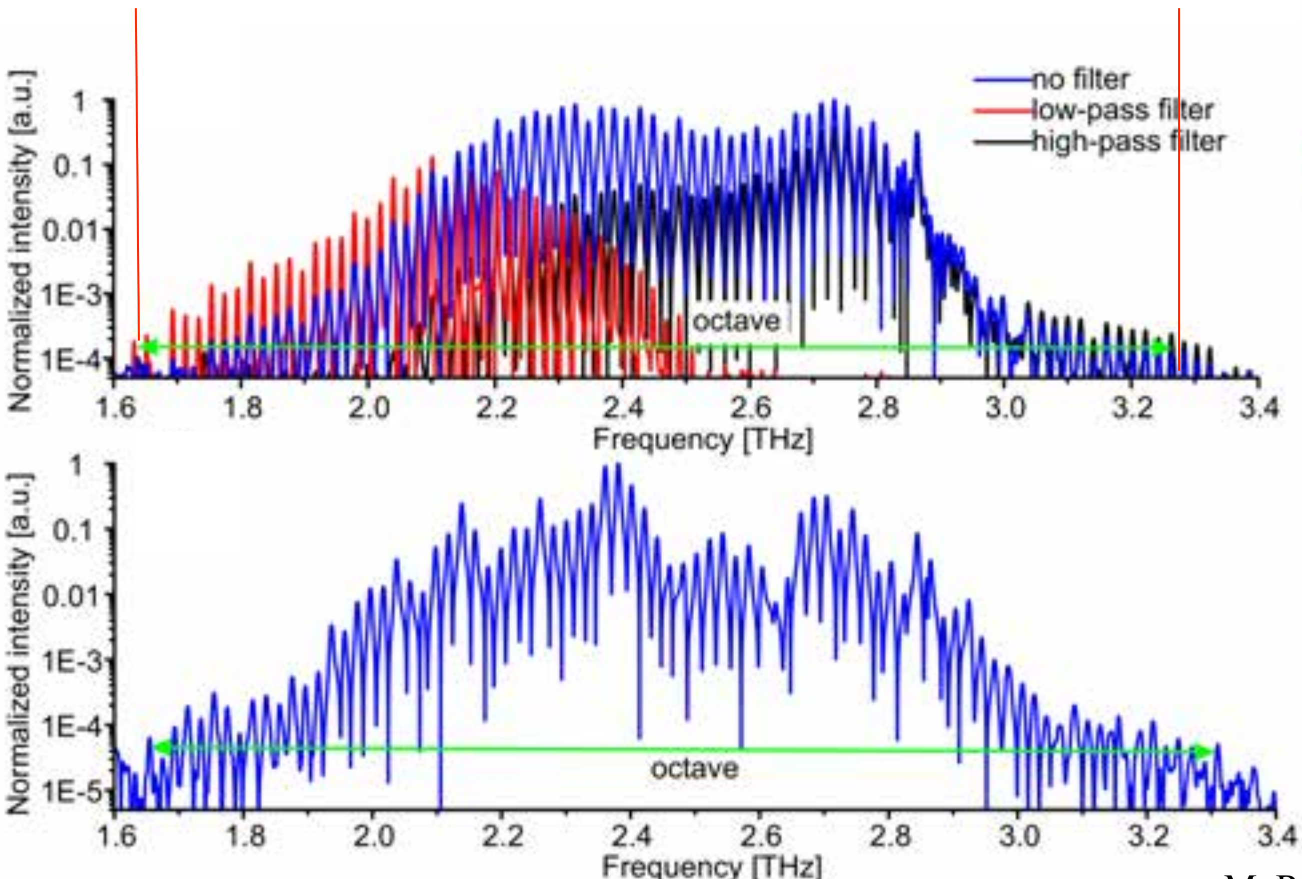
How broad can we make combs?

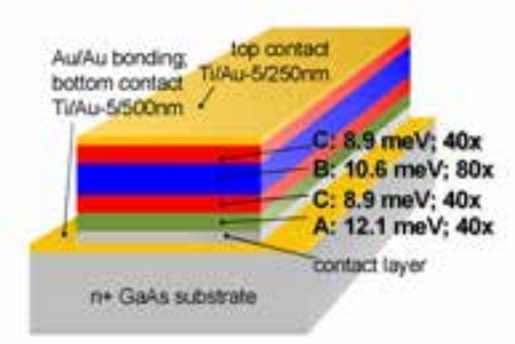
- A great feature is the self-referencing
- It needs an octave-spanning comb
  - or at least 2/3 of an octave

An octave-spanning semiconductor laser

1f

2f





C.w. operation
25K
Milliwatts output

->M. Rösch's talk

M. Roesch, G. Scalari et al., ArXiv (2014)

Outlook

- Shown that QCL can operate as FM combs
- Hard experimental proofs:
  - Beatnote and beatnote spetroscopy
  - Equidistance measurement
  - Quantum noise limit
- A numerical model agrees with the experiment
- Nice to have a theory that could quantify the locking criteria

Group in ETH

FIR team: G. Scalari, M. Geiser, C. Bonzon,
D. Turcinkova, G. Cerullo, M. Roesch

Metamaterial C. Maissen, F. Valmorra,P. Liu

MIR team: B. Hinkov, S. Riedi, J. Wolf,
G. Villares, A. Hugi

Growth Team: M. Beck, V. Liverini, K Ohtani