# Spectroscopy with Laser Frequency Combs

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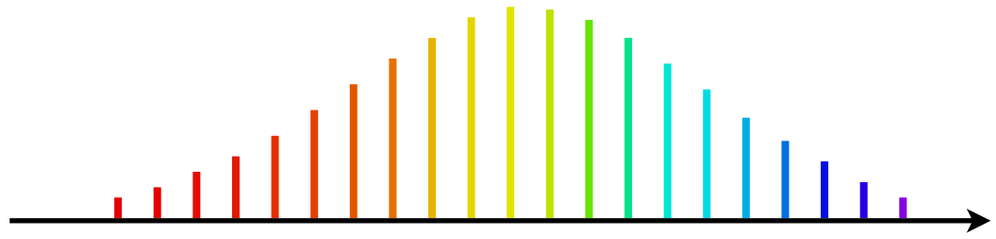
IQCLSW 2014

**International Quantum Cascade Lasers School & Workshop 2014**
Policoro (Matera) Italy, September 7th - 12th, 2014

"laser frequency comb"



Google Italia 268 000 entries

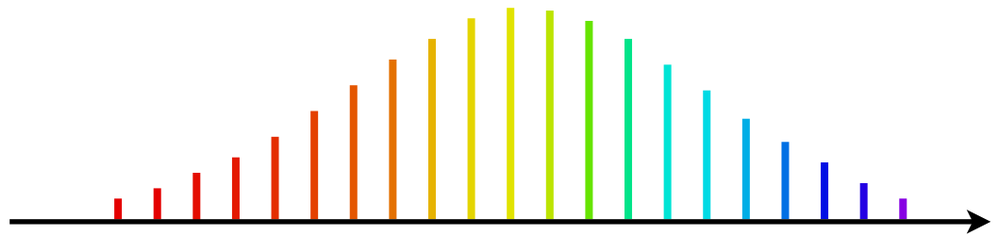
controlling radiation

from THz to XUV

IQCLSW 2014

4 talks

# laser frequency comb



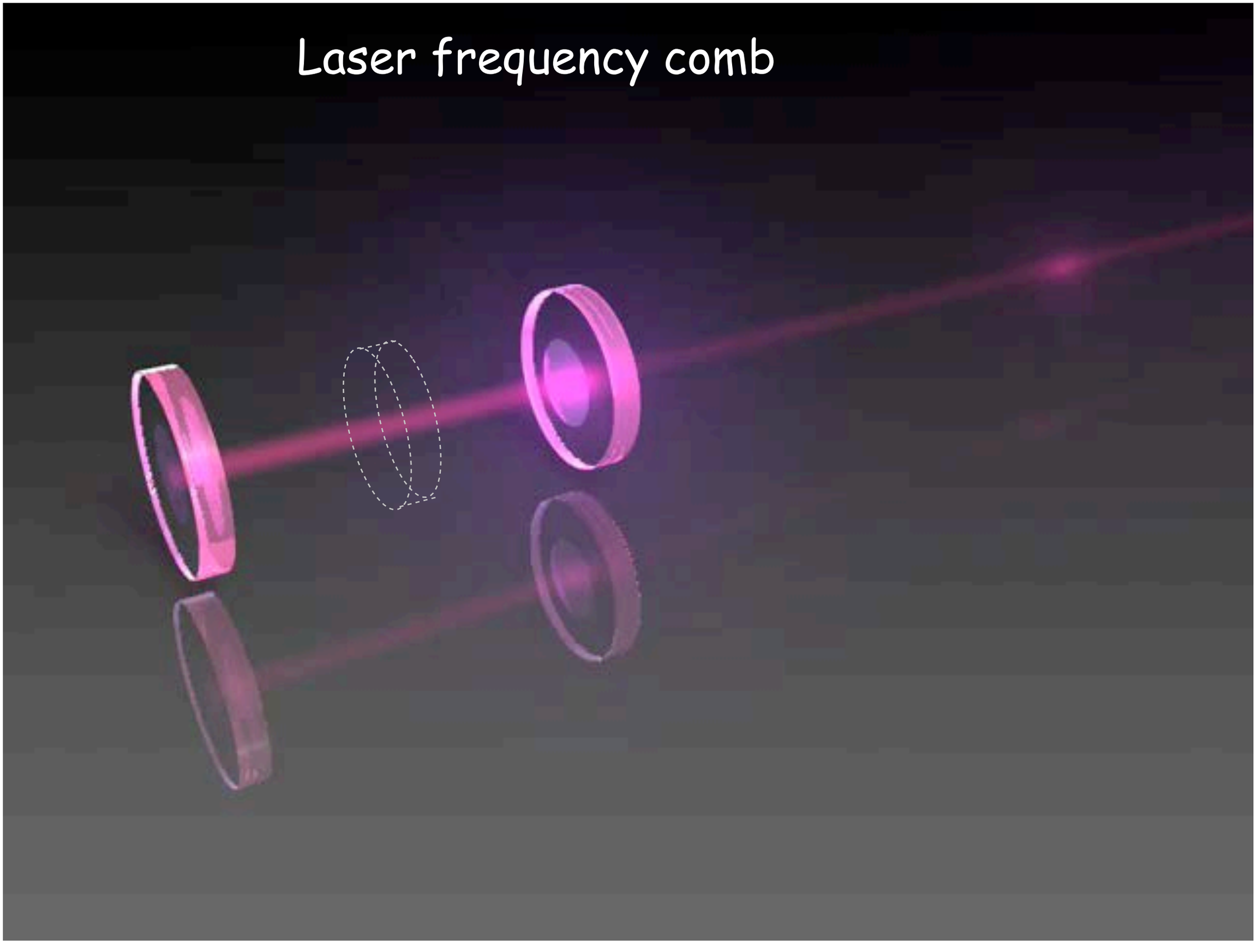
A simple tool for measuring optical
frequencies of 100's or even 1000's of THz.

A phase coherent link between
the optical and the radio-frequency region.

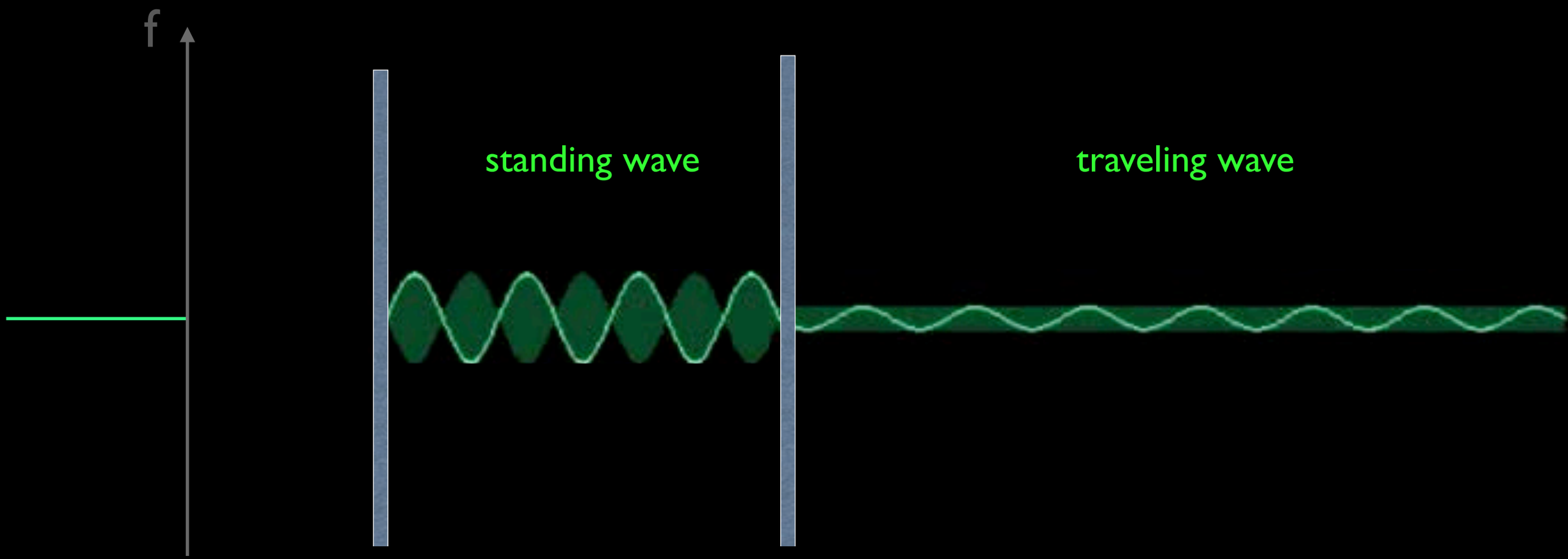
A clockwork mechanism for an
optical atomic clock.

T.W.H. **Passion for precision** (Nobel lecture), Rev. Mod. Phys. 78, 1297 (2006)

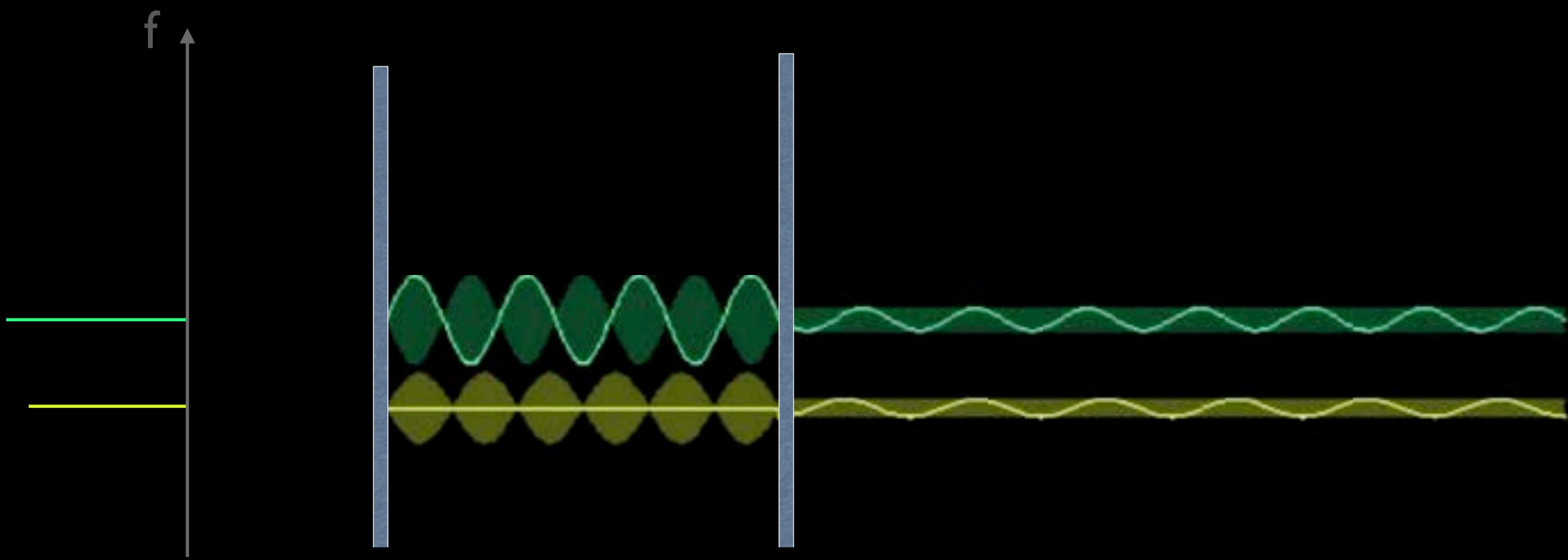
# Laser frequency comb



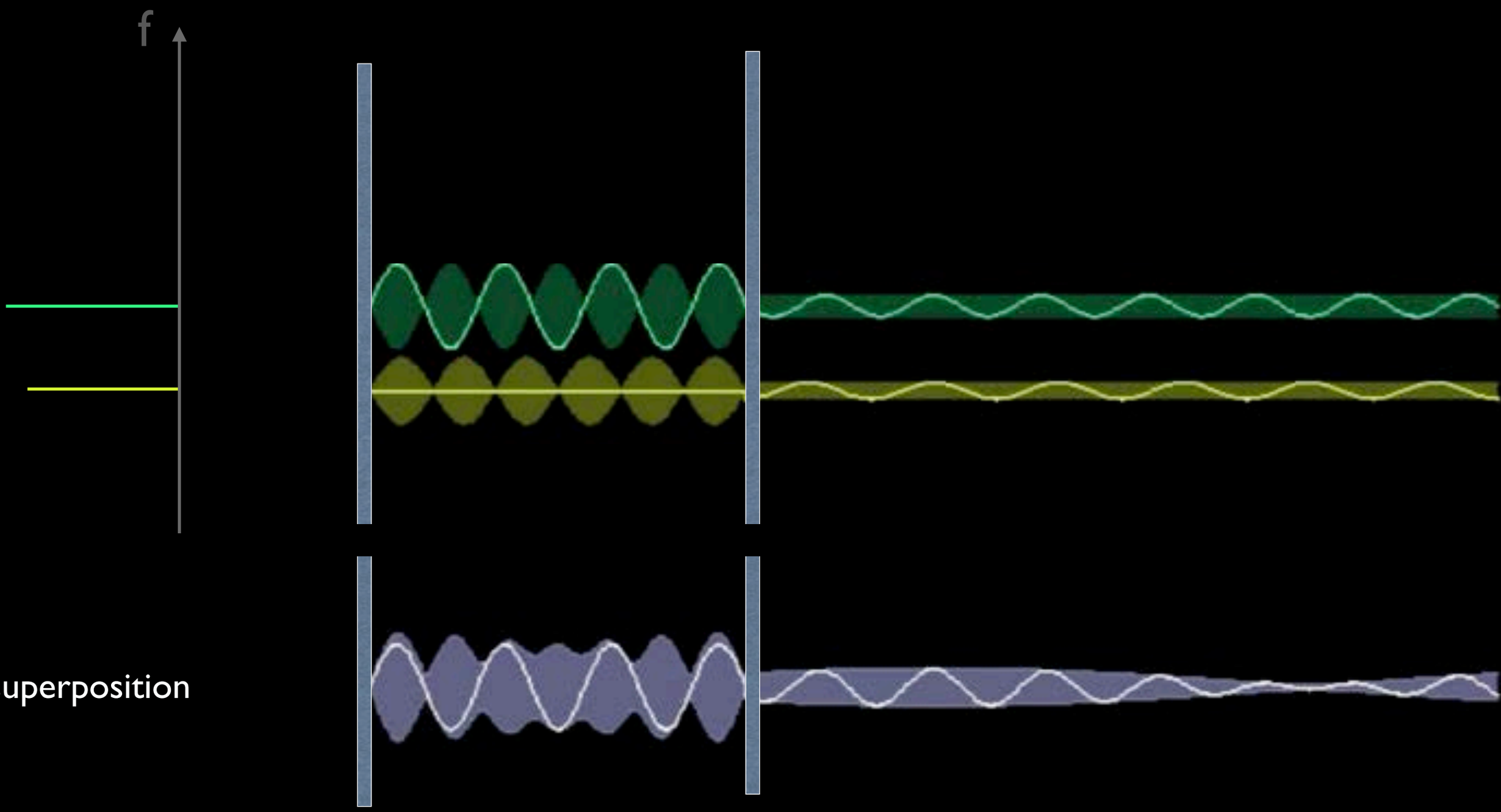
# single mode



# two modes

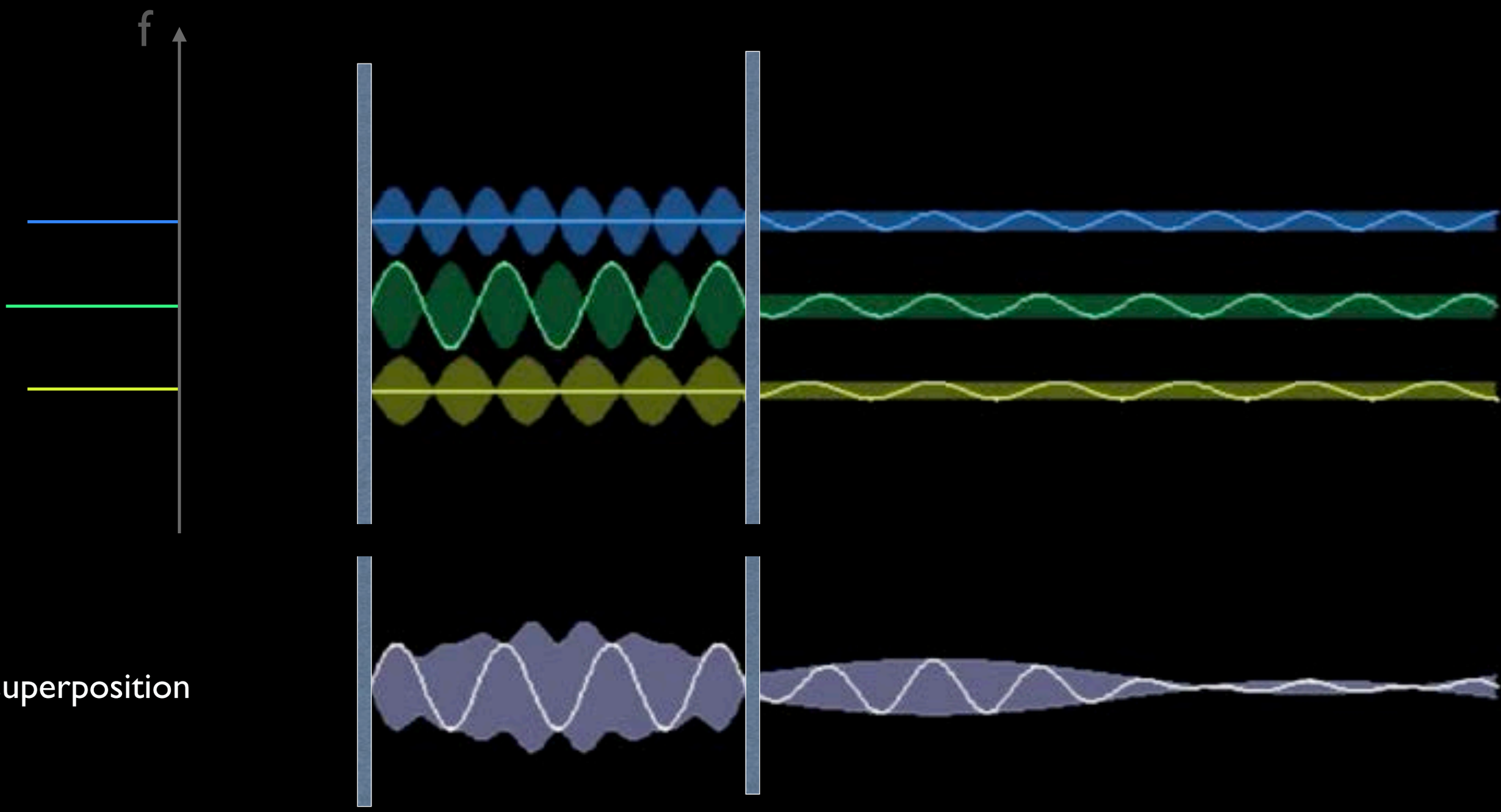


two modes



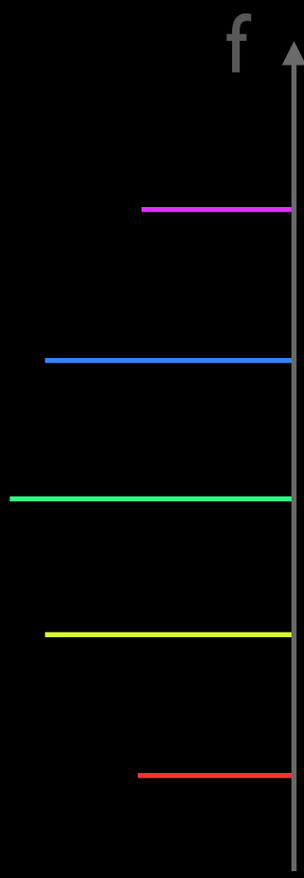

superposition

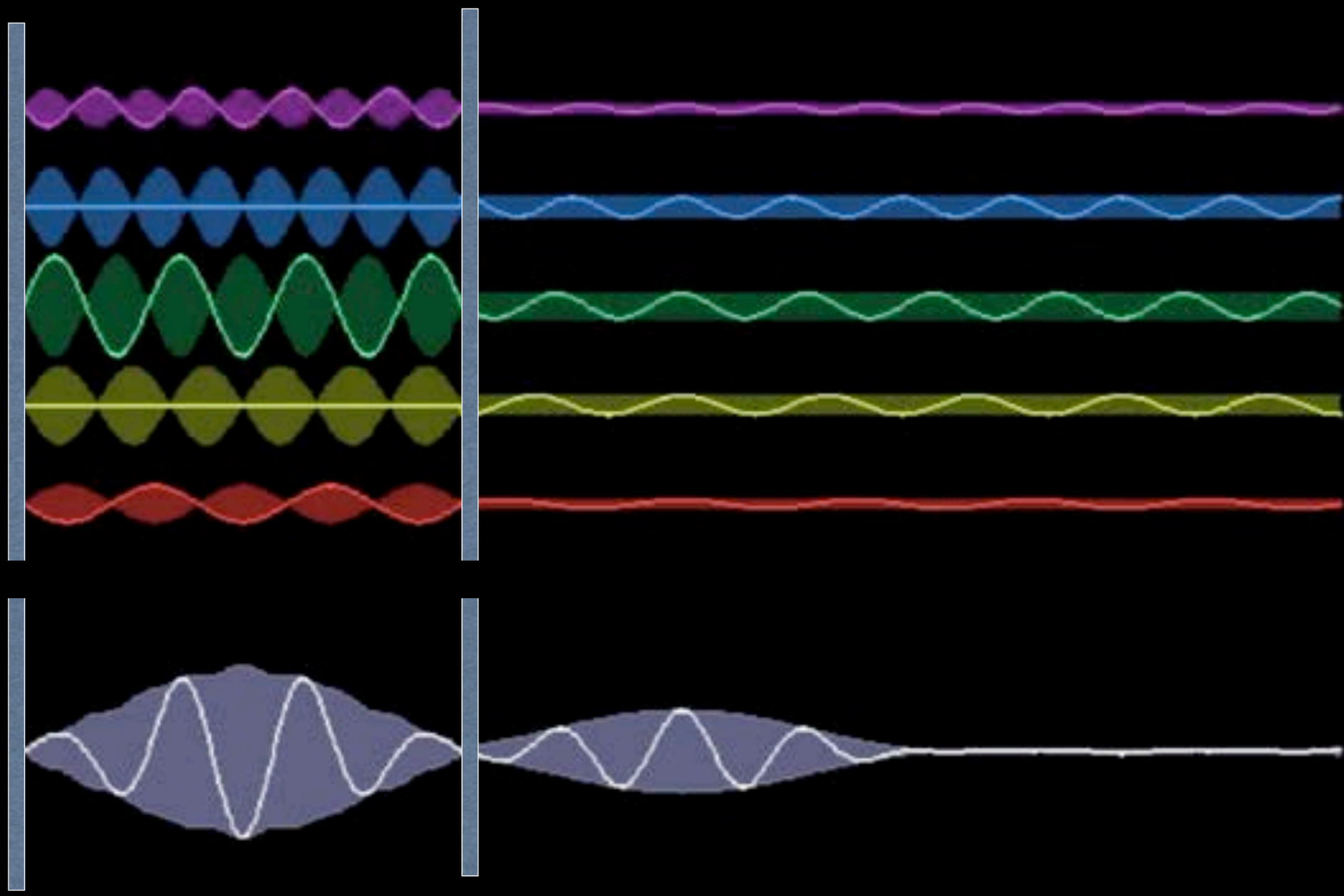
three modes




superposition

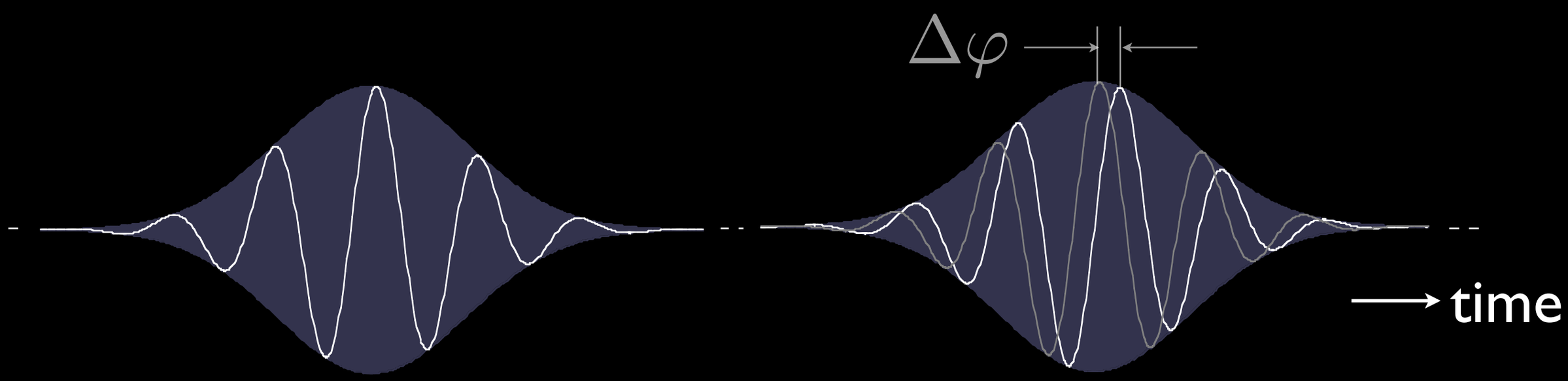
# many modes

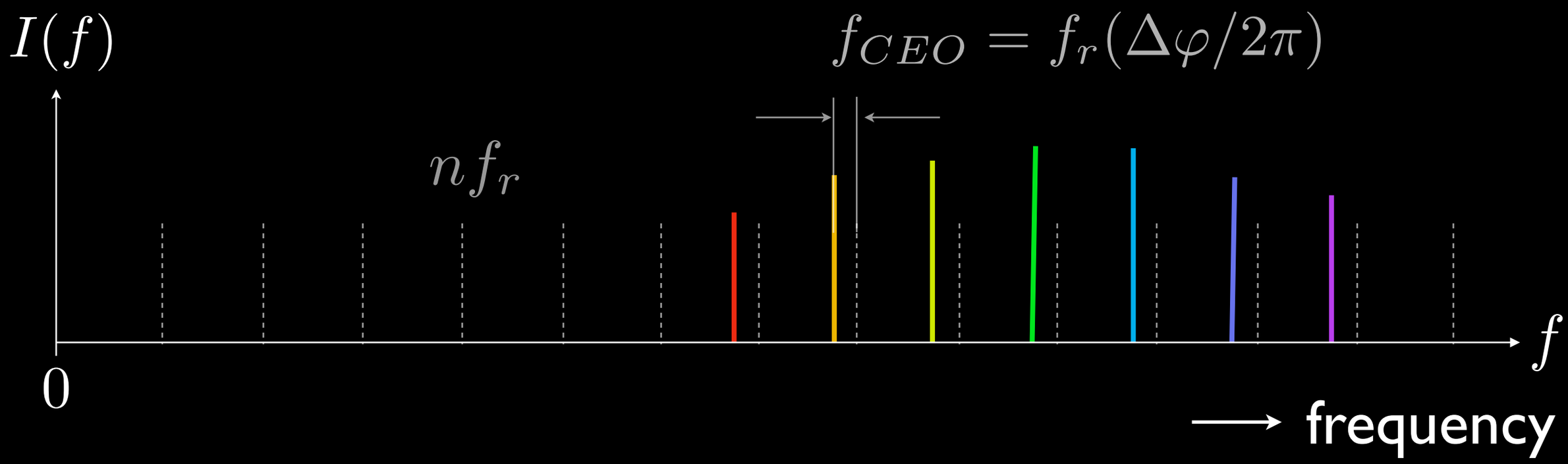


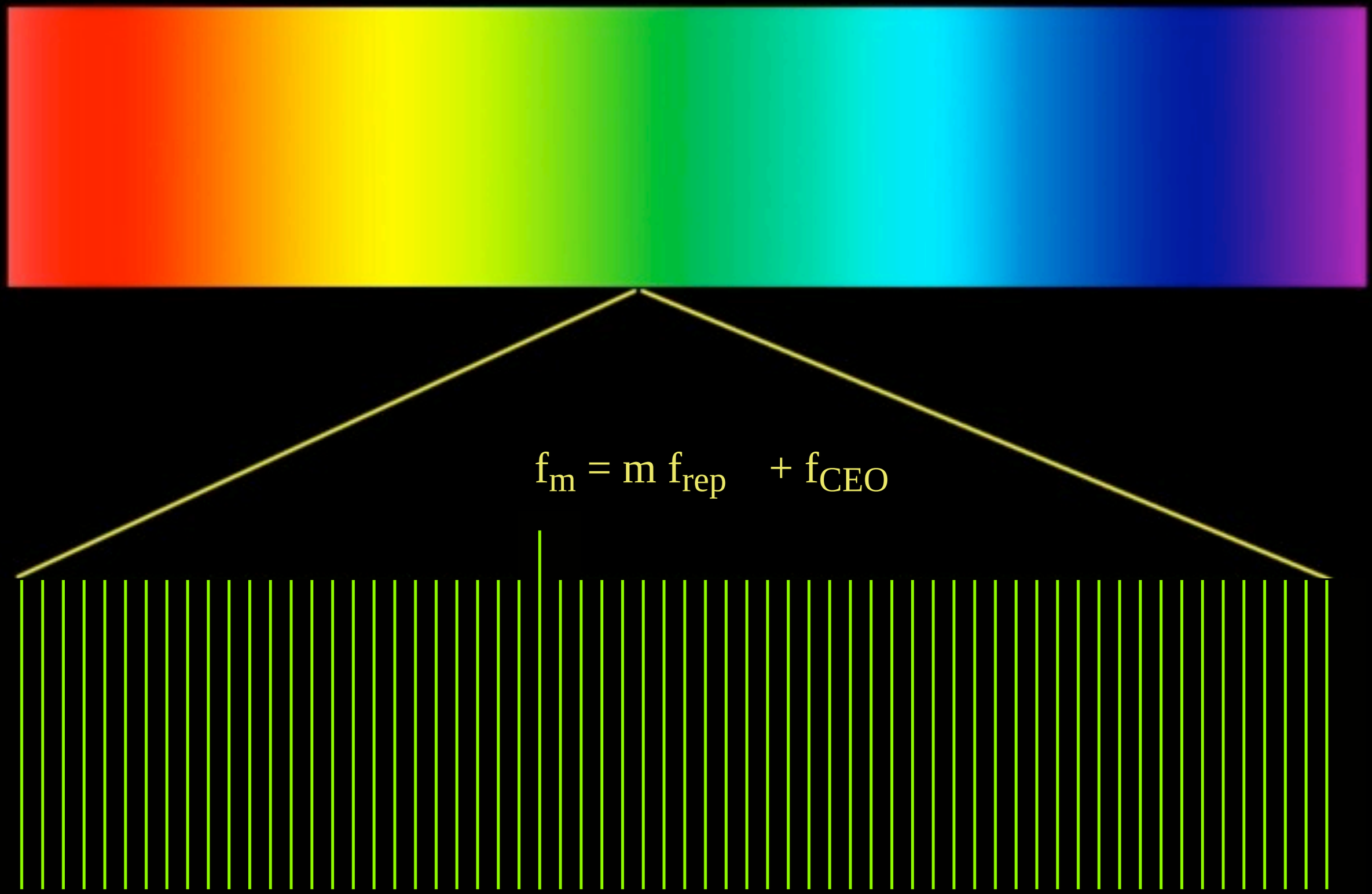
superposition

# carrier-envelope phase slips and offset frequency




J.N. Eckstein, *Ph.D. Thesis*, Stanford University, 1978


$f_m = m\, f_{rep} + f_{CEO}$

$f_m = m\, f_{rep}$

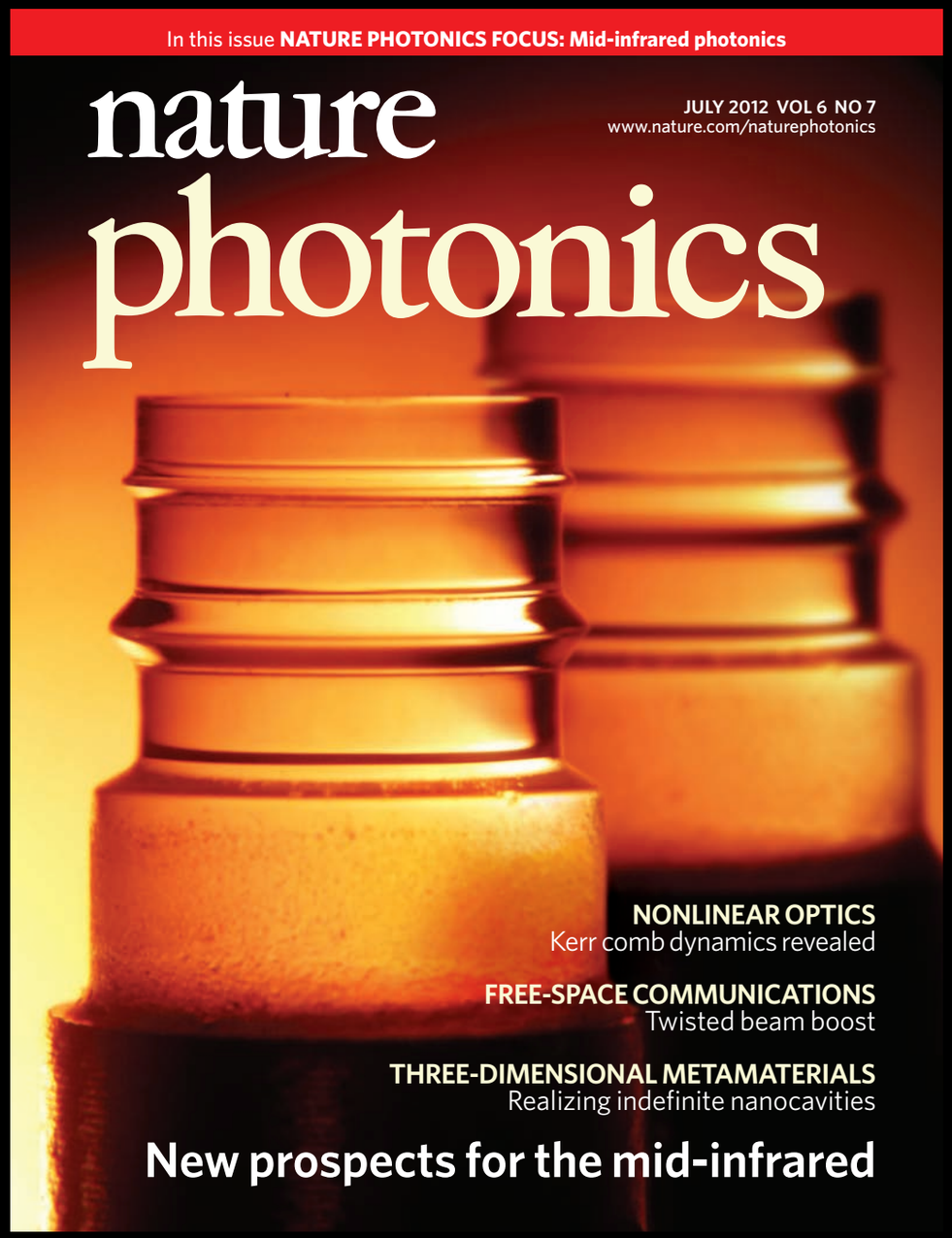
# Erbium doped fiber laser frequency comb

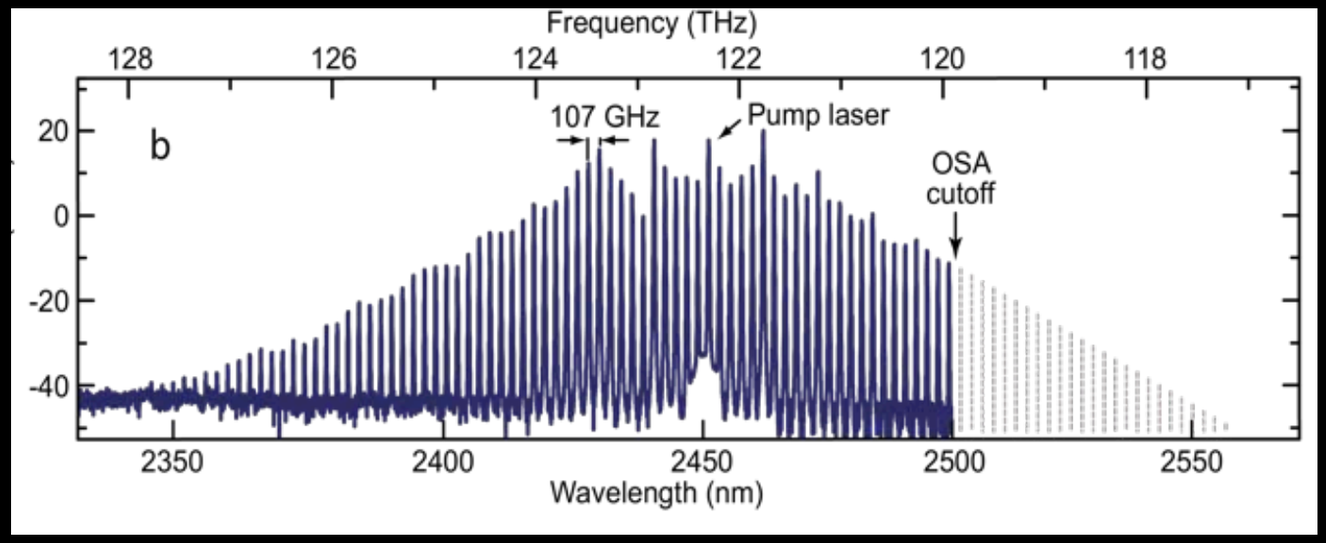
## Space-Comb

## Launch: November 2014

# Integrated Silicon Nitride Comb Sources

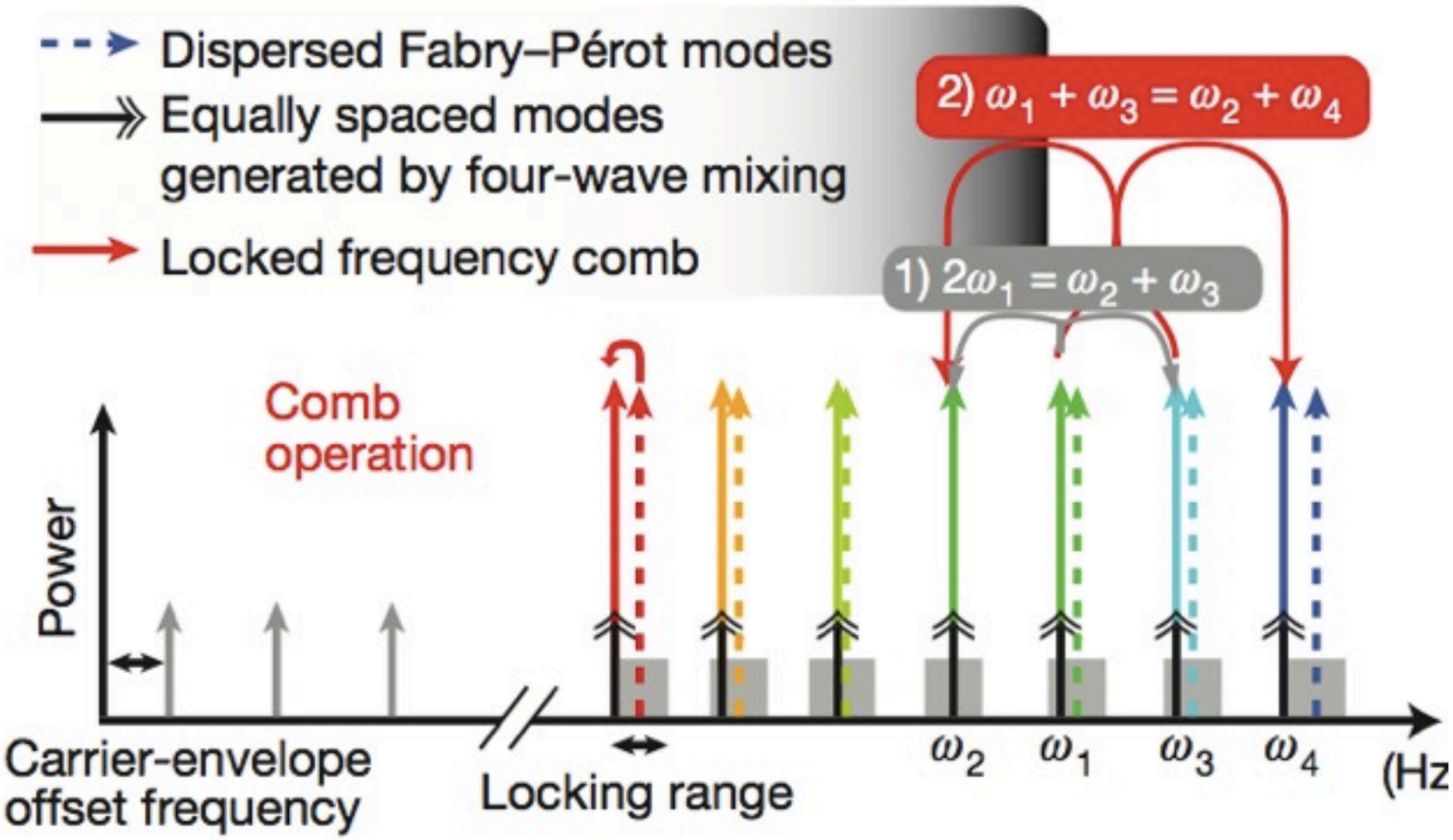
# Crystalline toroidal micro-resonators produce frequency combs in the mid-infrared

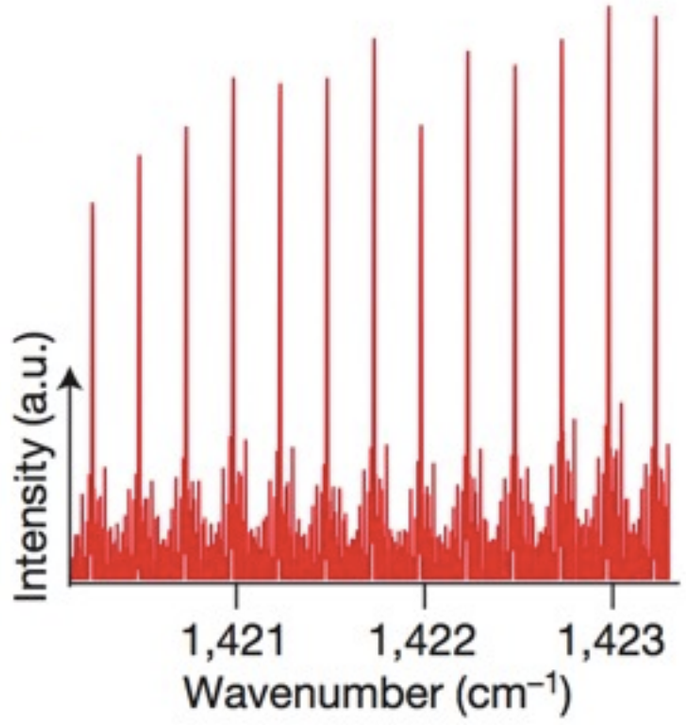



A. Schliesser et al, Nature Photonics 6, 440–449 (2012)
C.Y. Wang et al, Nature Communications 4, 1345 (2013)

# Towards mid-IR frequency combs from quantum cascade lasers




A. Hugi, G. Villares, S. Blaser, H. C. Liu & J. Faist, Nature DOI:10.1038/nature11620

# Frequency combs - evolutionary tree

trace gas sensing

XUV comb and harmonic generation

extrasolar planet searches

dual comb spectroscopy

coherent control

wave form synthesis

continuing expansion of universe ?

calibration of astronomical spectrographs

frequency comb spectroscopy

low-noise frequency synthesis

optical clocks

tests of fundamental physics

attosecond science

carrier-envelope phase control

self-referenced octave spanning frequency comb

frequency measurements

length measurements

time - frequency transfer

mode-locked femtosecond laser

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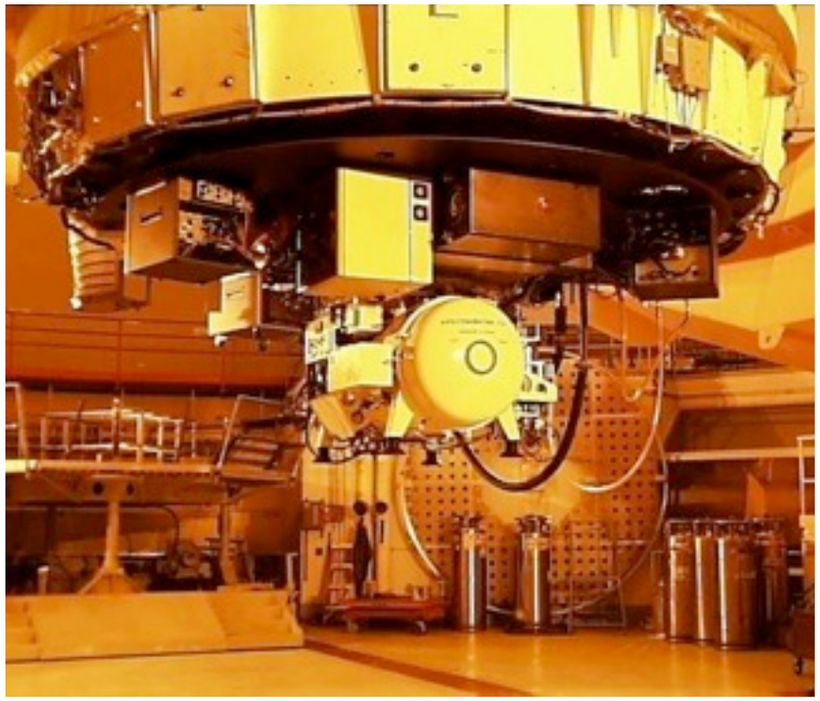
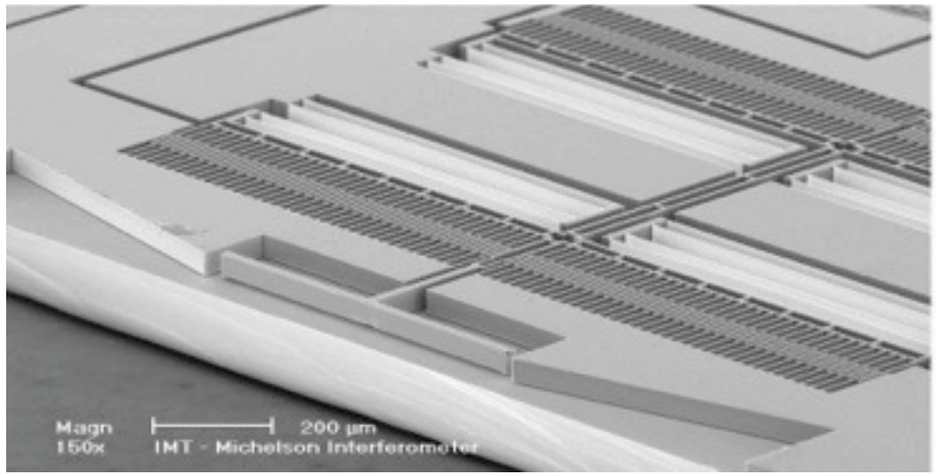
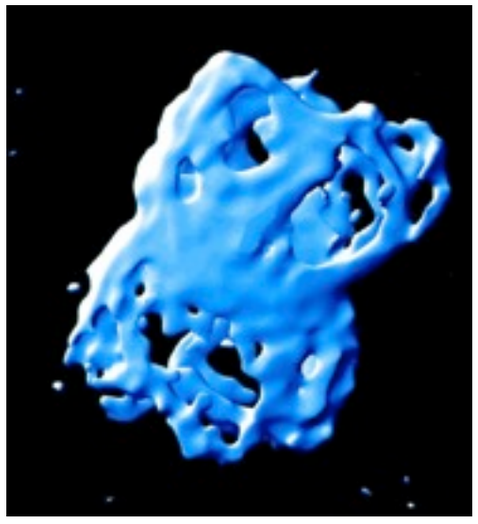
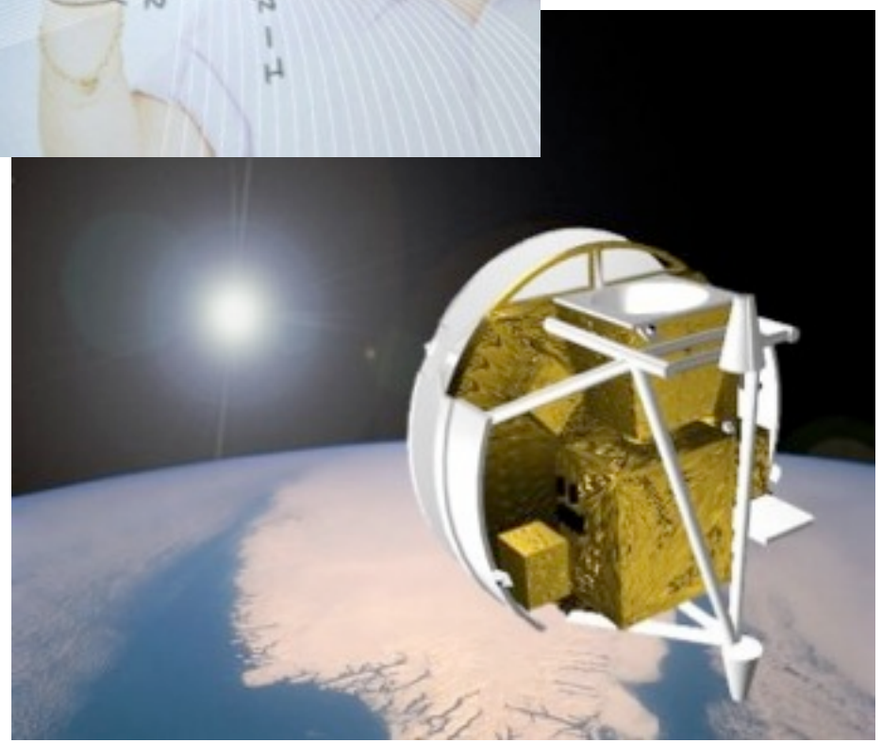
# Fourier transform spectroscopy



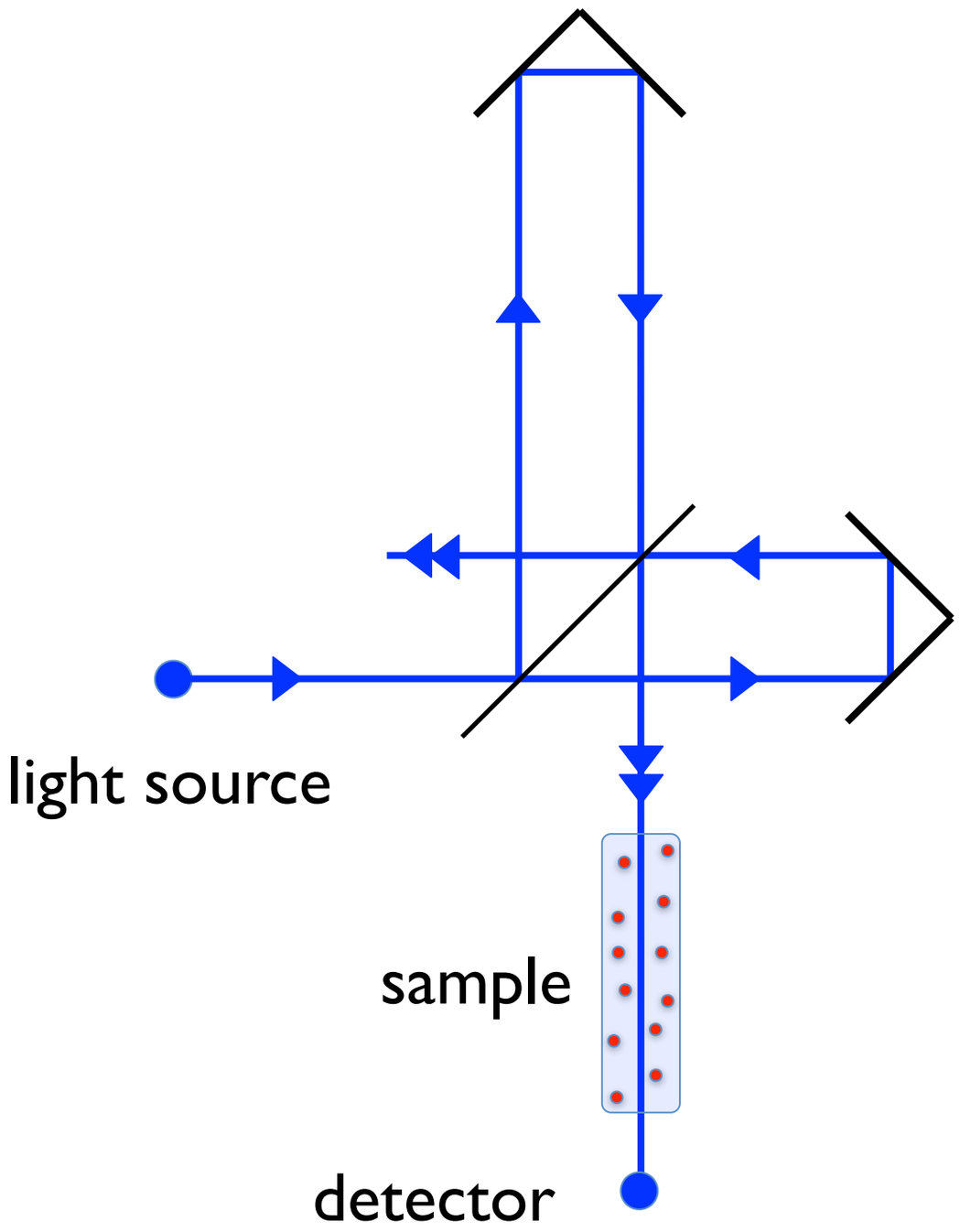



SOLEIL SYNCHROTRON





# Michelson-based Fourier transform spectroscopy



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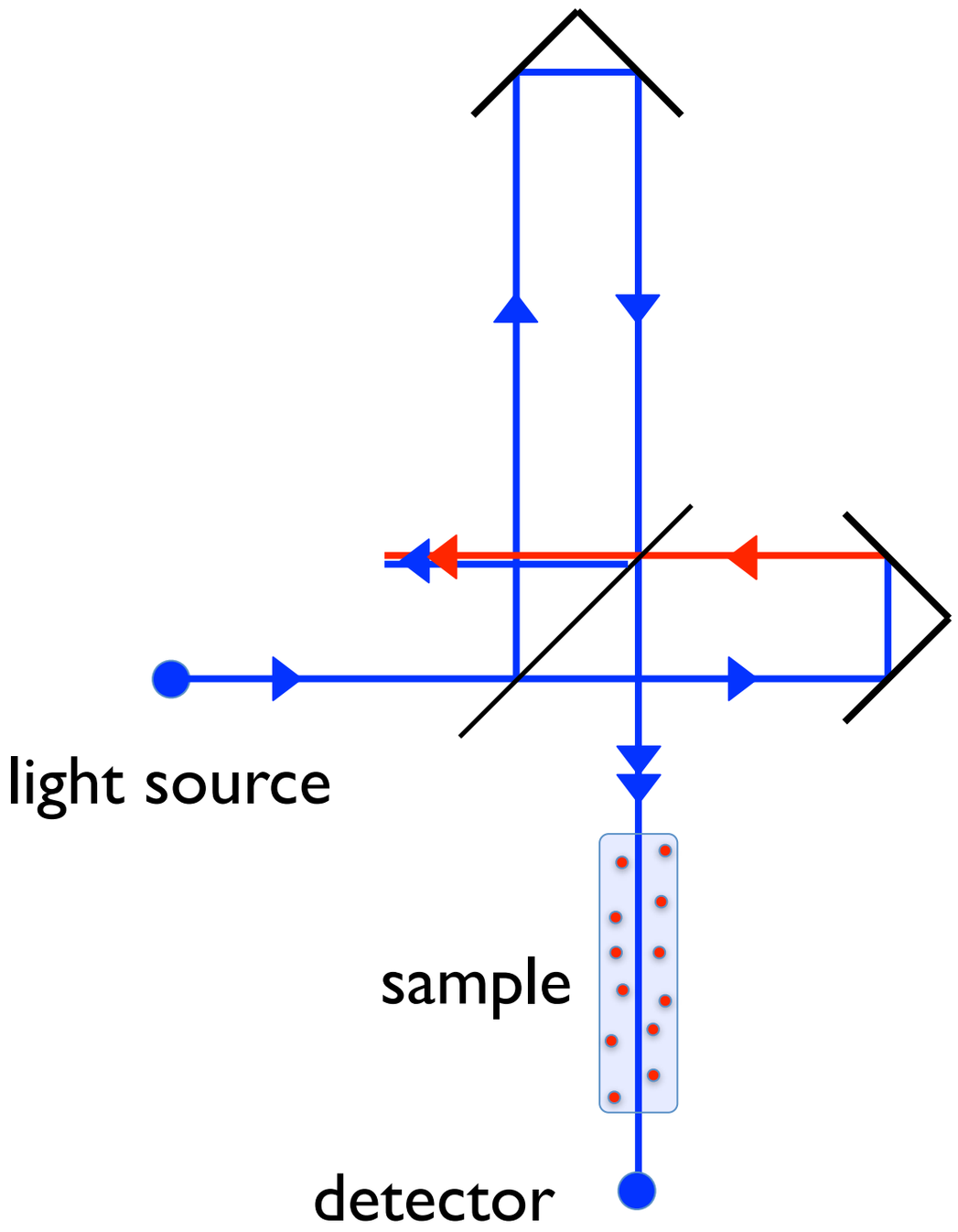

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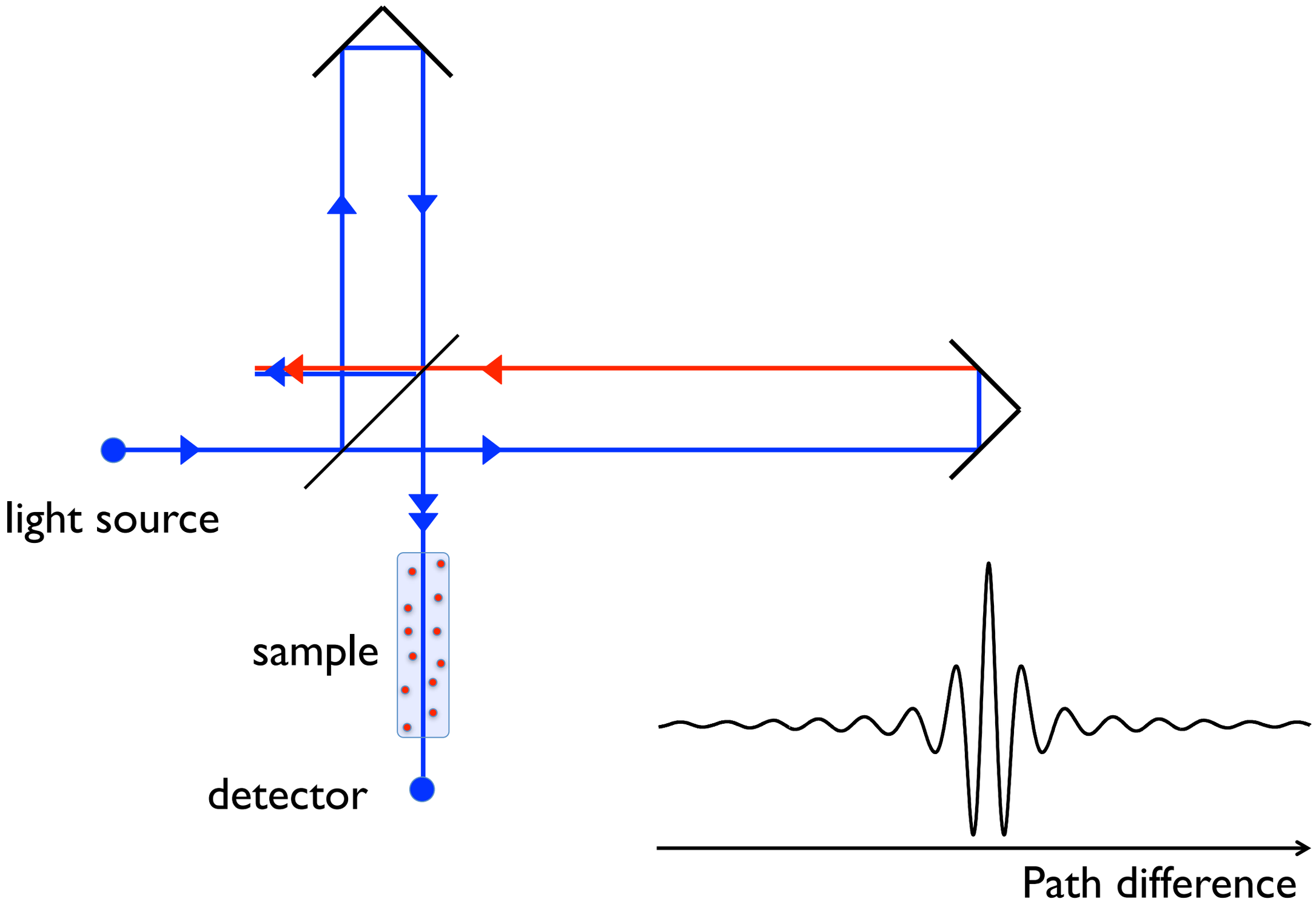

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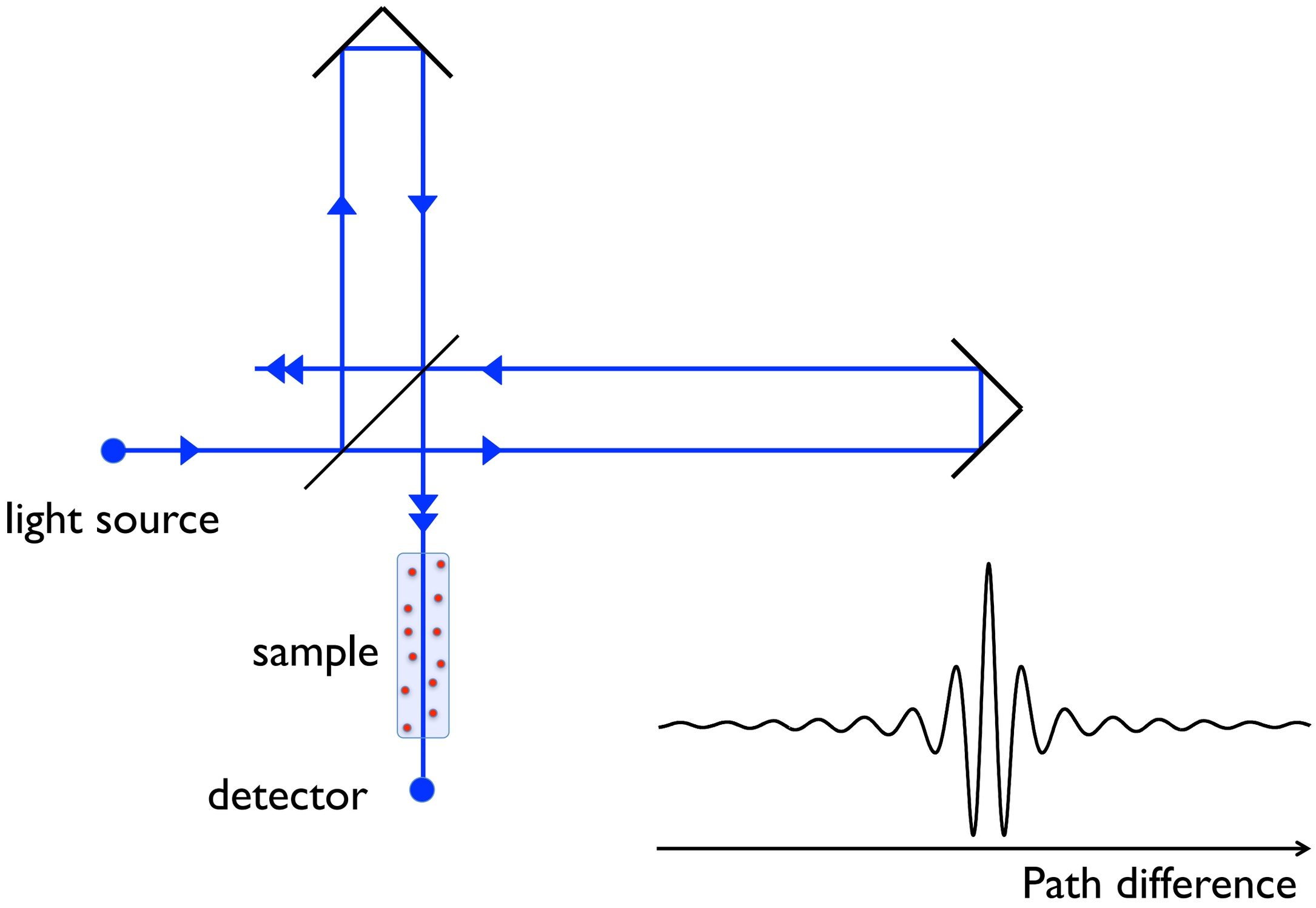

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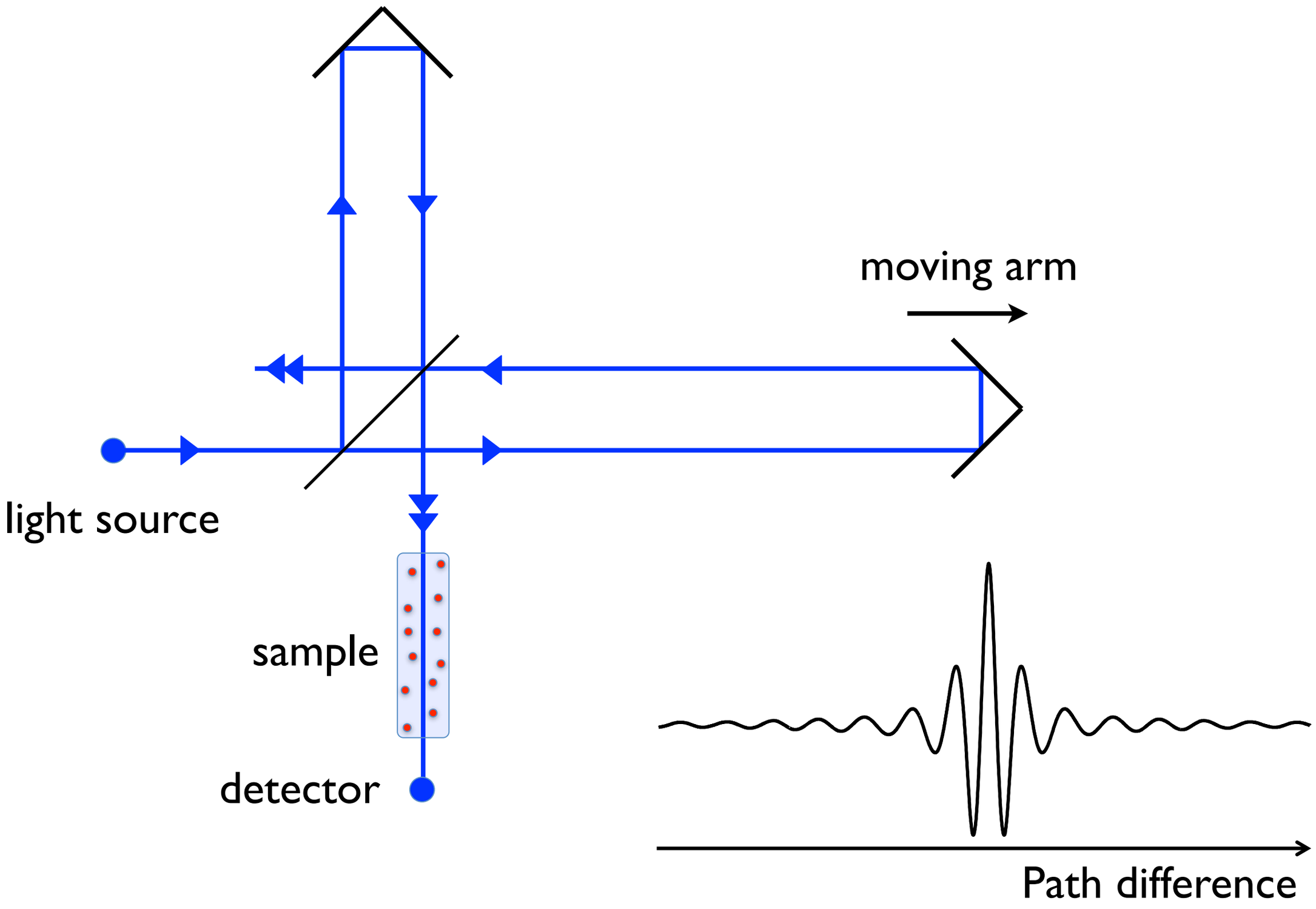
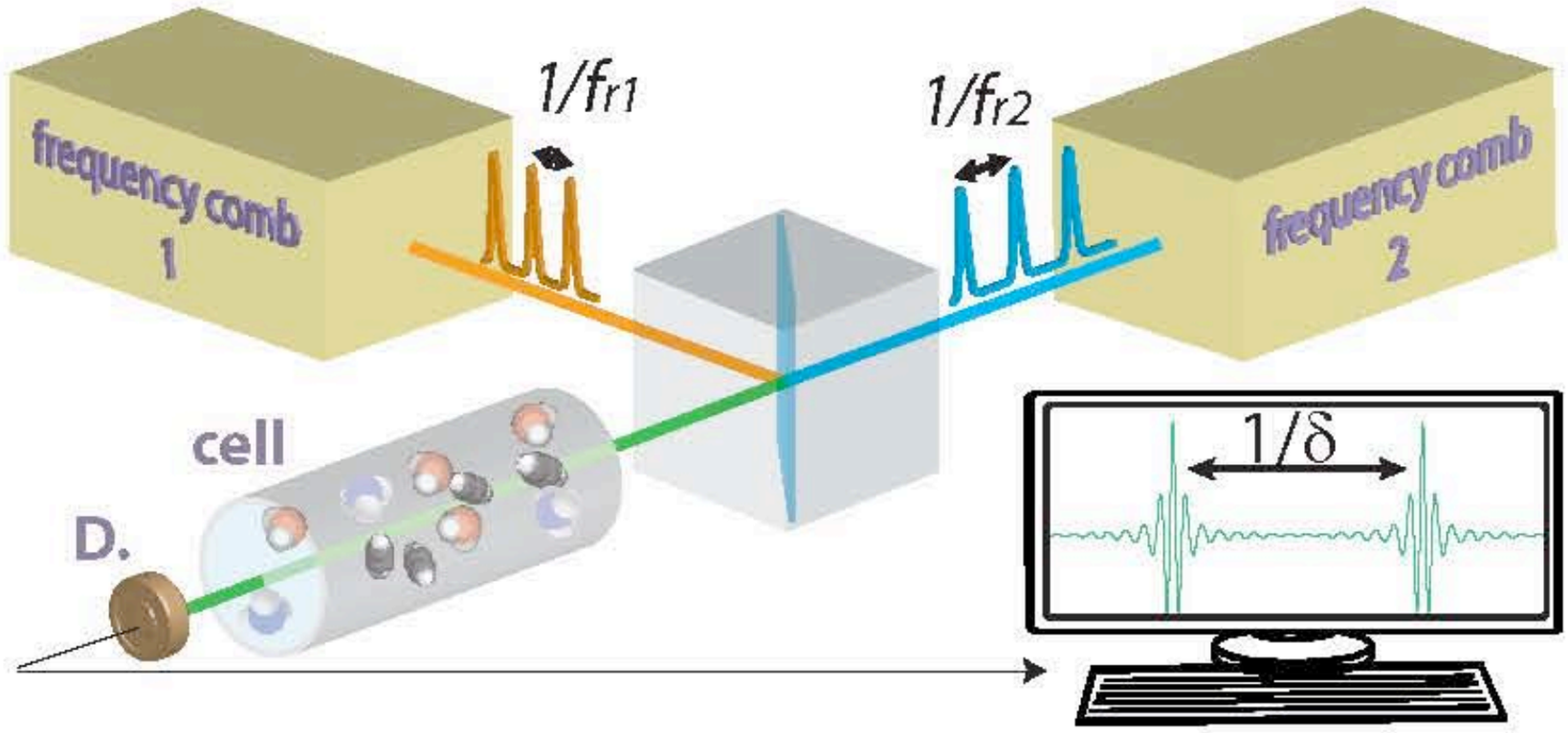
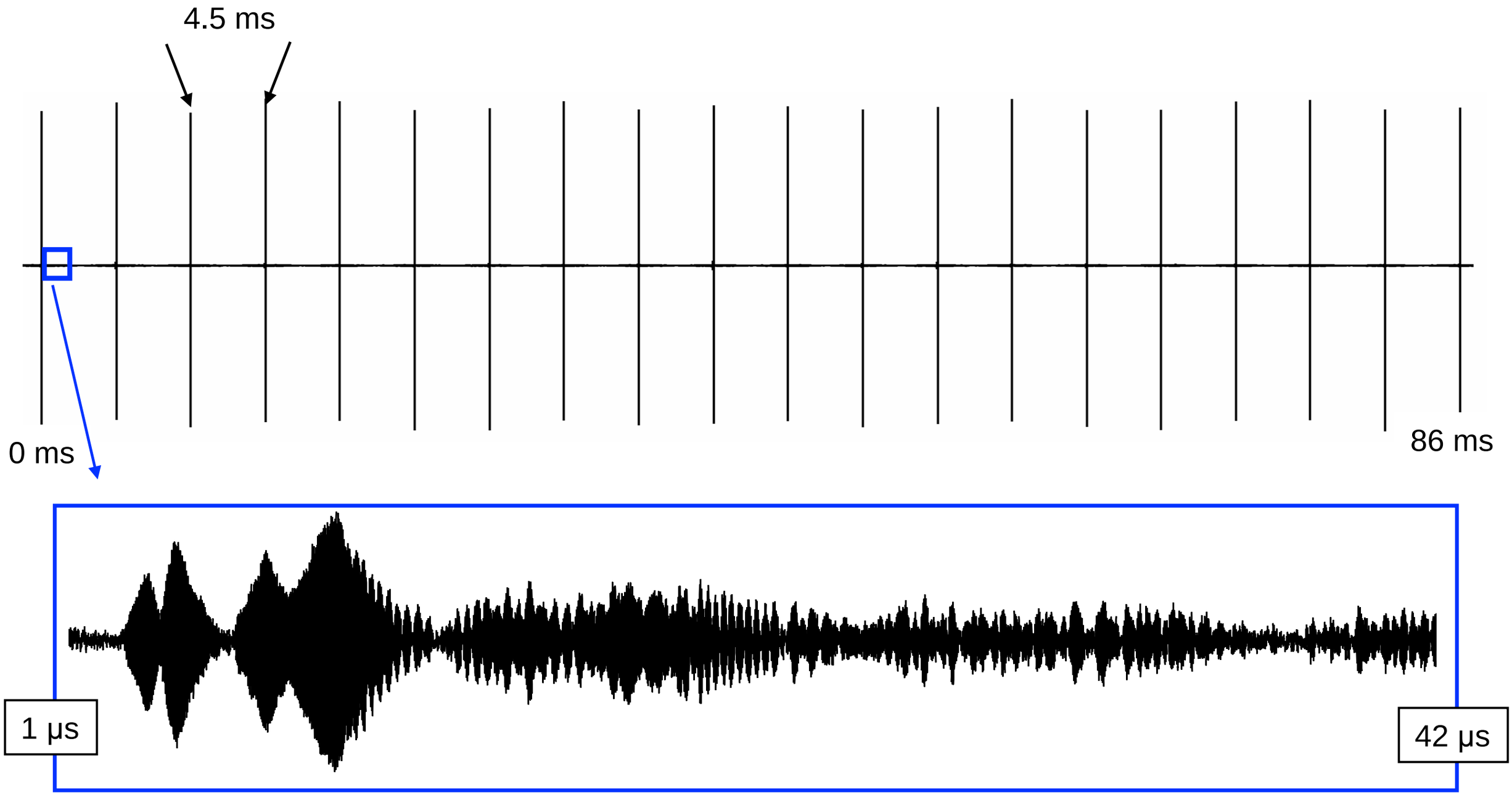


# Fourier Transform Spectroscopy with two frequency combs

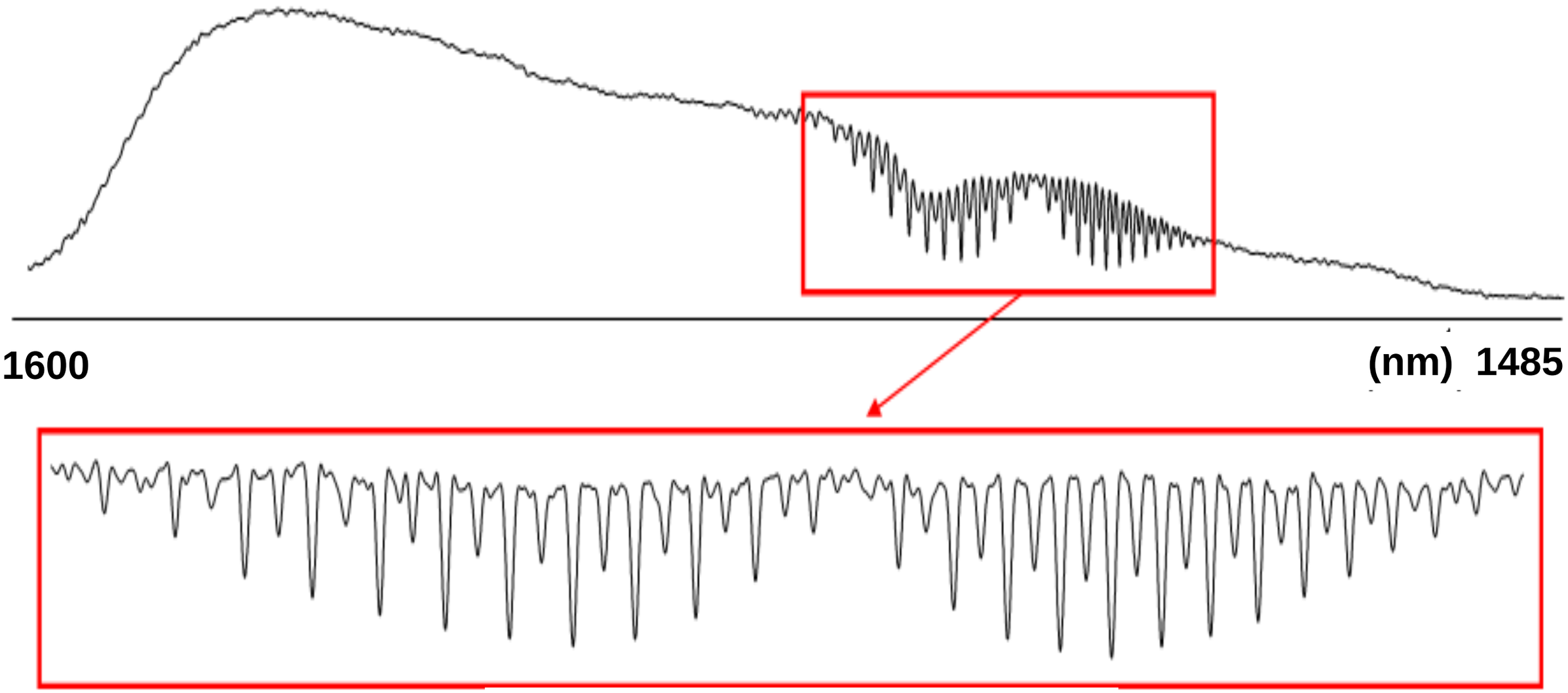



no moving parts!

# time domain interferometric signal



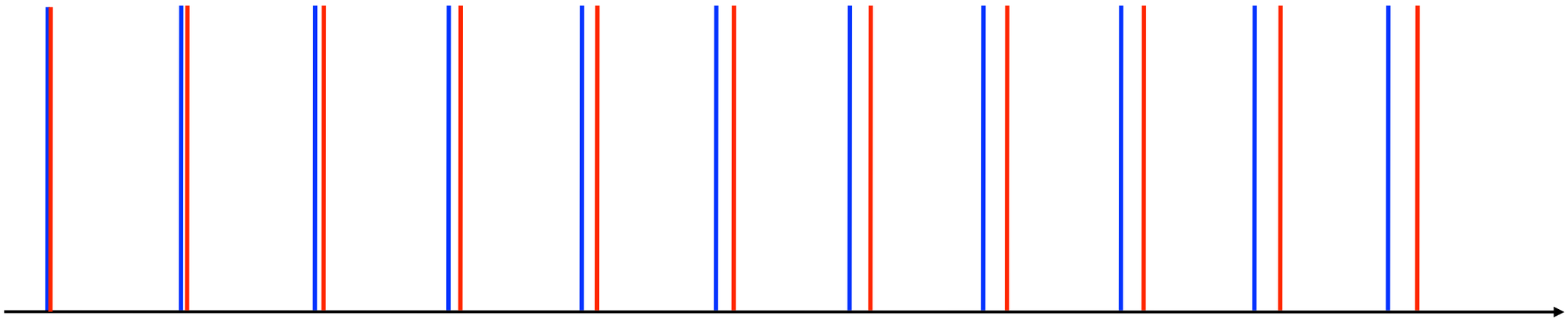
# after Fourier transformation




$C_2H_2$ $\nu_1+\nu_3$ band

Resolution: 3 GHz,  Measurement time: 42 μs (Single shot)

# pairs of comb lines produce radio frequency beat notes



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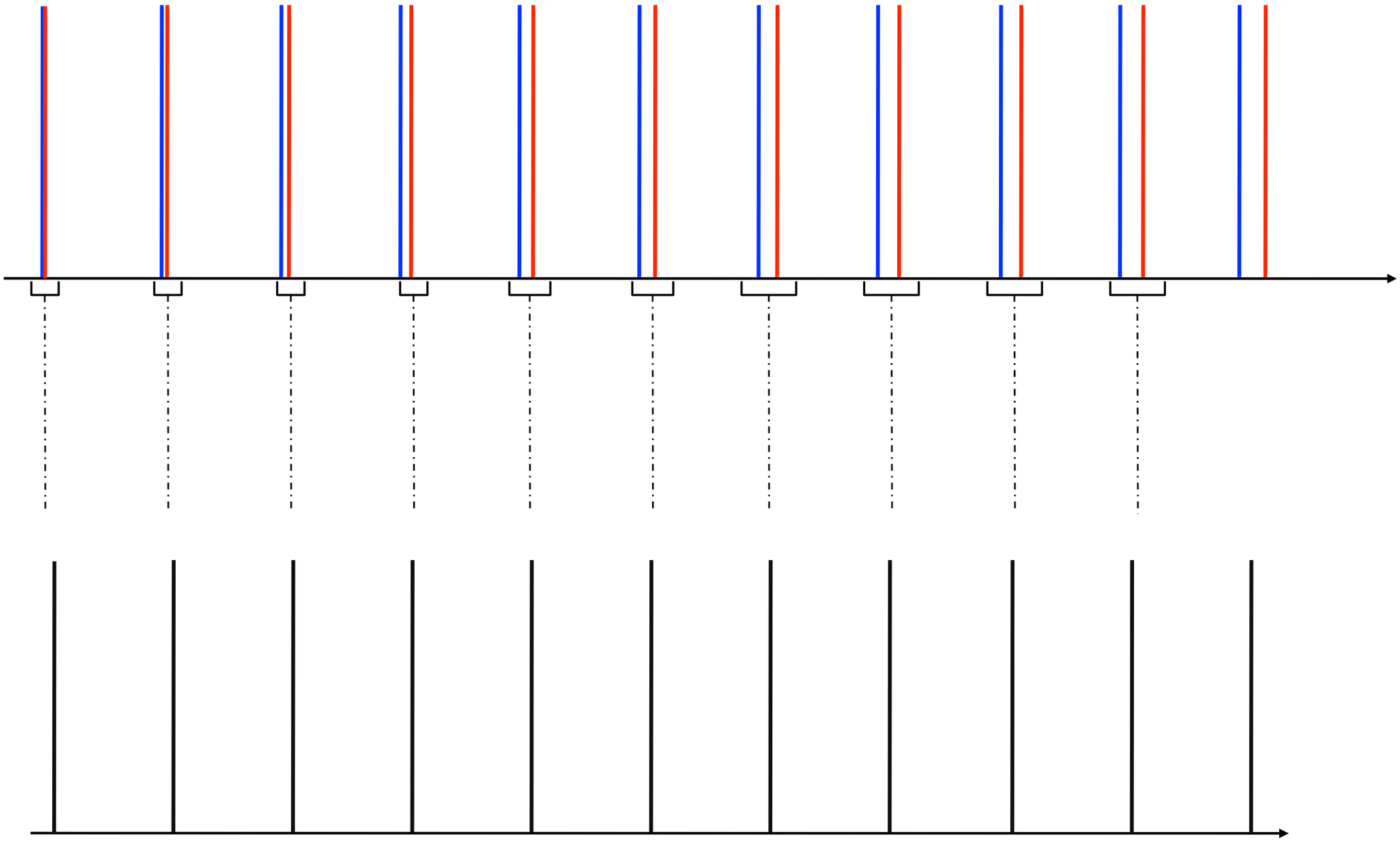

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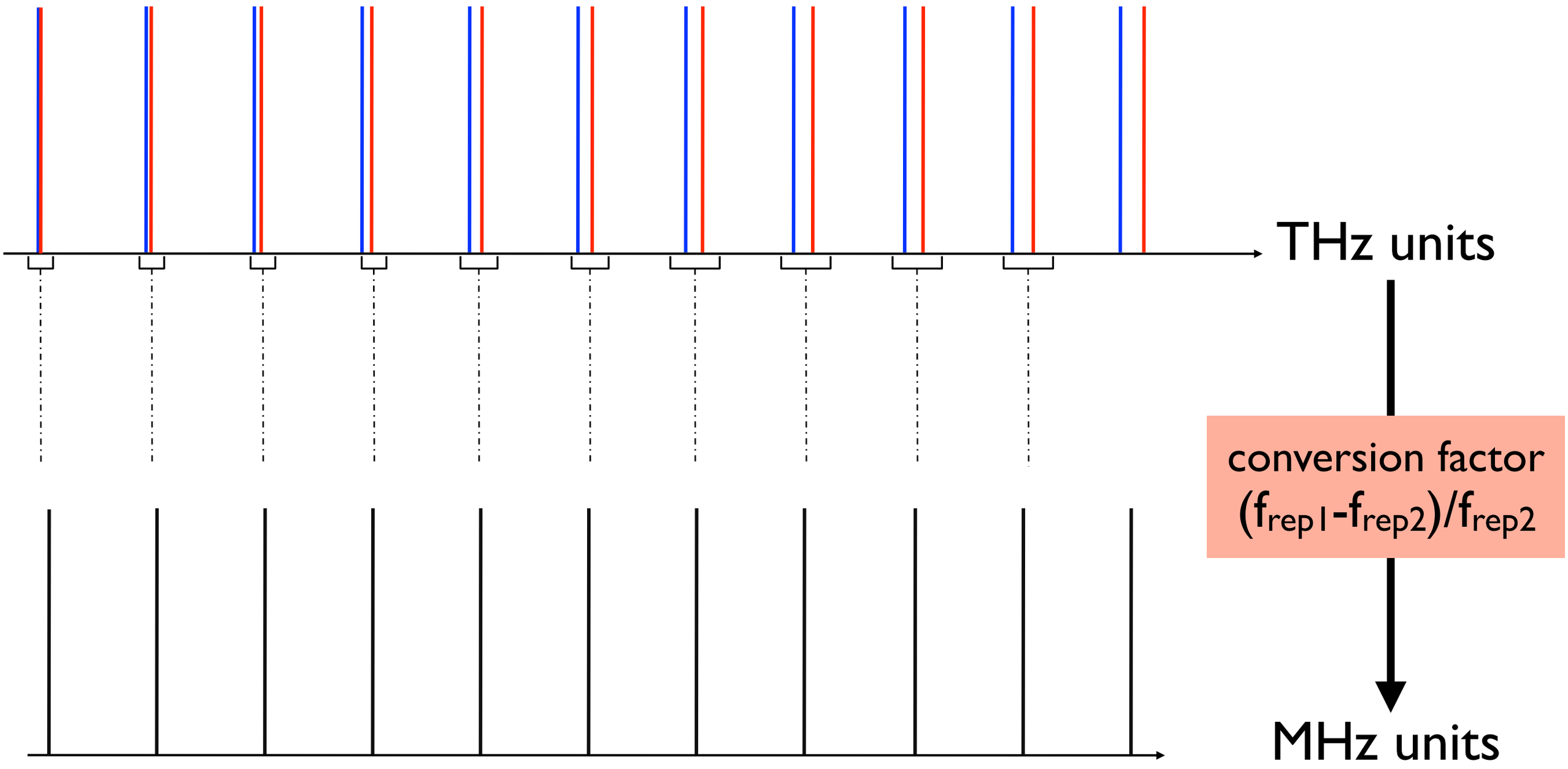
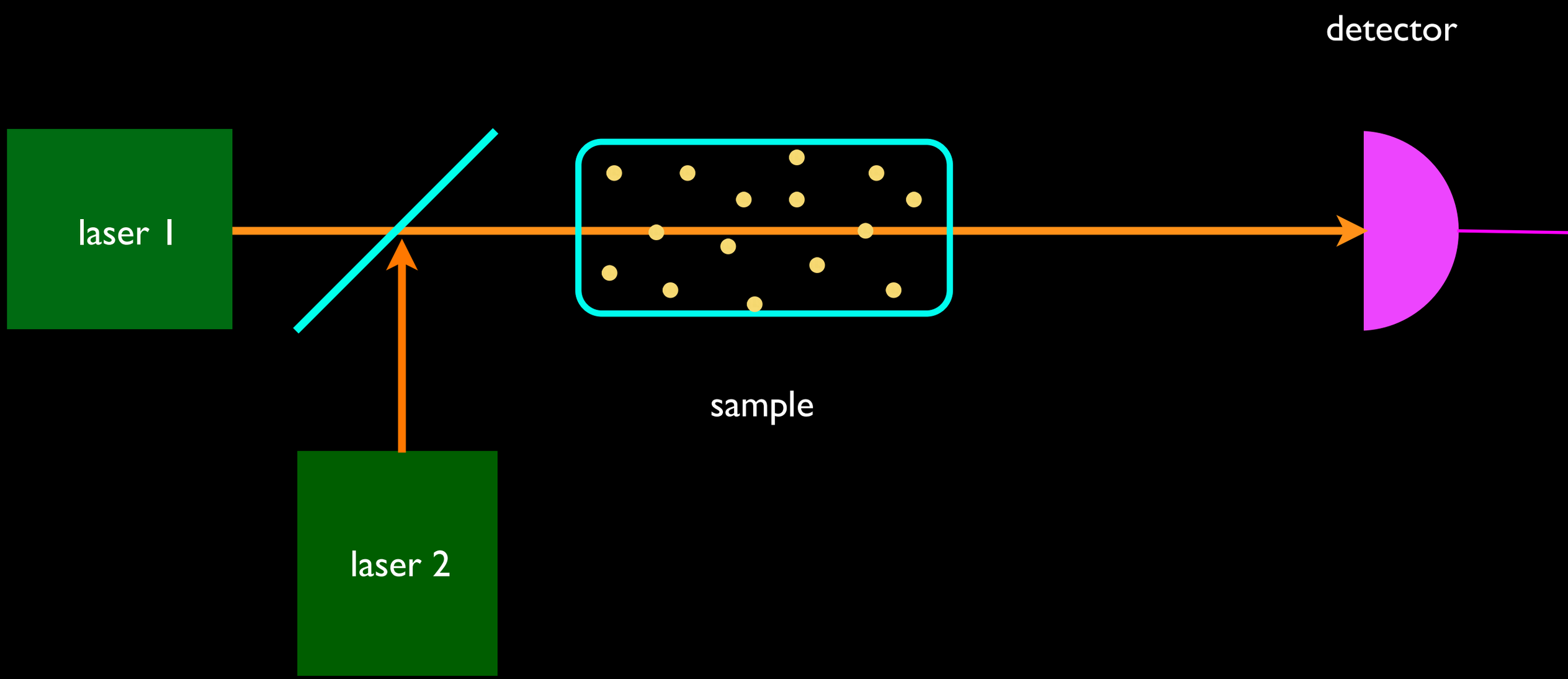
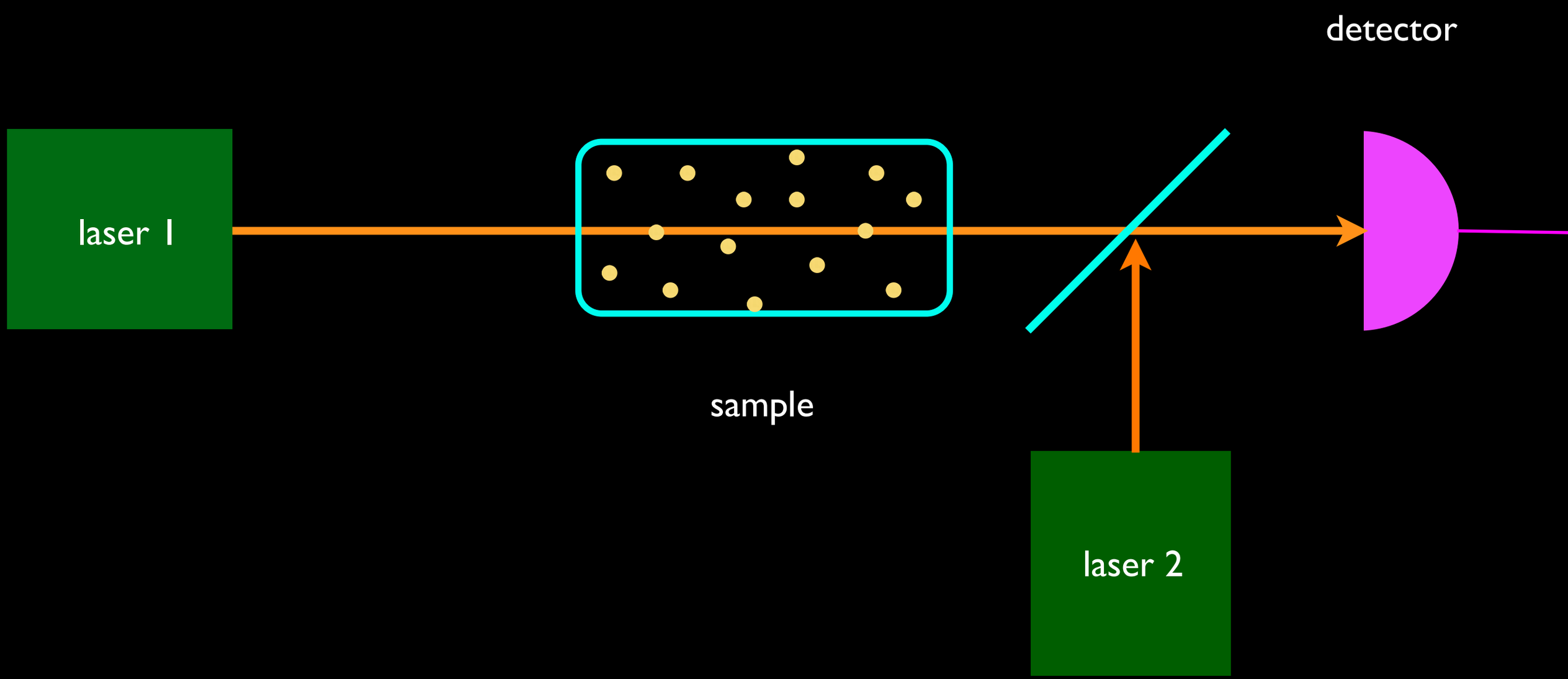


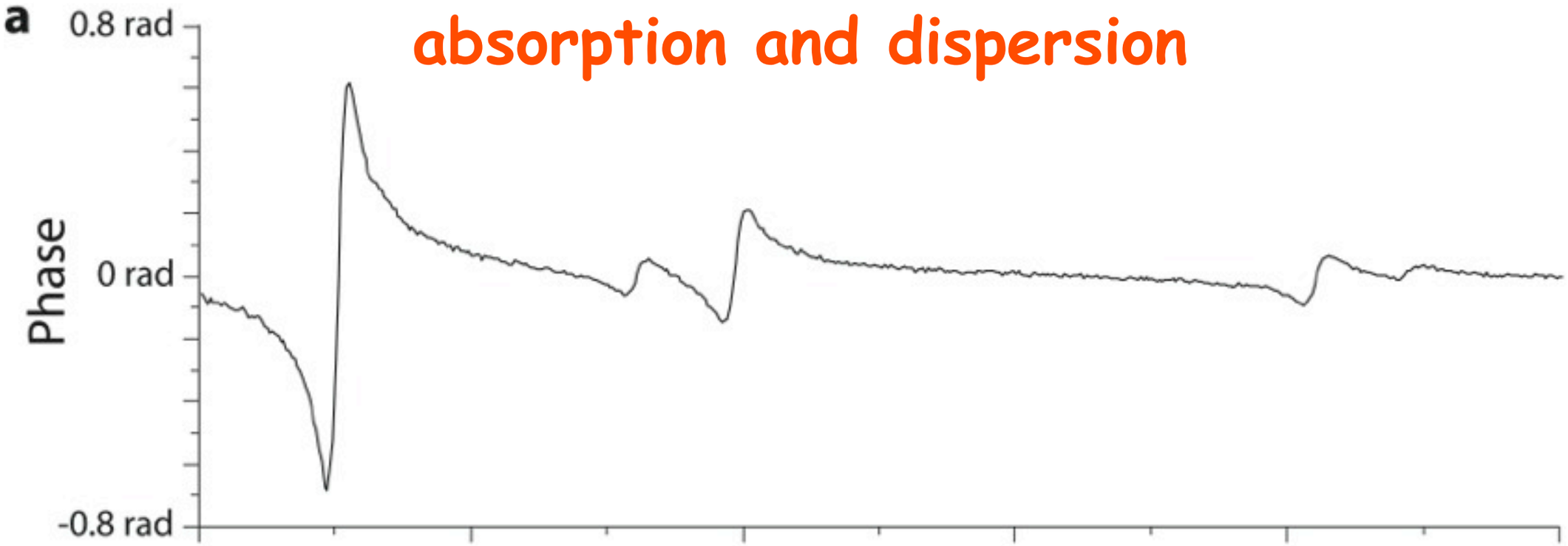
# Fourier Transform Spectroscopy with frequency combs

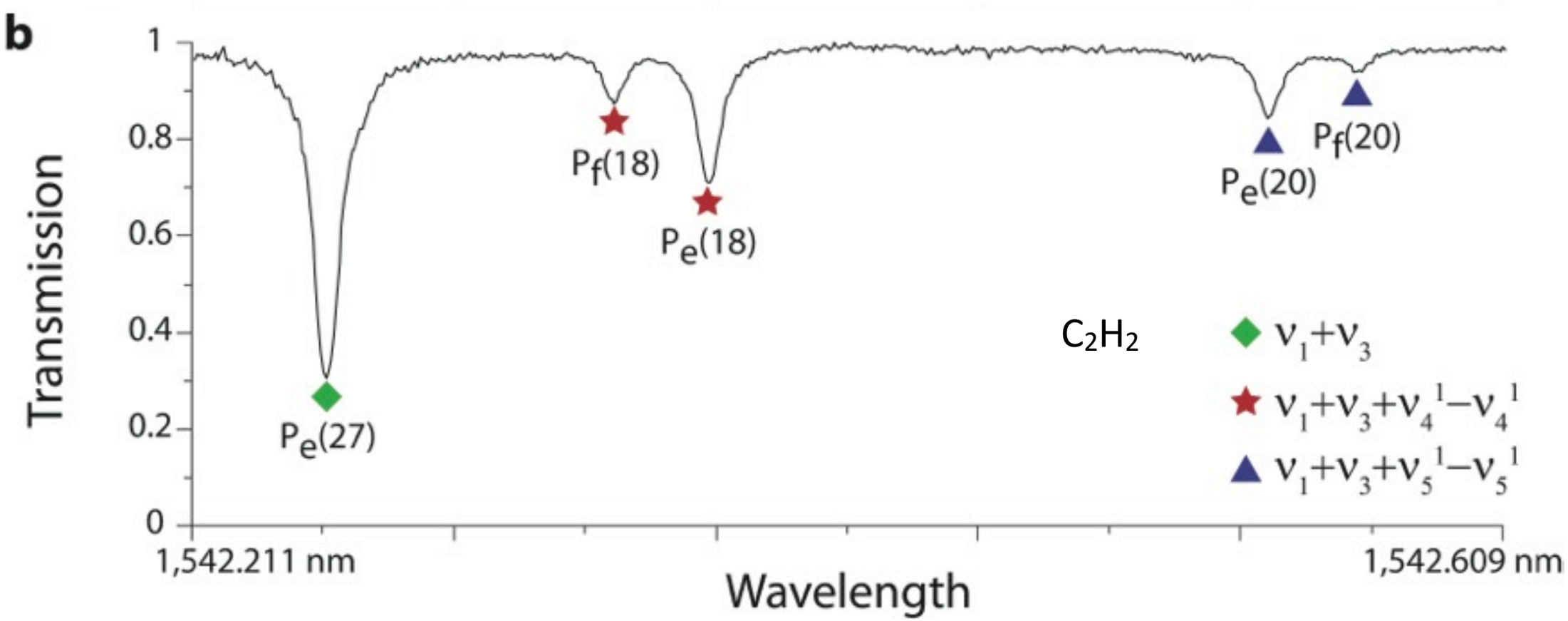


# Fourier Transform Spectroscopy with frequency combs

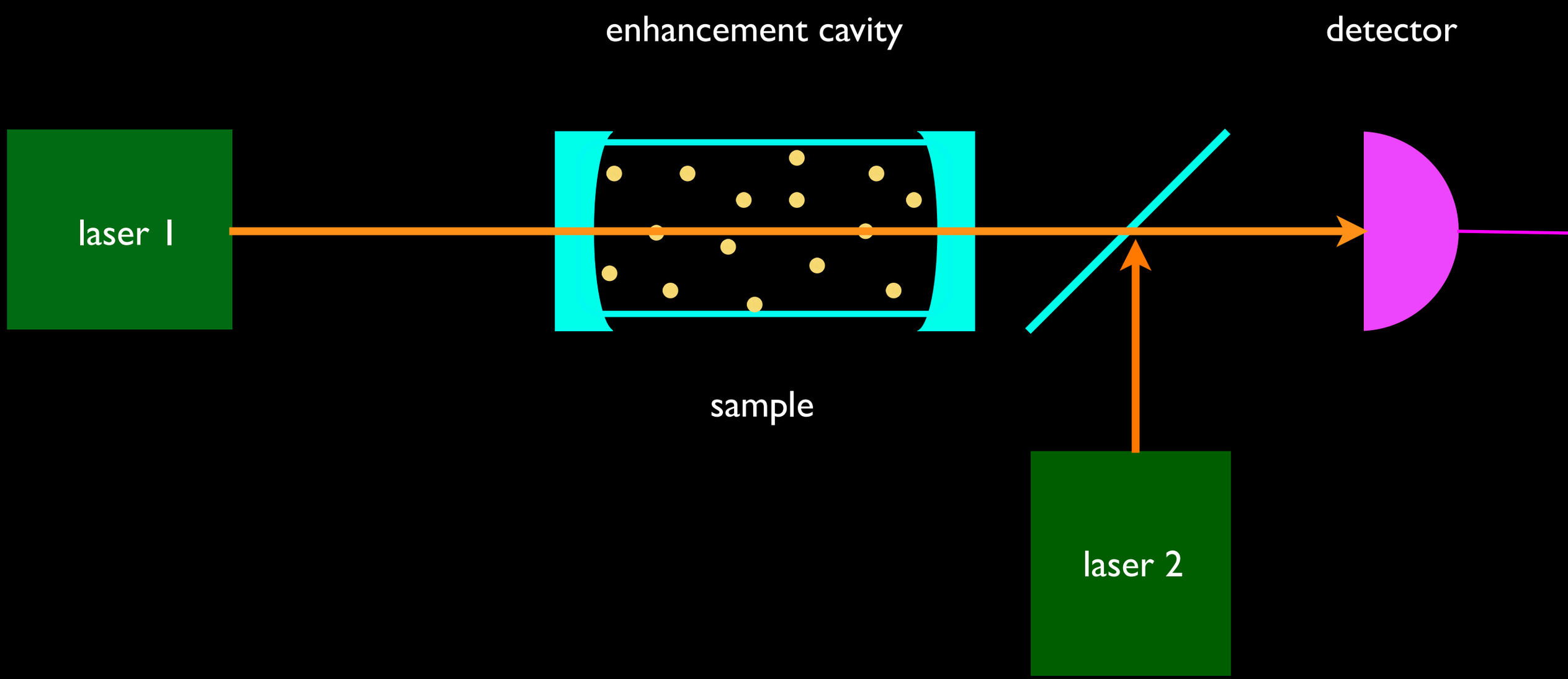


# absorption and dispersion



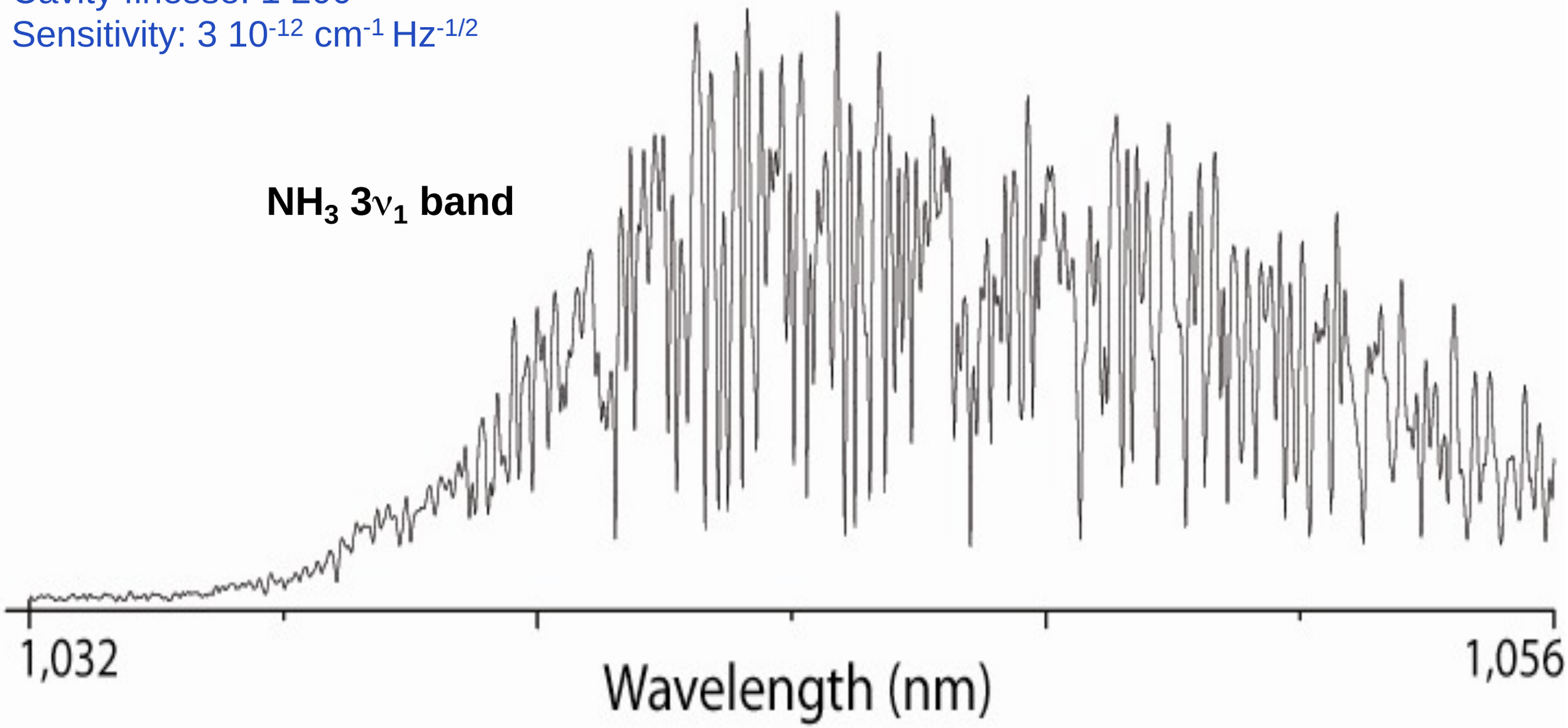



# Fourier Transform Spectroscopy with frequency combs



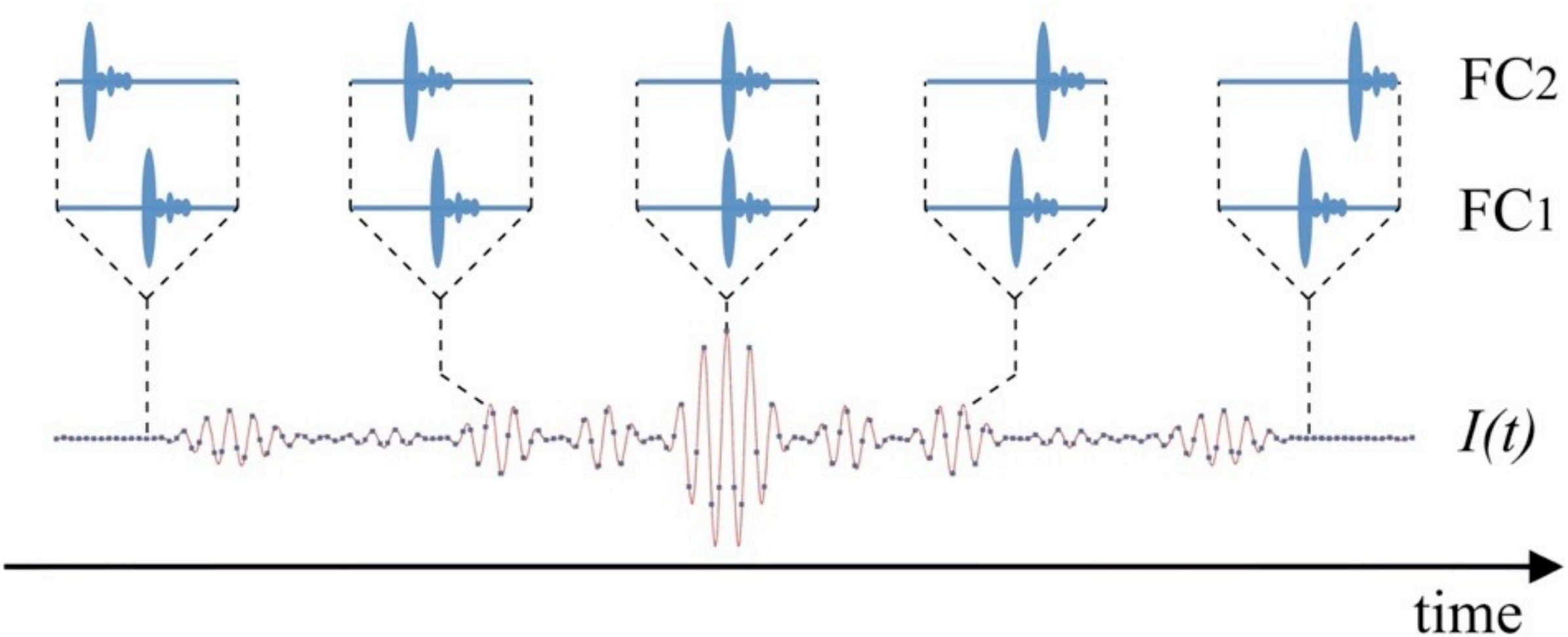
# Cavity-enhanced dual comb spectroscopy

Measurement time: 18 µs
Resolution: 4.5 GHz
Cavity finesse: 1 200
Sensitivity: 3 $10^{-12}$ $cm^{-1}$ $Hz^{-1/2}$

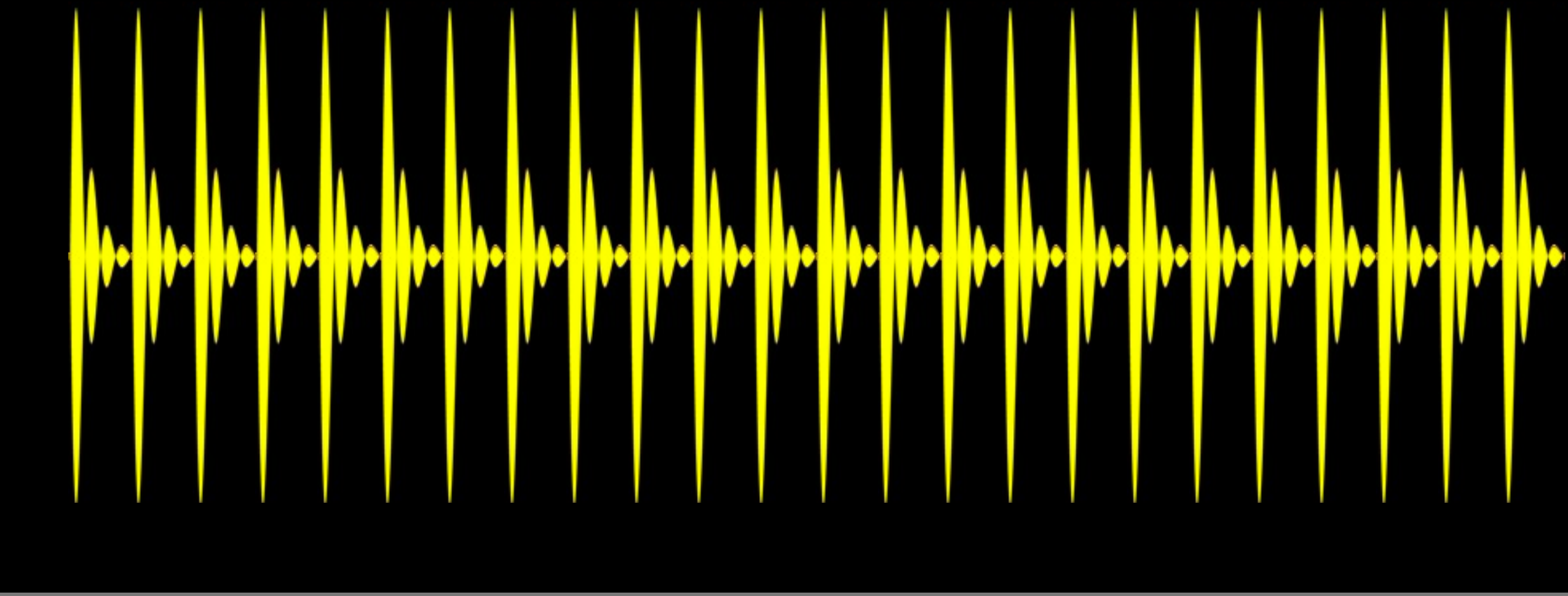



B. Bernhardt et al., Nature Photonics **4**, 55 (2010)

# asynchronous sampling



# repetitive waveform

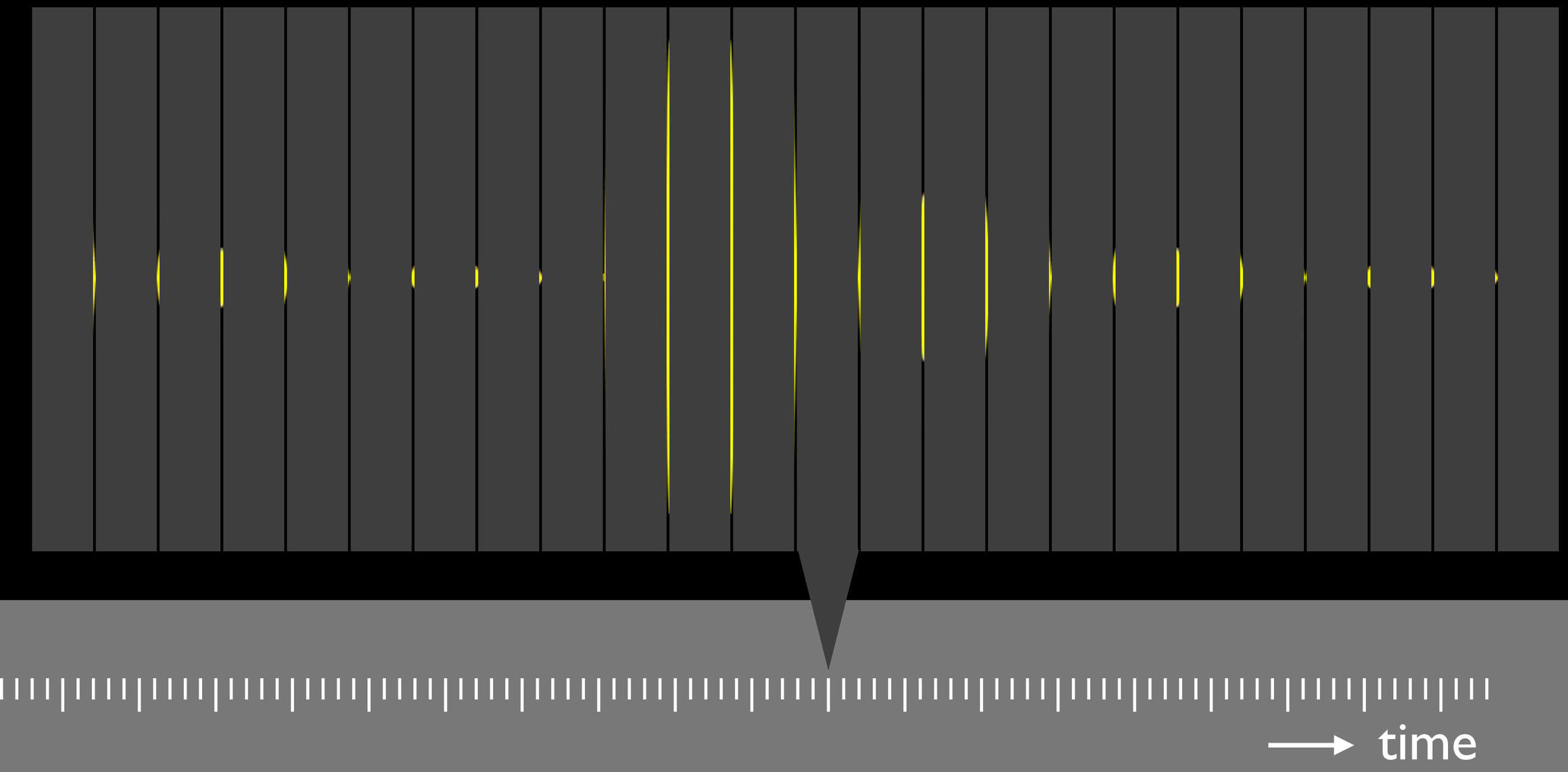


time

repetitive waveform

asynchronously sampled

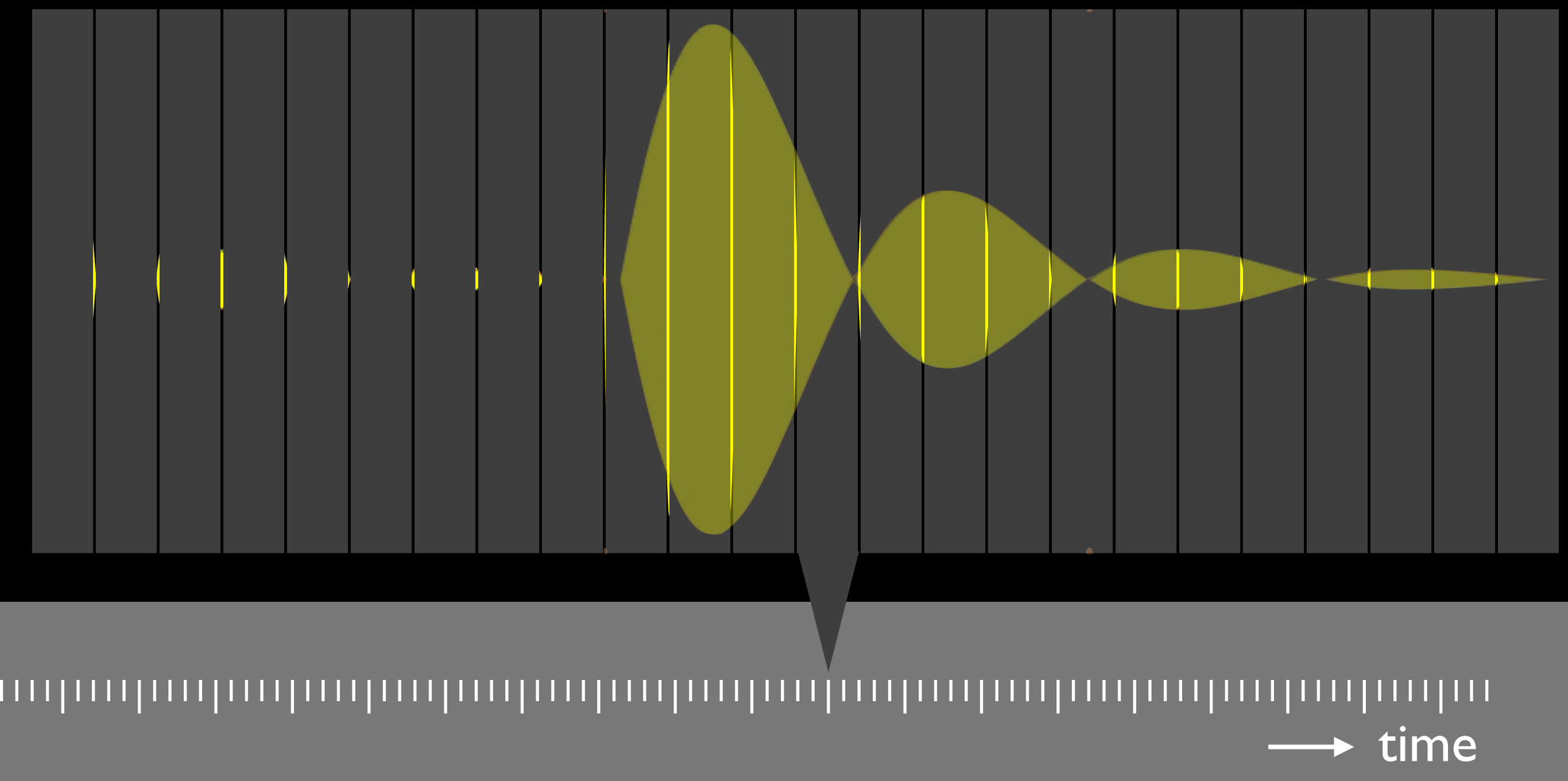
appears stretched in time



repetitive waveform

asynchronously sampled

appears stretched in time

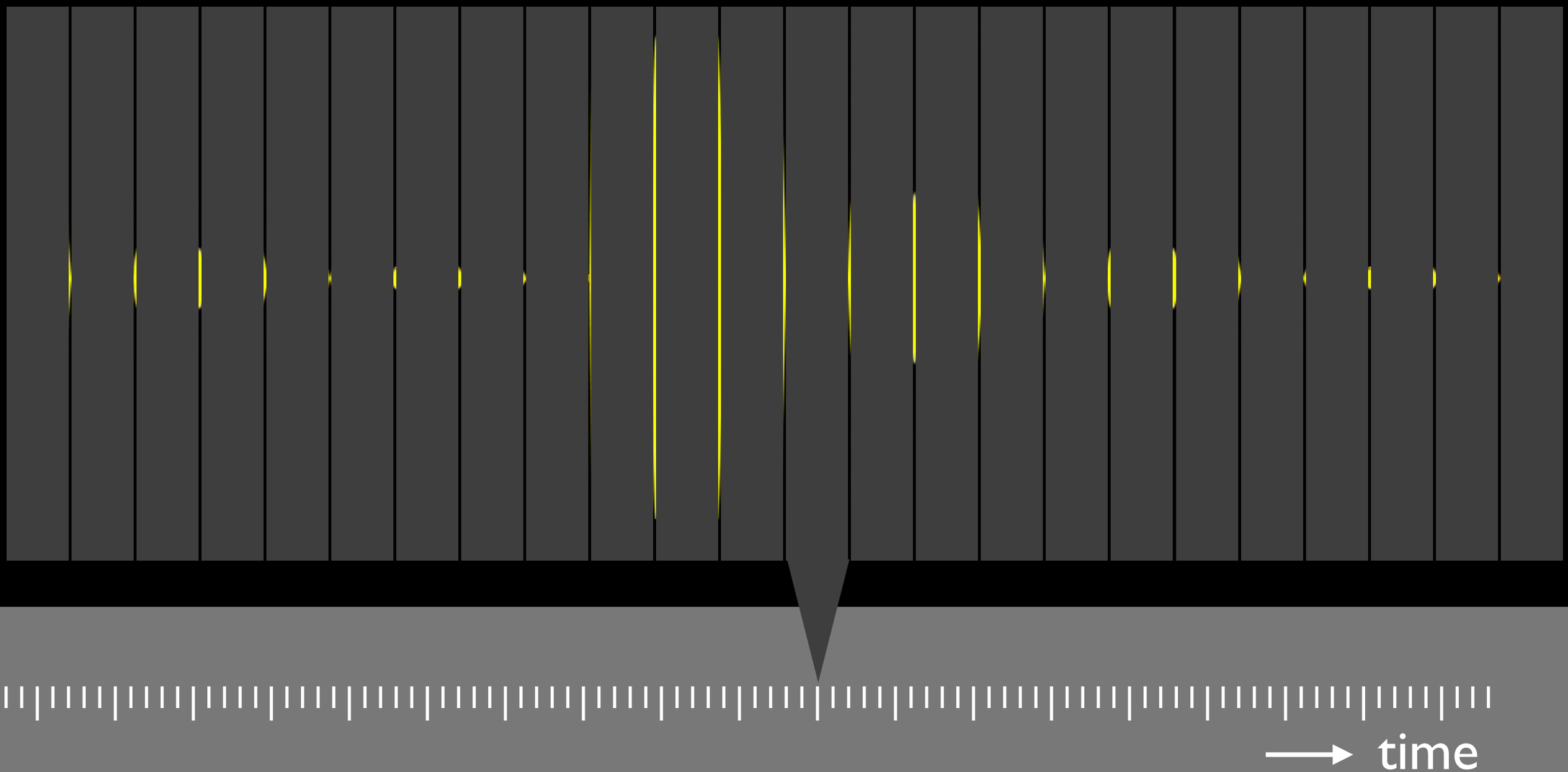


repetitive waveform

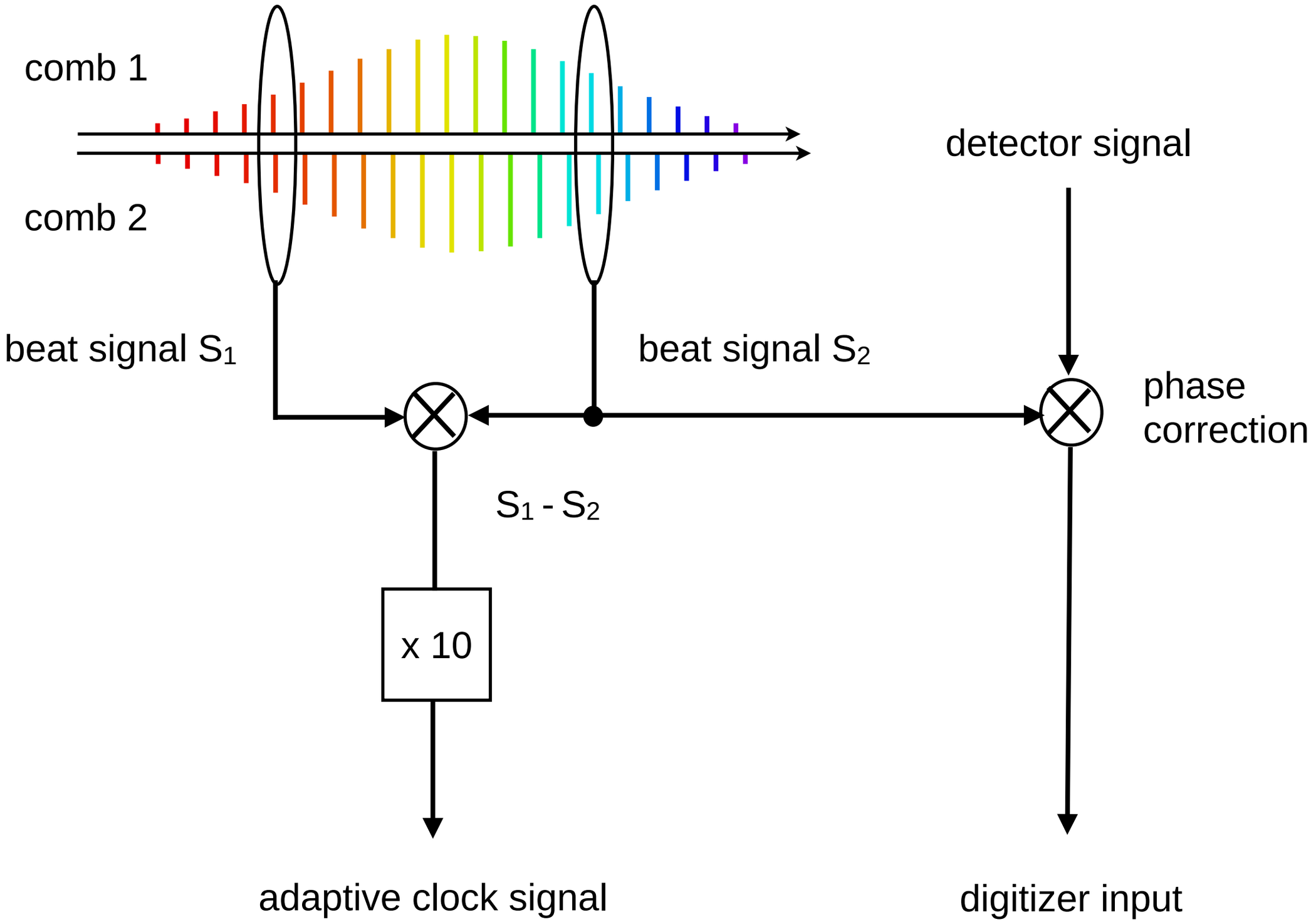
asynchronously sampled

appears stretched in time

timing fluctuations appear magnified



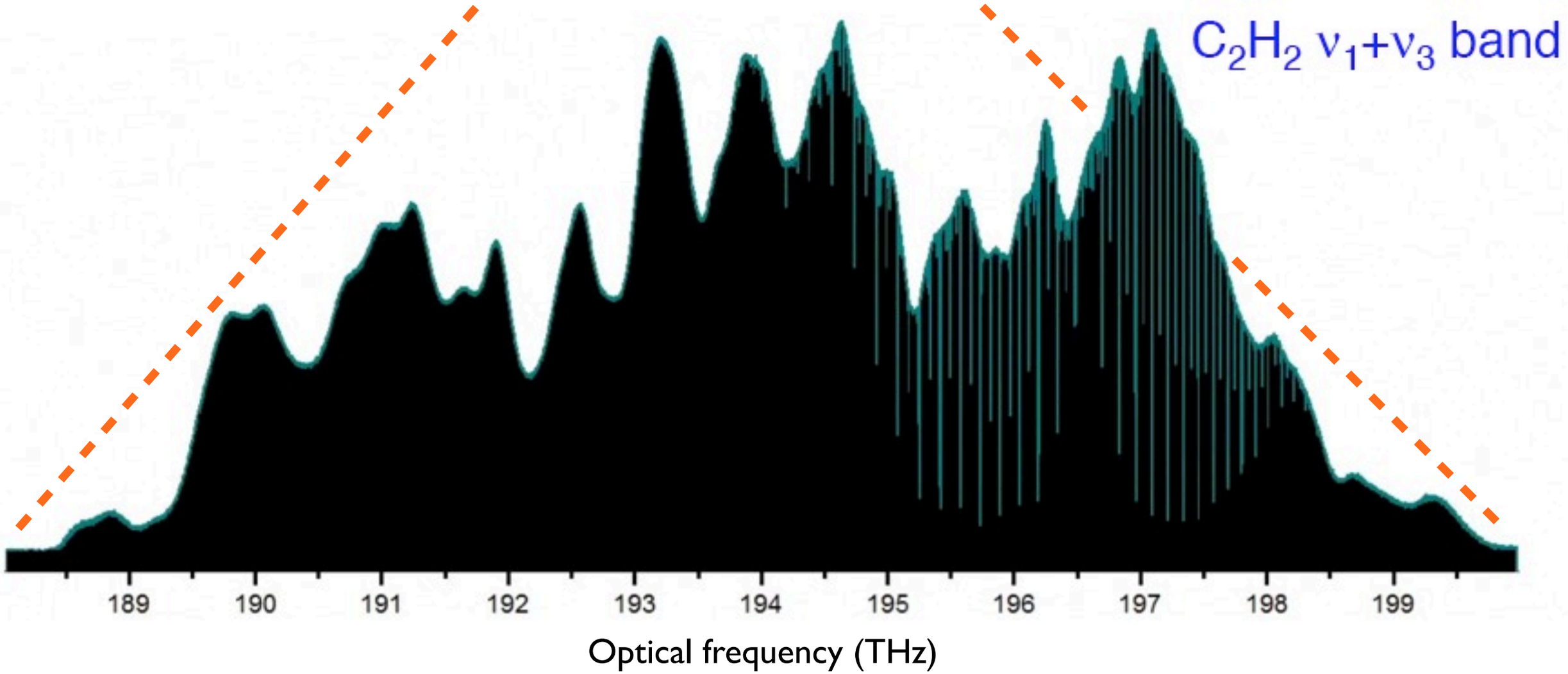
# Adaptive real-time dual-comb spectroscopy



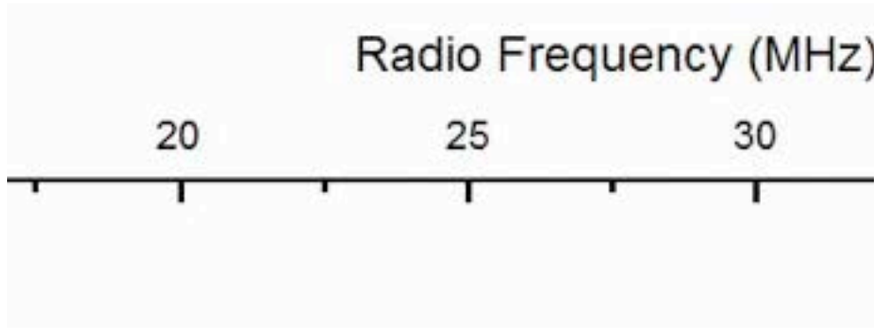

T. Ideguchi et al., Nature Communications 5, 3375 (2014)

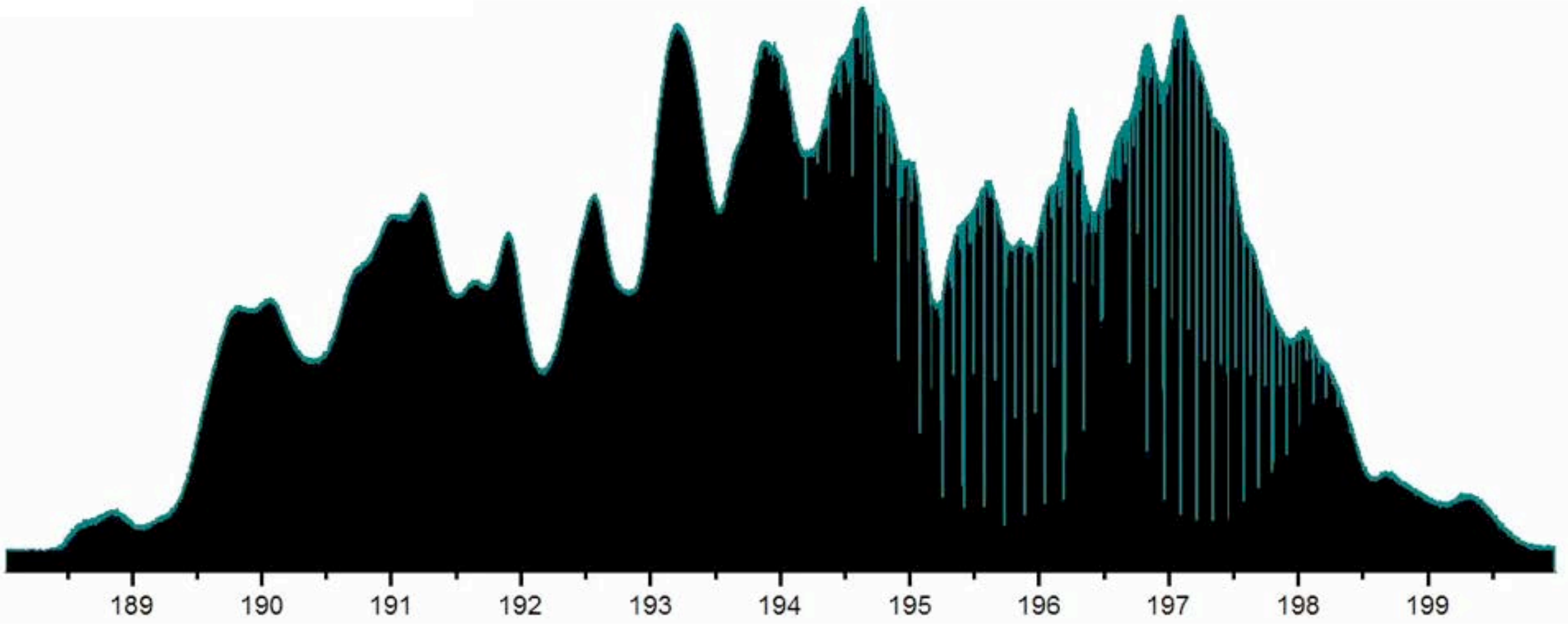
from "Powers of Ten" (YouTube)

Zooming into spectrum with resolved comb lines

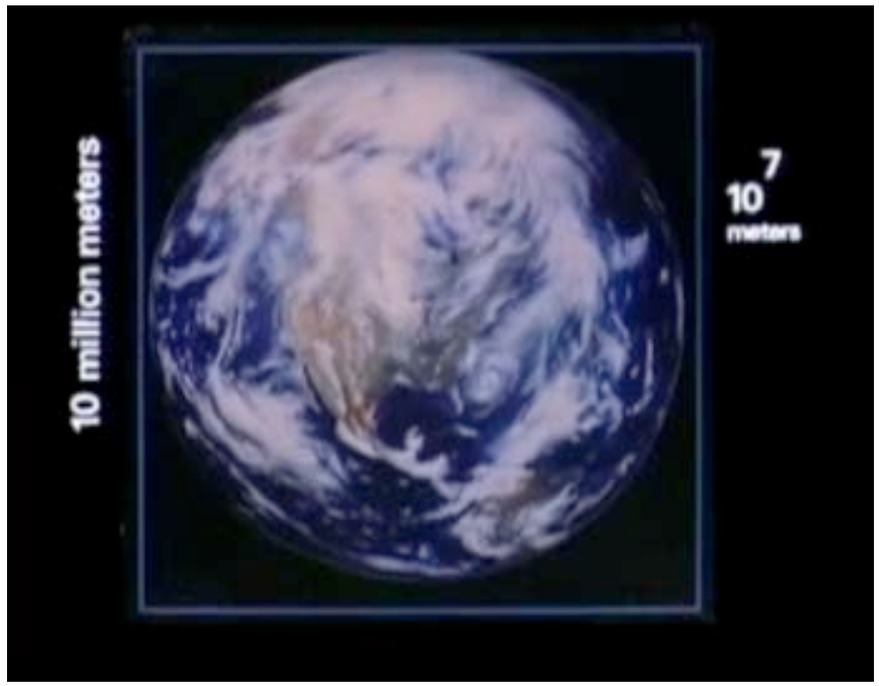



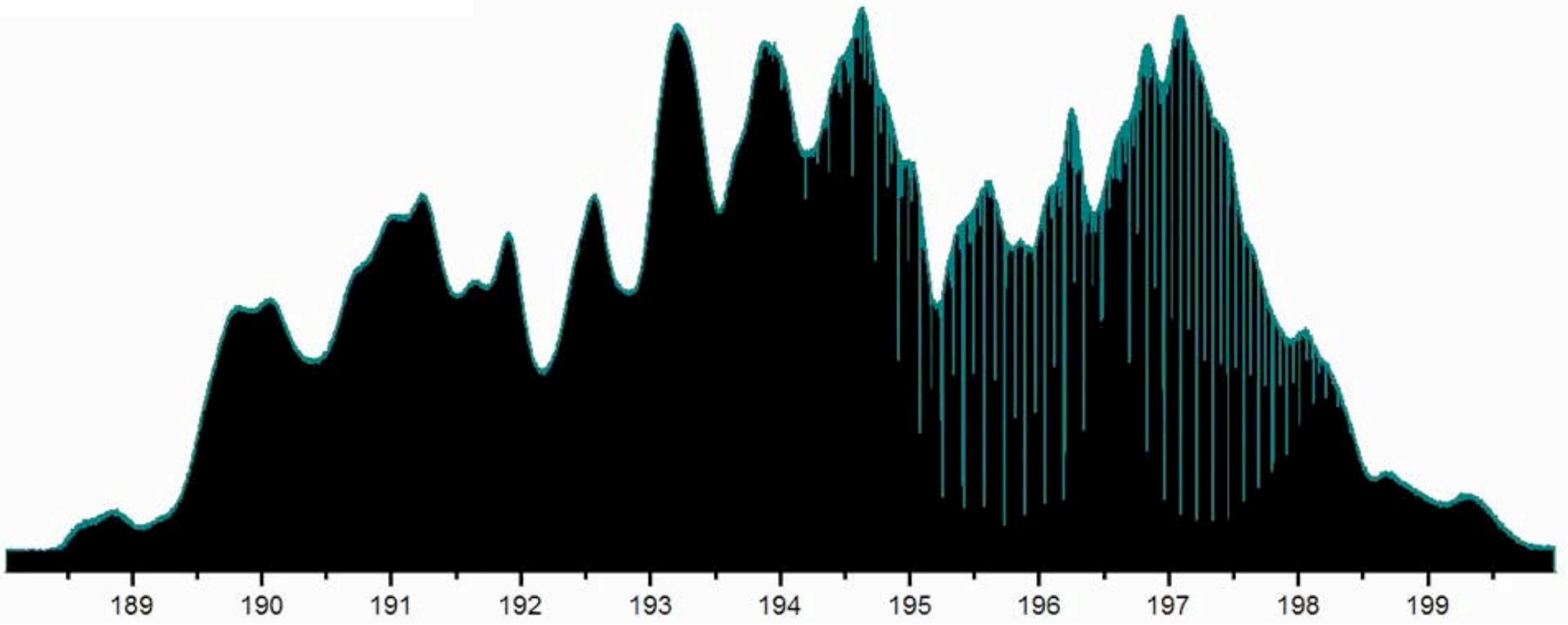
from “Powers of Ten” (YouTube)



from “Powers of Ten” (YouTube)



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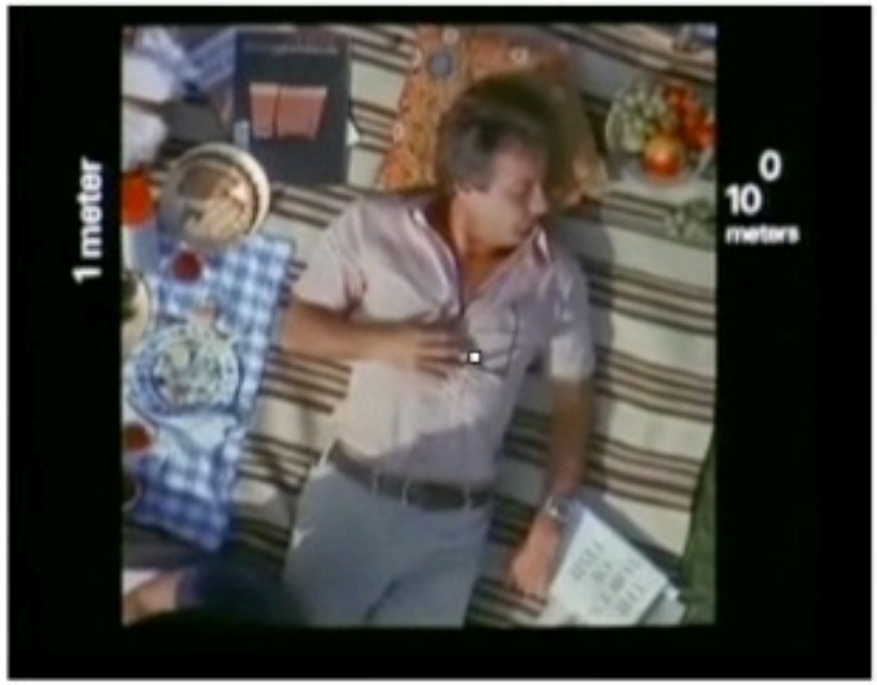
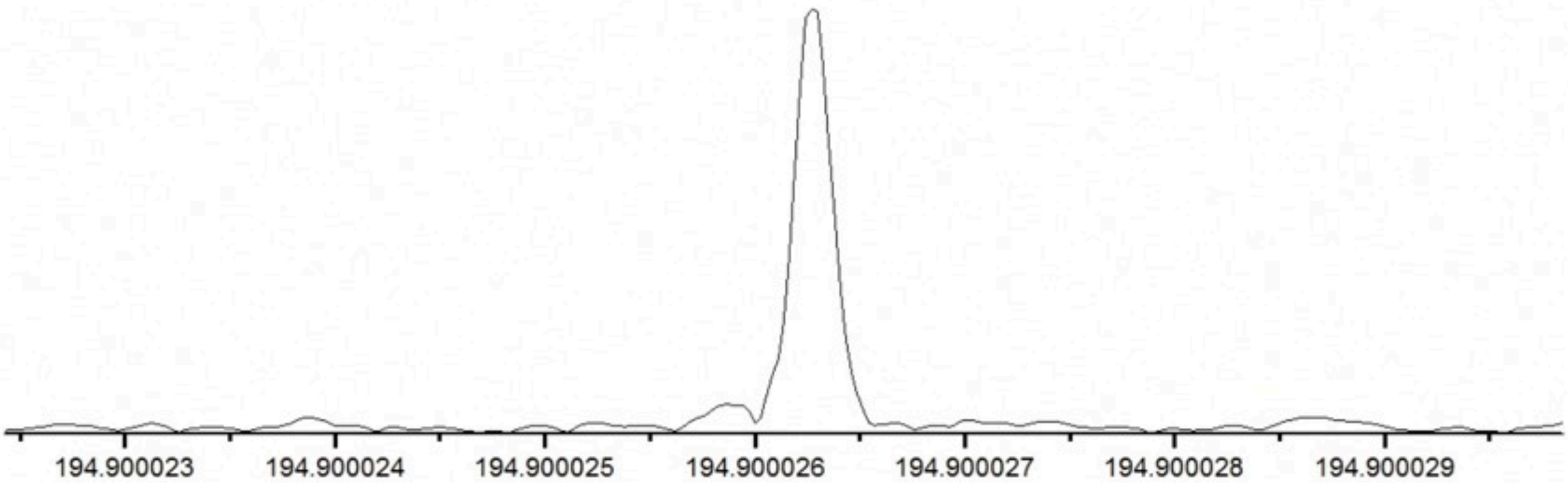
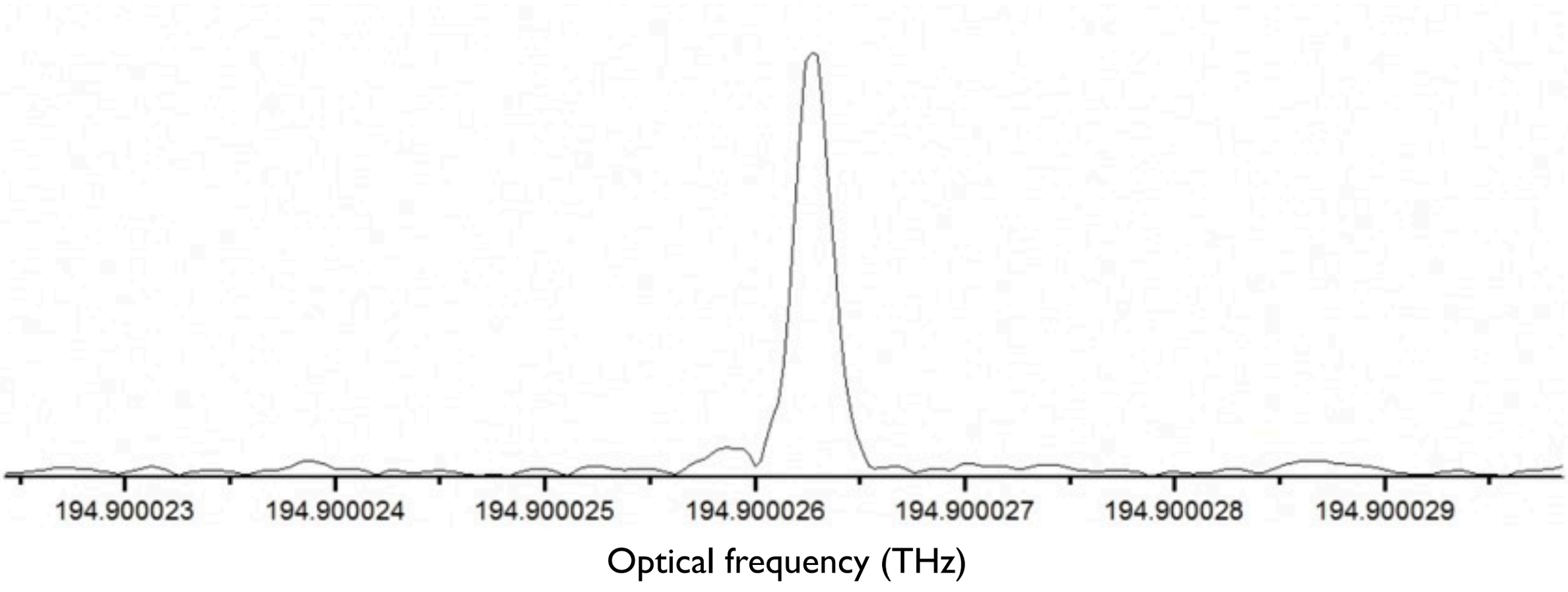



magnification: 2 000 000 x



| | |
|---|---|
| recording time: | 2.7 s |
| data points: | 268 000 000 |
| number of resolved comb lines: | 120 000 |
| resolution in the optical domain: | 202 kHz |

magnification: 2 000 000 x



# Dual comb spectroscopy

you need

- Two frequency comb sources
- A single fast photodetector
- A computer

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- Two frequency comb sources
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- A computer

you get

- Very short acquisition time
- Extreme sensitivity
- From low to extreme resolution
- Extreme accuracies
- Absorption and dispersion
- Spectral extension from THz to VUV

# Nonlinear dual comb spectroscopy

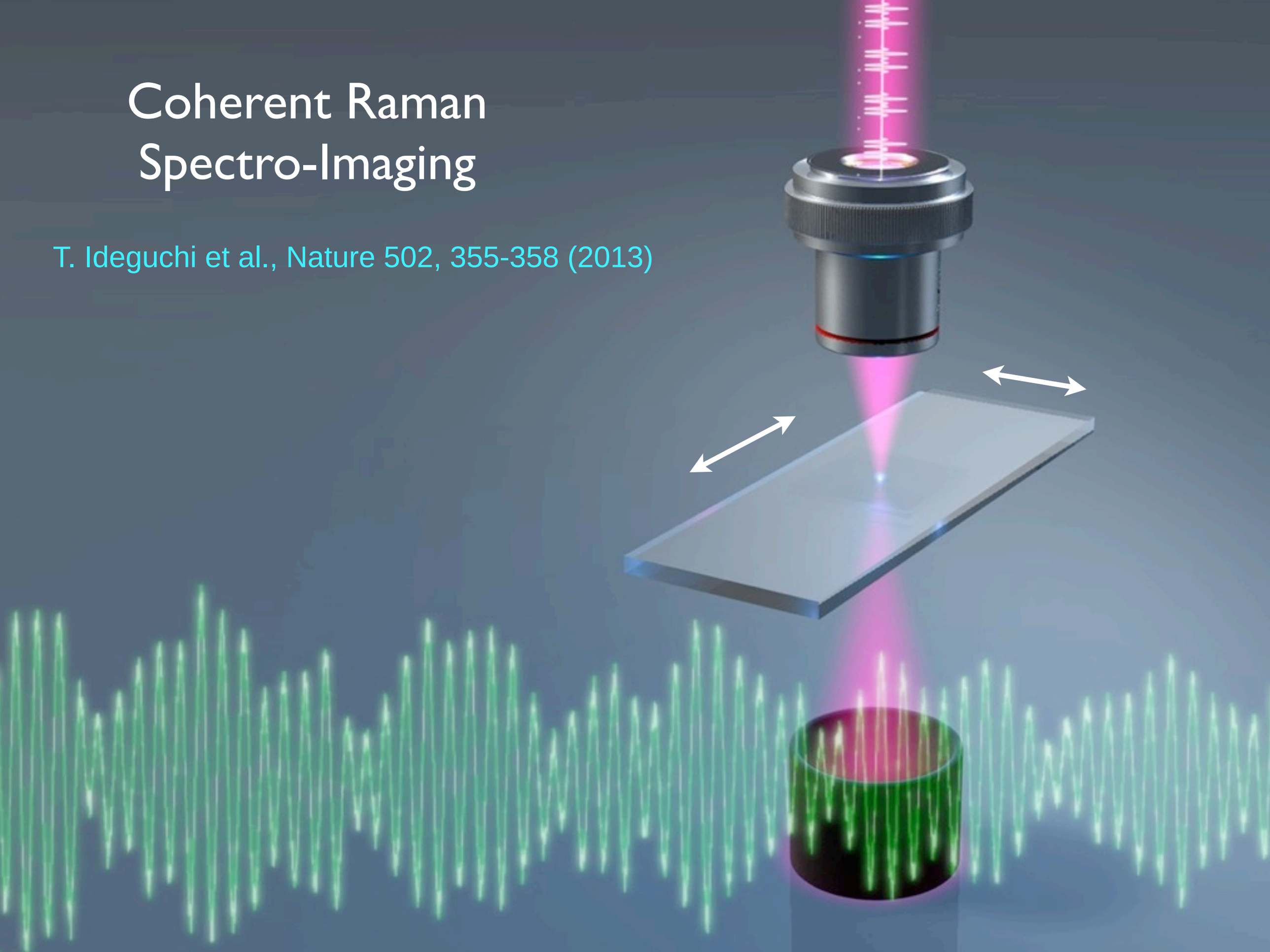
recently demonstrated:

two-photon spectroscopy
stimulated Raman spectroscopy
coherent Raman spectro-imaging

T. Ideguchi et al. Opt. Lett. 37, 4498-4500 (2012)
T. Ideguchi et al., Nature 502, 355-355 (2013)
A. Hipke at al., arXiv:1311.6138 (2013)
T. Ideguchi et al. arXiv:1403.3814 (2014)

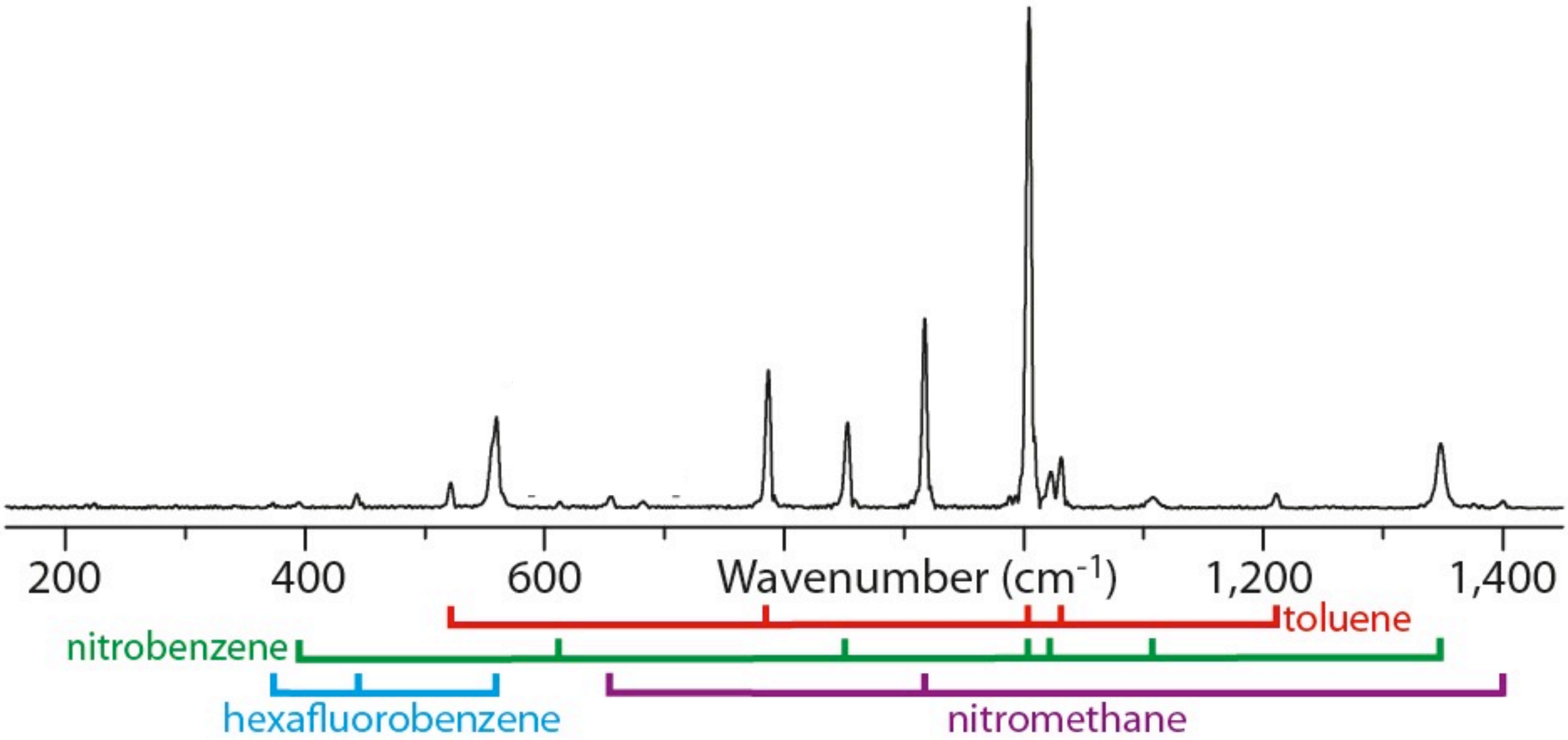
# Coherent Raman Spectro-Imaging

T. Ideguchi et al., Nature 502, 355-358 (2013)



# Dual-comb coherent anti-Stokes Raman spectroscopy

Measurement time: 295 μs
Resolution: 4 $cm^{-1}$



# Frequency combs - evolutionary tree

trace gas sensing

XUV comb and harmonic generation

extrasolar planet searches

dual comb spectroscopy

coherent control

wave form synthesis

continuing expansion of universe ?

calibration of astronomical spectrographs

frequency comb spectroscopy

low-noise frequency synthesis

optical clocks

tests of fundamental physics

attosecond science

carrier-envelope phase control

self-referenced octave spanning frequency comb

frequency measurements

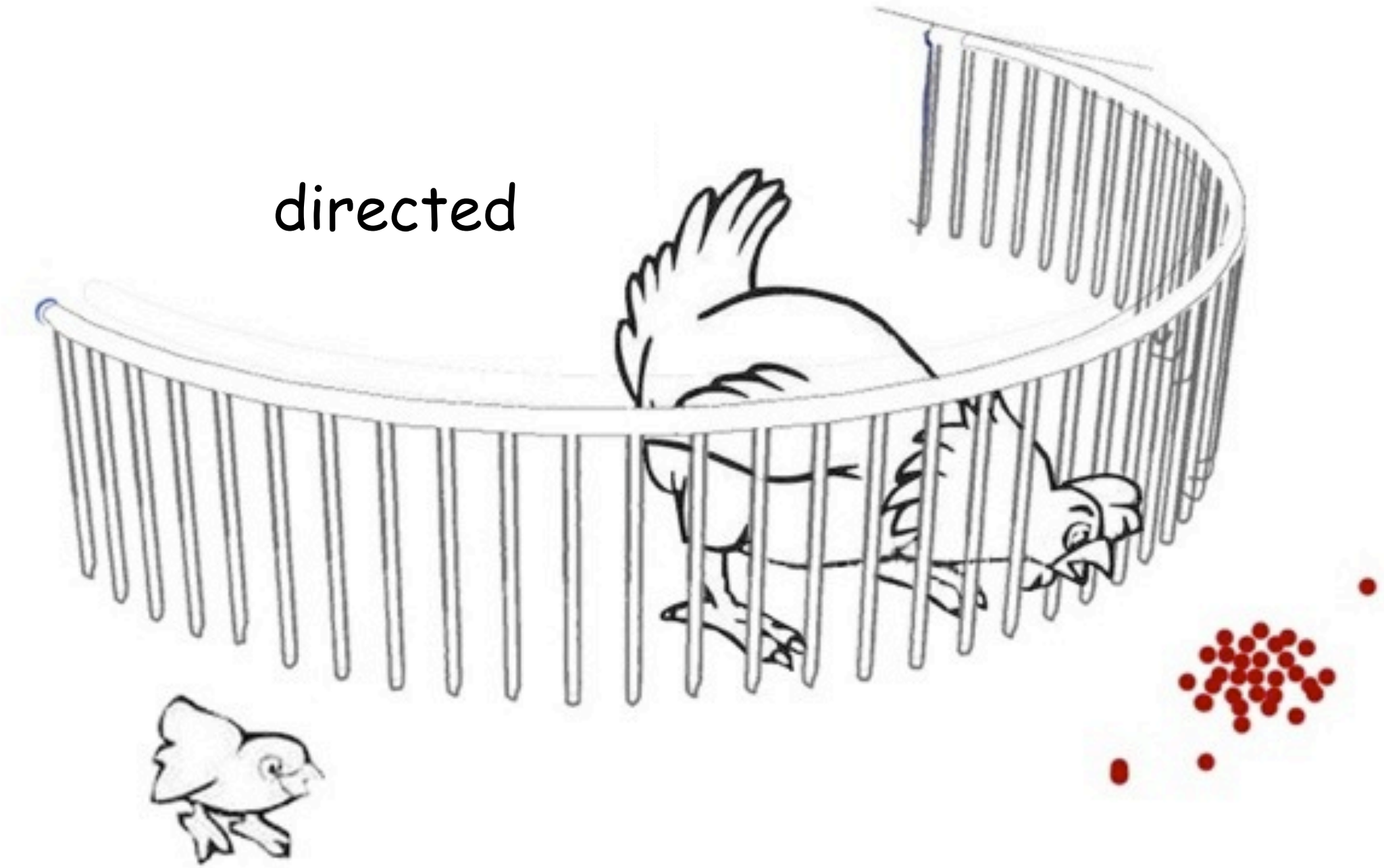
length measurements

time - frequency transfer

mode-locked femtosecond laser

curiosity driven research

directed



curious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# The End