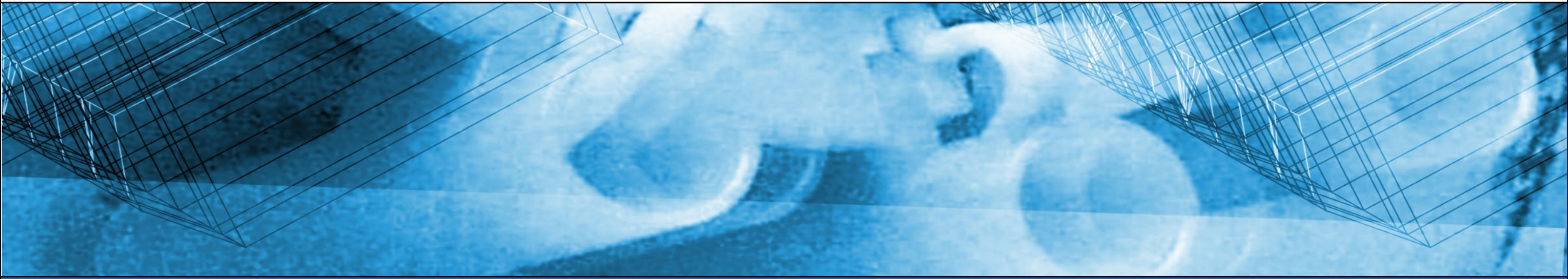


Chip-scale terahertz frequency combs and multiheterodyne spectroscopy

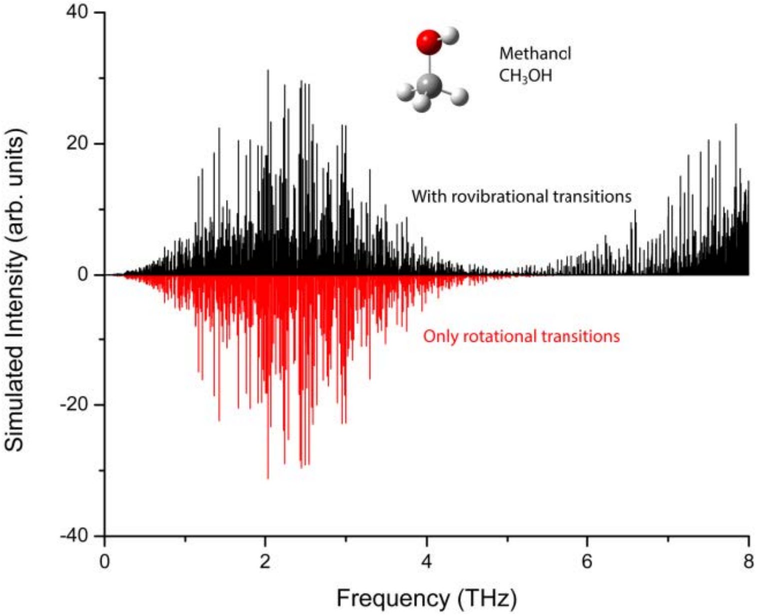
David Burghoff

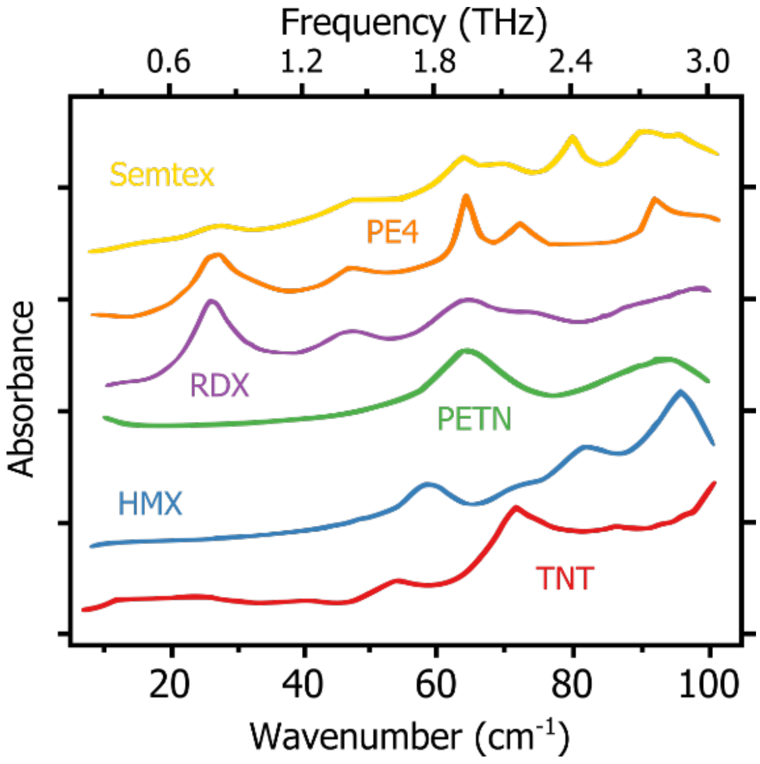
OSA Laser Systems Technical Group

December 6, 2017

Main motivation: broadband chip-scale spectroscopy

- Many molecules have distinctive spectral features at long wavelengths due to structure



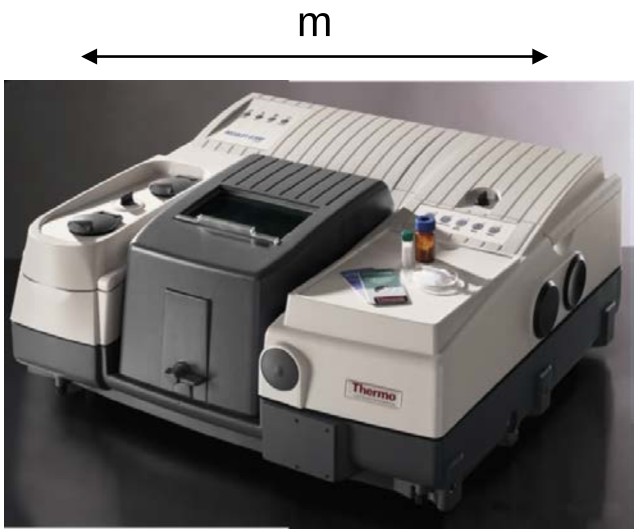


  - Terahertz: 30-300 µm (1-10 THz)
  - Mid-infrared: 3-30 µm

DARPA SCOUT program:
Broadband chip-scale spectroscopy

Spectral **Co**mbs from **U**V to **T**Hz

Basic problem: highest sensitivity systems are large and have moving parts



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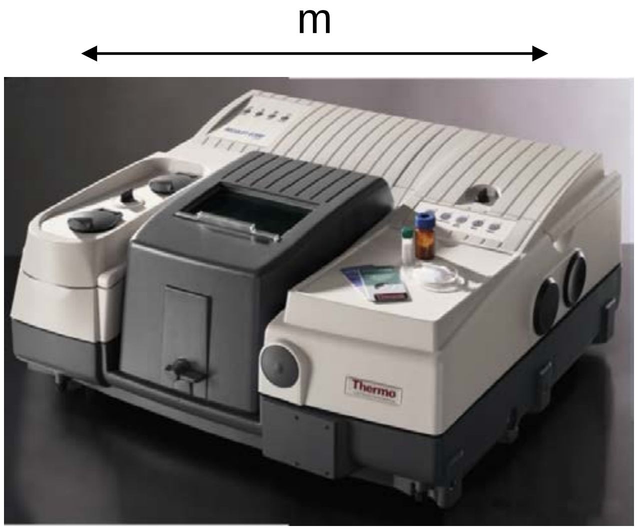

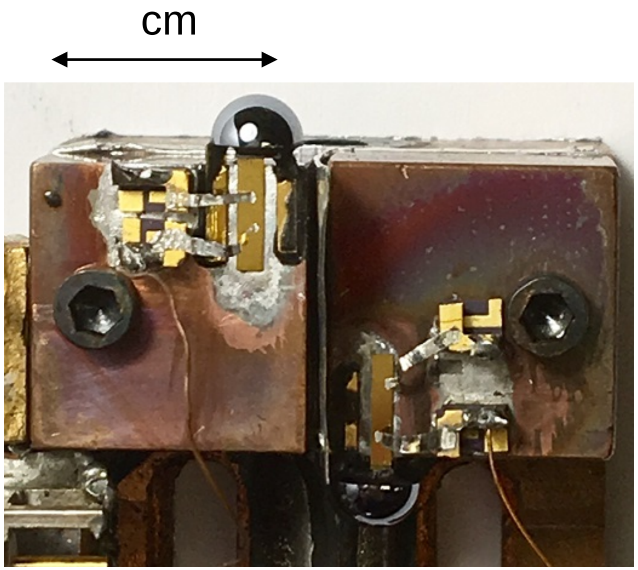

Semiconductor frequency combs can perform high-SNR spectroscopy without any moving parts!

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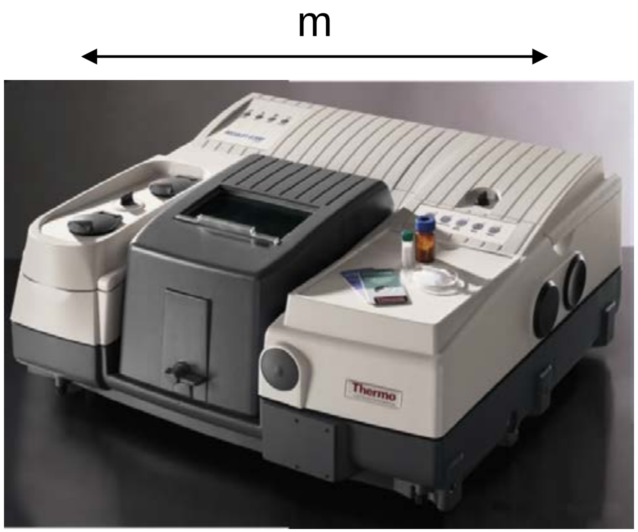

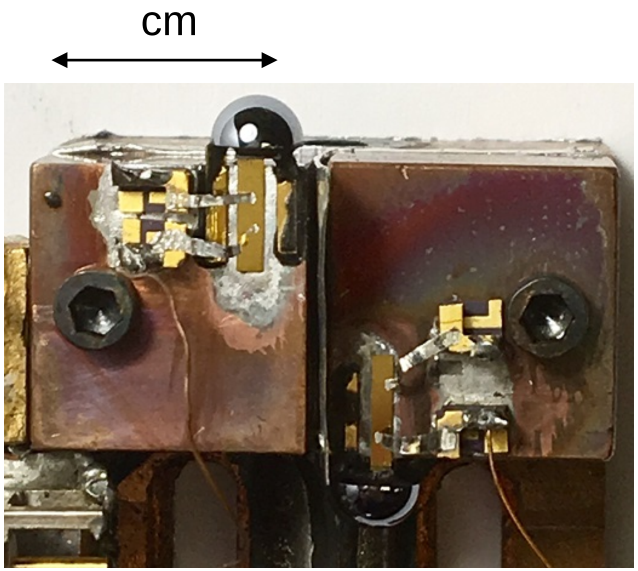

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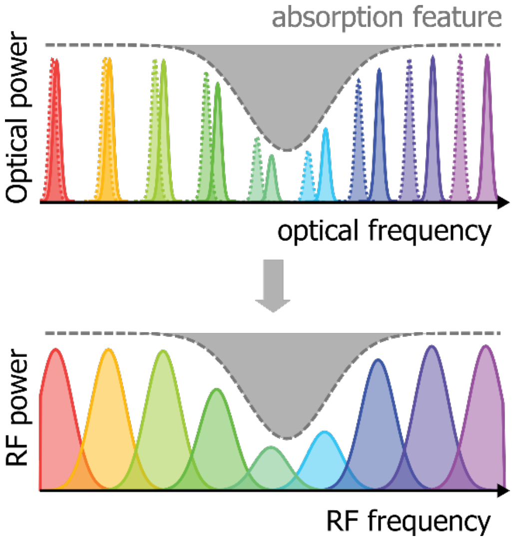
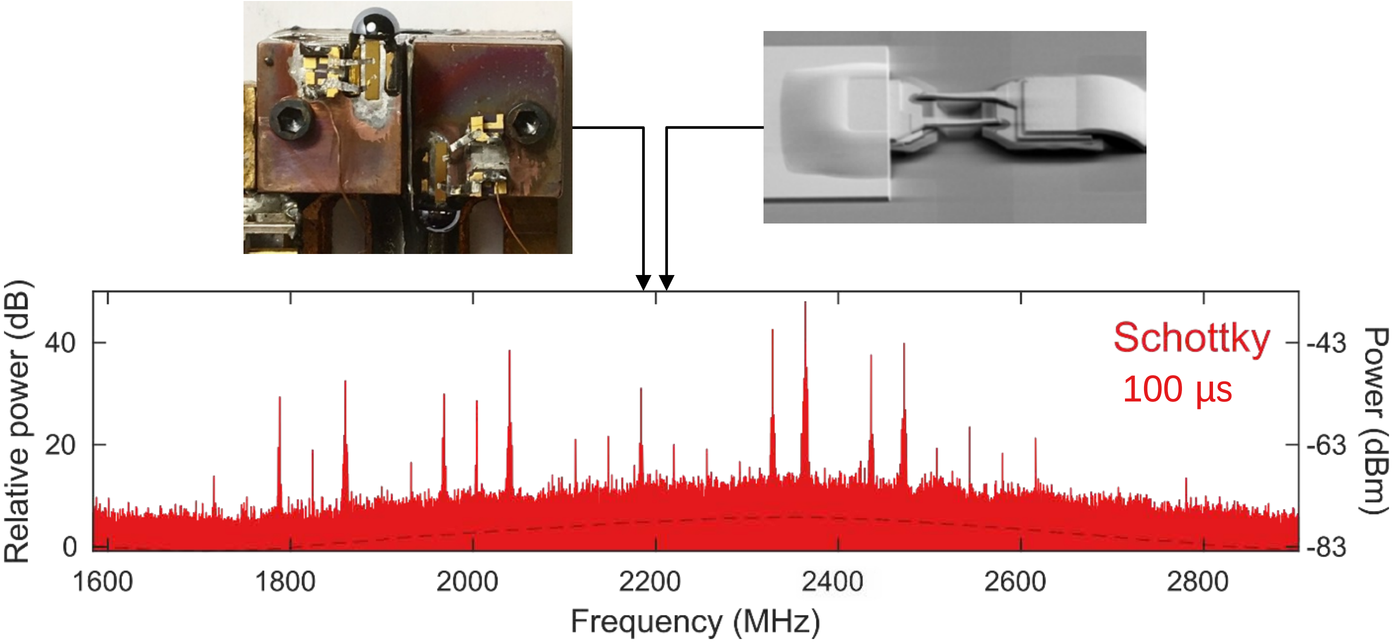


Dual comb spectroscopy

Terahertz QCL dual comb spectroscopy

- With dual comb spectroscopy, can perform broadband spectroscopy using chip-scale components. No spectrometer and no moving parts!



Yang, Burghoff et al., “Terahertz multiheterodyne spectroscopy using laser frequency combs,” *Optica* (2016).

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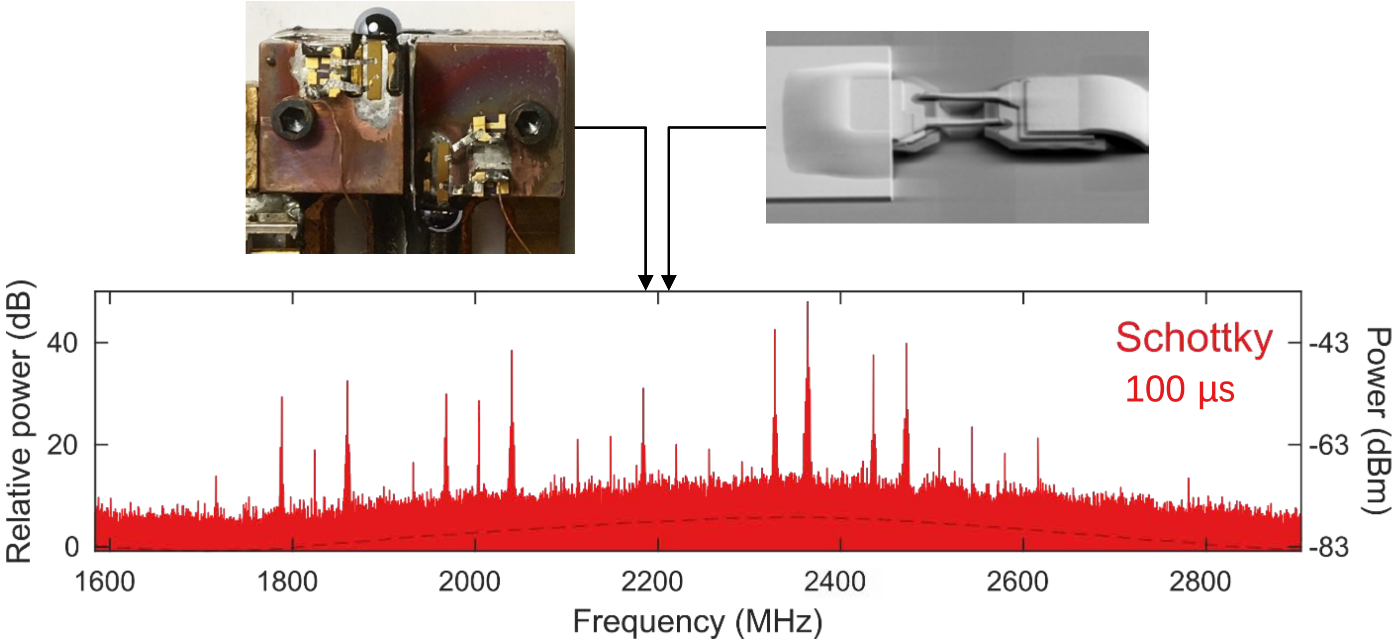

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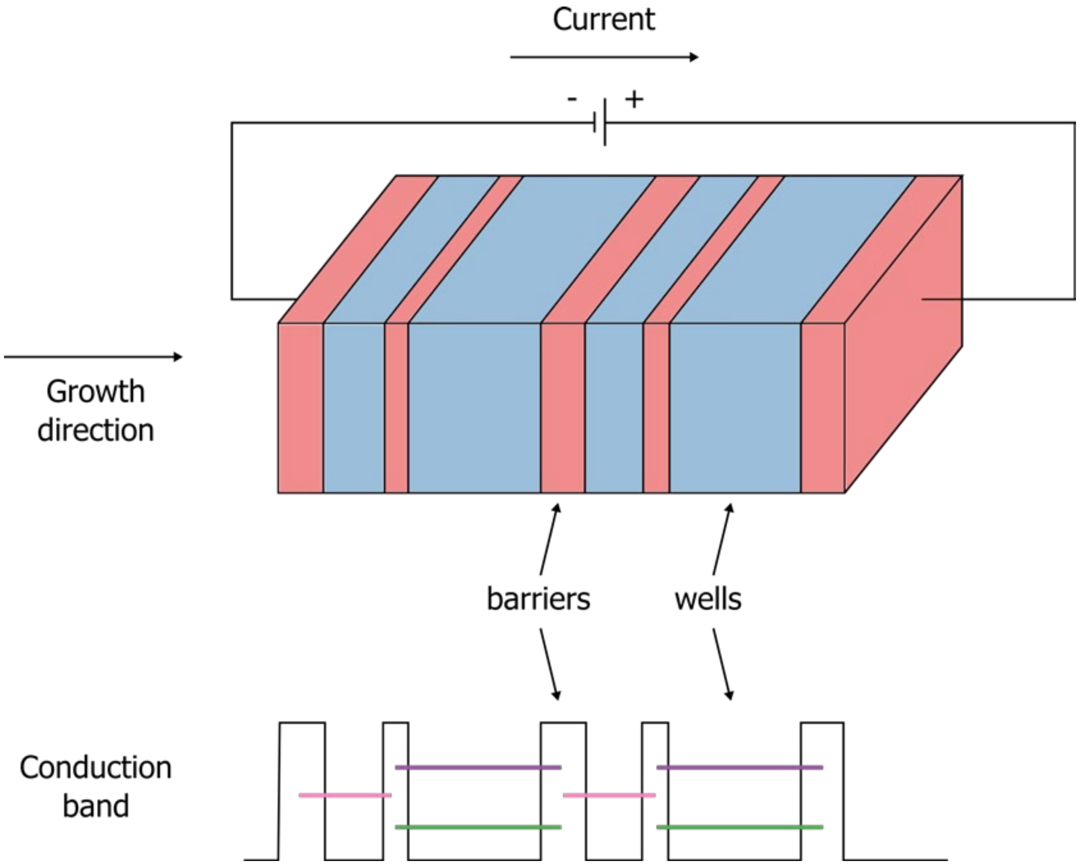
- **Part I** : Quantum cascade laser (QCL) basics
- **Part II** : Comb formation in QCLs
- **Part III** : Dual comb spectroscopy based on THz QCLs

Part I

Quantum cascade laser basics

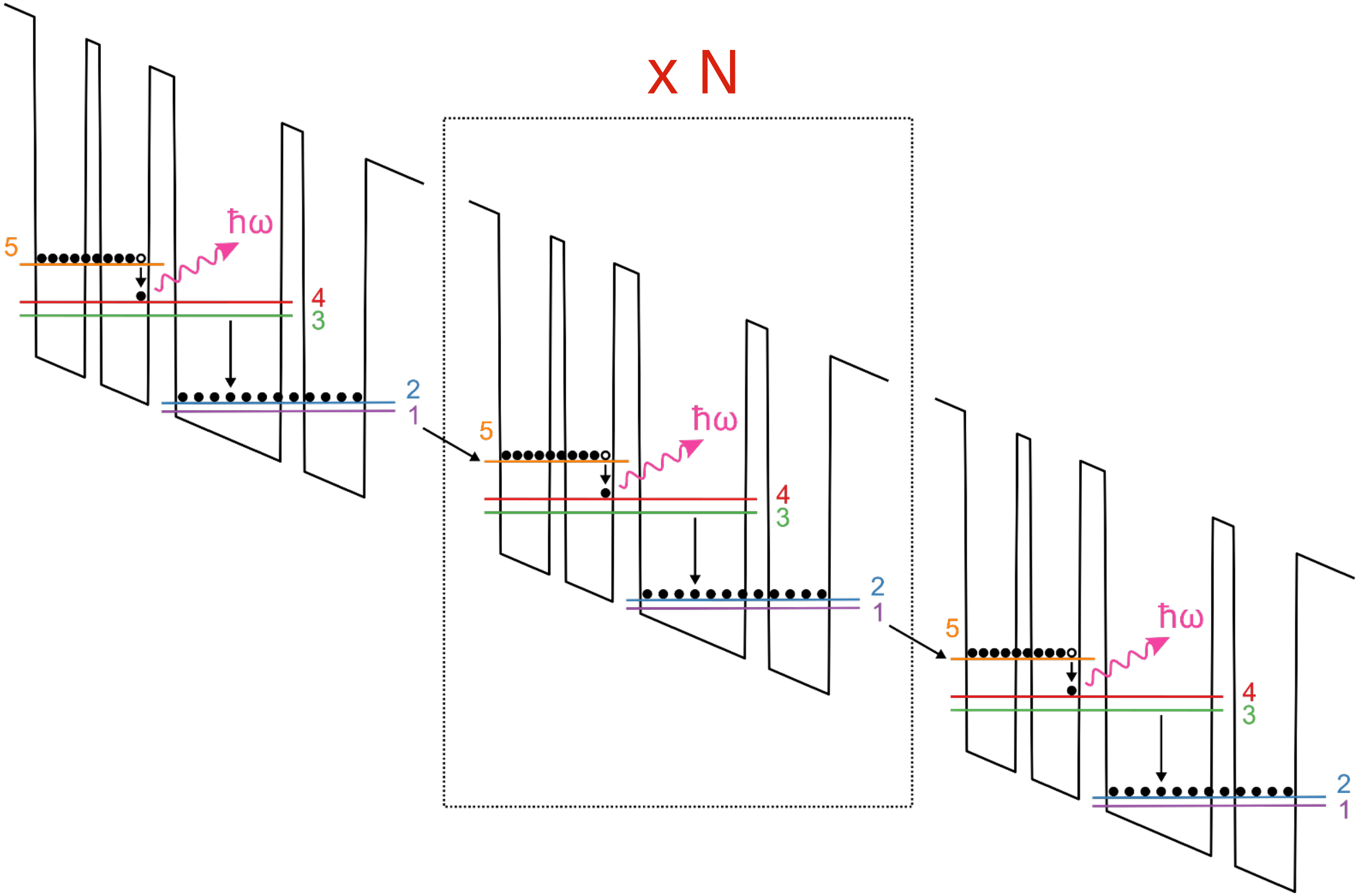
Quantum cascade lasers: a triumph of band structure engineering

- Basic idea: grow a series of quantum wells and barriers on a semiconductor substrate



Can make “artificial atoms”

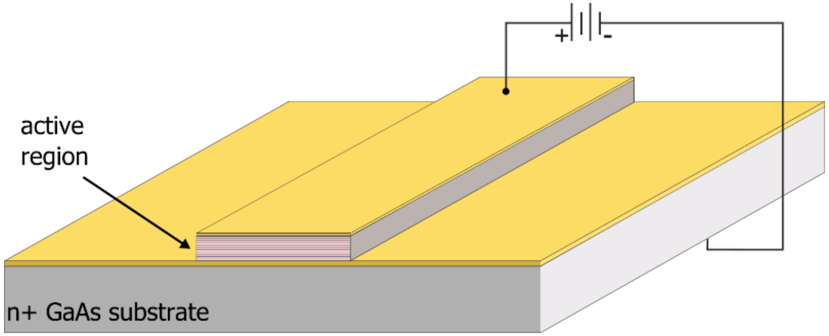
Electrically-pumped intersubband lasers



- $\hbar\omega$ chosen by design—not nature
- No electron-hole recombination. One electron cascades down N identical modules, generating N photons.

Terahertz quantum cascade lasers (1-5 THz)

- Main focus of this talk: THz quantum cascade lasers (GaAs/AlGaAs)
  - Metal-metal waveguide: gain medium sandwiched between two gold layers



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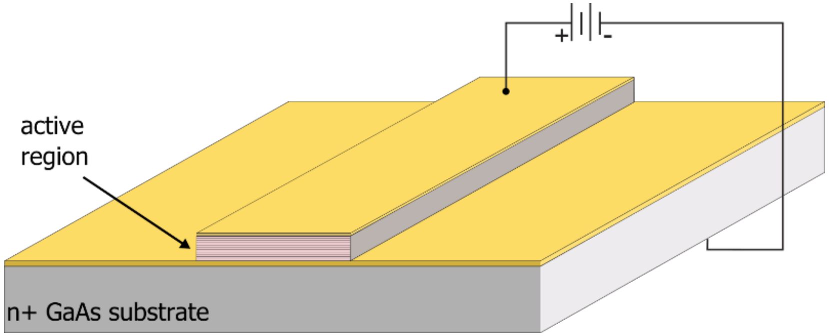
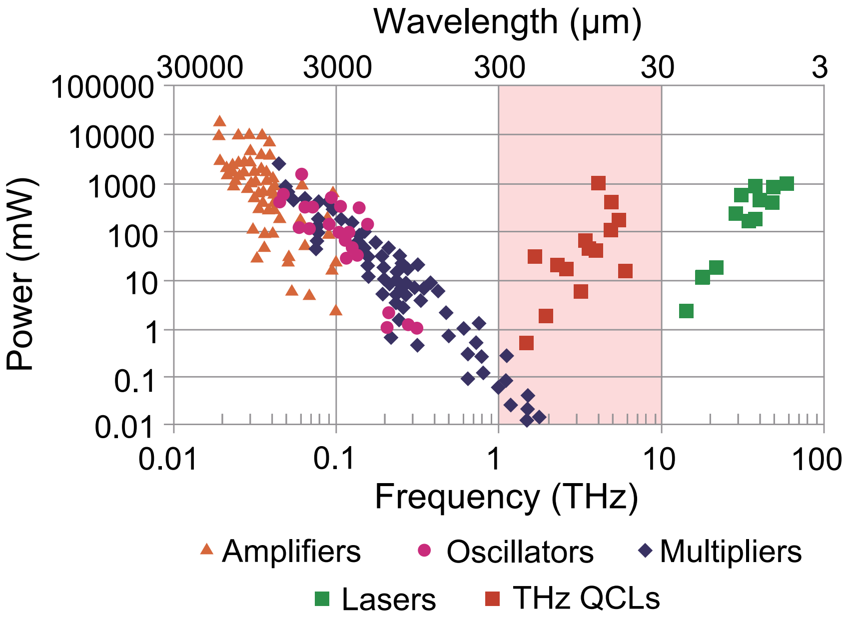




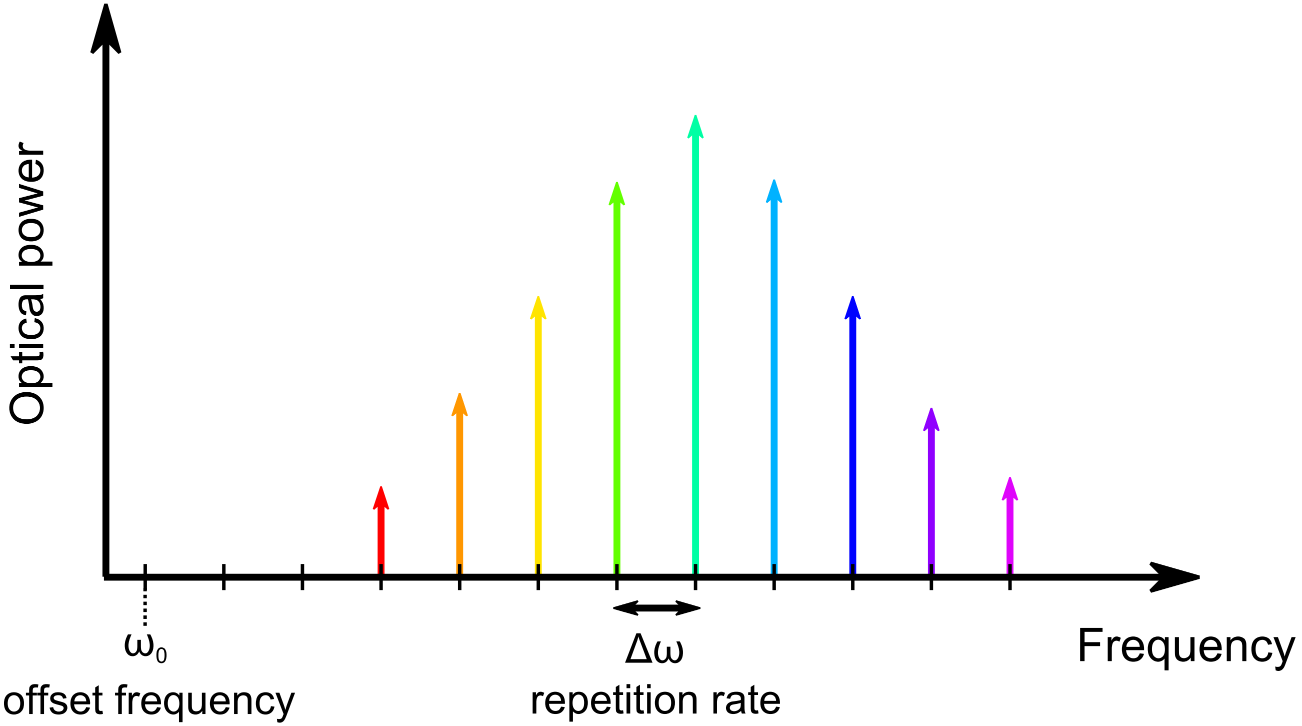
- Milliwatt power, low cost and size
- **Excellent platform for nonlinear optics**
  - large intersubband nonlinearity, intracavity power (W)

Part II

Comb formation in QCLs

What is a frequency comb?

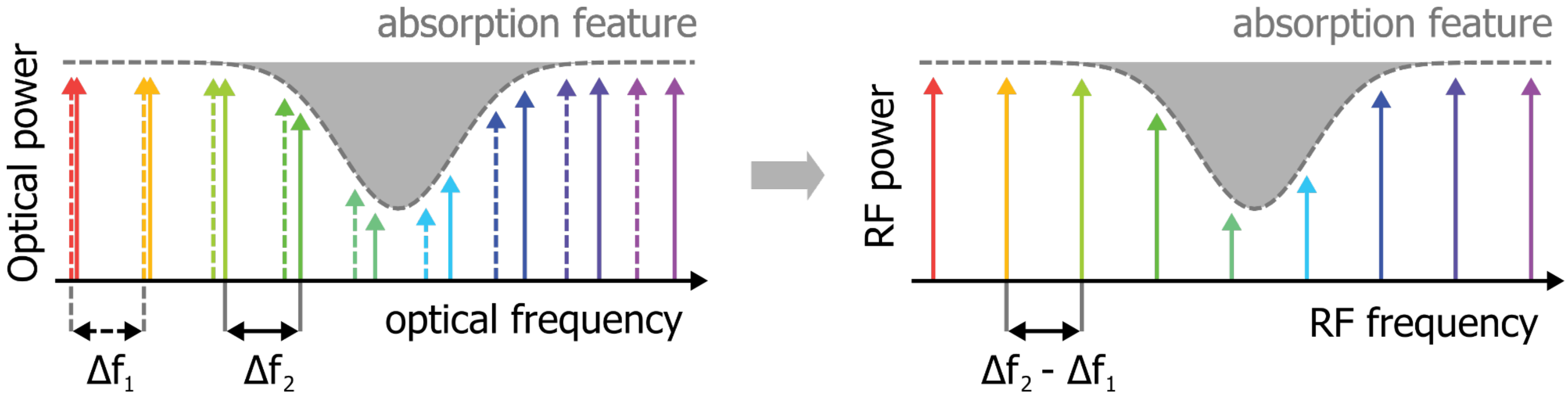
- **Frequency combs**: Light sources that consist of a large number of **evenly-spaced** laser lines



- First laser combs based on pulsed mode-locked lasers (e.g., Ti:Sapphire)

Frequency combs for frequency discrimination

- How are combs relevant for spectroscopy?
  Dual comb spectroscopy (multi-heterodyne)
  - Two combs, with slightly different spacings, shined on fast detector



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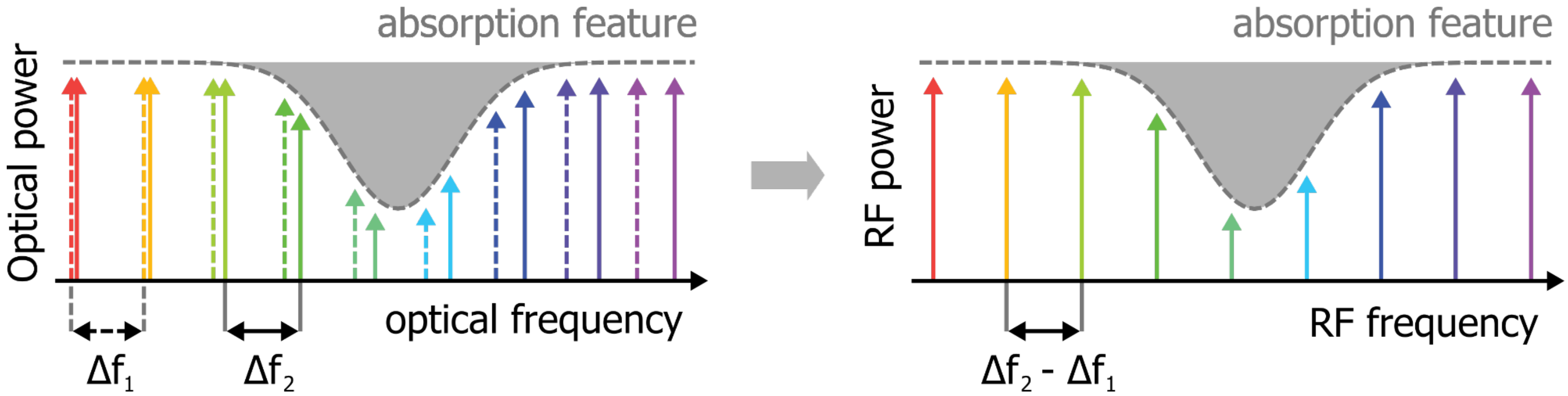
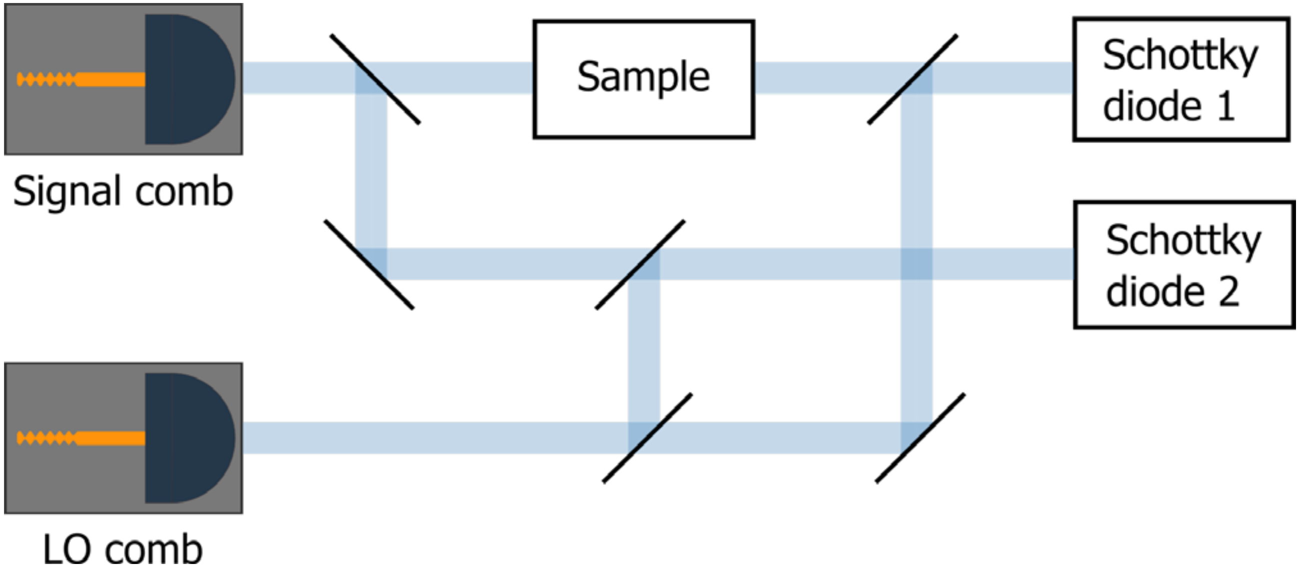


- **Optical** spectrum encoded onto **electrical** spectrum
- Can have broadband capabilities and high sensitivity, without moving parts!

Dual comb spectroscopy can be very cheap and small



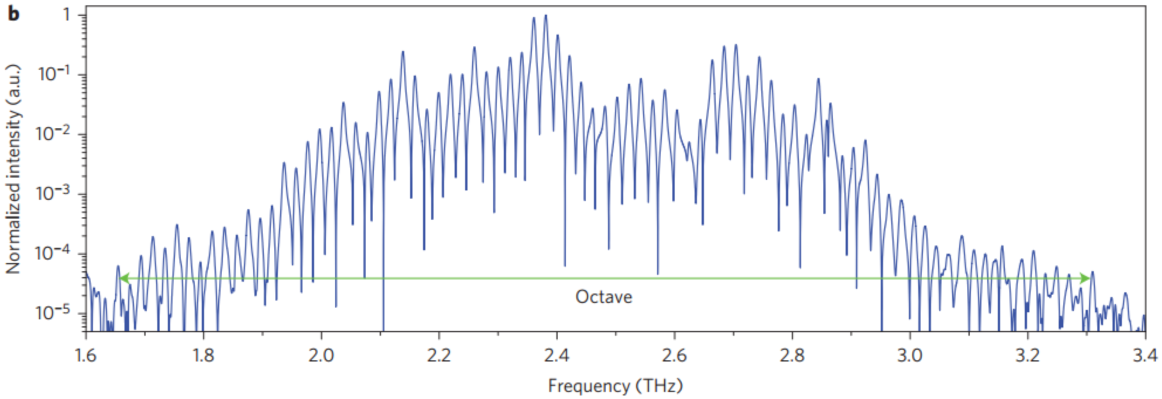
- Can make very compact THz spectrometers
  - Lasers: could be $
  - Schottky diodes (Si): ¢

- Coherence requirements not that severe

But how do you get a quantum cascade laser to form a comb?

QCLs are intrinsically broadband devices

- Spatial & spectral hole burning → broad spectrum
- Can mix different gain media for even broader spectrum



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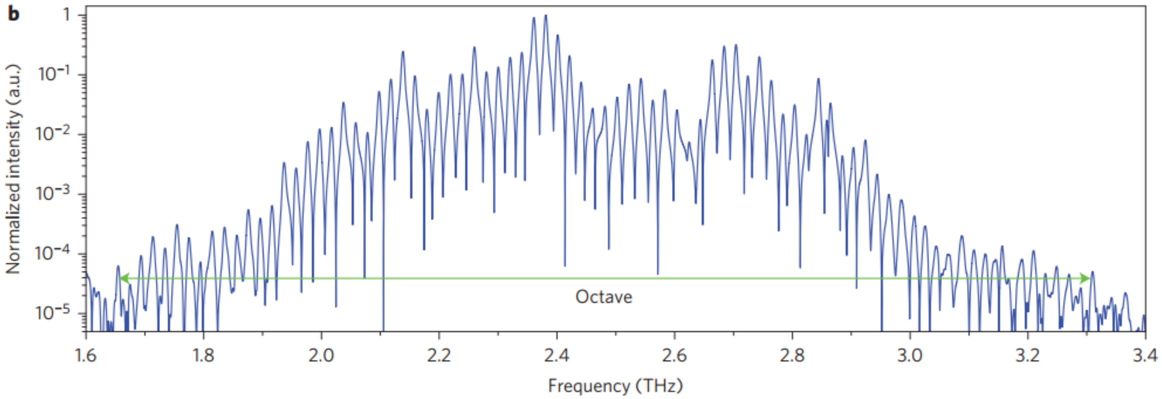

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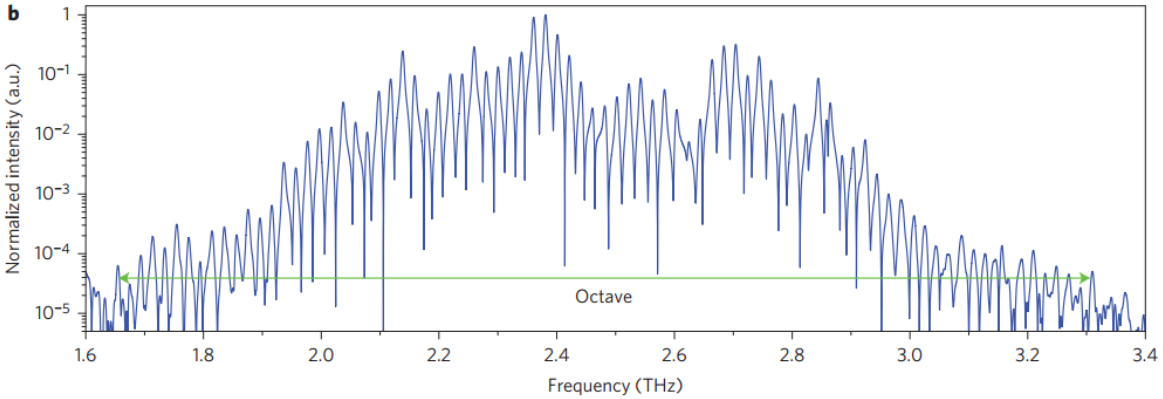

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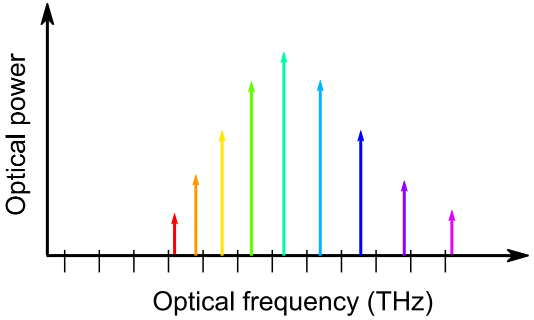

non-comb

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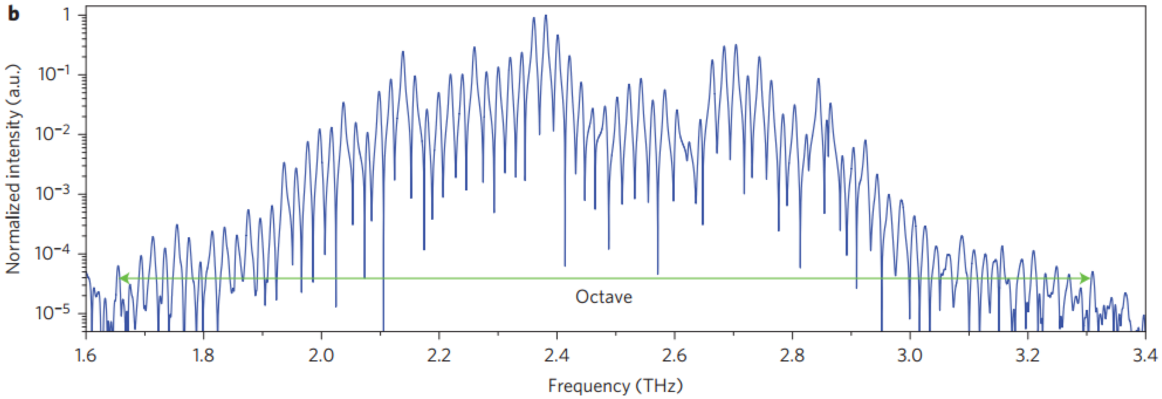

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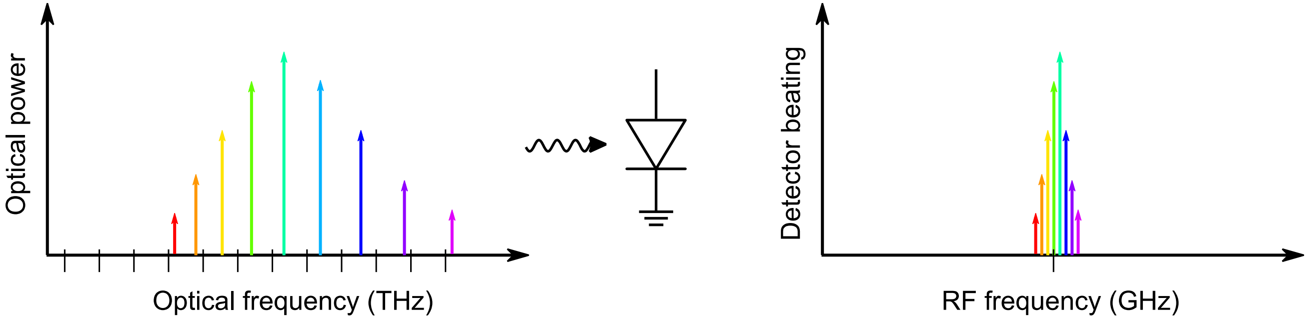

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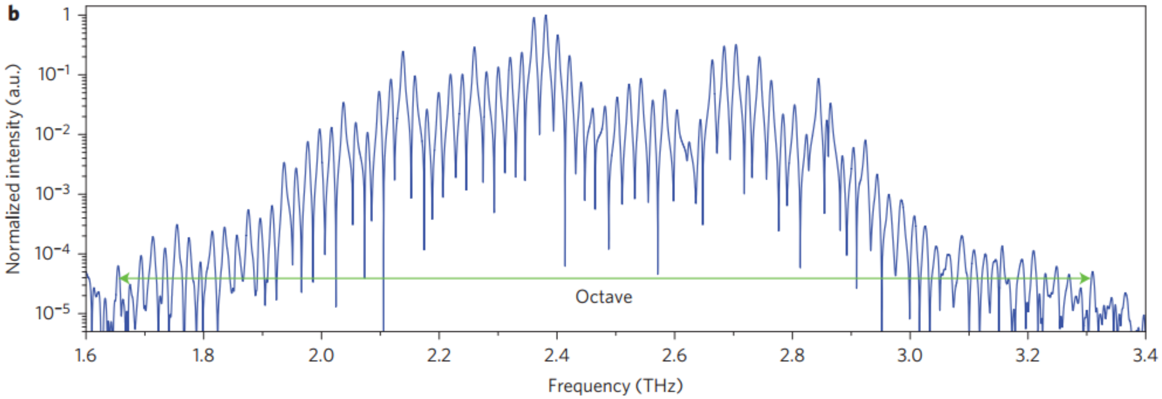

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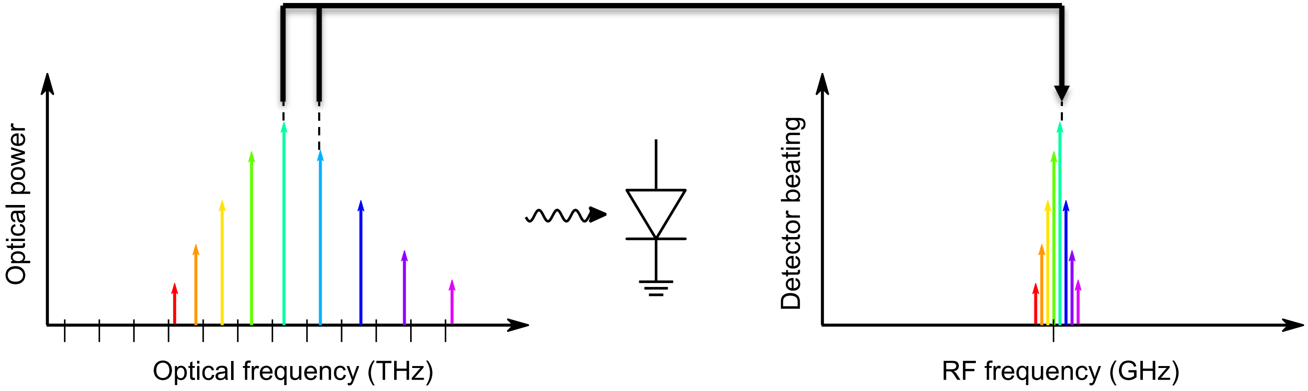

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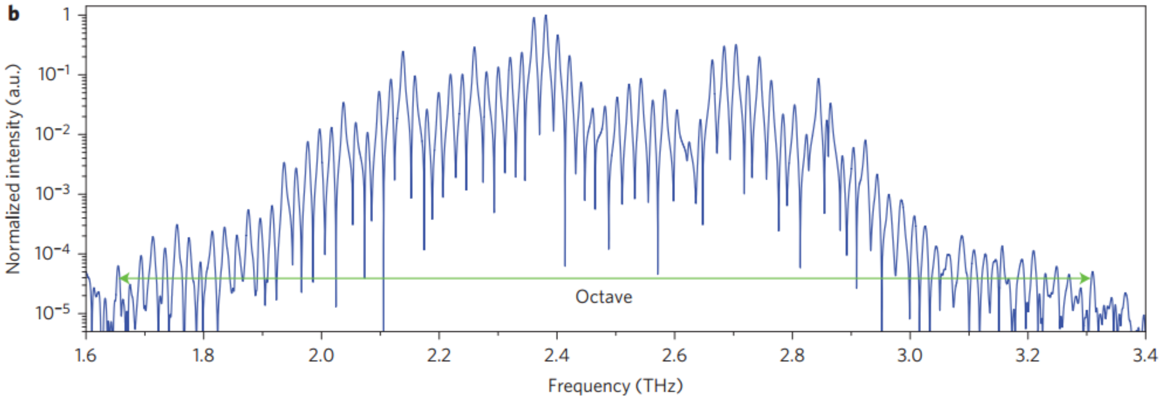

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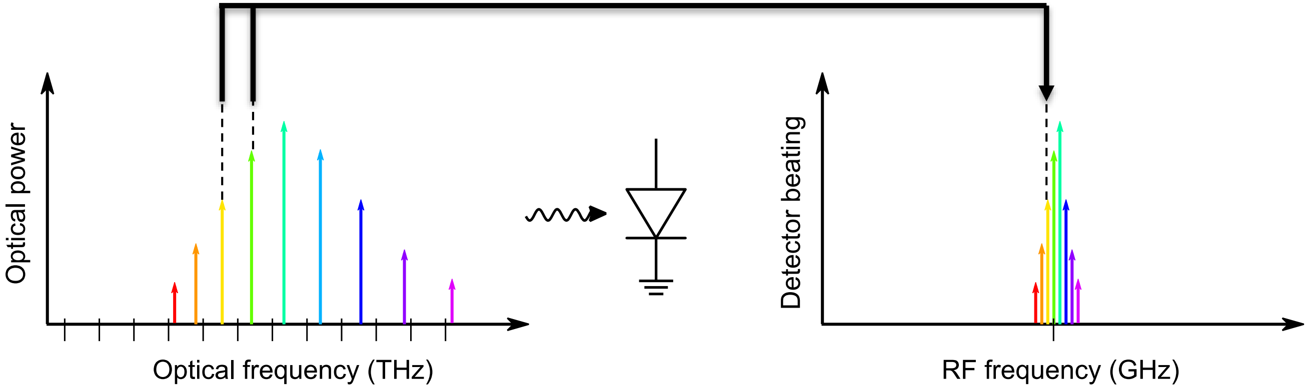

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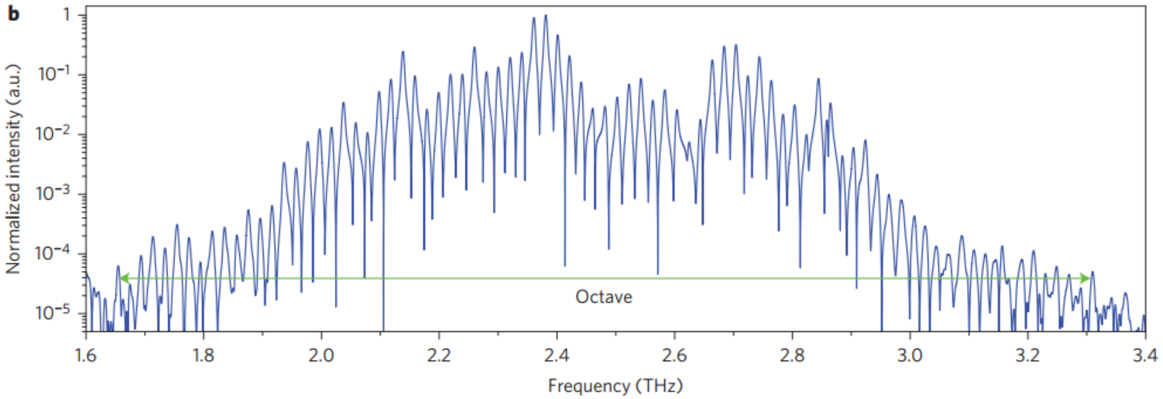

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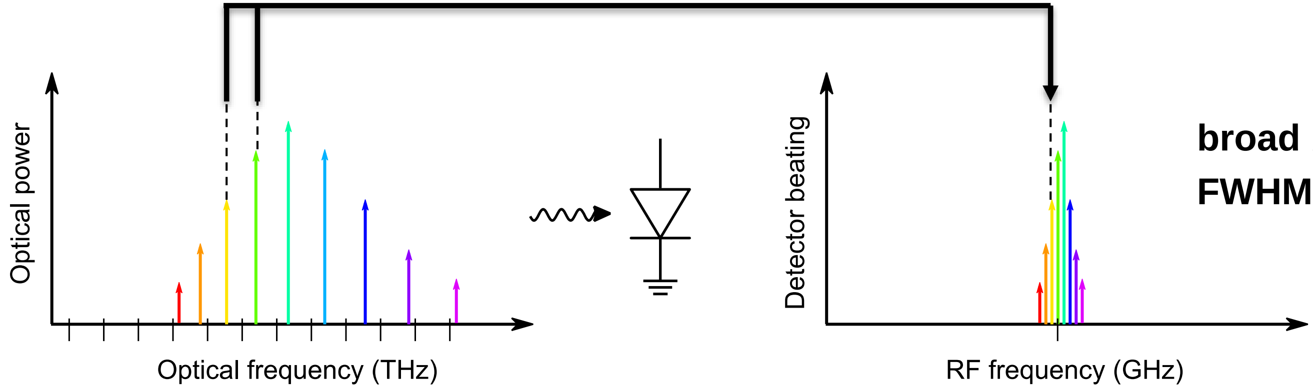

broad beatnote

FWHM >> MHz

non-comb

Spontaneous comb formation in QCLs

- Long been known that the beatnote can spontaneously narrow (100 MHz to 10 kHz)

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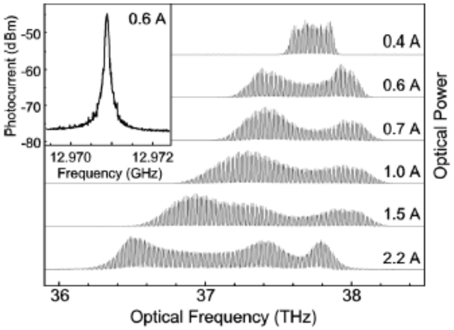

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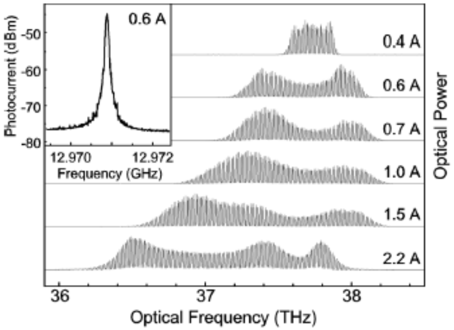

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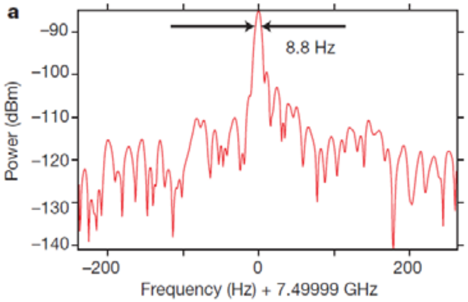

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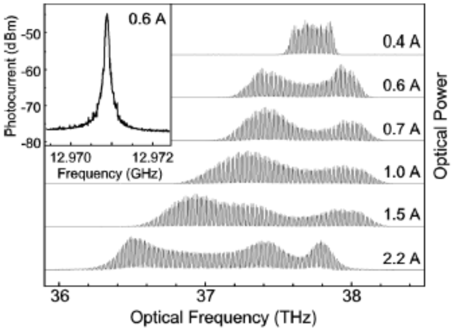

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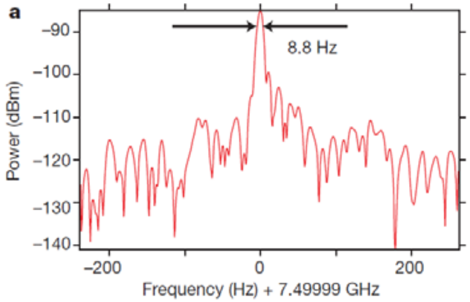

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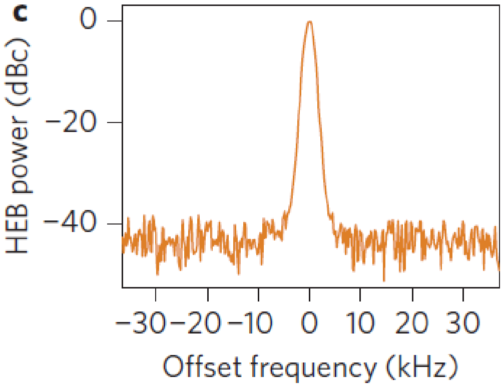

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Spontaneous comb formation in QCLs

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Mid-IR

  - 2000: First observed by Capasso group, attributed to pulse formation
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THz

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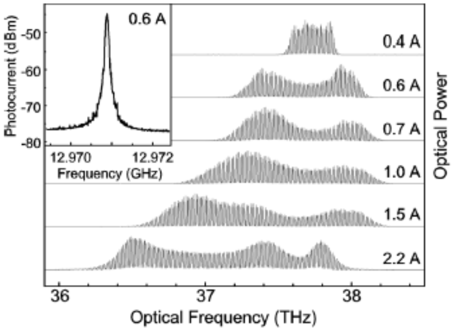

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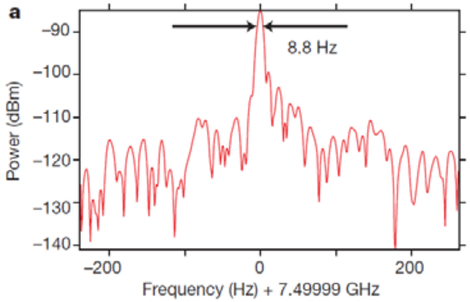

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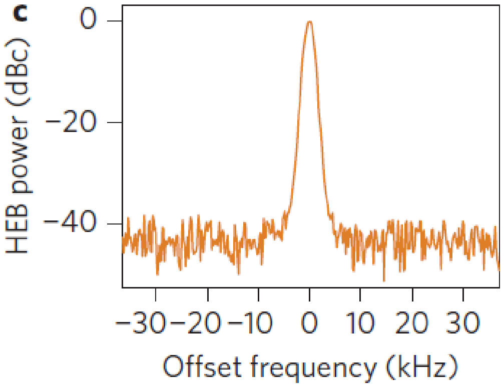

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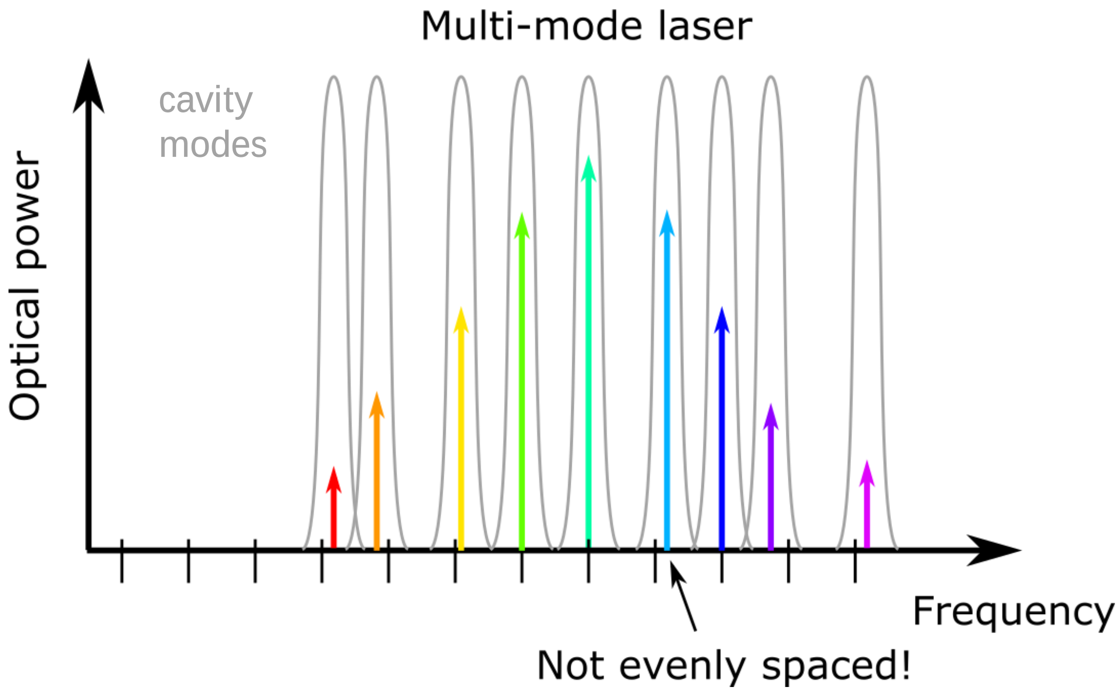
Quantum cascade laser combs: basic principles

- QCL lasing is naturally broadband, but uncorrelated
- Four-wave mixing allows lines to synchronize, pull new lines into comb
- Can collapse into a comb state



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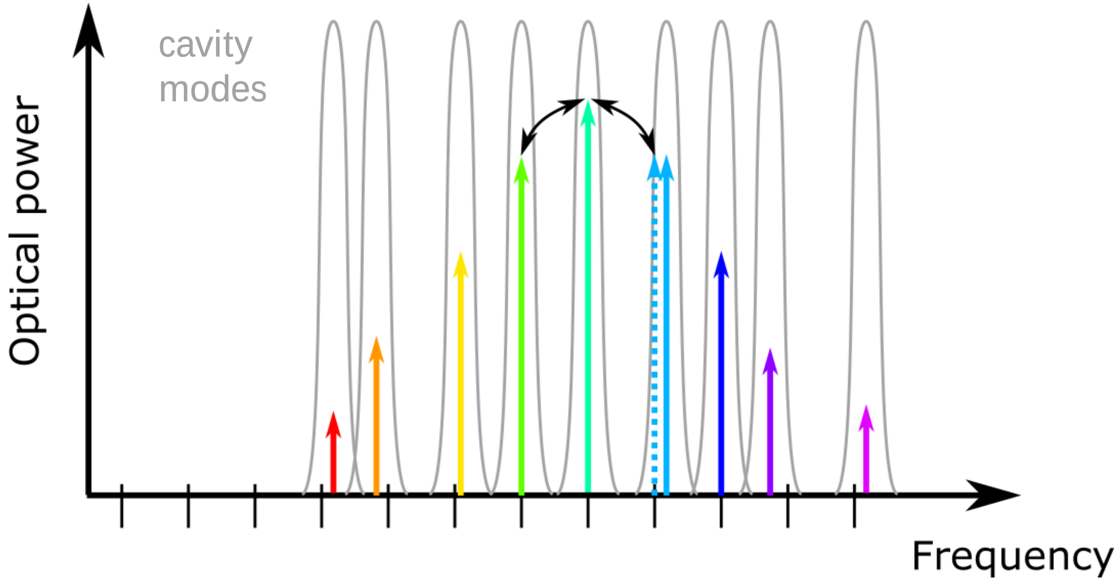
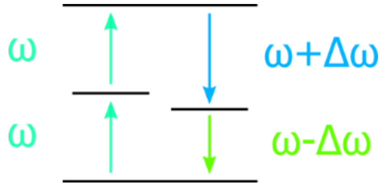
Degenerate four-wave mixing generates sidebands



Not evenly spaced!

Degenerate four-wave mixing ($\chi^{(3)}$)



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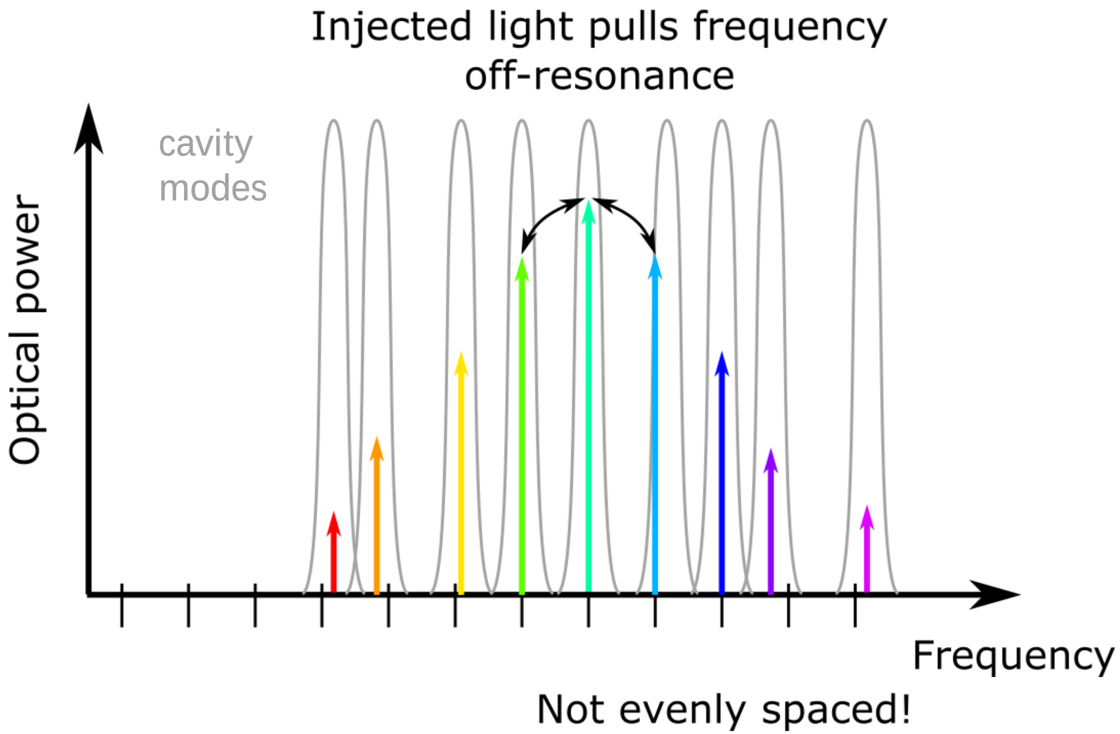

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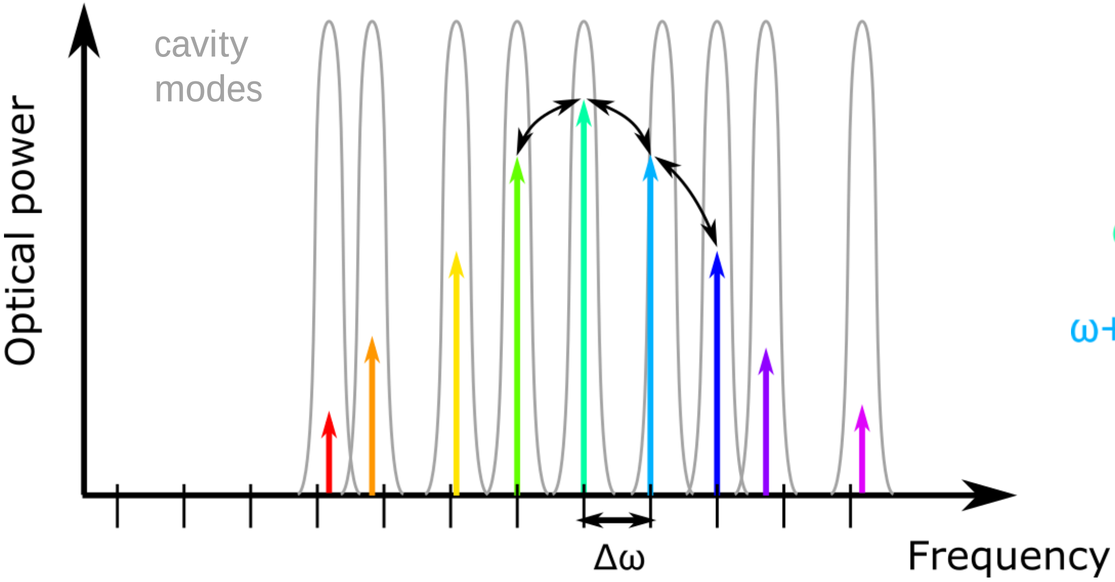
Nondegenerate four-wave mixing
more lines off-resonance



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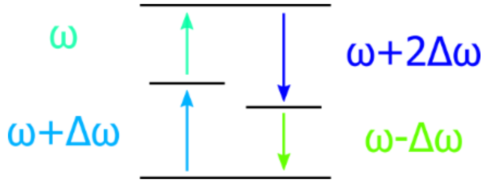

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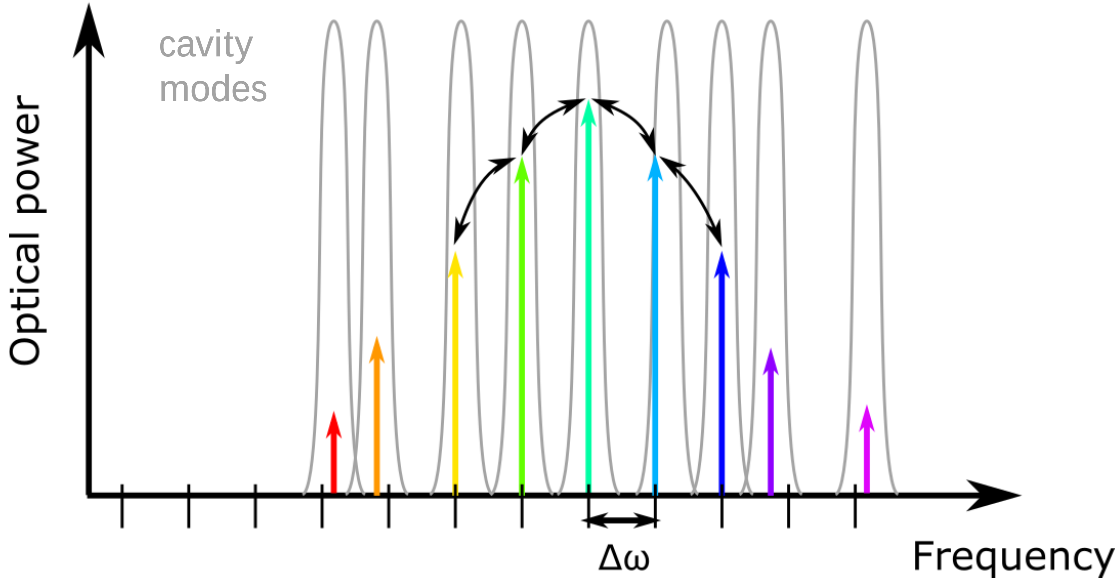

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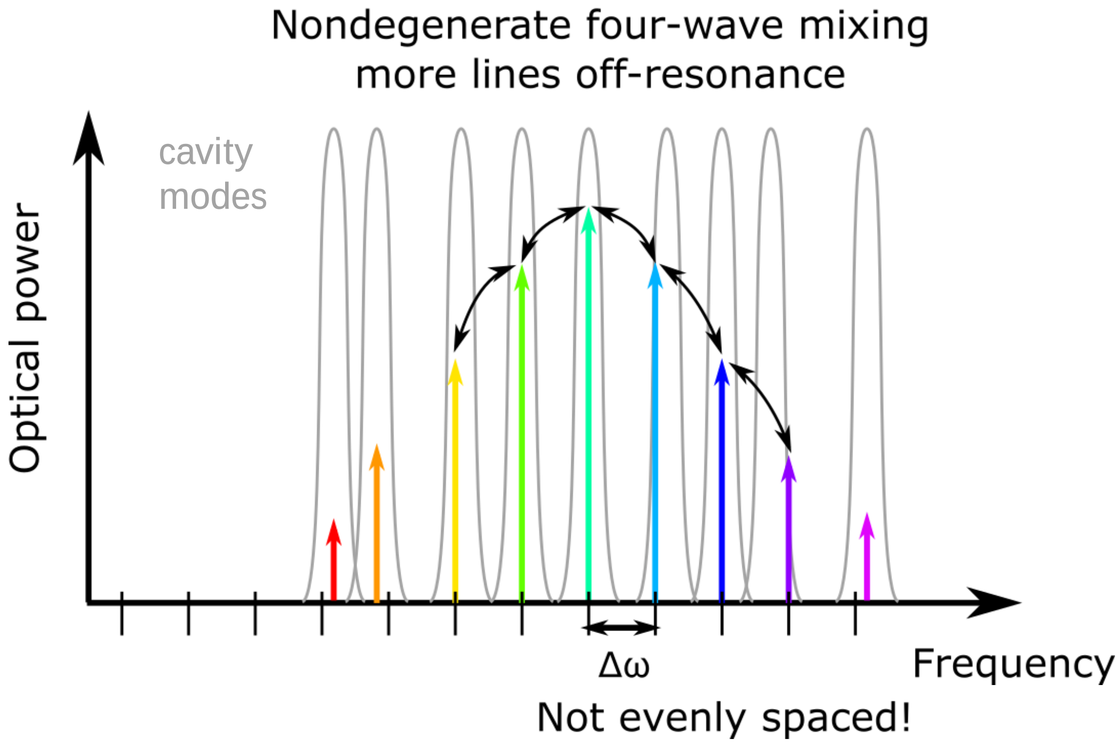

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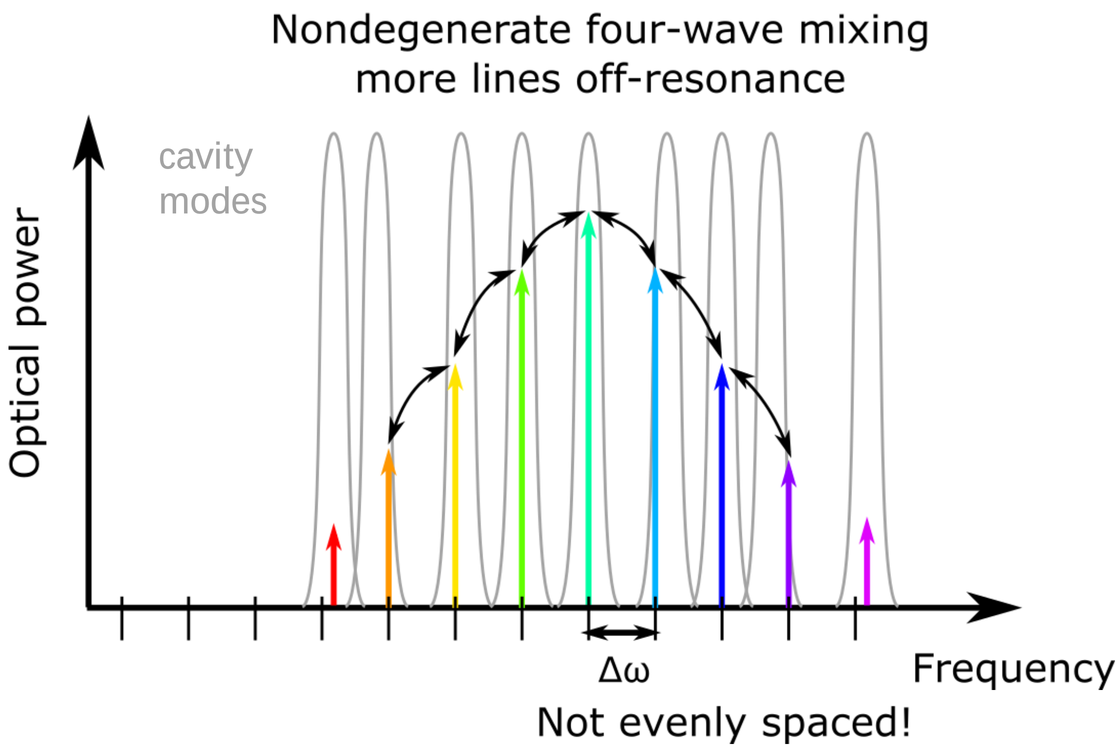

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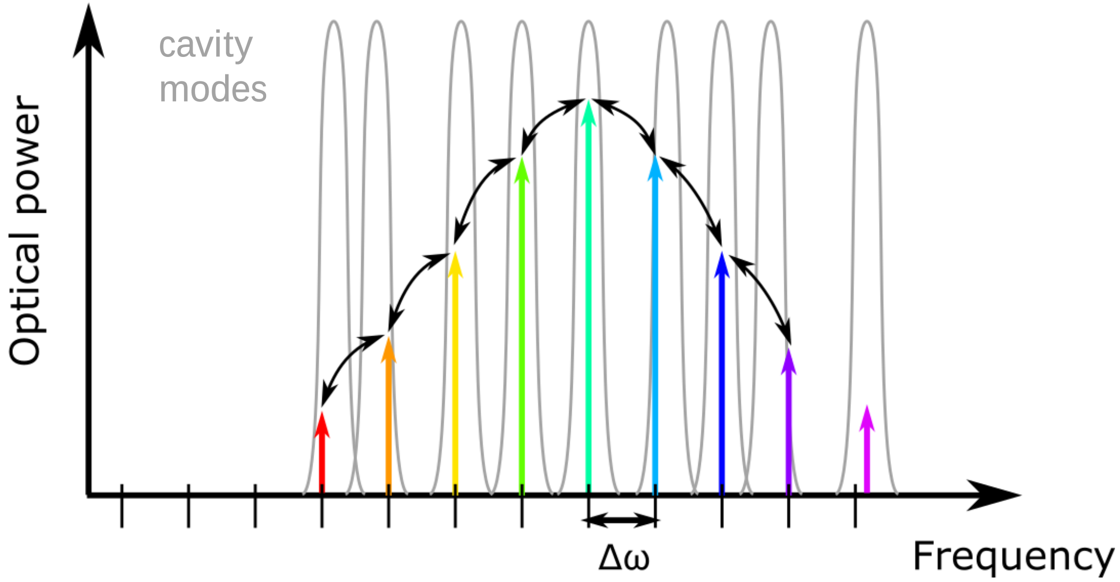

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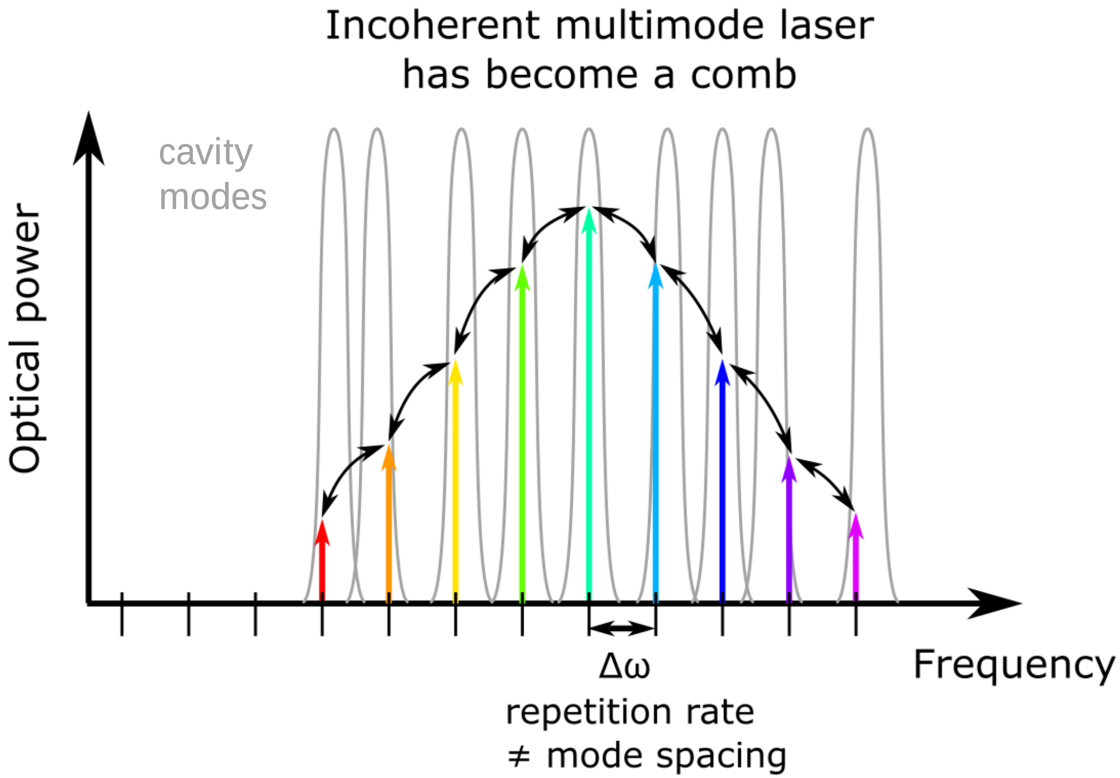

Dispersion: the enemy of comb formation

- Spontaneous comb formation is difficult to observe in terahertz QCLs. Why?

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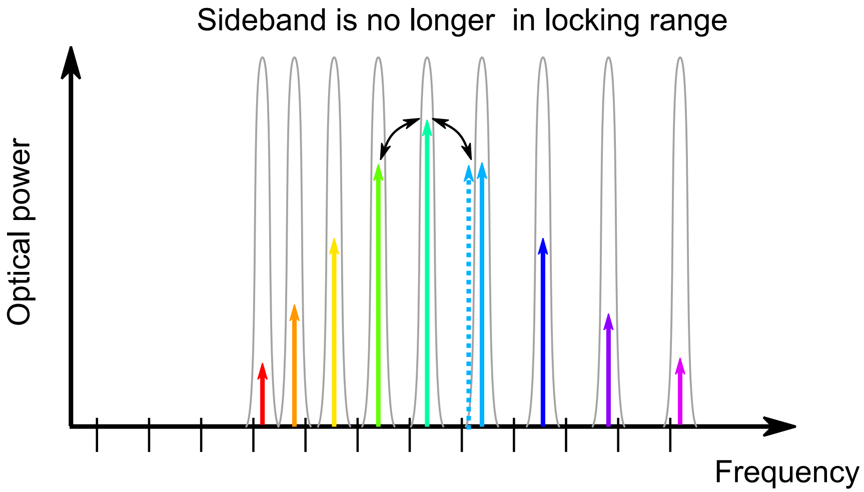
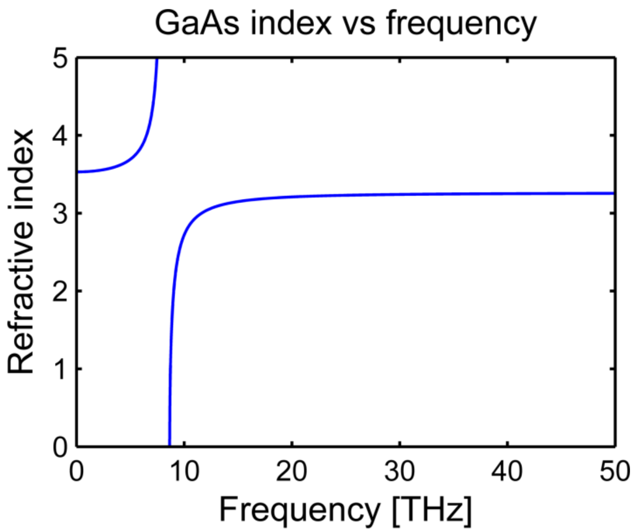


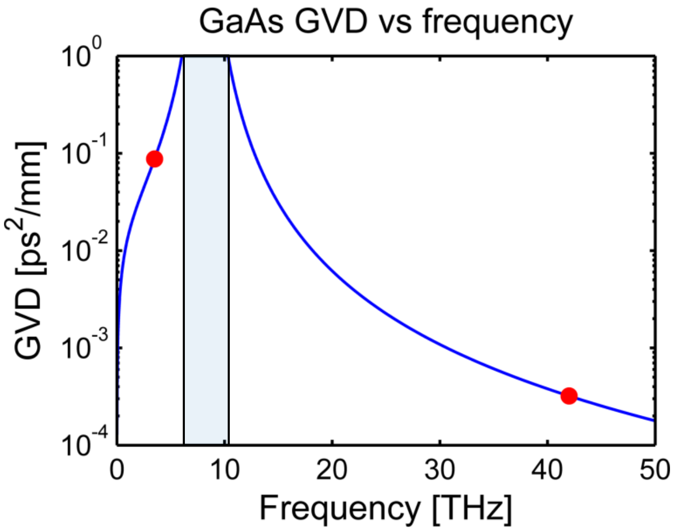
- As group velocity dispersion (GVD) increases, modes are more and more non-uniform
- Four-wave mixing is too far off-resonance, injection locking cannot occur

Dispersion: the enemy of comb formation

- Spontaneous comb formation is difficult to observe in terahertz QCLs. Why?

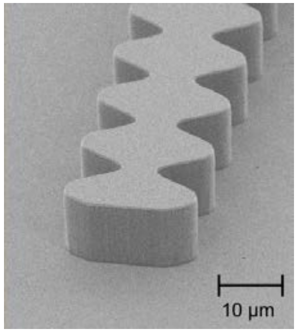
- III-V materials are particularly dispersive in THz
  - GaAs at 3.5 THz: 87,400 fs^2/mm
  - Frequencies separated by 1 THz will slip by λ/4 after only 130 μm!

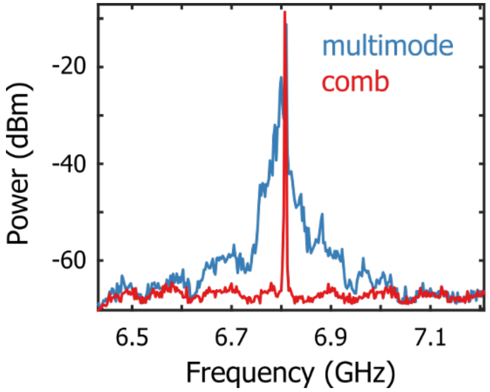




Combs enabled by dispersion engineering

- Spontaneous comb formation is unpredictable and hard to reproduce between material growths, so compensate dispersion instead





Burghoff et al., “Terahertz laser frequency combs,” *Nat. Photon.* (2014)

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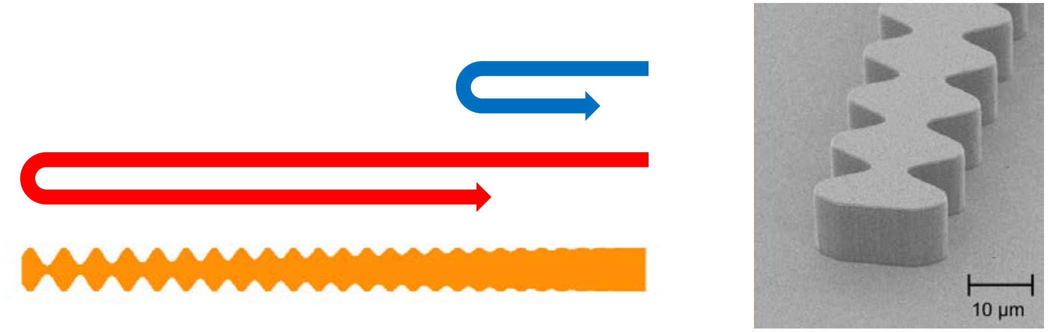
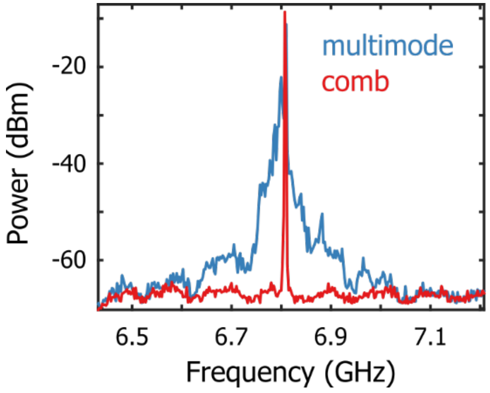
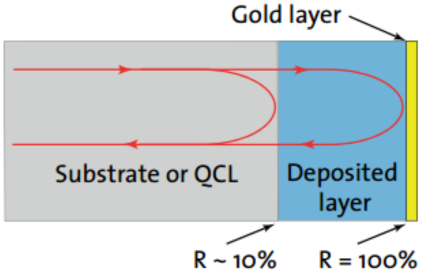


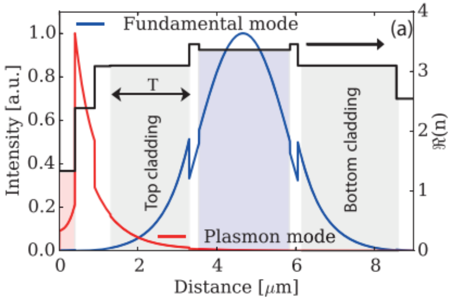


Burghoff et al., “Terahertz laser frequency combs,” *Nat. Photon.* (2014)

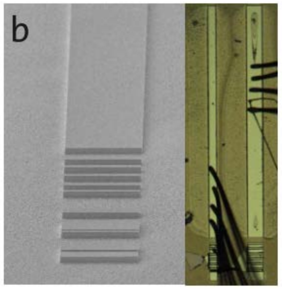
- Other GVD compensation schemes in QCLs:



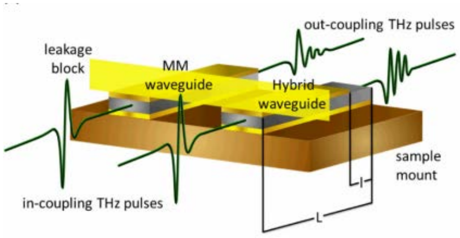
Villares et al., *Optica* (2016)



Bidaux et al., *Opt. Lett.* (2017)



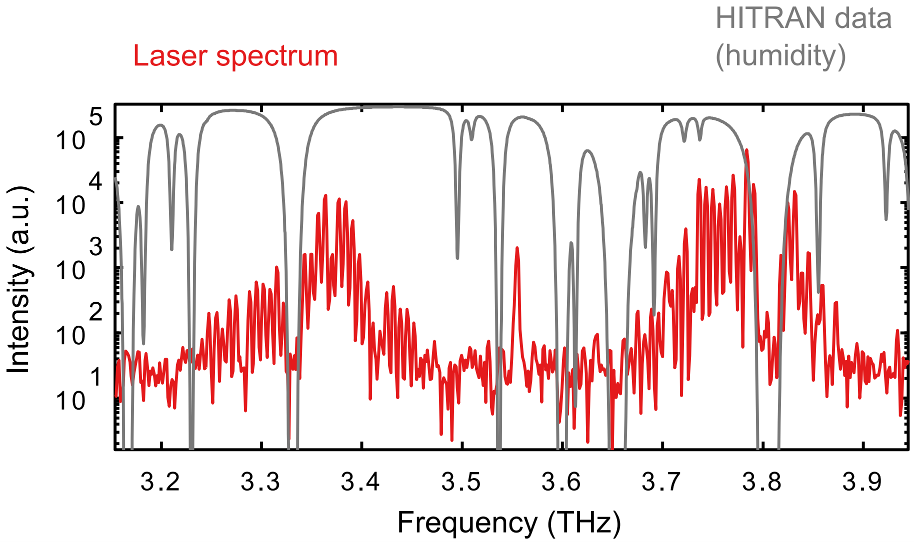
Faist et al.,
Nanophotonics (2015)

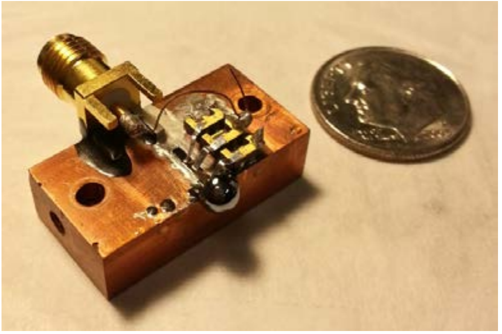


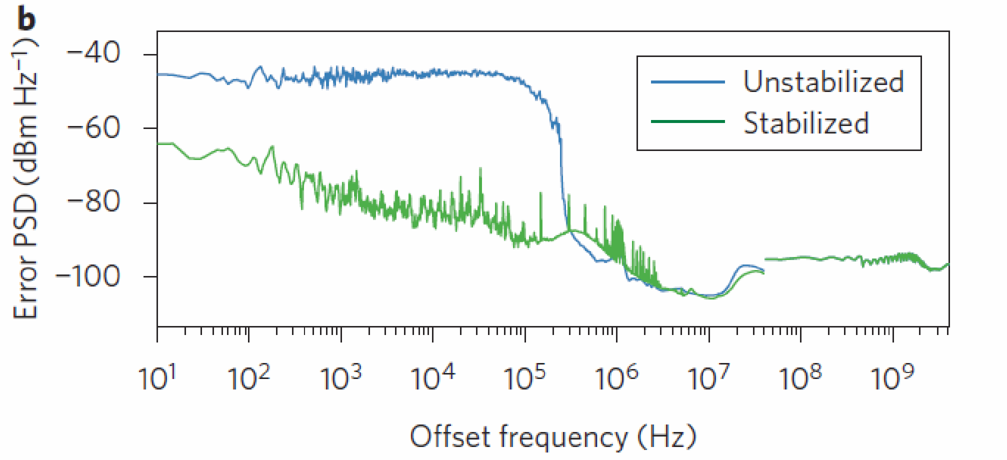
Fobbe et al.,
Opt. Express (2016)

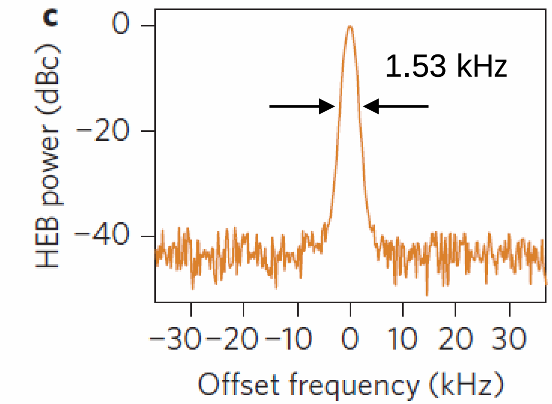
Basic results: spectra

- Properly-compensated laser generates broadband spectrum, narrow beatnote when CW-biased









Burghoff et al., *Nat. Photon.* (2014)

Spectrally-resolved coherence measurement

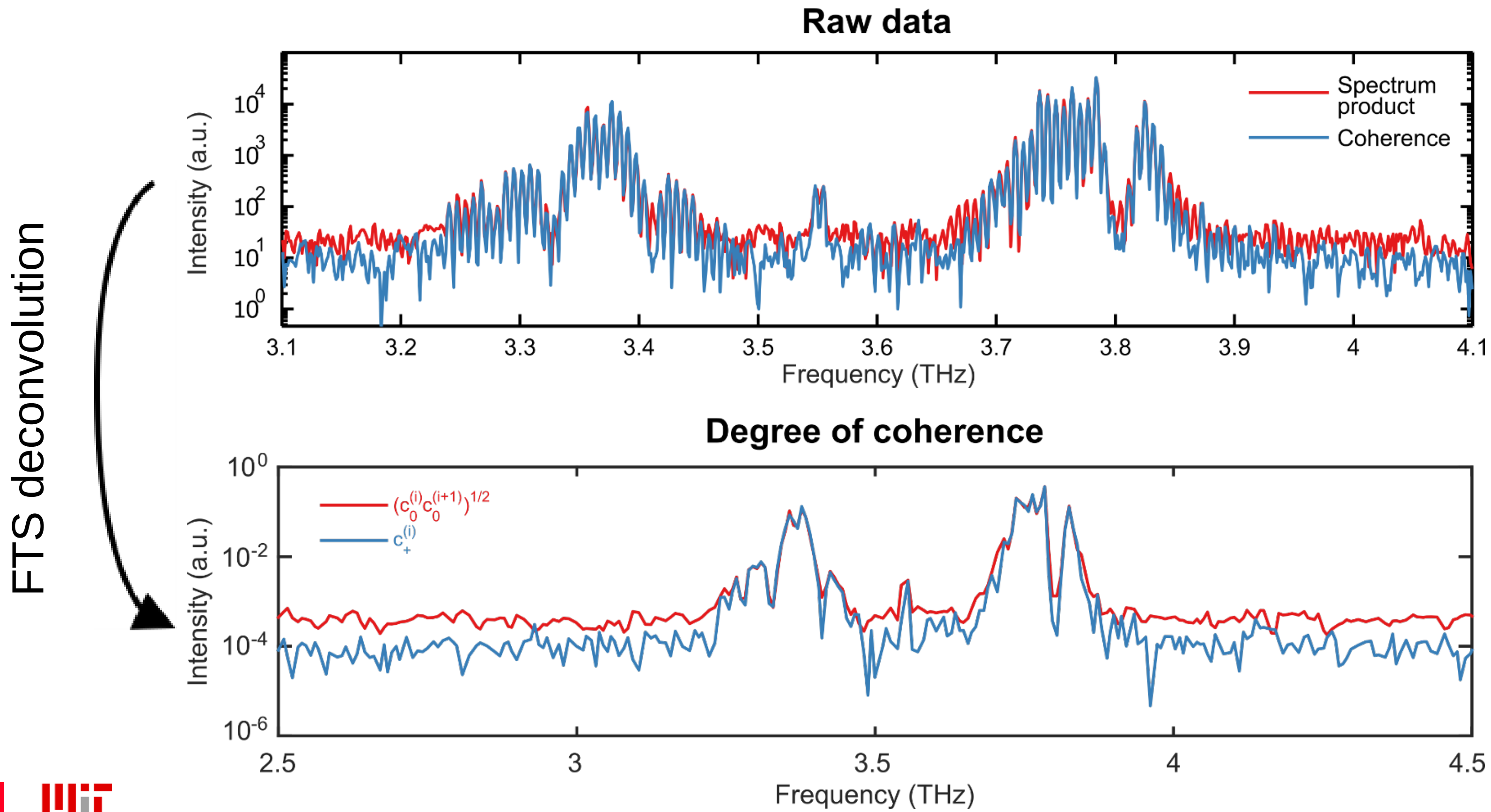
- Can use an interferometer to directly measure first-order coherence...

$$g_{\pm}(\omega) \equiv \frac{\left|\langle E^*(\omega)E(\omega \pm \Delta\omega)\rangle\right|}{\sqrt{\langle|E(\omega)|^2\rangle\,\langle|E(\omega \pm \Delta\omega)|^2\rangle}}$$

← coherence

← spectrum

- Dispersion-compensated THz QCL comb

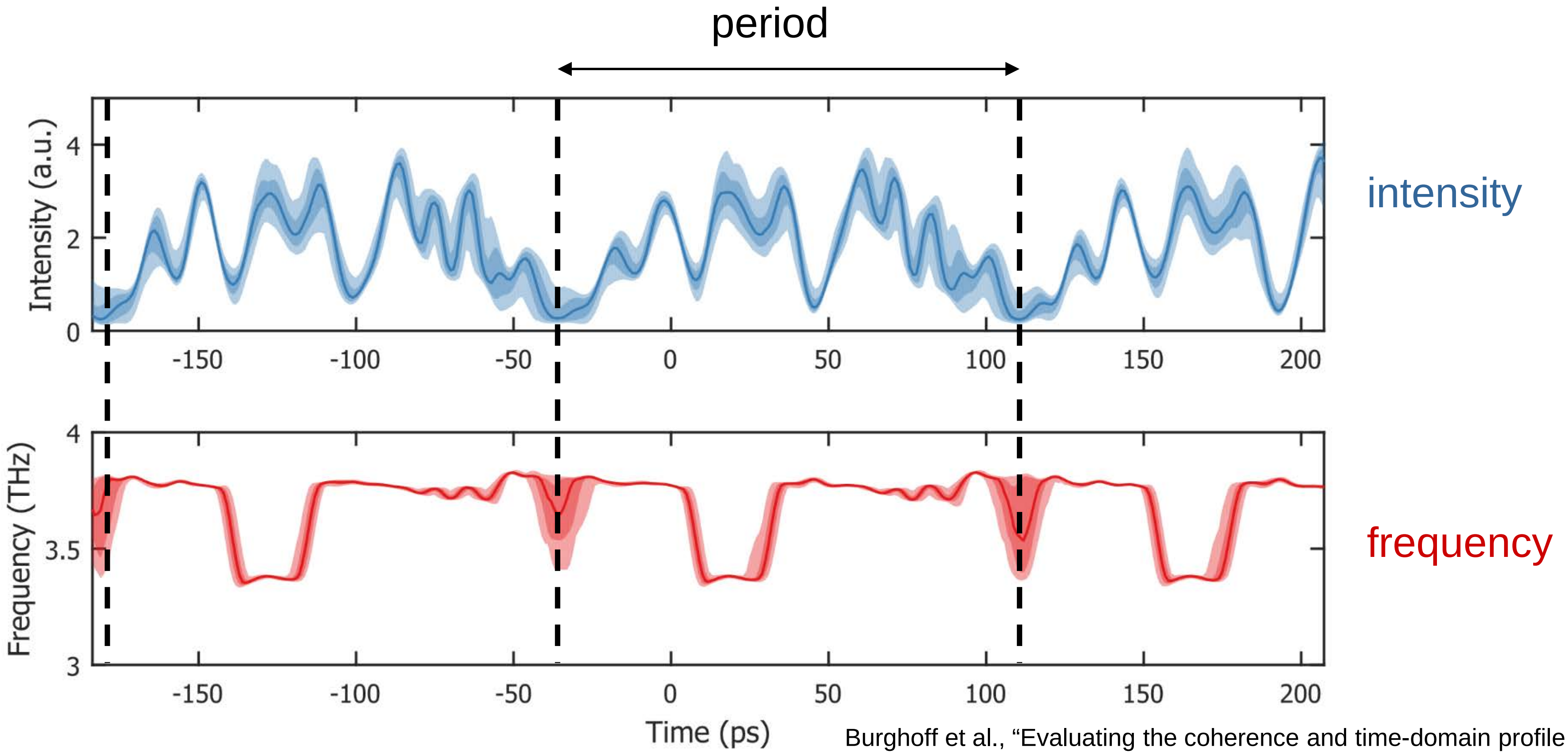


Raw data

Degree of coherence

$g^{(2)}$: Benea-Chelmus, Faist et al., PRA (2016)

Temporal properties of QCL combs



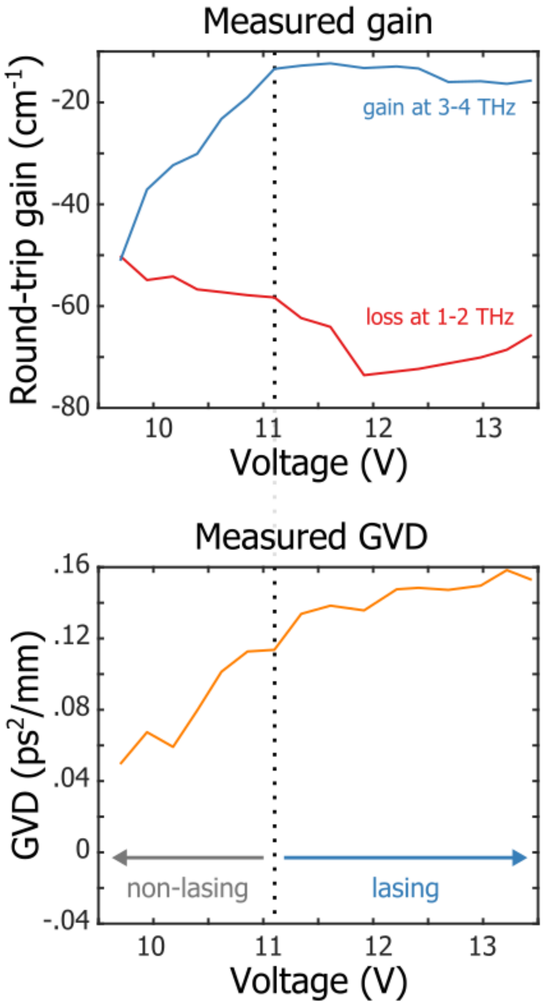
Burghoff et al., “Evaluating the coherence and time-domain profile of quantum cascade laser frequency combs,” *Opt. Express* (2015).

- Instantaneous intensity and frequency are **periodic**, but not a pulse
- Simultaneous AM and FM, not conventional mode-locking
    - QCL dynamics preclude mode-locking (see Khurgin et al, APL (2014))

Theory:

Tzenov, Jirauschek et al., Opt. Express (2016)
Tzenov, Jirauschek et al., IEEE THz (2017)
Henry, Khurgin et al., Opt. Eng. (2017)

Bias dependence of dispersion above threshold



Gain: clamps to constant value above threshold

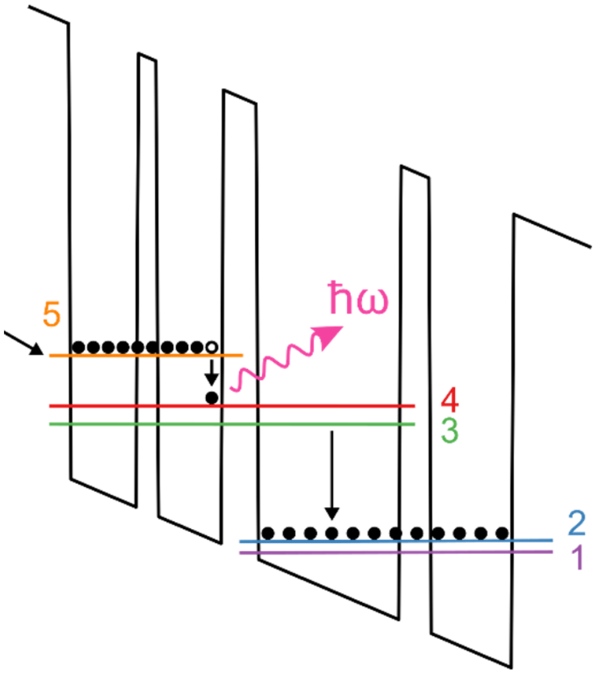
Dispersion: does *not* clamp above threshold in QCLs
Lineshape changes with bias, changing dispersion

Limits the dynamic range of comb formation

Burghoff et al., “Dispersion dynamics of quantum cascade lasers,” *Optica* (2016)

Intracavity detection

- QCLs can act as detectors as well as lasers
  - Current through QCL changes due to stimulated emission



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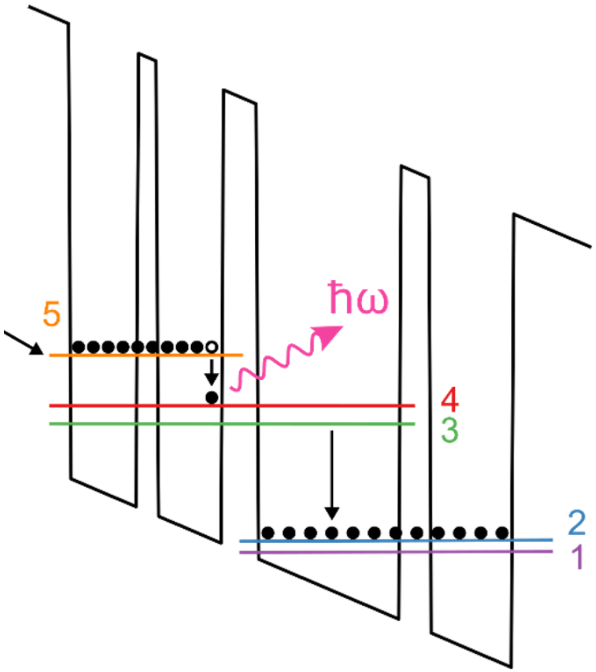
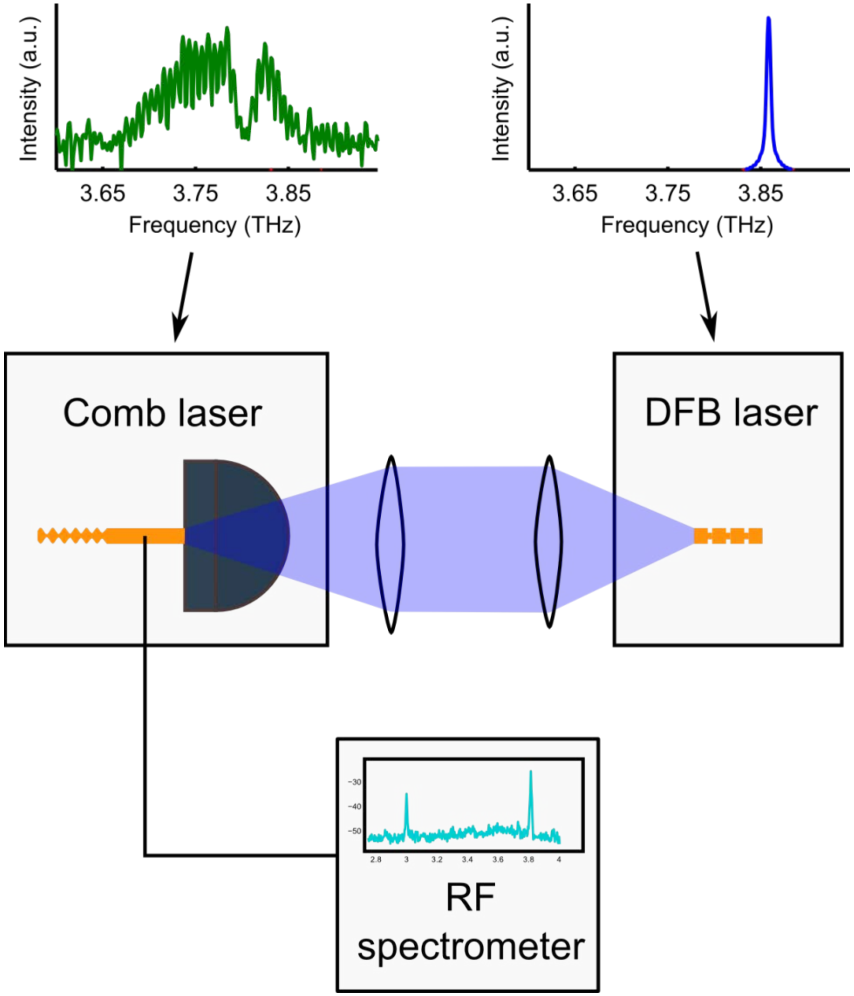
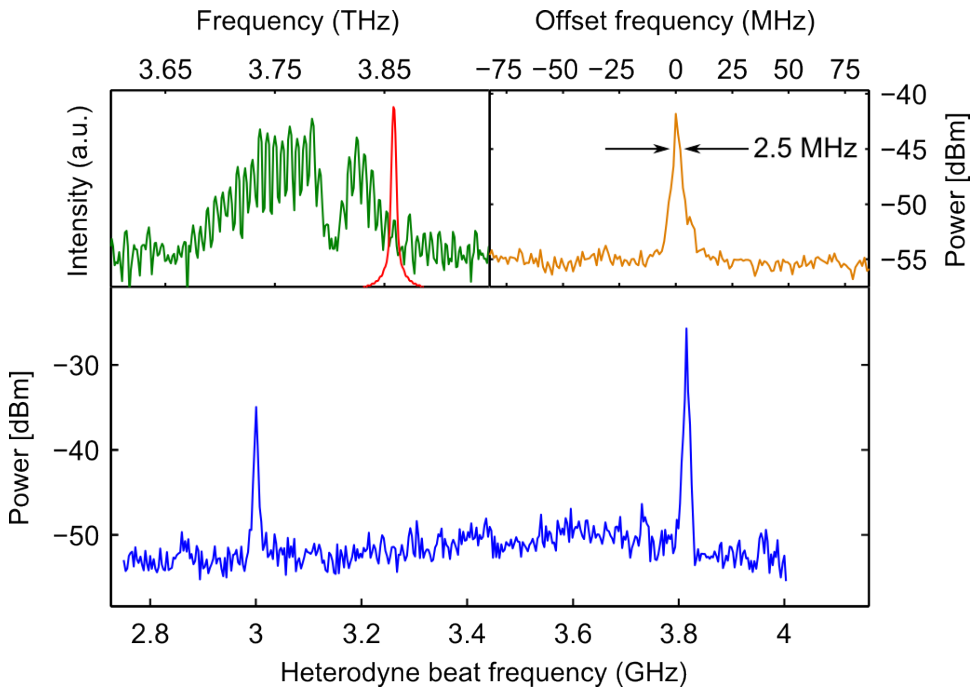


- Injecting another laser allows the difference frequency to be detected



Intracavity detection (2)

- Inject light from a narrowband (DFB) laser into comb cavity and measure intracavity beating between them
- Two beatnotes sum to the repetition rate, tune with DFB
- SNR limited by shot noise of QCL
  - NEP~nW/√Hz (similar to pyroelectric detection)
  - Equivalent joint power here ~10 uW



Burghoff et al., “Terahertz laser frequency combs,” *Nat. Photon.* (2014)

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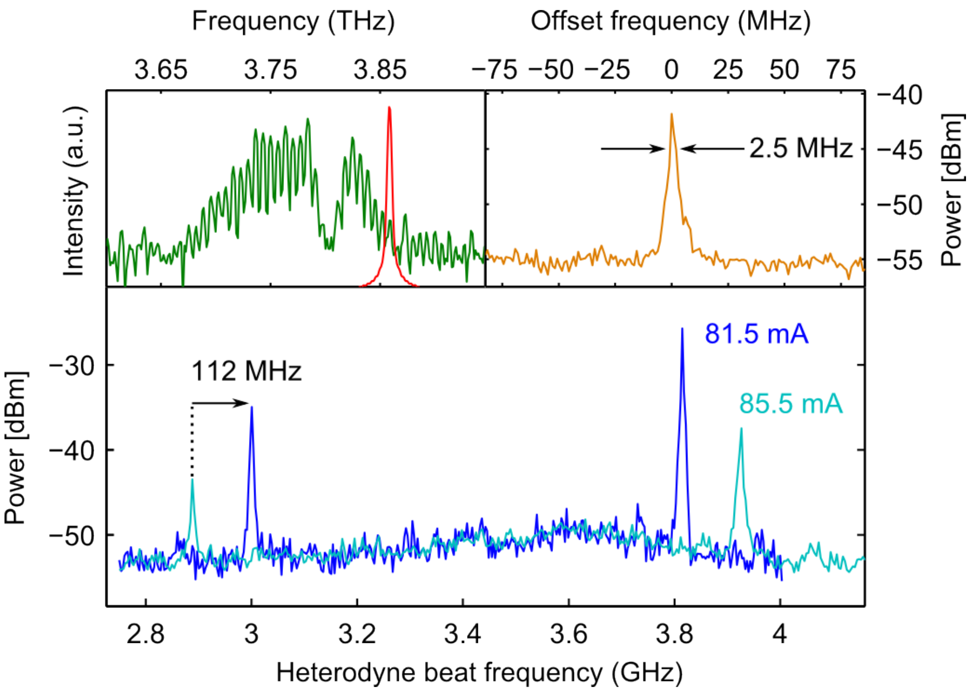


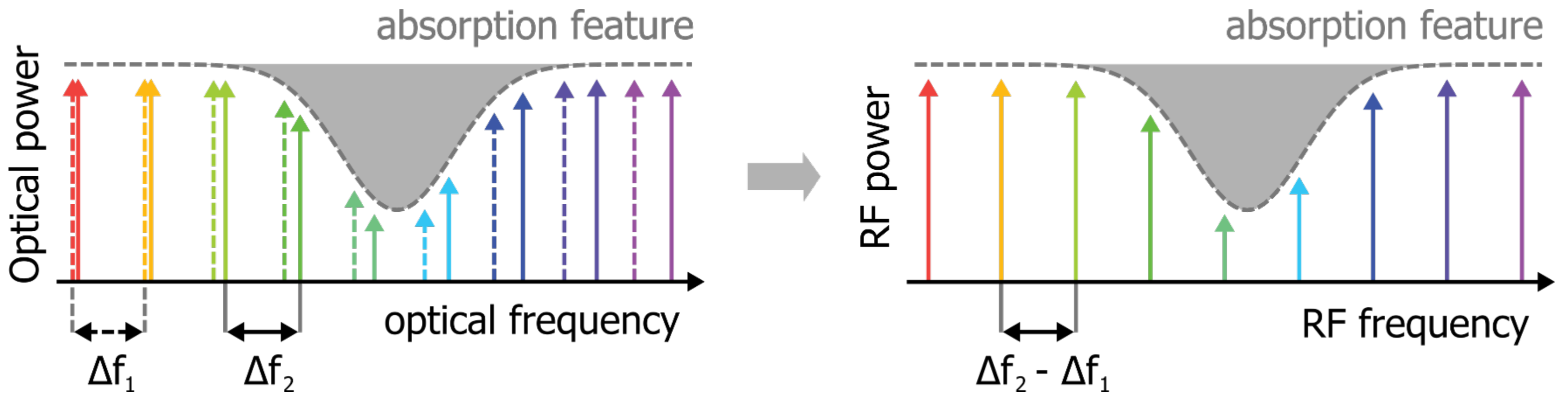
Burghoff et al., “Terahertz laser frequency combs,” *Nat. Photon.* (2014)

Part III

Multiheterodyne spectroscopy based on terahertz QCLs

Dual comb spectroscopy reminder

- Two combs, with slightly different spacings, shined on a detector. **Electrical** spectrum encodes **optical** spectrum:



- Two optical combs beat together to produce a comb at radio frequencies (MHz)
  - Typically, Δf_i~10 GHz and Δf_1-Δf_2~10 MHz

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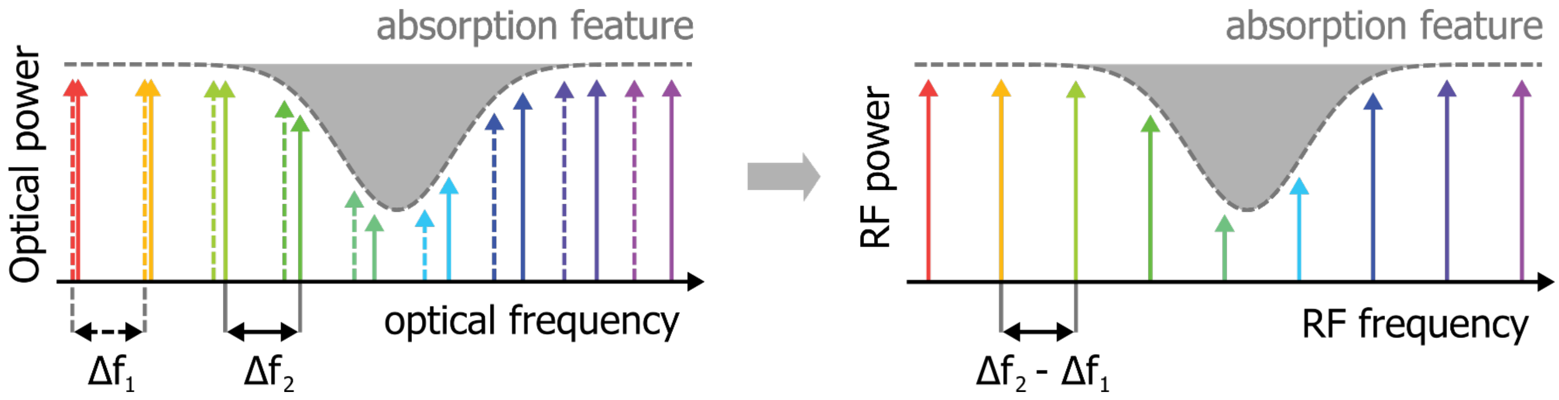

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  - van der Weide, Murakowski, Keilmann, *IEEE TMTT* (2000)

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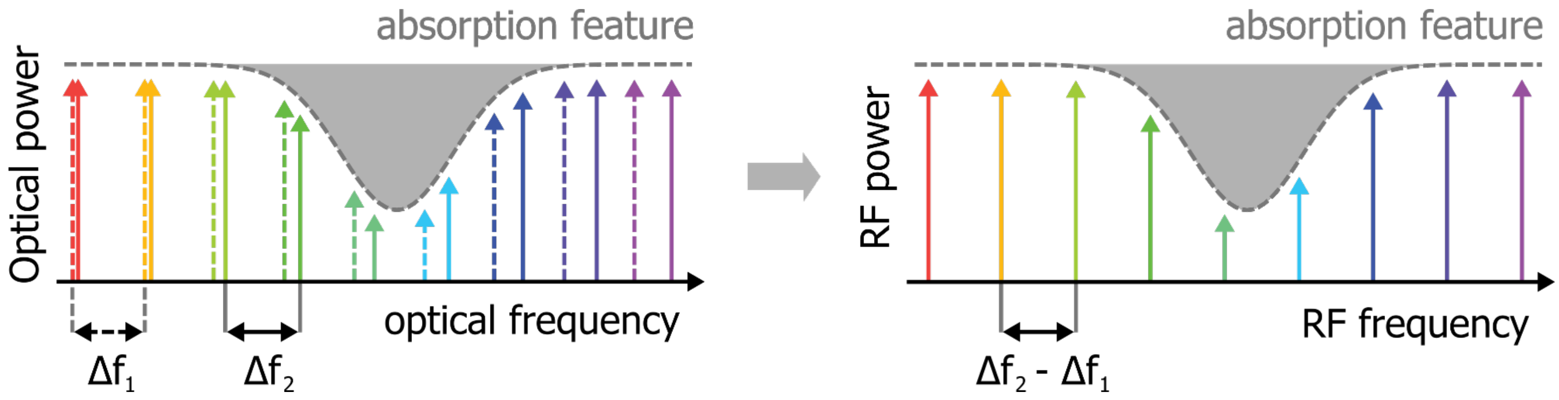

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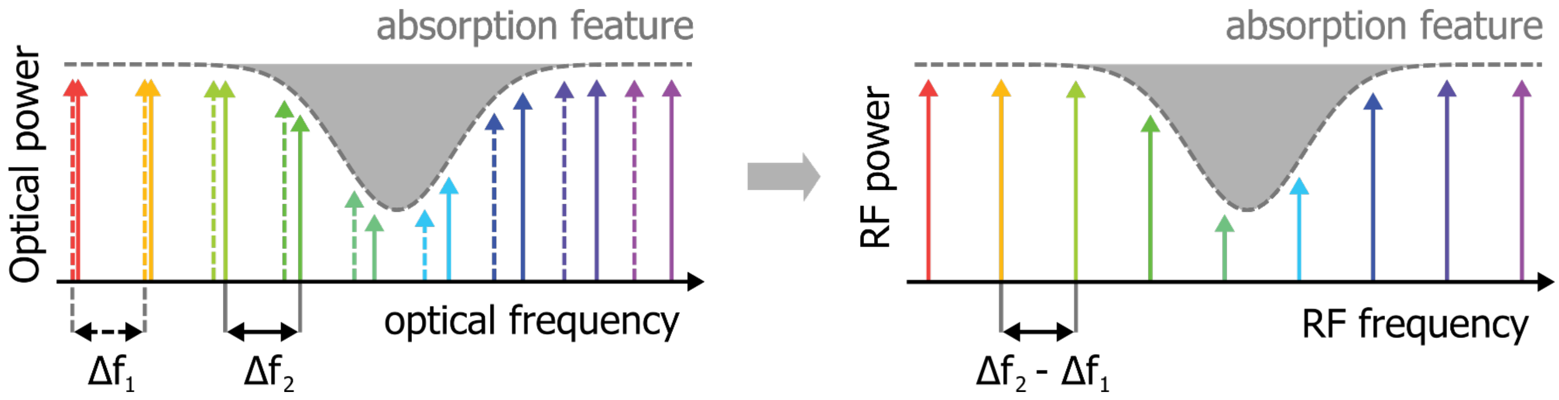

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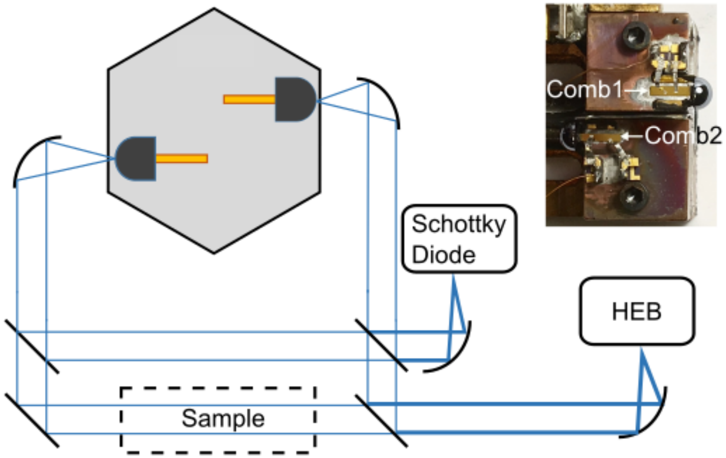
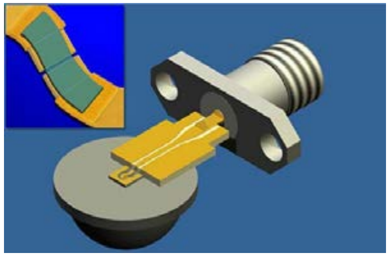
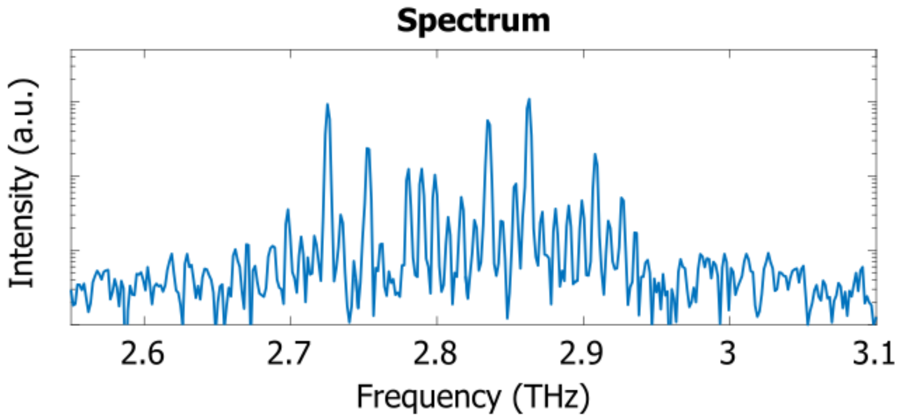
  - Review from NIST: Coddington et al., Optica (2016)
- Mid-IR QCLs:
  - Wang, Wysocki, et al., *APL* (2014)
  - Villares, Faist, et al., Nat. Comm. (2014)

THz QCL dual comb spectroscopy

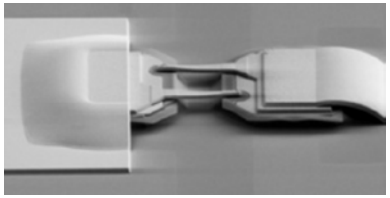


Detector 1: HEB (superconducting bolometer)



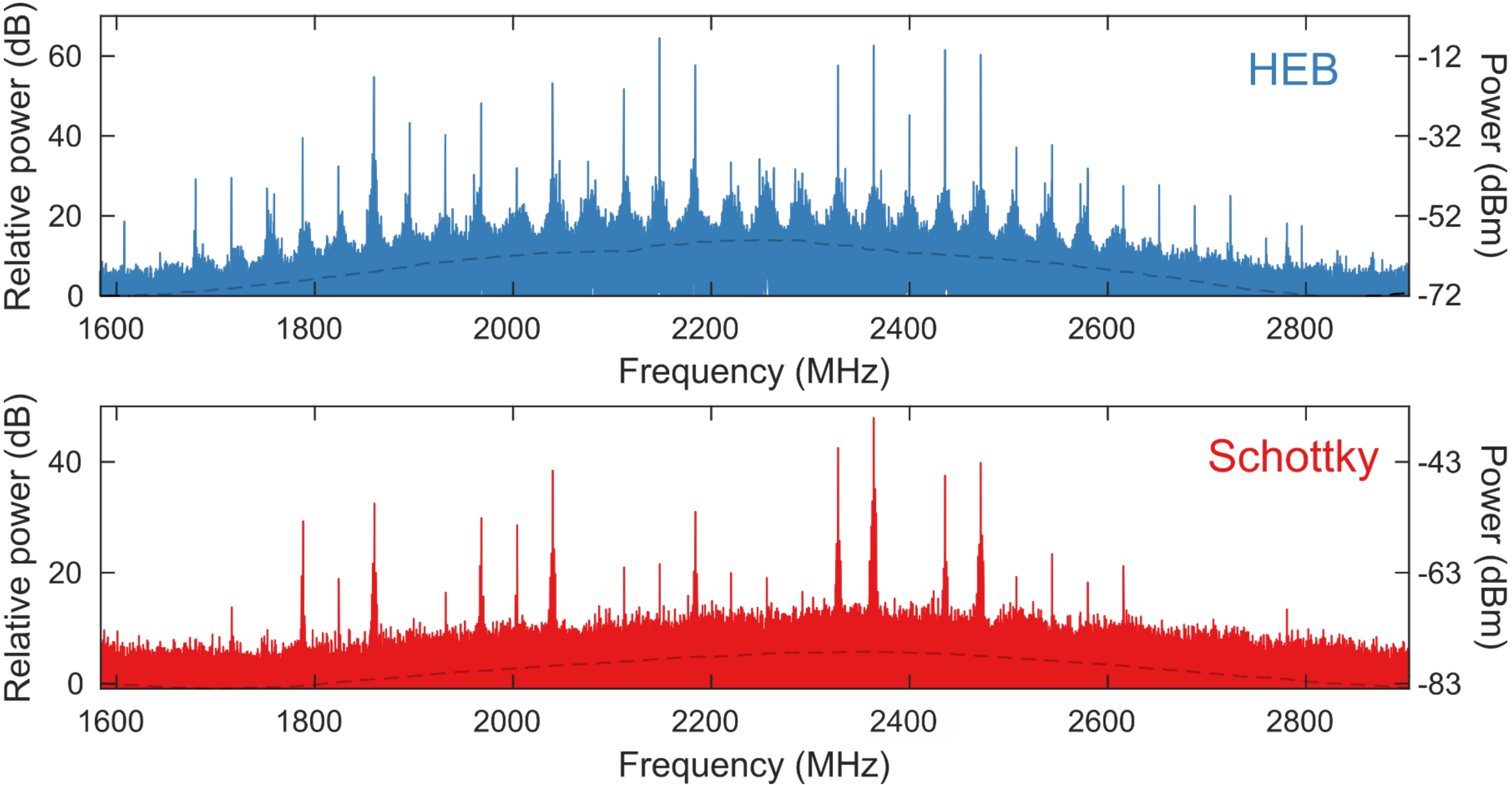


Detector 2: Schottky mixer (room-temperature)



Yang, Burghoff et al., “Terahertz multiheterodyne spectroscopy using laser frequency combs,” *Optica* (2016)

Example dual comb signals, 100 μs integration

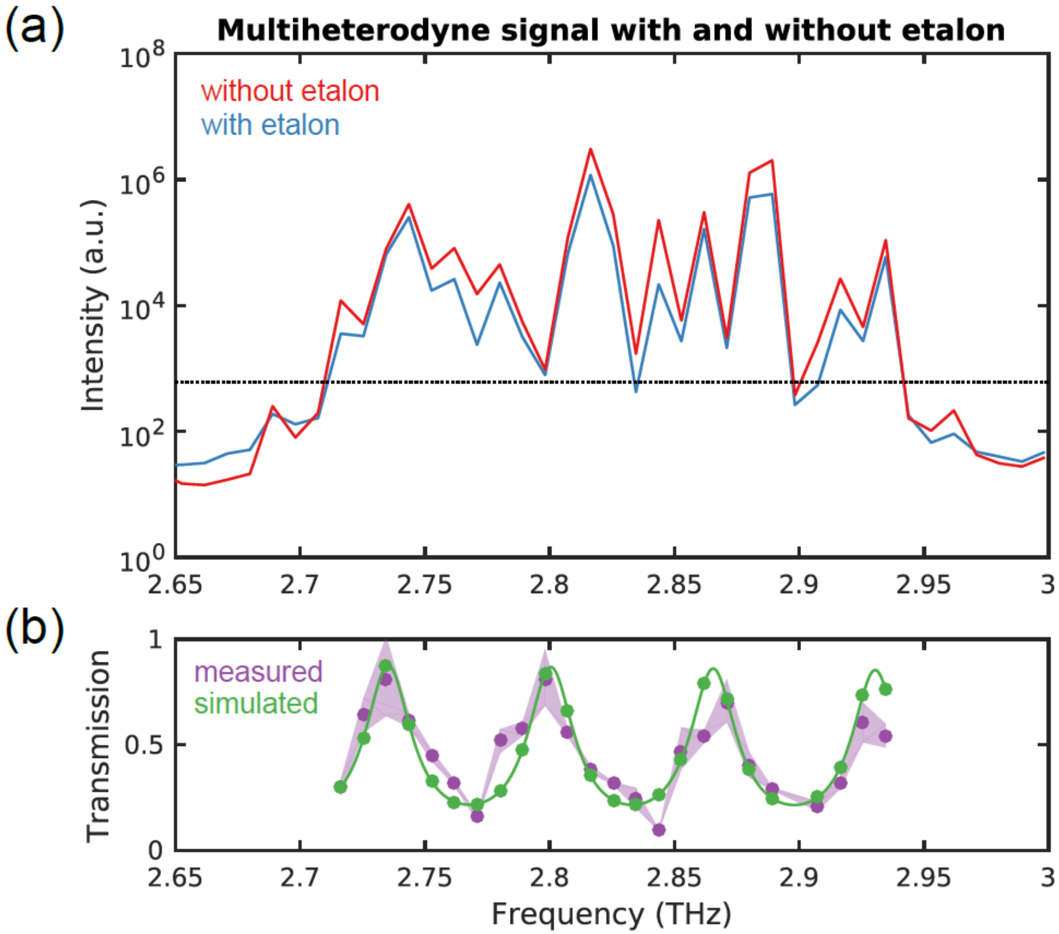


Yang, Burghoff et al., *Optica* (2016)

- HEB: Hot electron bolometer from J.R. Gao
  - Zhang, Gao, et al, “Quantum noise in a terahertz hot electron bolometer mixer,” APL (2010)
- Schottky mixer: Virginia Diodes WR-0.34HM

Broadband etalon sample

- Etalon transmission measurement
  - 625 um thick undoped GaAs etalon



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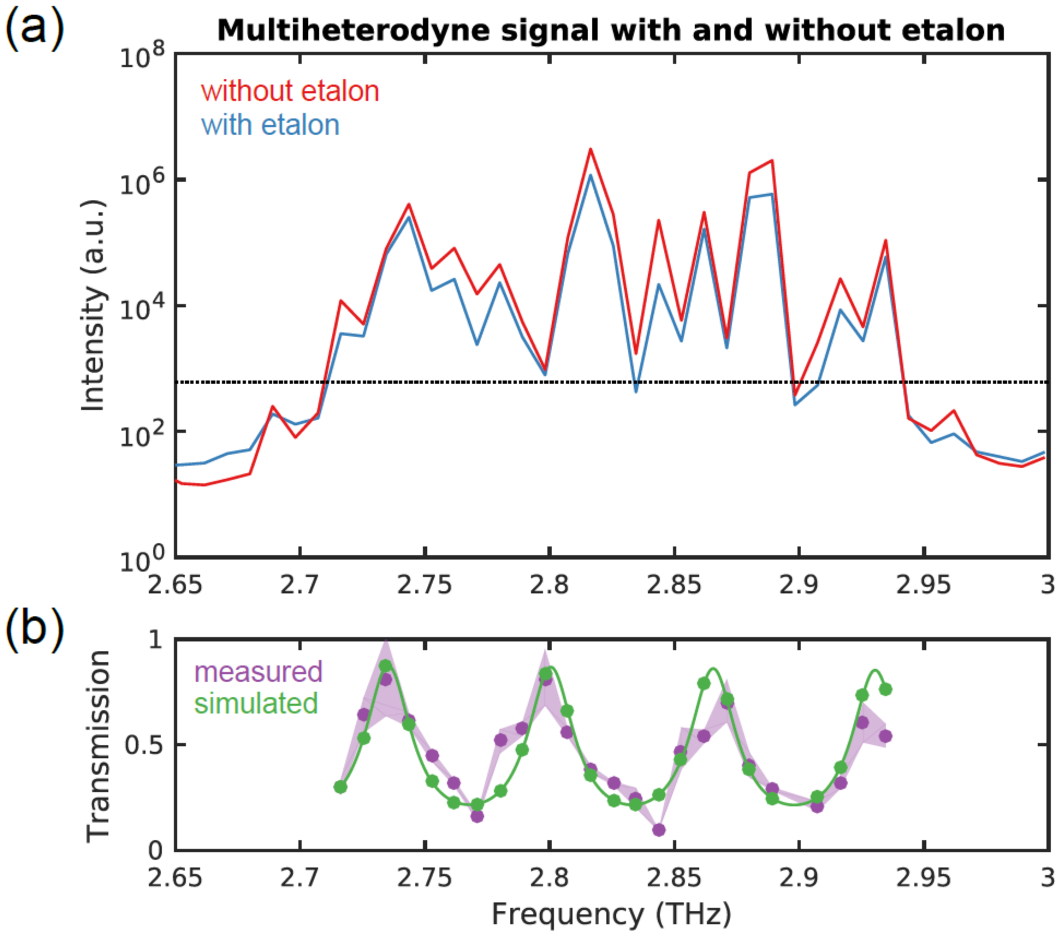


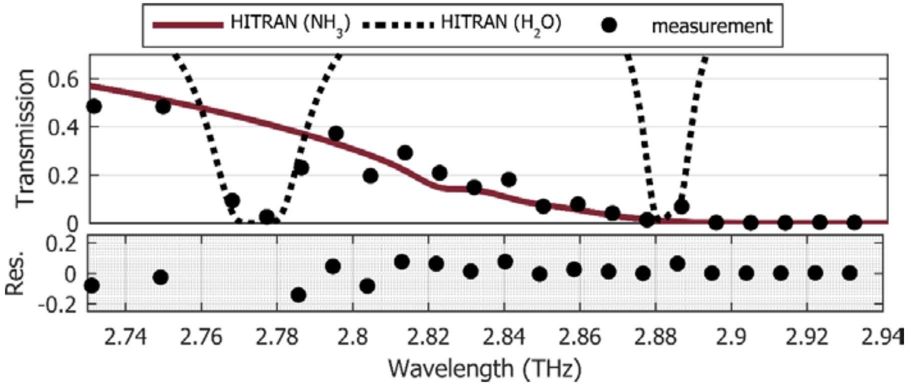
Current limitations:
non-uniformity of comb
+ detector nonlinearity

Yang, Burghoff et al., *Optica* (2016)

Gas spectroscopy

- Collaboration with Gerard Wysocki's group at Princeton (Jonas Westberg and Lukasz Sterczewski)

- Ammonia
- 460 Torr
- 500 µs acquisition time
- 220 GHz of coverage at 2.84 THz
- Water absorption highlighted



Sterczewski, Wysocki *et al.,* "Terahertz multiheterodyne spectroscopy with quantum cascade lasers – a feasibility study," in *International Conference on Infrared, Millimeter and Terahertz Waves* (2017).

Westberg, Wysocki, et al., "Broadband mid-infrared and THz chemical detection with quantum cascade laser multi-heterodyne spectrometers" in Proc. SPIE 10210, Next-Generation Spectroscopic Technologies (2017).

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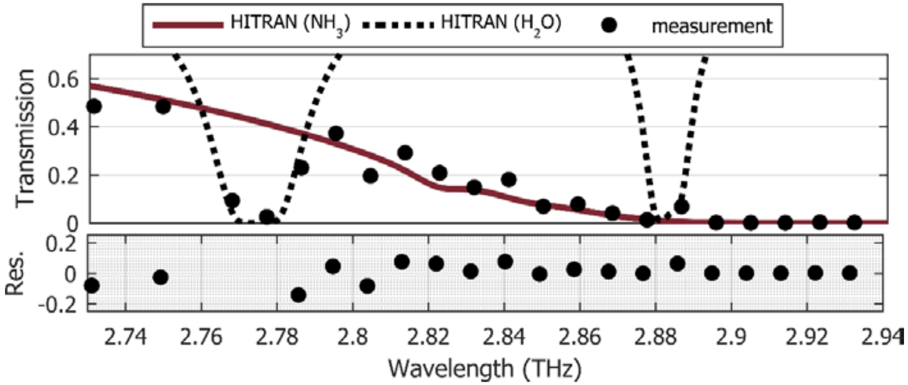

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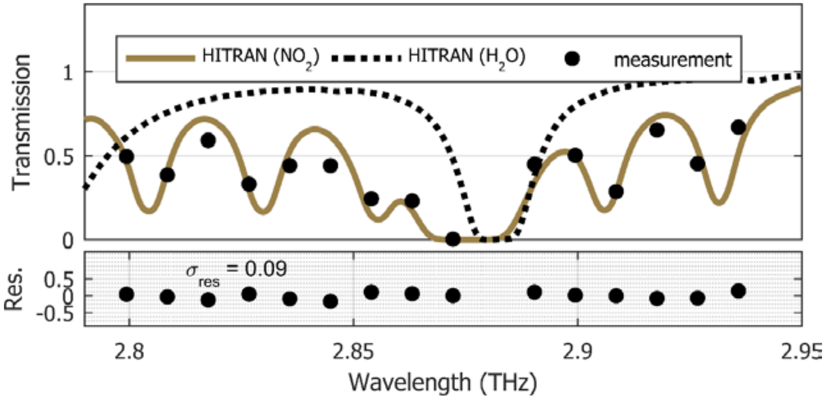

- Nitrogen dioxide (NO_2)
- 840 Torr
- 500 µs acquisition time

Sterczewski, Wysocki *et al.*, "Terahertz multiheterodyne spectroscopy with quantum cascade lasers – a feasibility study," in *International Conference on Infrared, Millimeter and Terahertz Waves* (2017).

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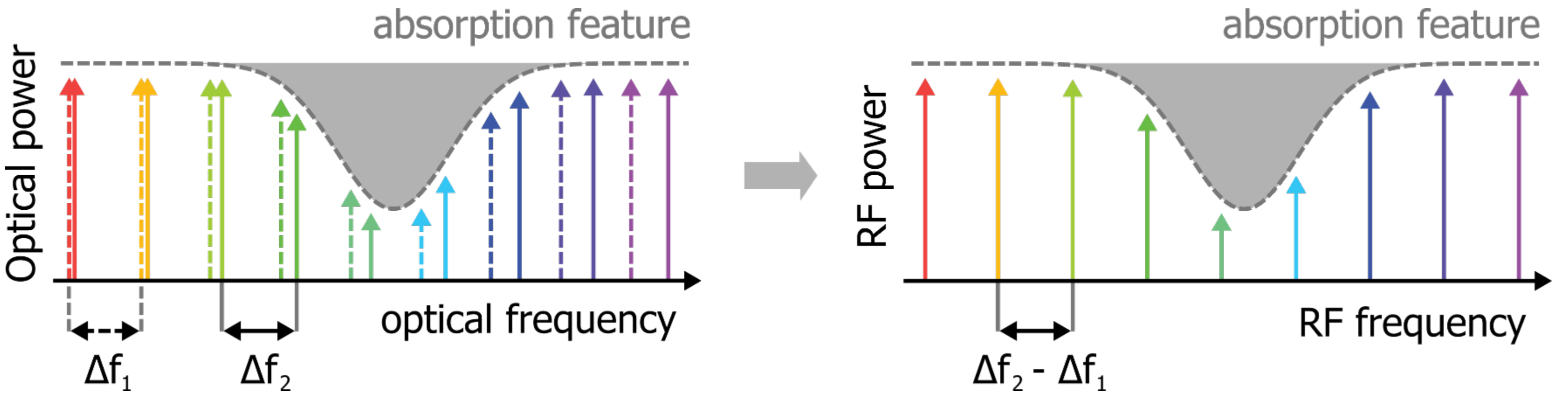
Influence of phase noise on dual comb measurement

- Cartoon picture of dual comb spectroscopy:



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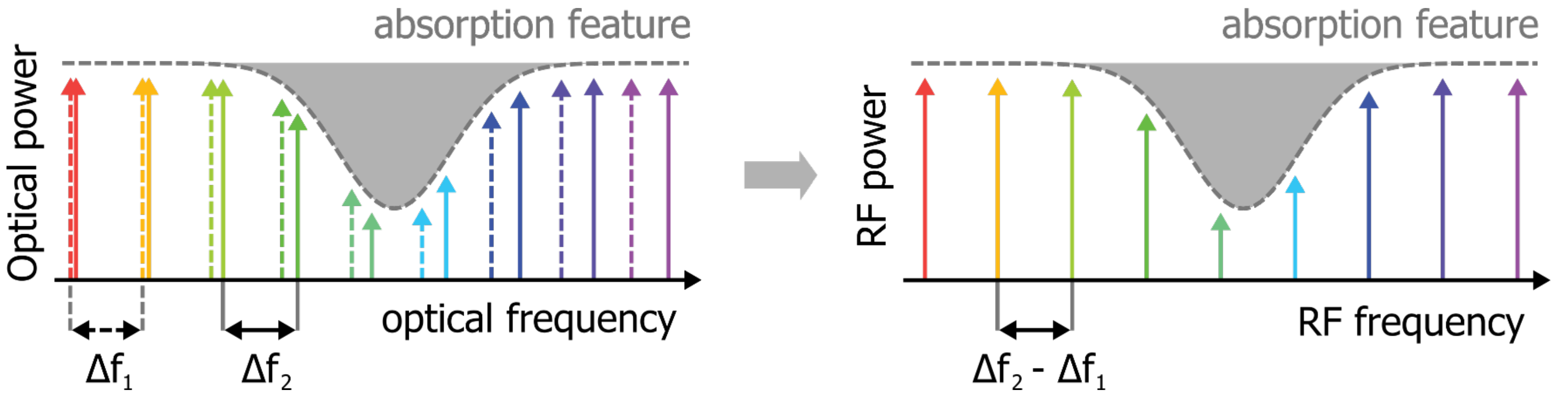
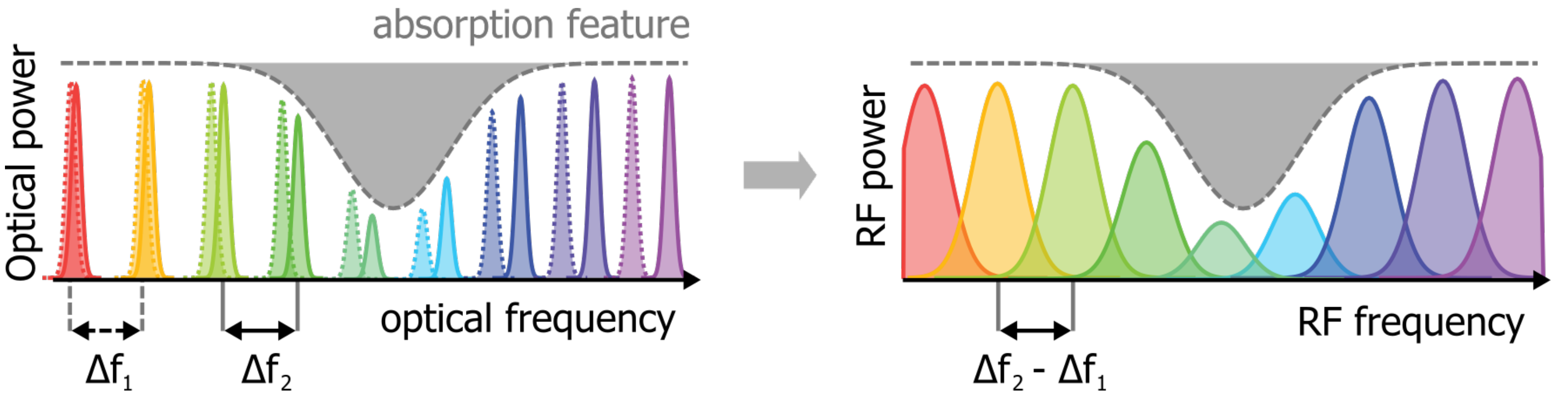


- Real life: some phase noise (feedback, environmental fluctuations, etc.)



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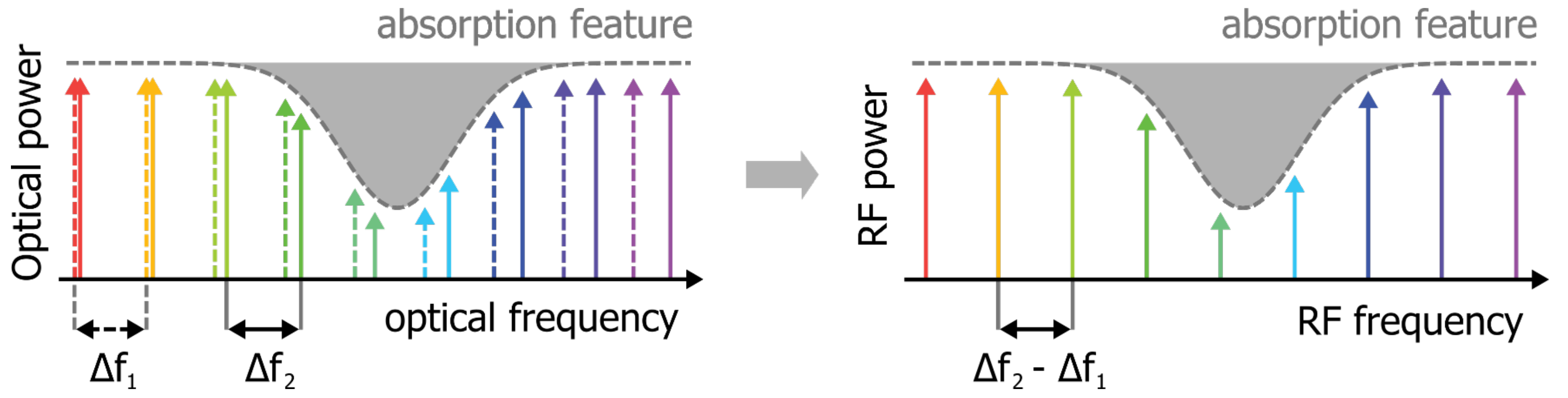

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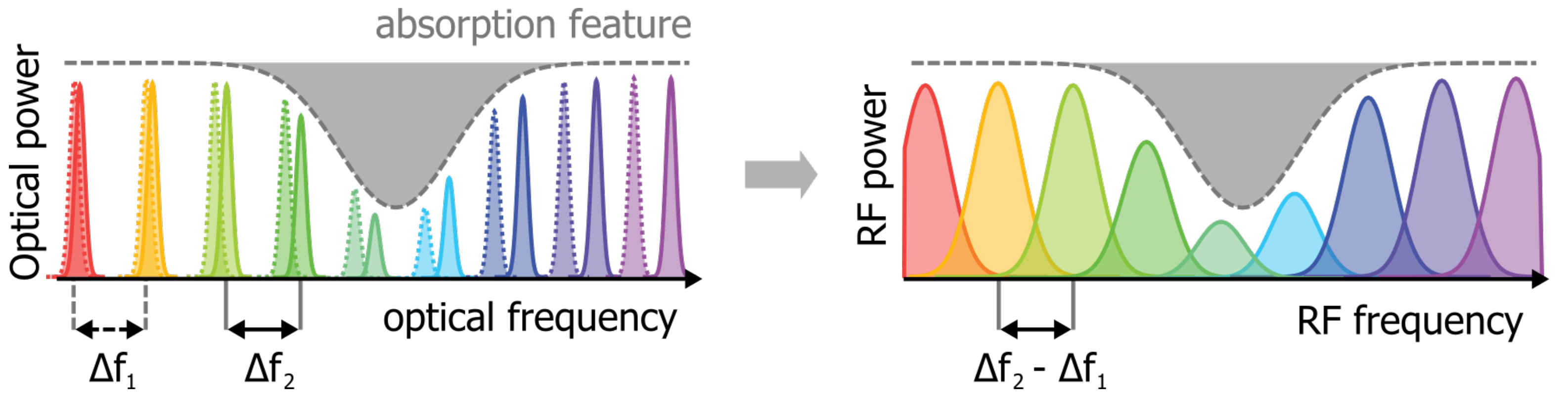
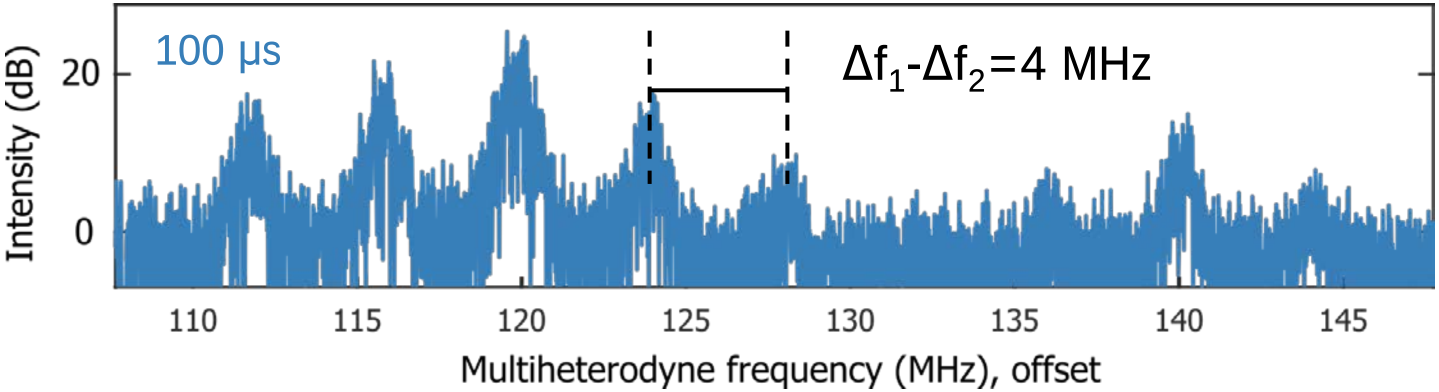


What happens when there is too much noise?

Effect of phase noise

- Low noise state



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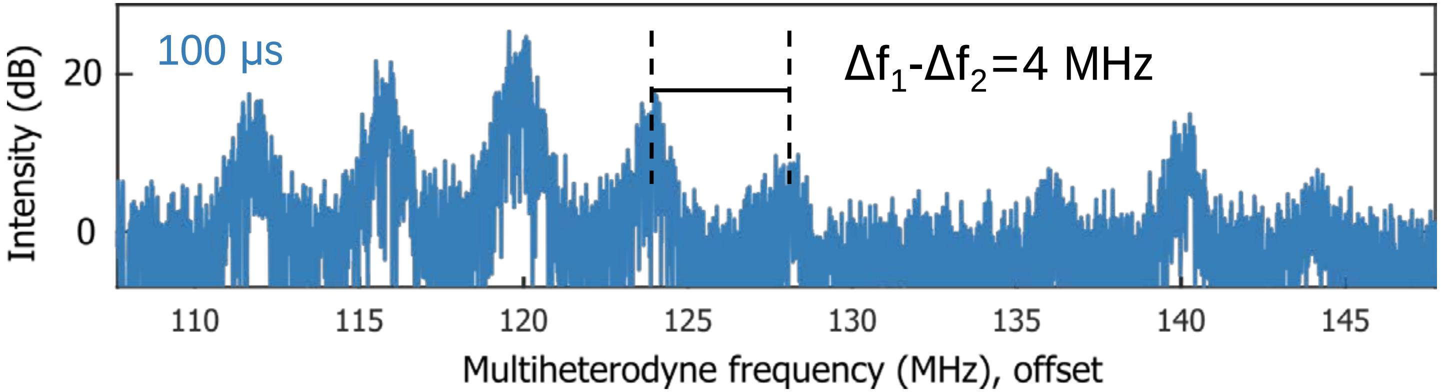
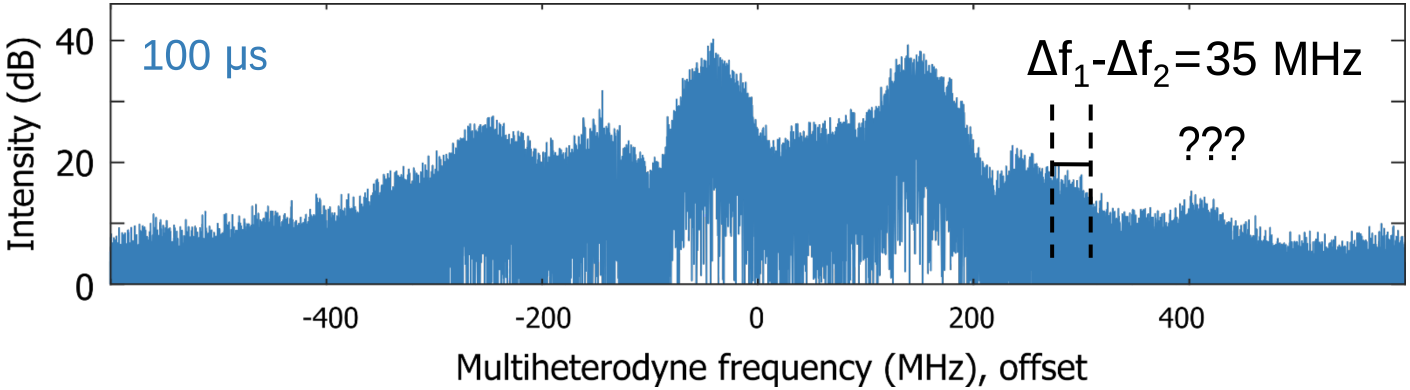
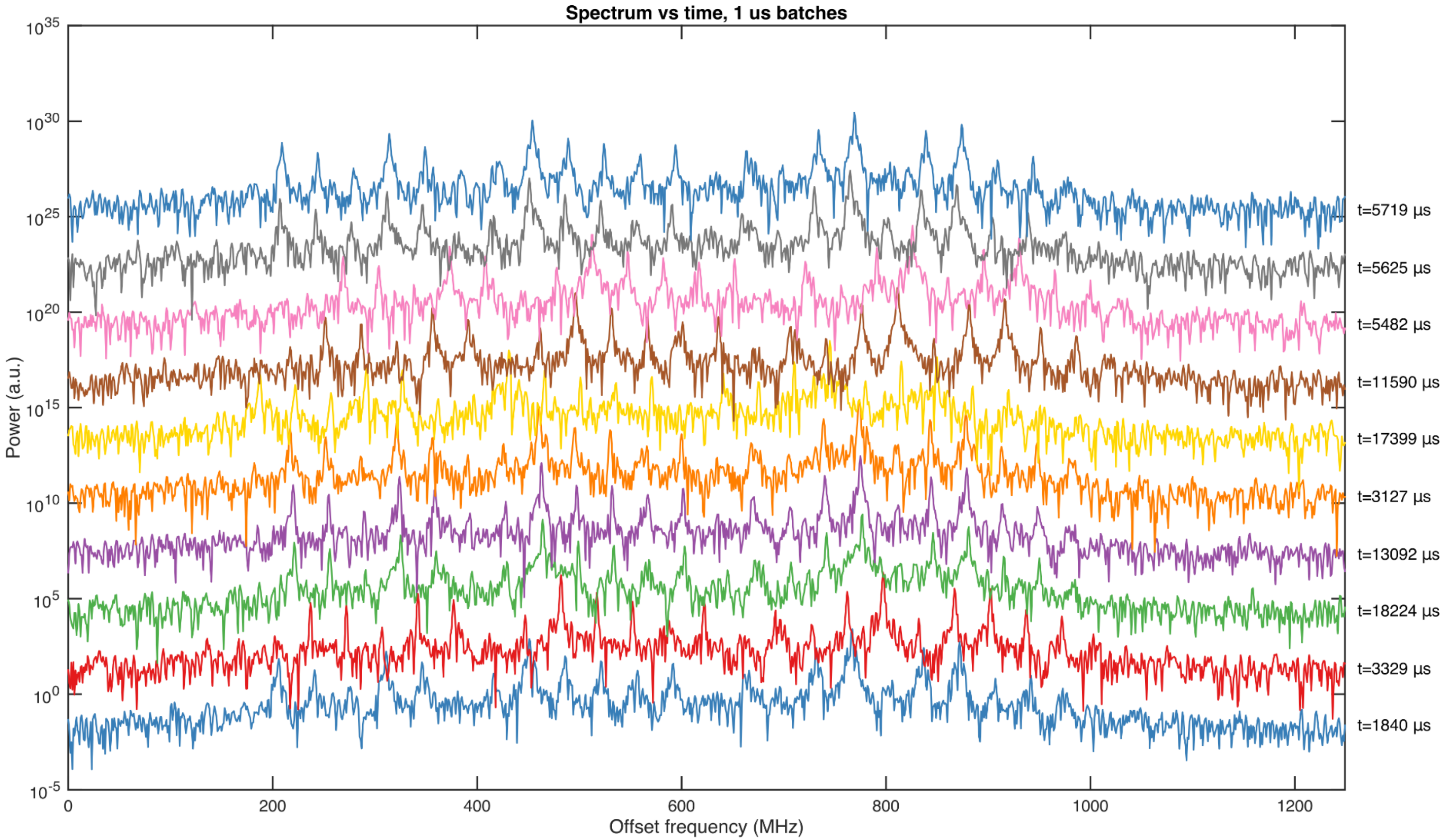


- High noise state (but still comb!)



High noise short timescale spectra



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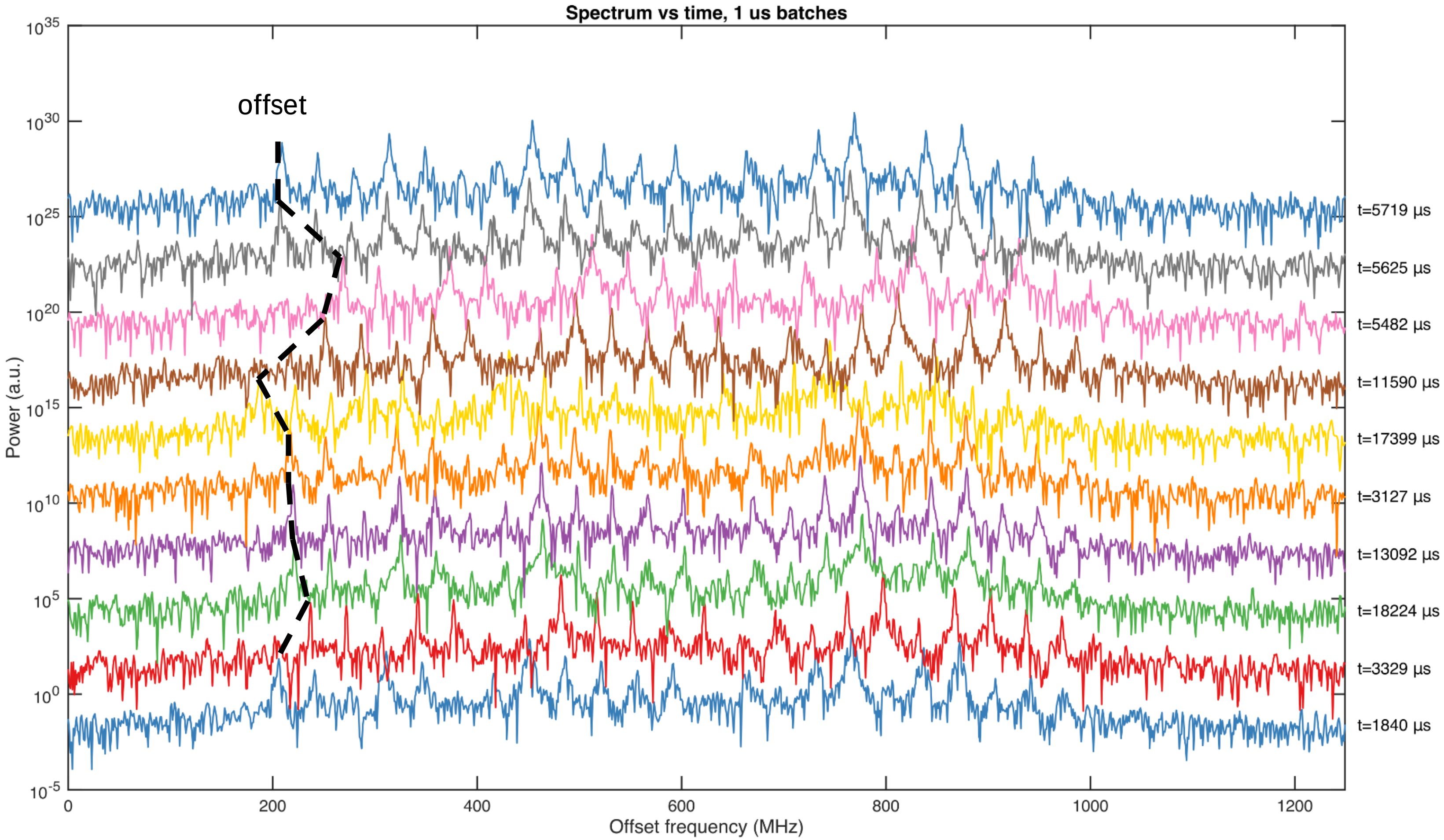

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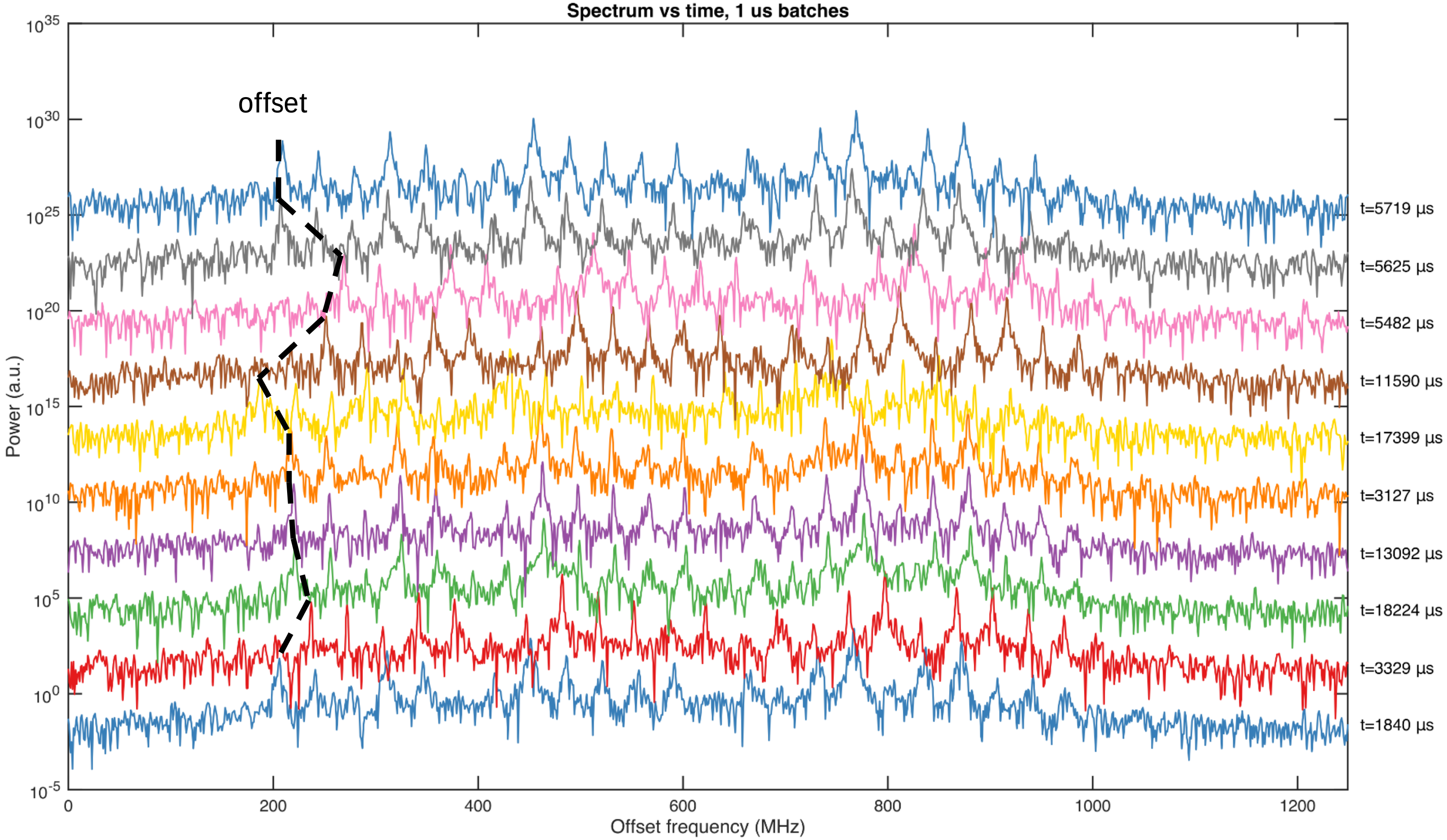
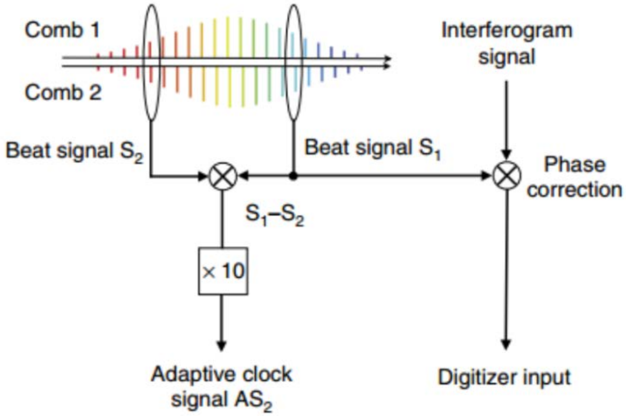
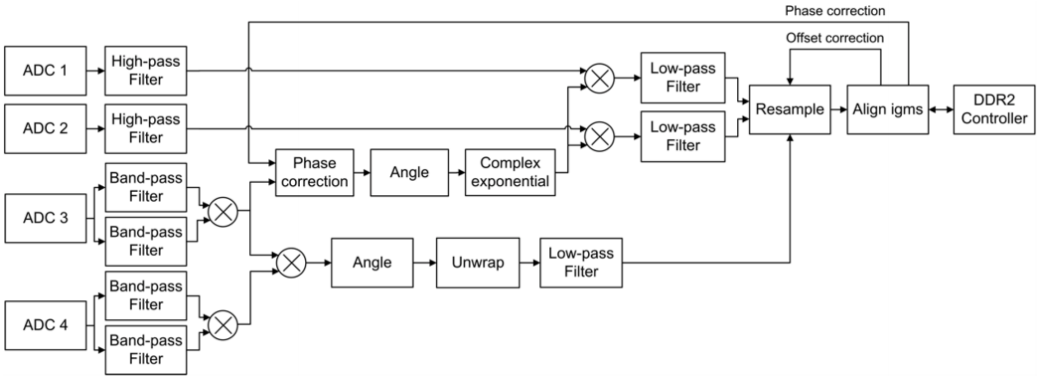


Offset fluctuations ~ 3*rep rate spacing

Adaptive sampling approaches



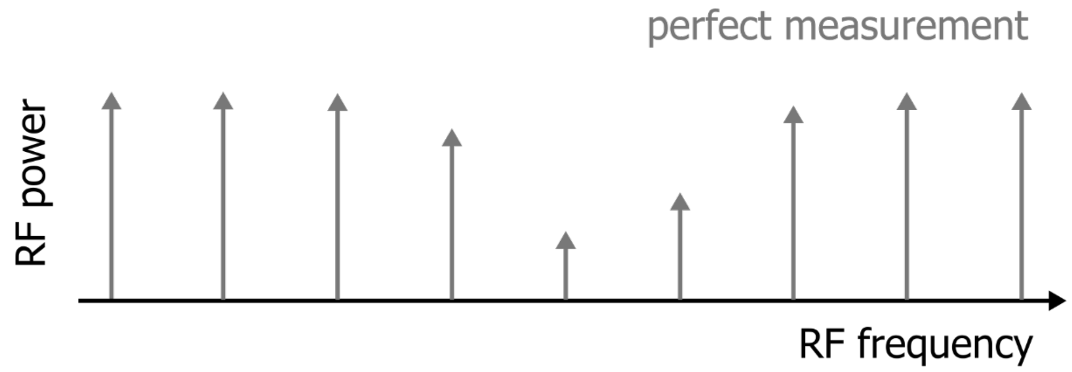
Ideguchi et al., *Nat. Comm.*, (2014)



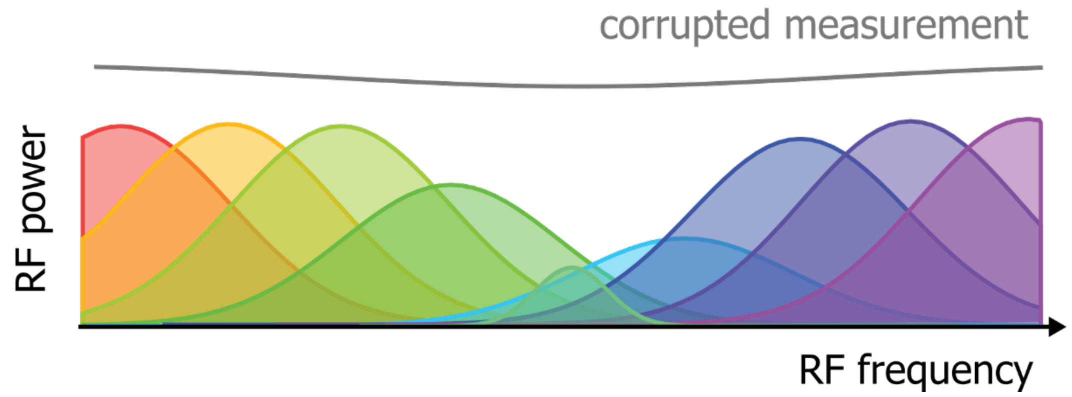
Roy et al., *Opt. Express*, (2012)

- Mode-locked laser community has done a lot of work on adaptive sampling techniques that correct these effects, but they’re challenging at long-wavelengths:
    - Usually detector-noise limited, so no optical power to spare
    - Would like to avoid extra lasers, other optical components

Can corruption of signal be inverted digitally?

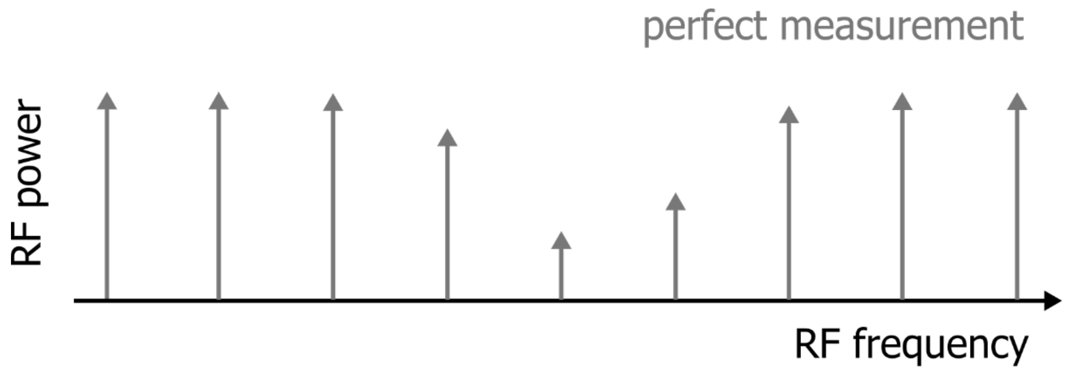


No or low
phase noise

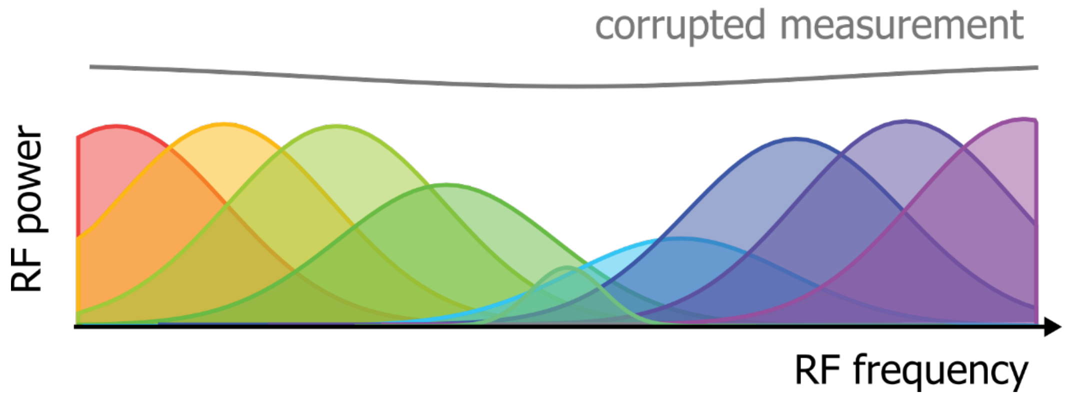


Phase noise large,
but still correlated

Can corruption of signal be inverted digitally?



No or low phase noise



Phase noise large, but still correlated

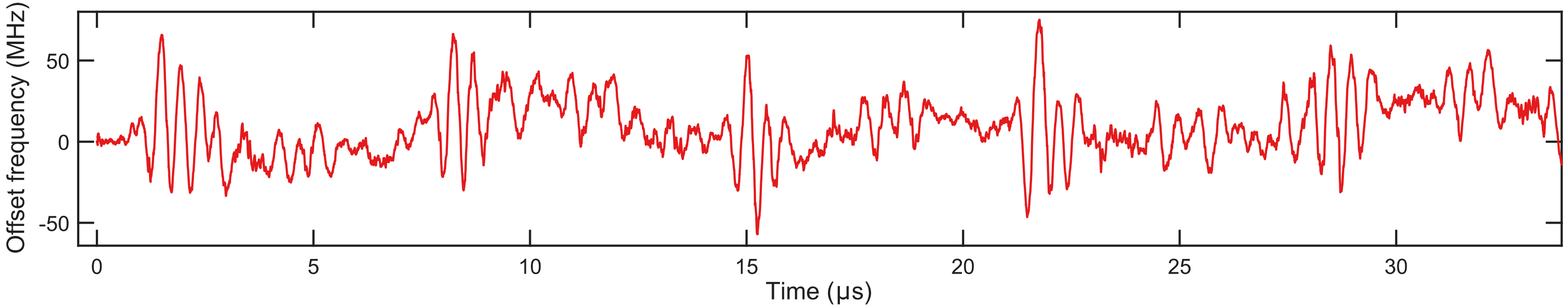
Can this corruption be extracted digitally, without any optical components?

Extended Kalman filter frequency tracking

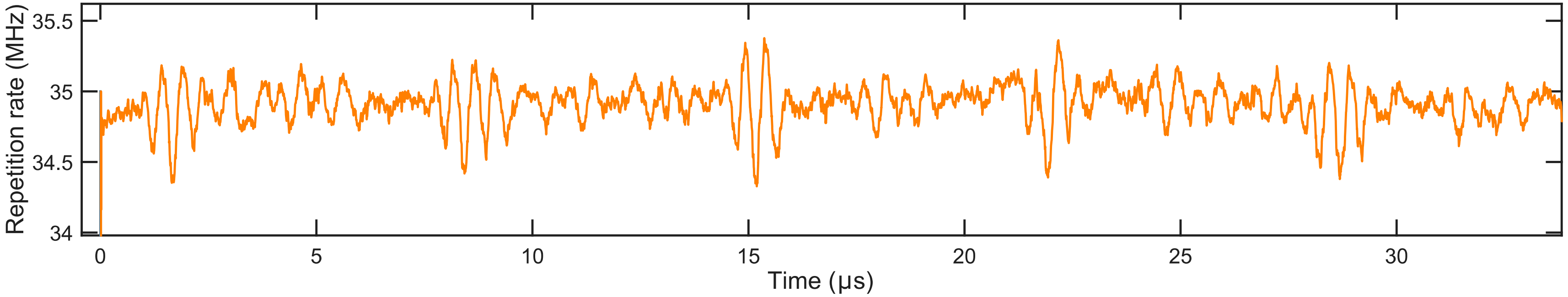
Use Kalman filters to track frequencies (solve a regularized fitting problem):

$$J(\phi_0, \Delta\phi) = \int dt \, \frac{1}{\sigma^2} \left| y(t) - \sum_n A_n e^{i(\phi_0(t)+n\Delta\phi(t))} \right|^2 + \frac{1}{Q_0}\left(\frac{df_0}{dt}\right)^2 + \frac{1}{Q_\Delta}\left(\frac{d\Delta f}{dt}\right)^2$$

Offset frequency



Repetition rate



Burghoff, Yang, et al., "Computational multiheterodyne spectroscopy," *Science Advances* (2016)

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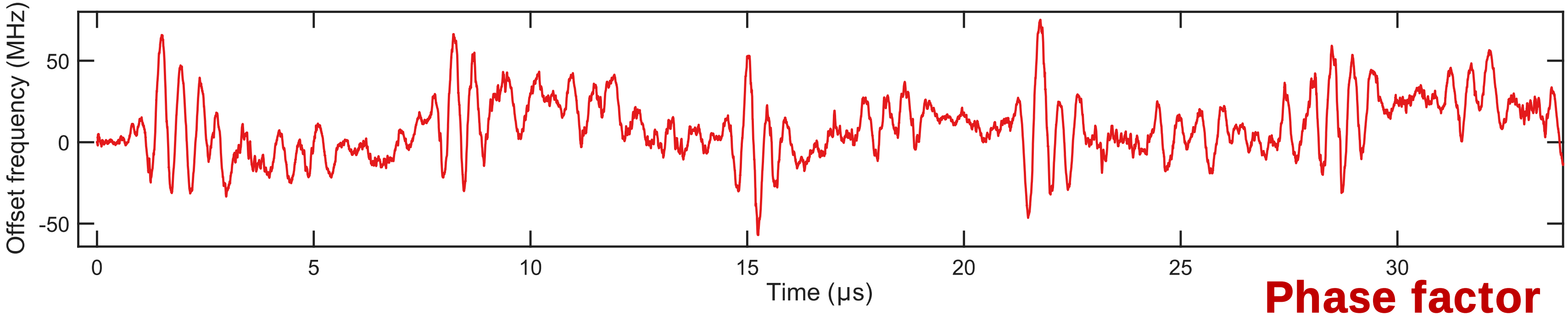
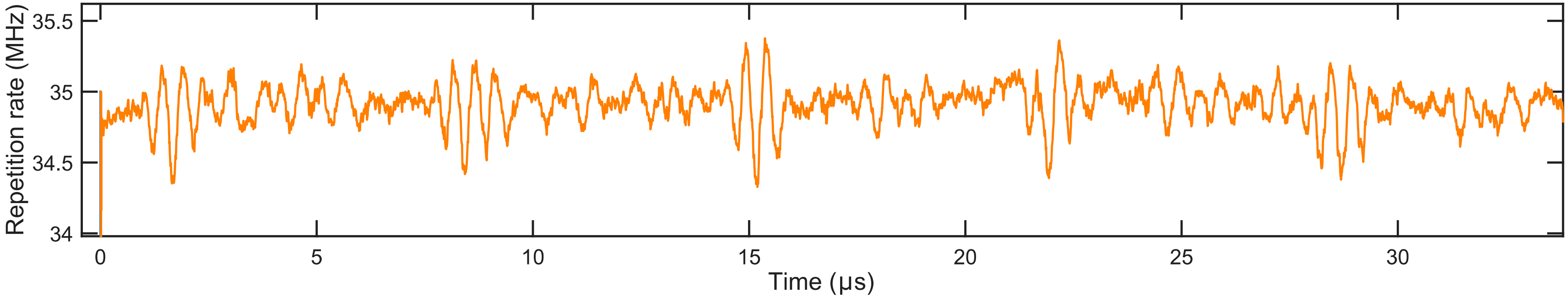


Phase factor

Repetition rate



Burghoff, Yang, et al., “Computational multiheterodyne spectroscopy,” *Science Advances* (2016)

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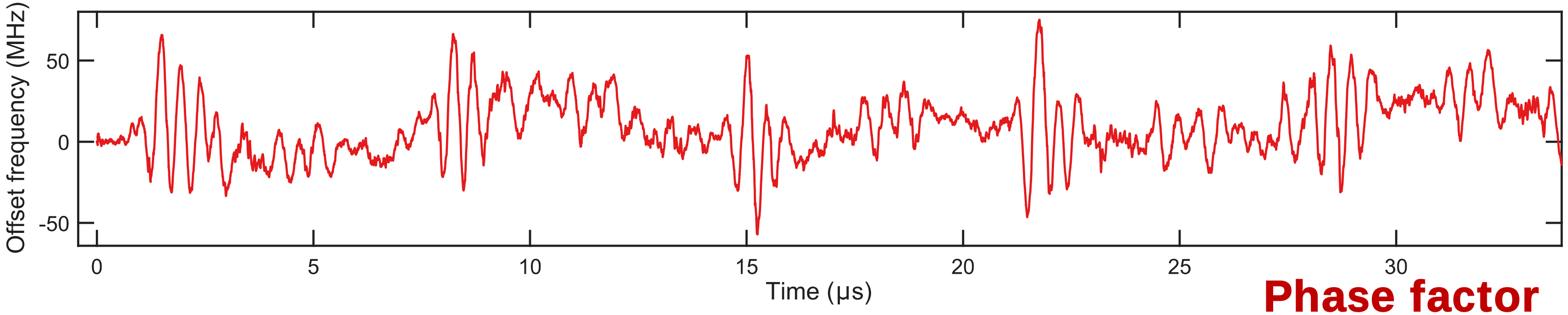
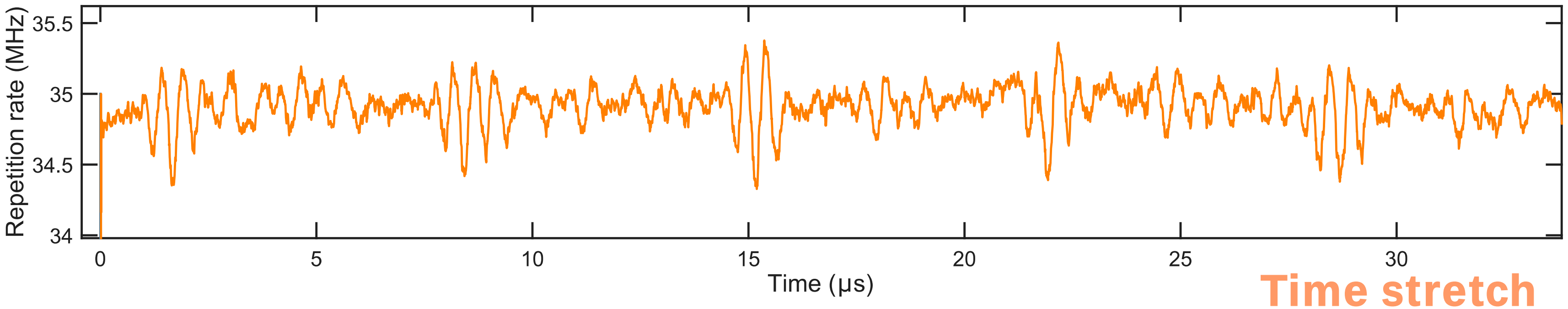
Offset frequency



Phase factor

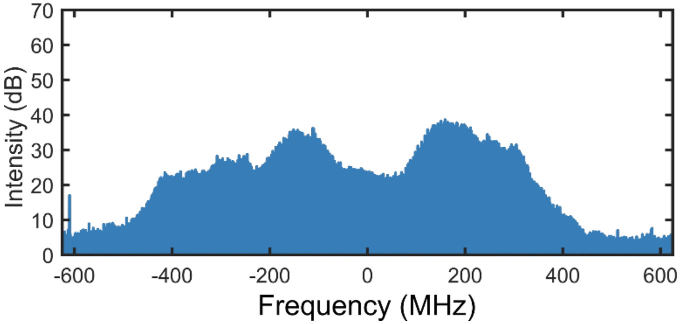
Repetition rate



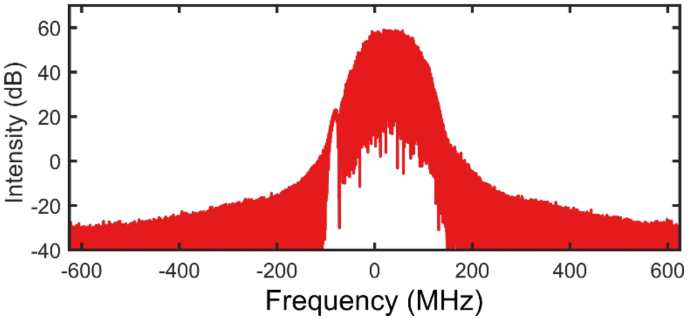
Time stretch

Burghoff, Yang, et al., “Computational multiheterodyne spectroscopy,” *Science Advances* (2016)

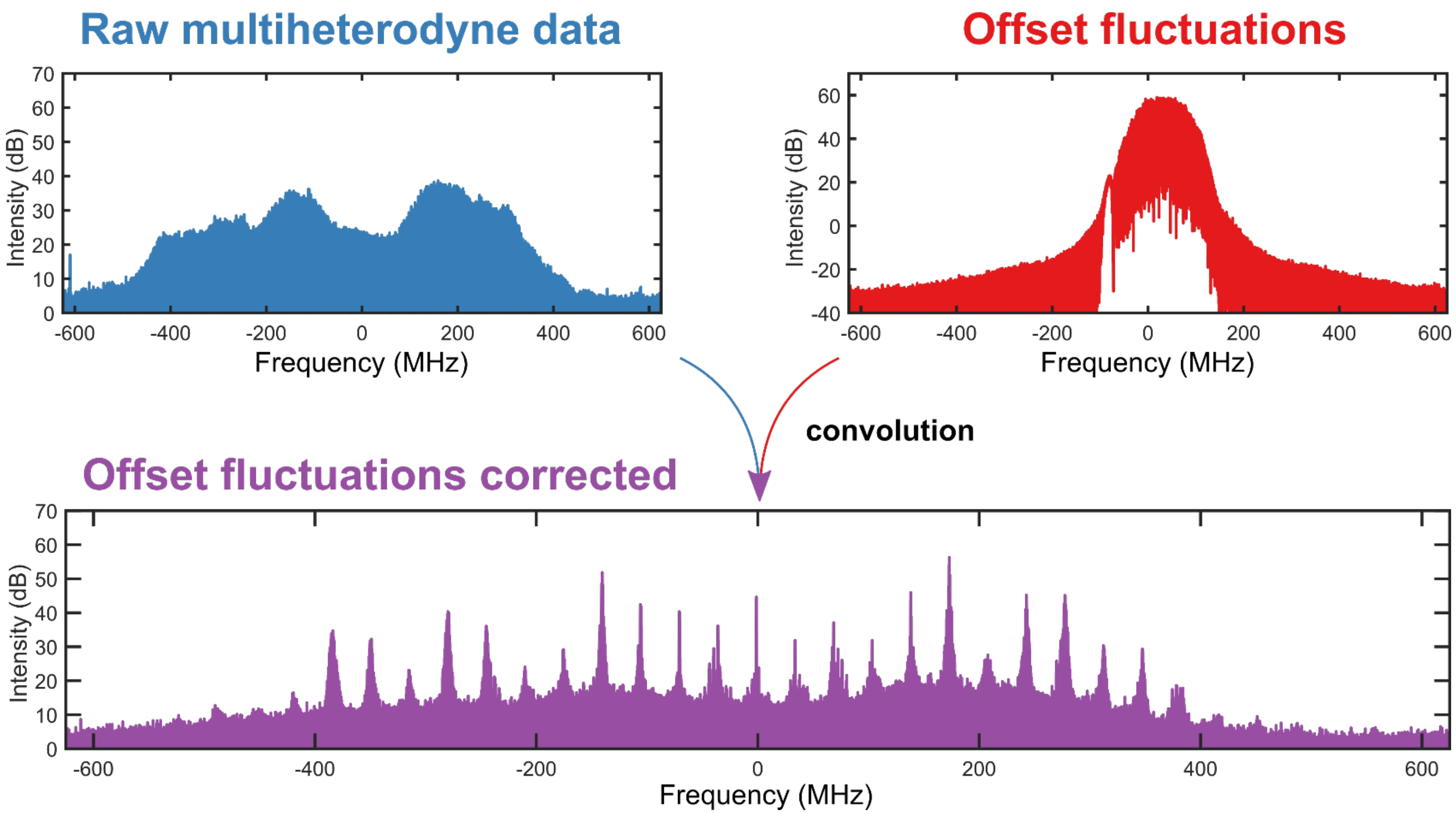
Raw multiheterodyne data



Offset fluctuations



Burghoff, Yang, et al., *Science Advances* (2016)



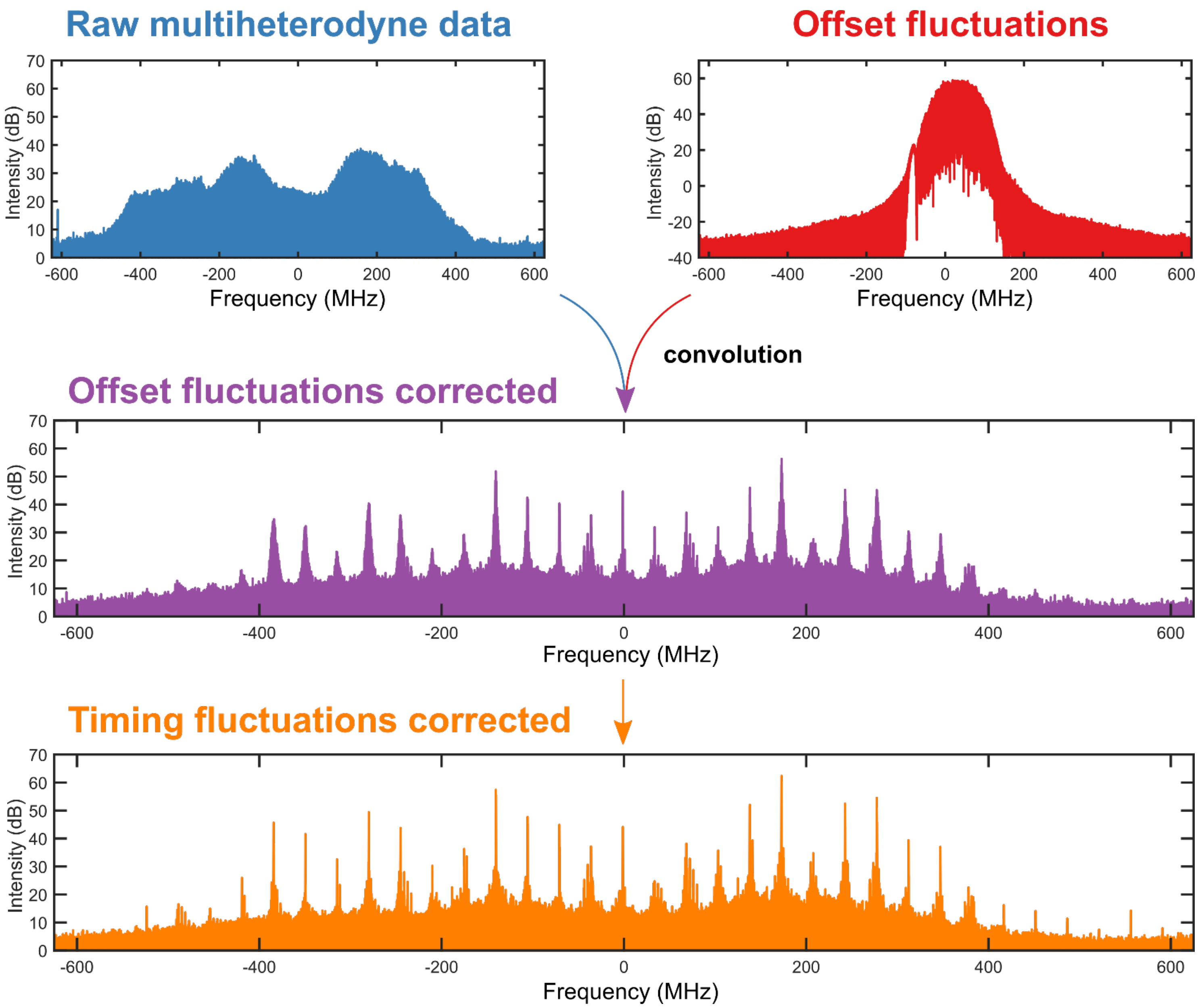
Raw multiheterodyne data

Offset fluctuations

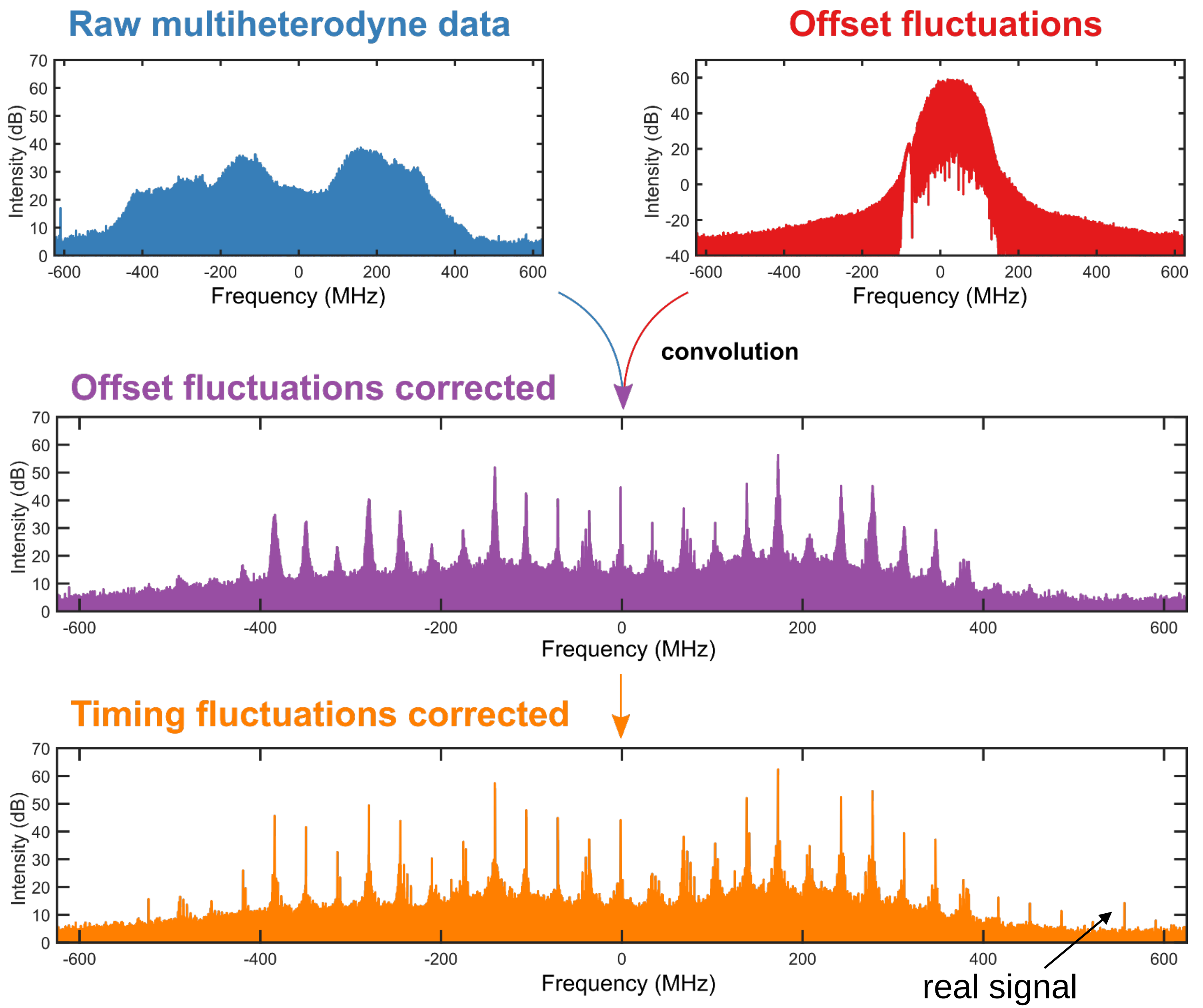
convolution

Offset fluctuations corrected

Burghoff, Yang, et al., *Science Advances* (2016)



Burghoff, Yang, et al., S*cience Advances* (2016)



Raw multiheterodyne data

Offset fluctuations

convolution

Offset fluctuations corrected

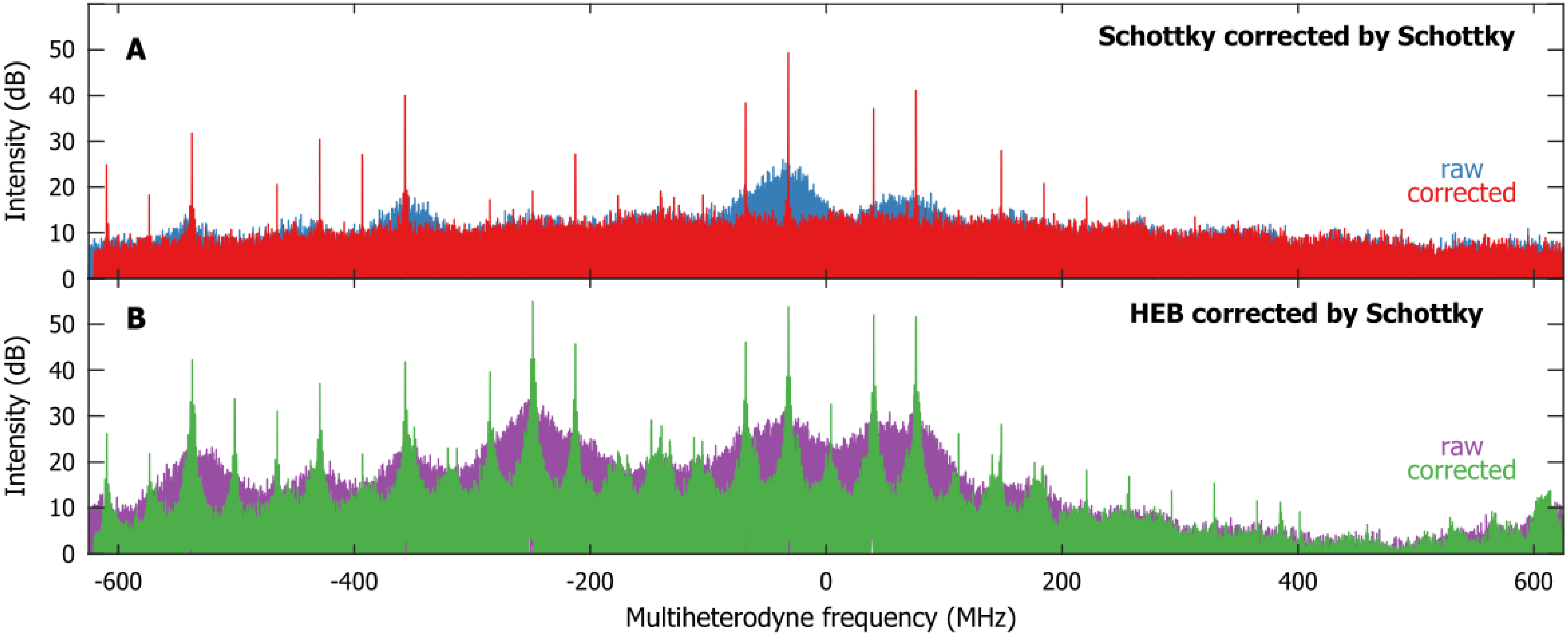
Timing fluctuations corrected

real signal recovered from the noise floor

Burghoff, Yang, et al., *Science Advances* (2016)

Cross-correcting different spectroscopic channels

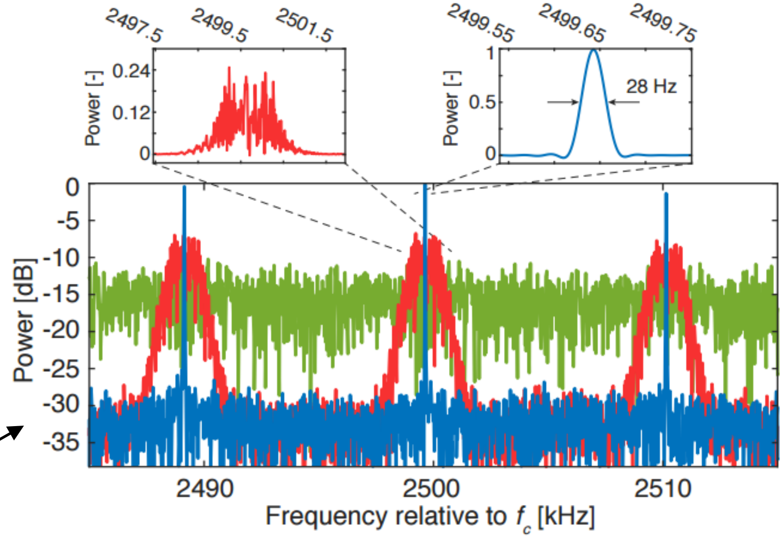
- Can use dual-comb signal from one detector to correct dual-comb signal of another



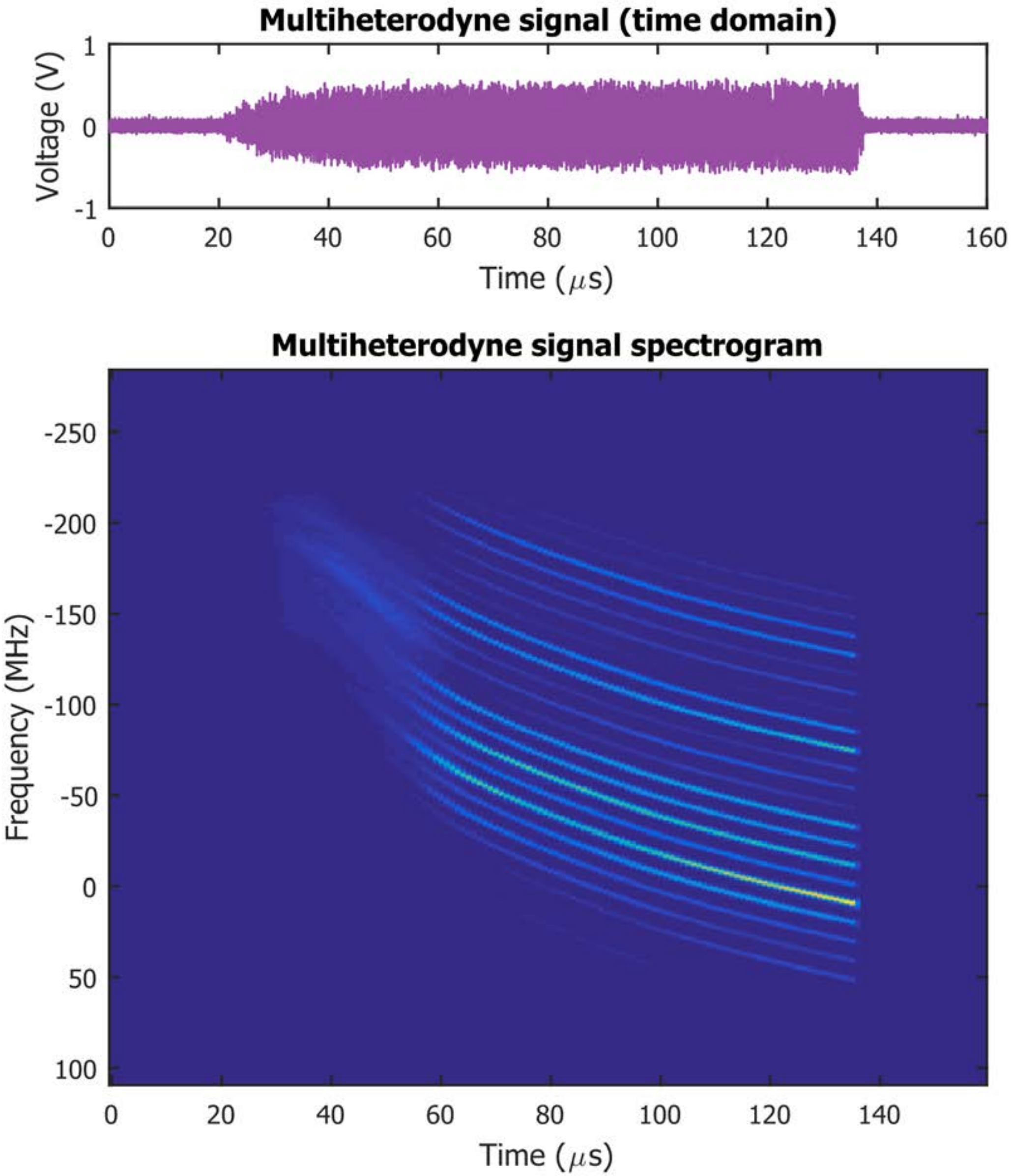
Burghoff, Yang, et al., “Computational multiheterodyne spectroscopy,” *Science Advances* (2016)

Advantages of computational correction

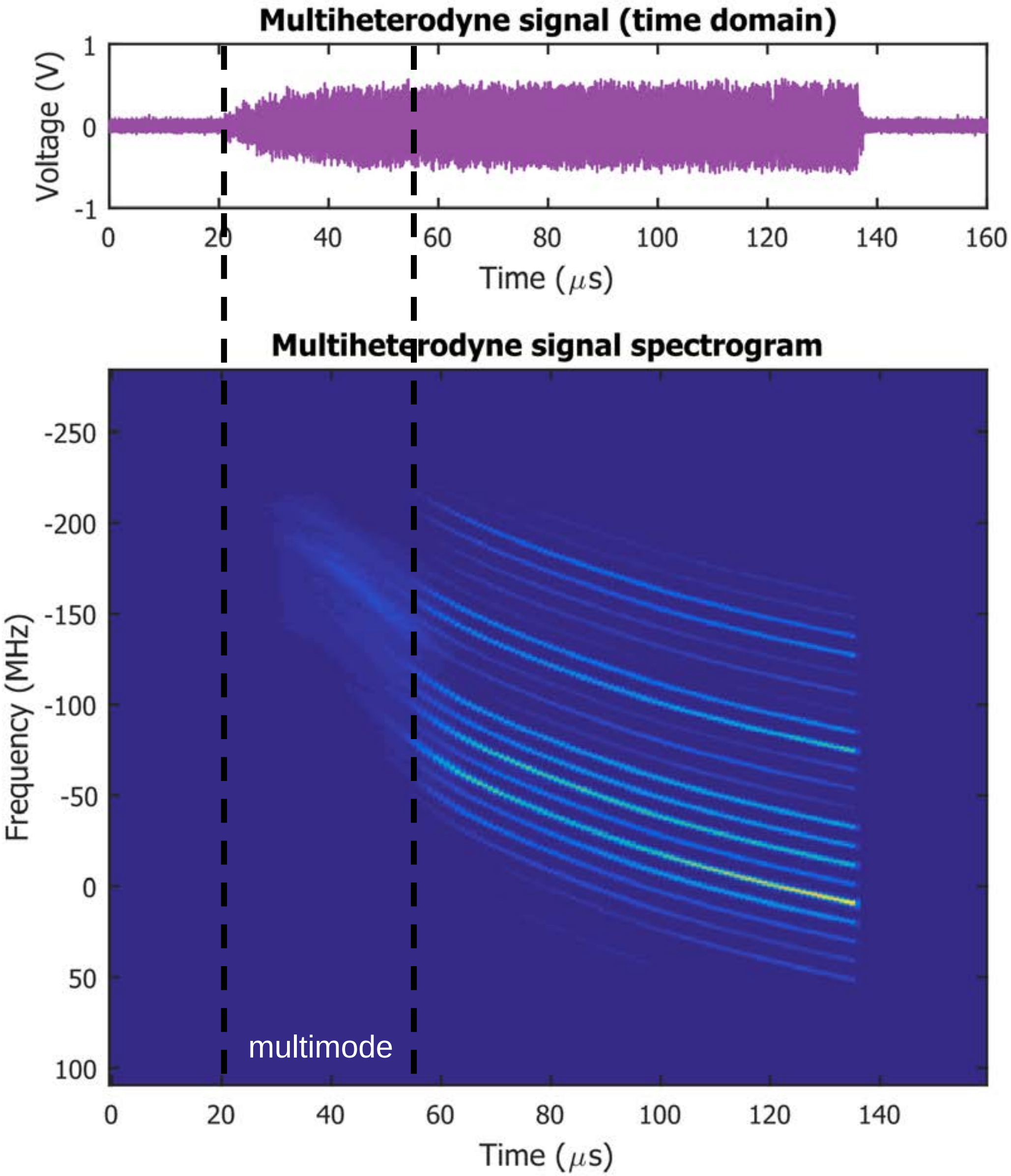
- Removes the need for temperature stabilization, bias stabilization, etc.
- Enables pulsed mode dual comb spectroscopy, which greatly reduces laser power consumption and makes portable sensing more viable.
- Computational approach viable for mode-locked lasers, interband cascade lasers:
  - Interband Cascade Lasers: Sterczewski, Westberg, Wysocki, et al., Opt. Eng (2017)
  - Mode-locked lasers: Hebert, Genest, et al., Optics Express (2017)



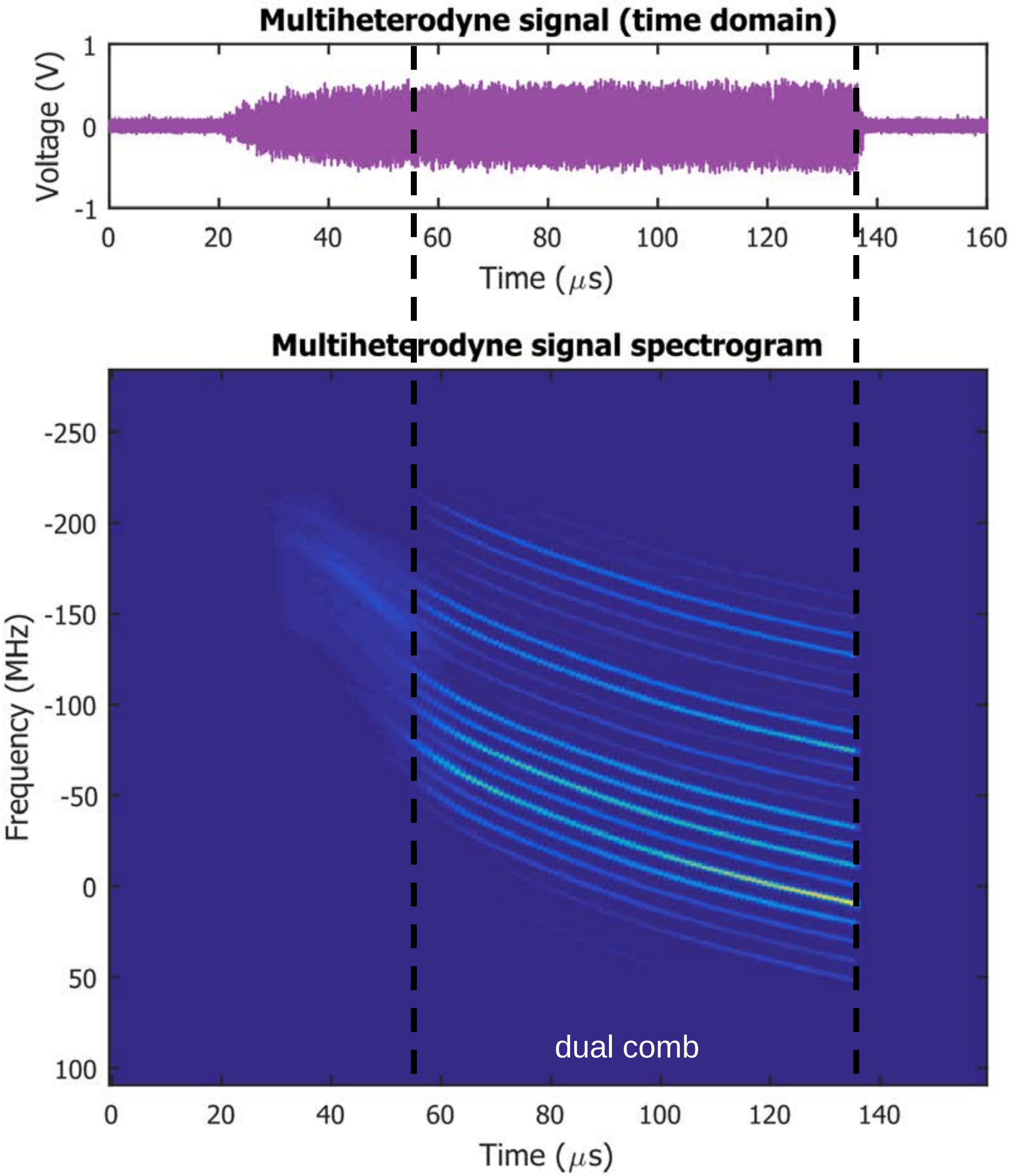
Dual comb signal in pulsed mode



Dual comb signal in pulsed mode



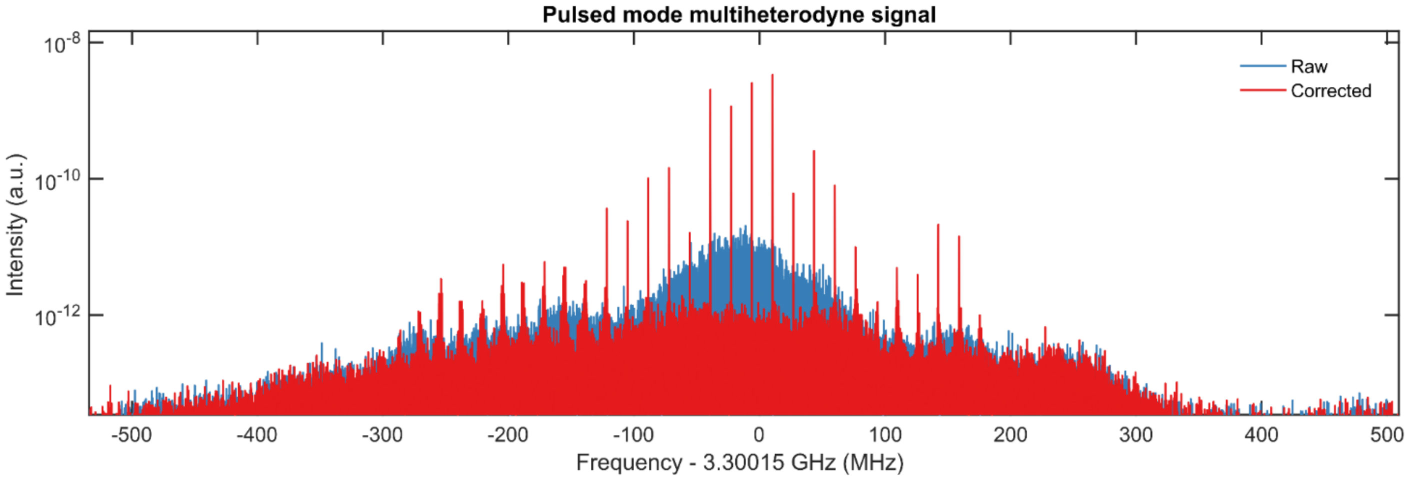
Dual comb signal in pulsed mode



signal is chirped
due to laser
heating

Pulsed mode dual comb spectroscopy

Spectrum from a single 120 us electrical pulse



- Computational phase correction allows for chirp to be measured and continuously corrected
- Reduces cooling requirements on QCLs (~10 W in continuous wave mode)

K561 Integral Stirling
170 mW @ 80 K

Conclusions

- Showed how by using dispersion engineering, terahertz quantum cascade lasers can be forced to operate in a comb regime
- Showed that dual comb (multiheterodyne) spectroscopy was possible in both pulsed and continuous-wave mode
- Showed that signal processing could be used to digitally reverse phase and timing corruption inherent to many dual comb systems, making systems more practical