



State of progress in THz and broadband spectrum manipulation group

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**Minoshima ERATO 2nd group meeting in 2014
(Dec. 22, 2014@AIST)**

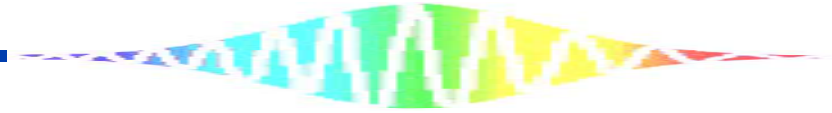
Research topics

(1) Intelligent THz instrumentation and application

- Adaptive sampling dual THz comb spectroscopy (M2)
- Broadband THz synthesizer (M2)
- THz-comb-referenced spectrum analyzer (M2)
- THz digital holography (B4, just start)
- THz ghost imaging (M1, not start)

(2) Novel optical comb measurement and application

- Full-field confocal optical-comb microscopy (M2)
- Discrete Fourier transform infrared spectroscopy (Hsieh)
(in collaboration with AIST)
- CARS gas analysis (Harsono, not start)
- Sensing comb for photoacoustic imaging (M1, just start)



(1) Adaptive sampling dual THz comb spectroscopy

THz gas spectroscopy

(1) Rotational transitions of polar molecules

- ✓ Rich spectral fingerprints
- ✓ High selectivity and high sensitivity
- ✓ High discrimination at low pressure due to narrow Doppler linewidth ($\sim 1\text{MHz}$)

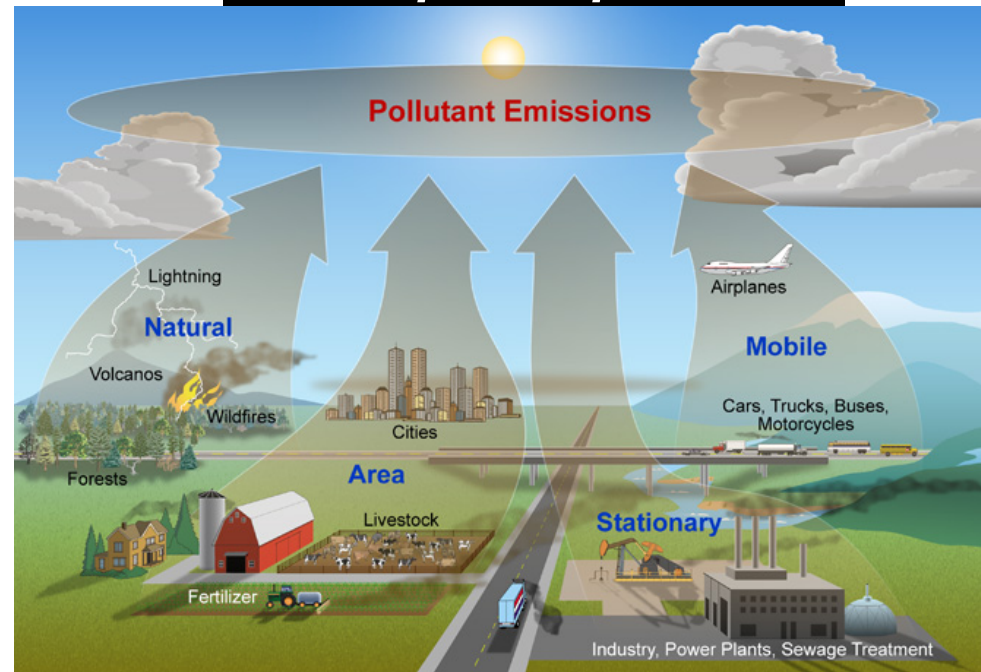
Atmospheric pollution

☞ **Multiple gas analysis**

(2) Less scattering

- ✓ $\lambda_{\text{THz}} \gg$ particle diameter

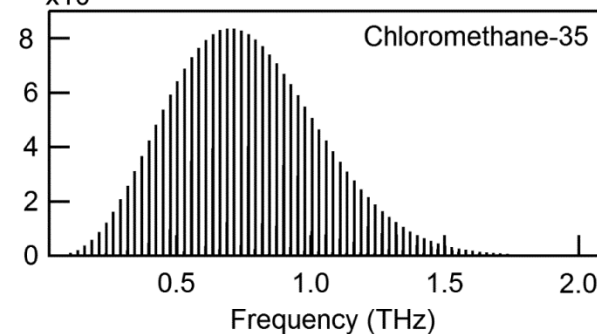
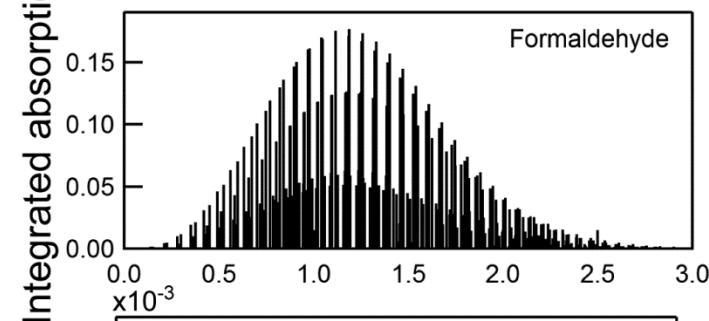
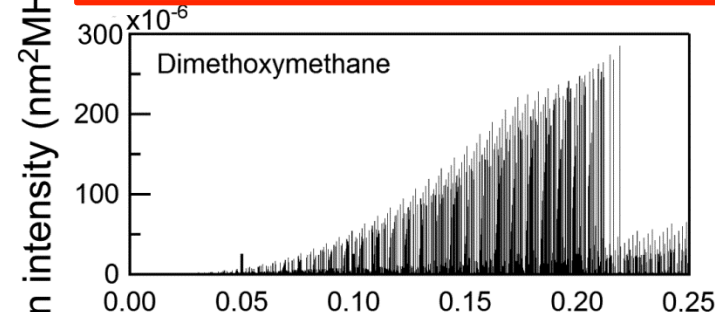
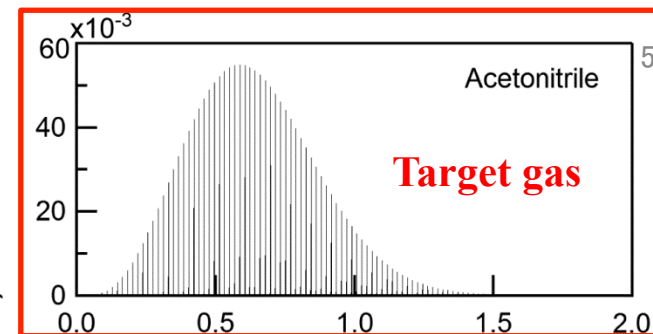
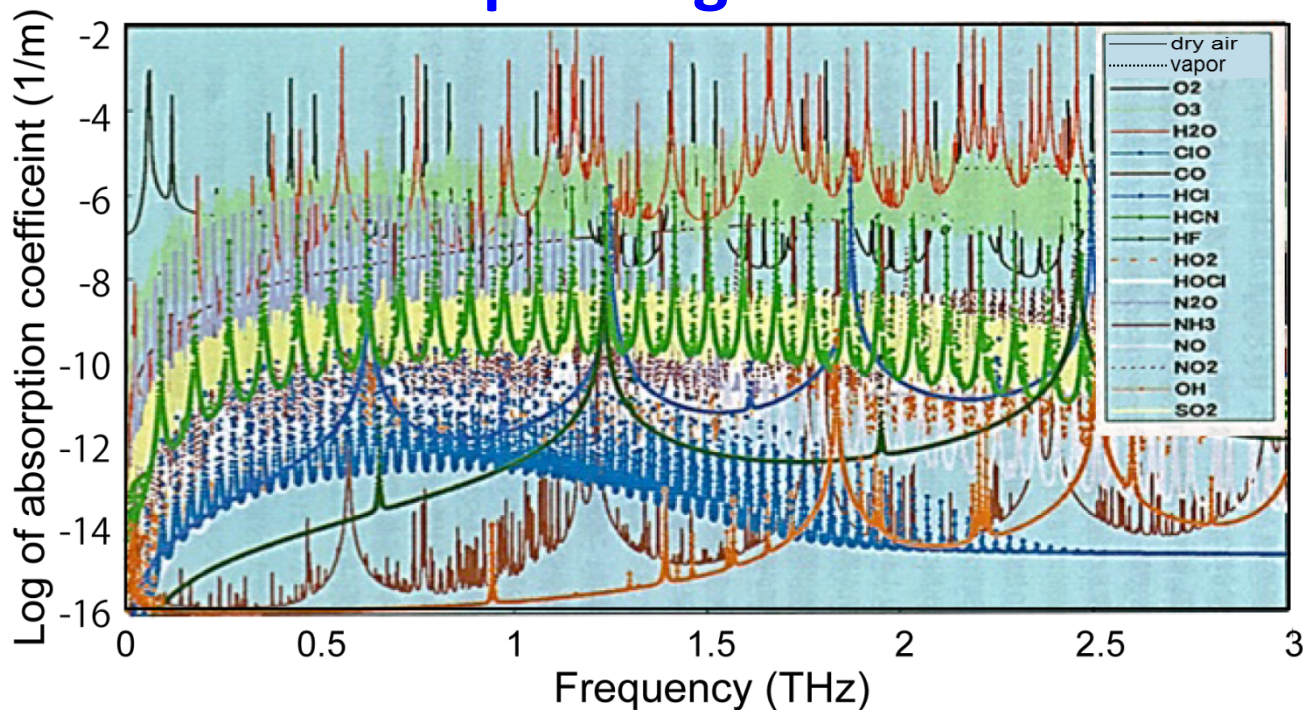
☞ **Possible to analyze gas mixed with aerosols (fog, cloud, smoke, soot, etc)**



Ref) <http://www.nature.nps.gov/air/aqbasics/sources.cfm>

THz spectral fingerprints

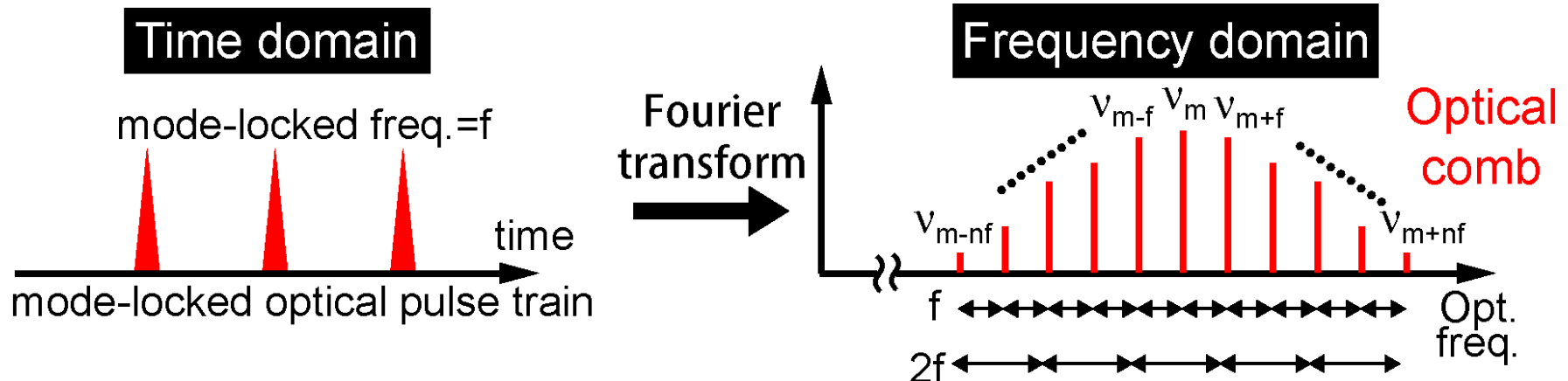
Atmospheric gas molecule



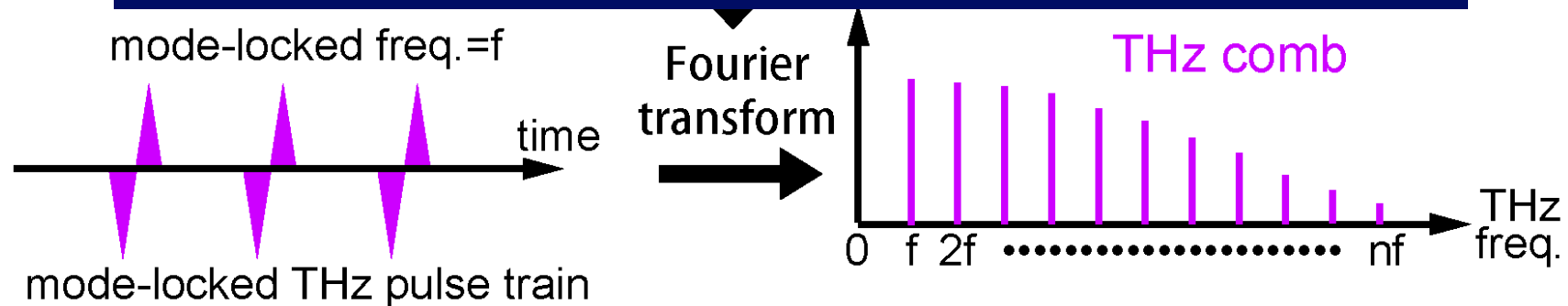
Volatile organic compound (VOC) gas

To discriminate the target gas correctly, high resolution, high accuracy, and broadband spectrum are required!!

Optical comb and THz comb



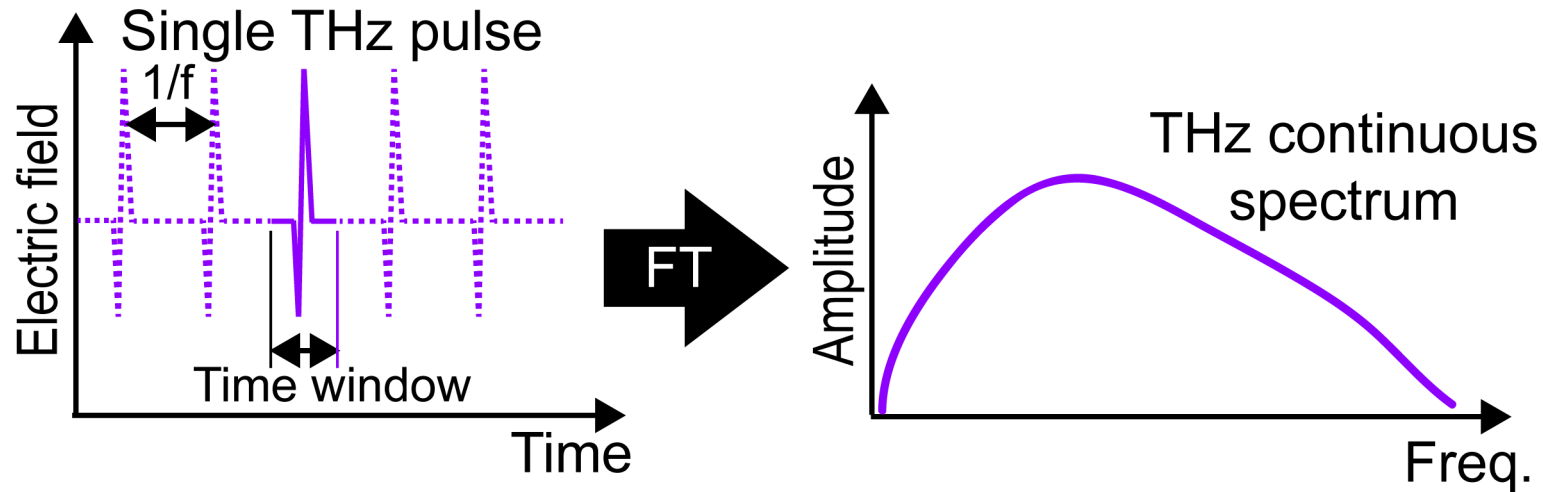
*Precise frequency marker
for broadband THz spectrum*



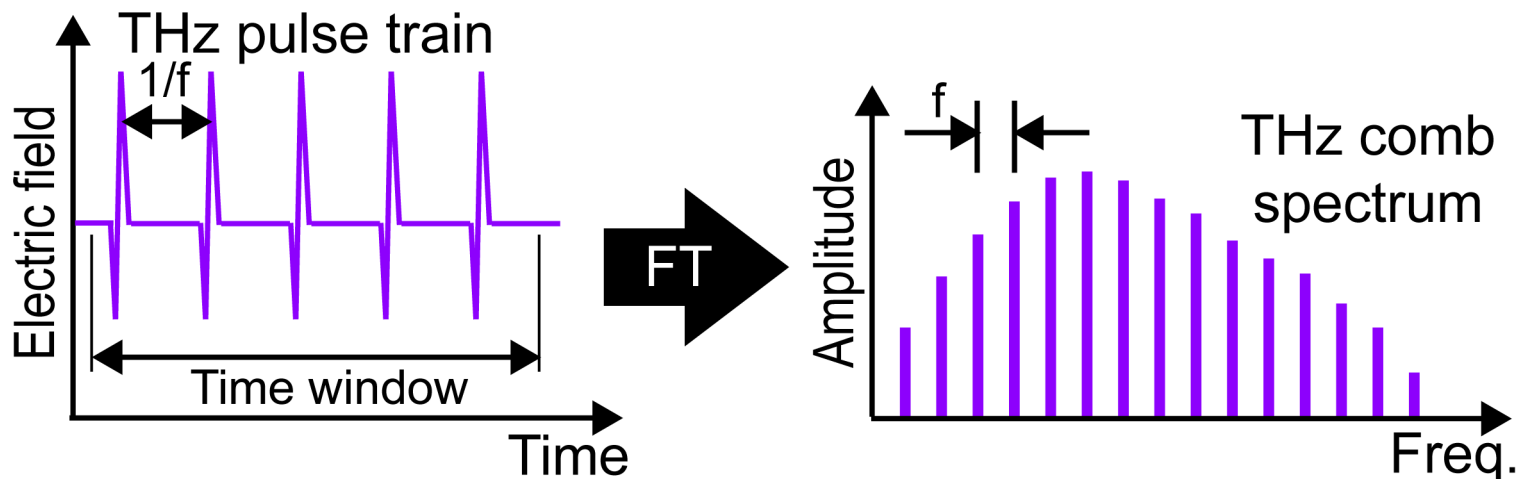
Simple, broadband selectivity, high spectral purity,
and absolute frequency calibration

How to measure mode-resolved THz comb spectrum

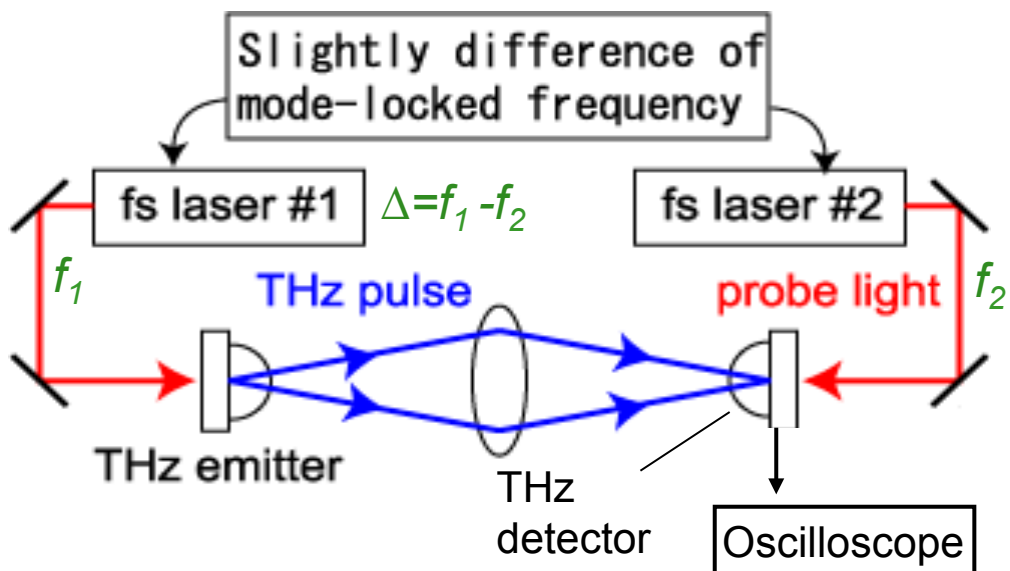
Traditional THz-TDS equipped with mechanical time-delay scanning



Time-window-extended THz-TDS



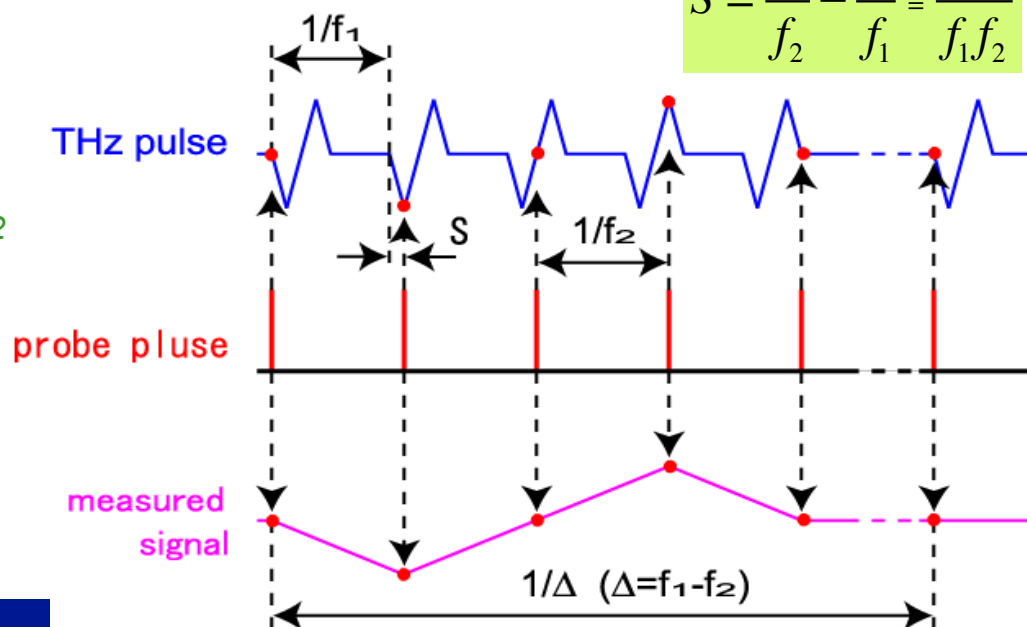
Asynchronous-optical-sampling THz-TDS (ASOPS-THz-TDS)



ref) T. Yasui, *Appl. Phys. Lett.* 87, 061101 (2005).

Overlap timing between THz and probe pulses is automatically shifted every pulse

$$S = \frac{1}{f_2} - \frac{1}{f_1} = \frac{\Delta}{f_1 f_2}$$

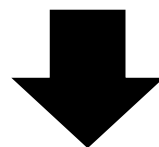


Time scale of ps THz pulse is linearly expanded to μ s order

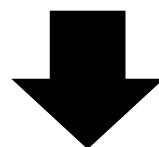
Temporal magnification factor (TMF) = $f_1/\Delta f$

- No need for mechanical time-delay scanning
- No limitation for size of time window

Use of **free-running**, dual fs lasers
in dual THz comb spectroscopy

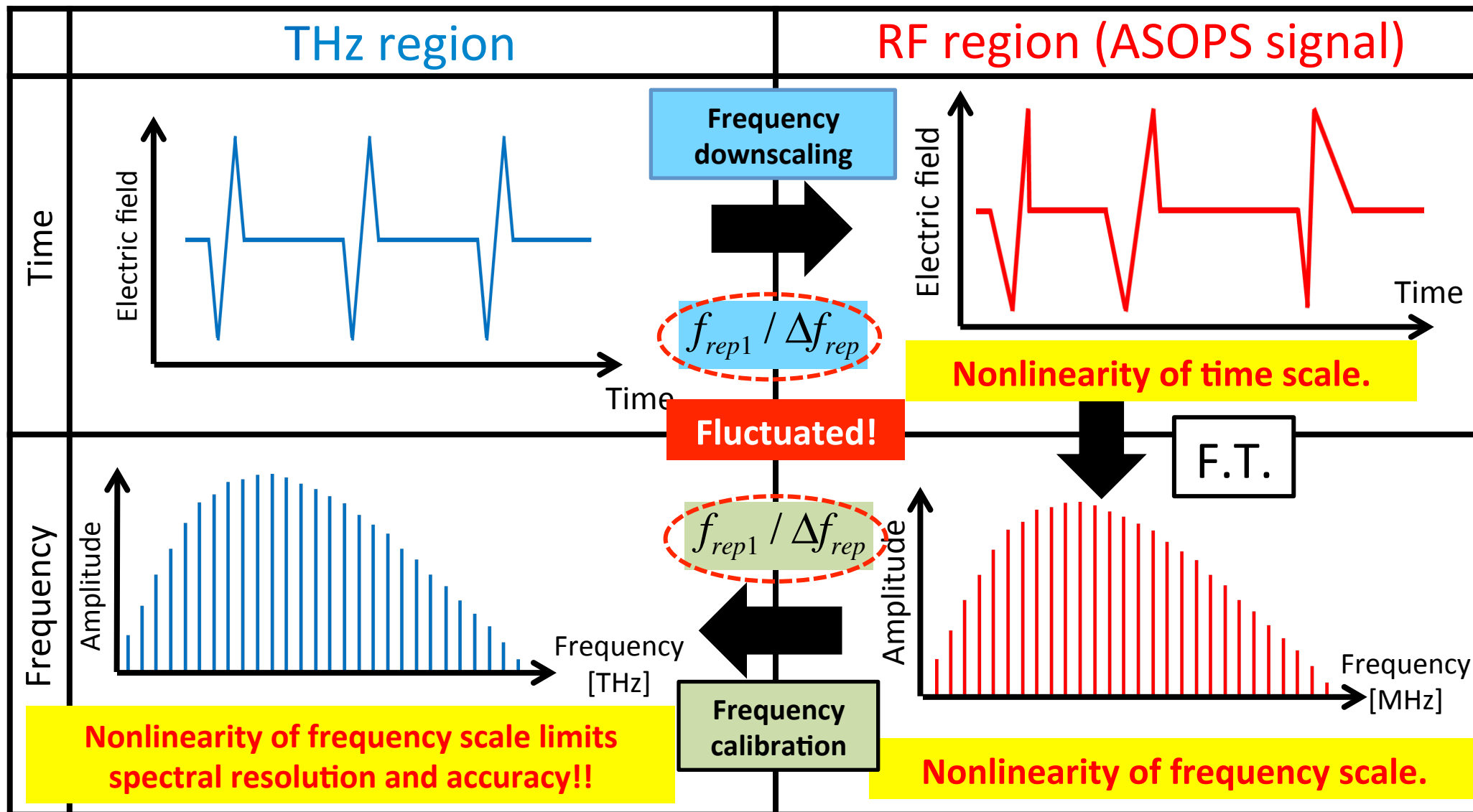


**Expand the application fields of
dual THz comb spectroscopy**



However, timing jitter between free-running dual fs lasers distorts the linearity of time and frequency scales due to fluctuation of TMF!

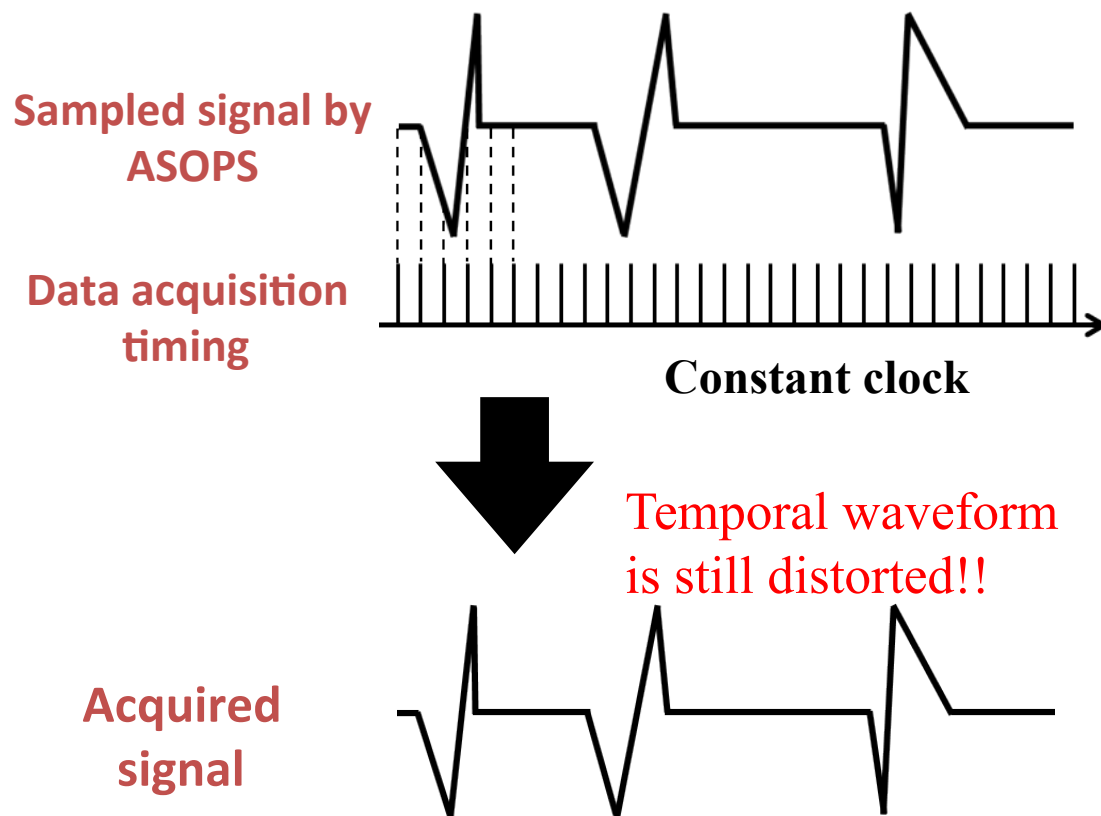
Influence of timing jitter in ASOPS



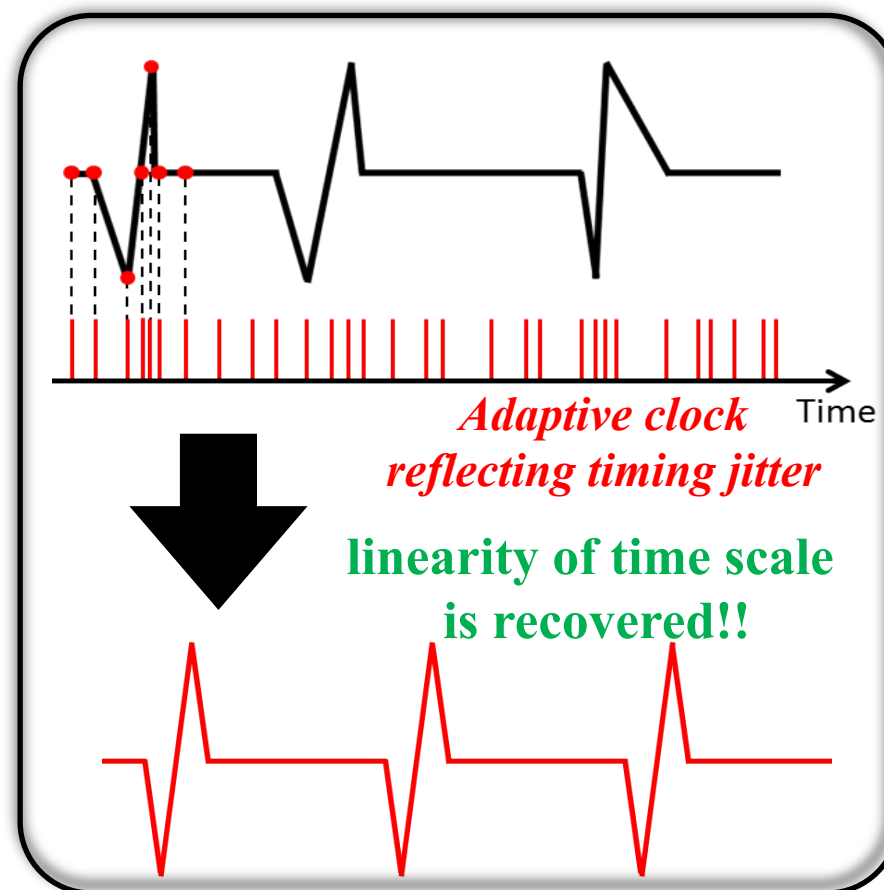
Adaptive sampling method

Ref) T. Ideguchi, Nat. Comm., 5, 3375 (2014).

**Conventional method
(constant sampling)**

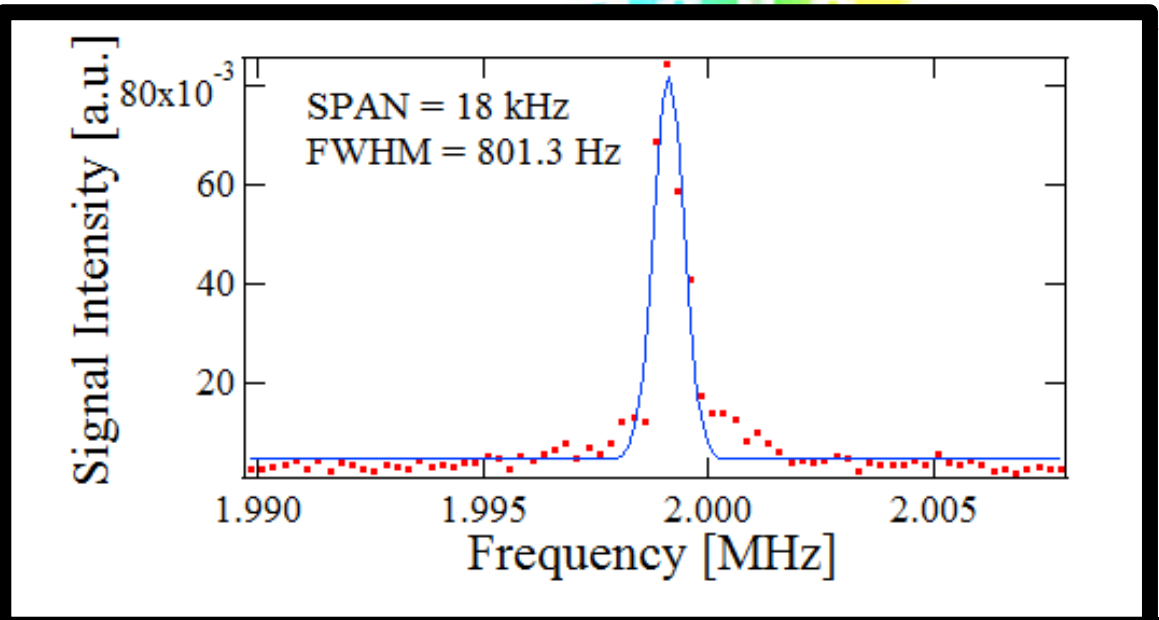


**Proposed method
(Adaptive sampling)**

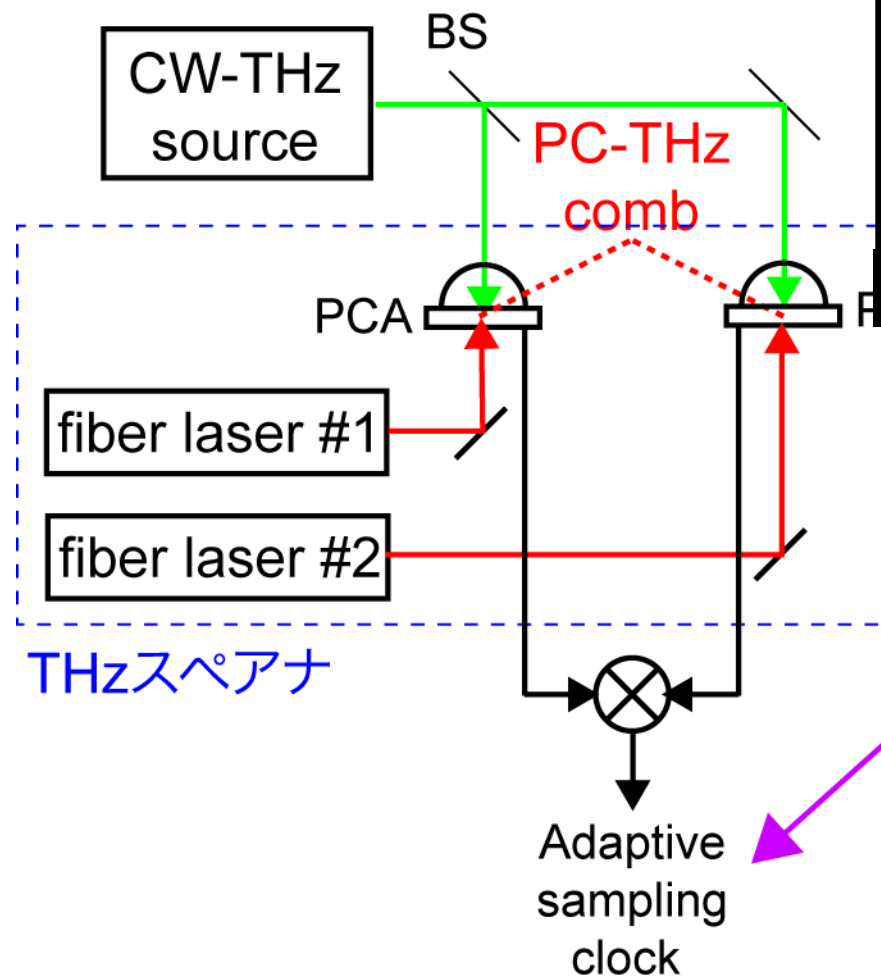


Adaptive clock can be generated by beat signal between dual THz comb modes!!

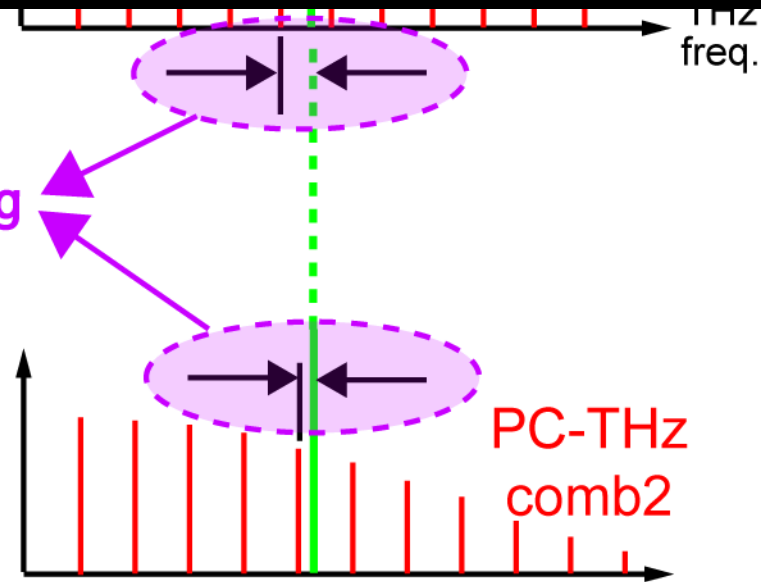
Generation of THz THz-comb-refere



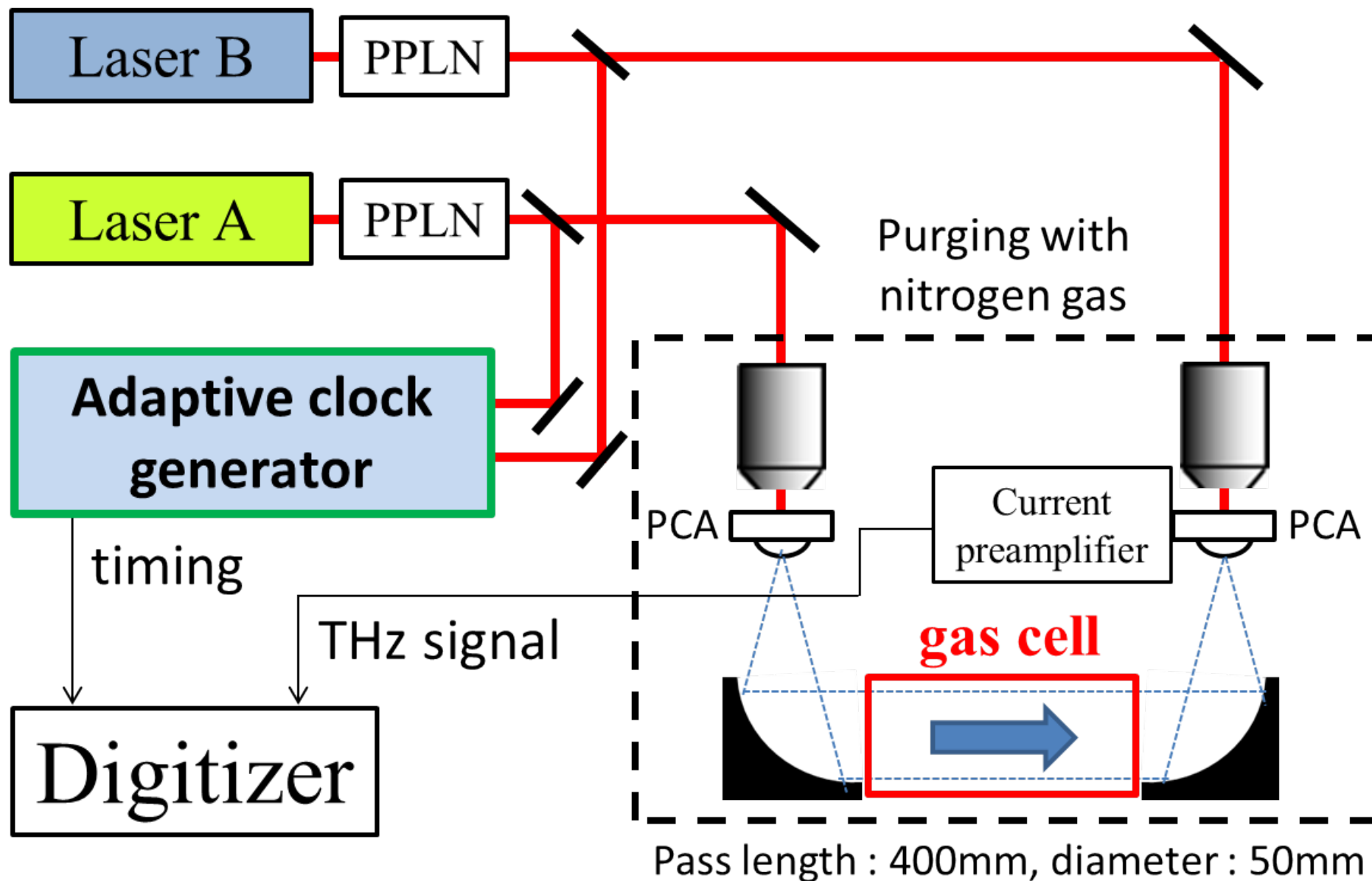
Beat signal between dual THz comb modes at 4 THz



Mixing



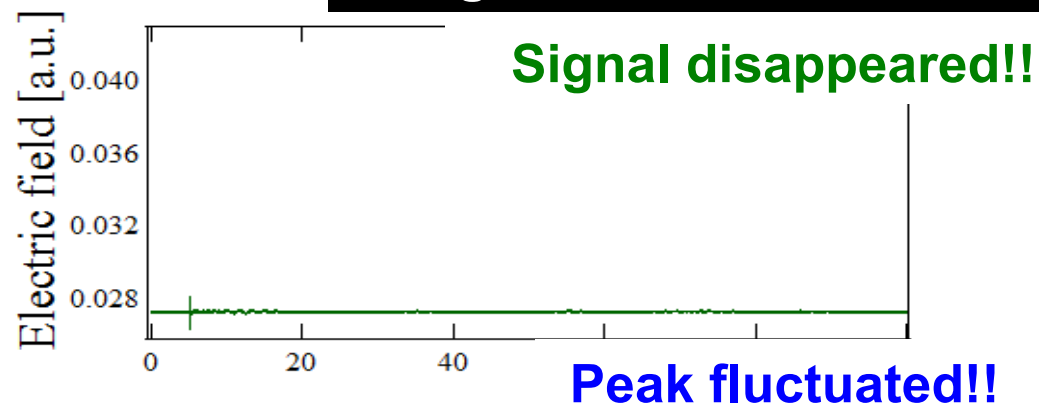
Setup for dual THz comb spectroscopy



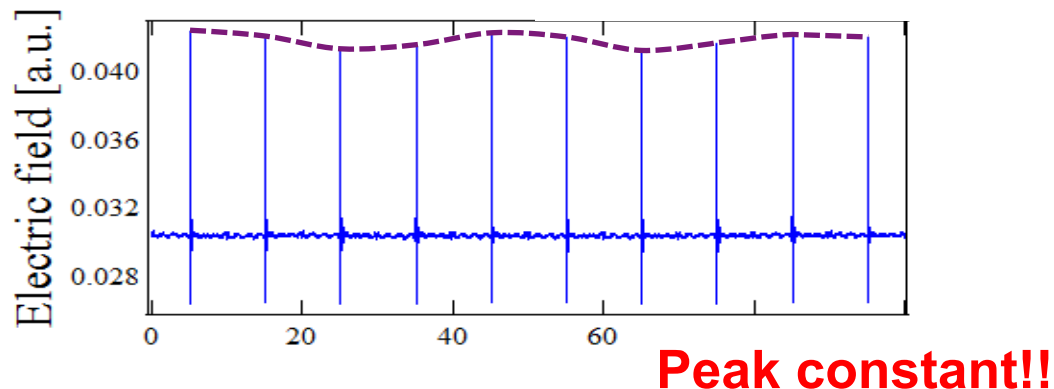
Integrated temporal waveform of THz pulses

Integration number=10000

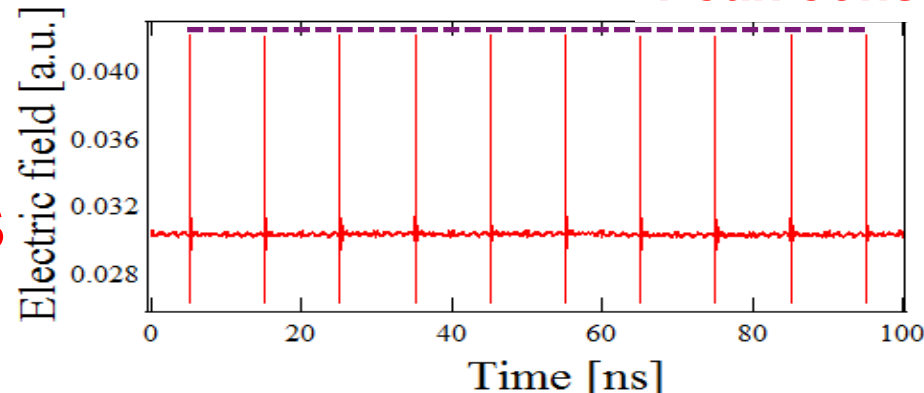
Constant clock
with free-running lasers



Constant clock
with stabilized lasers



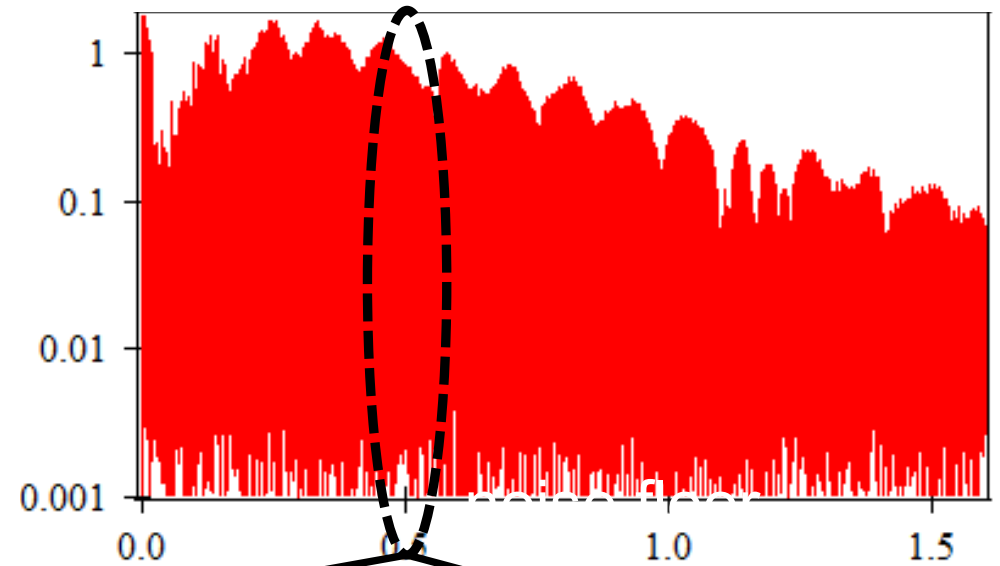
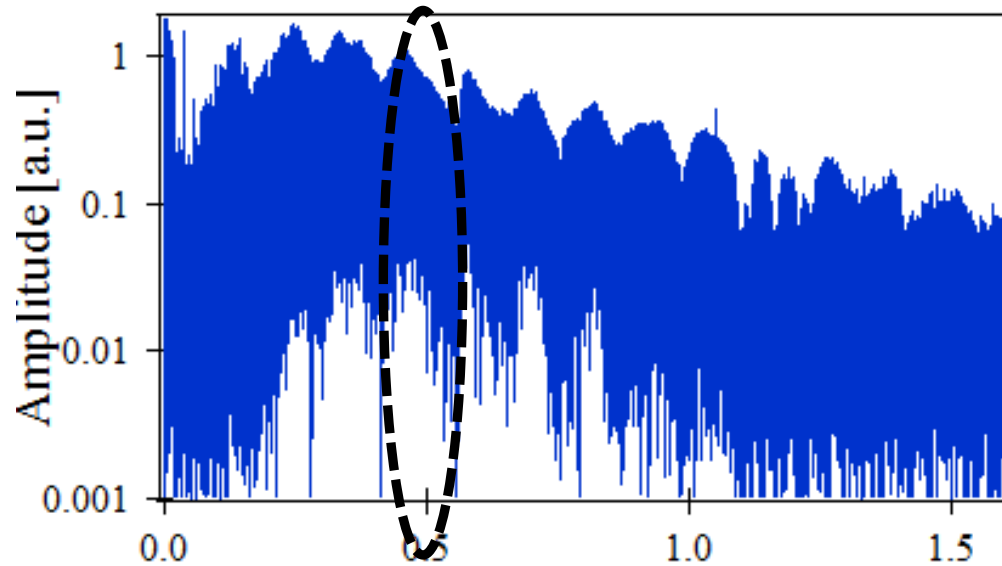
Adaptive clock
with free-running lasers



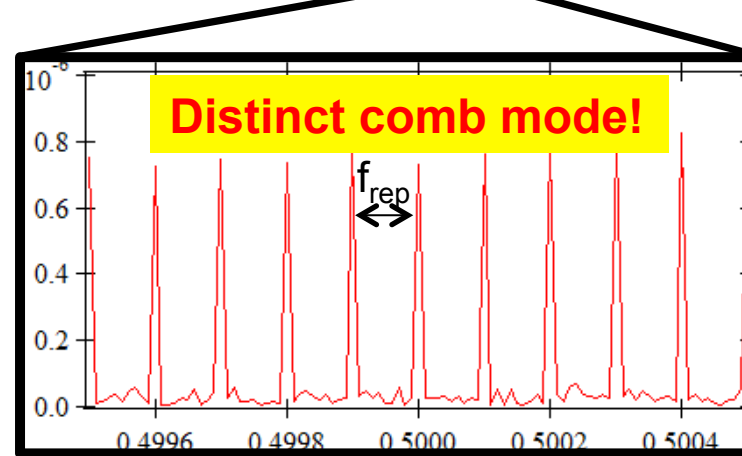
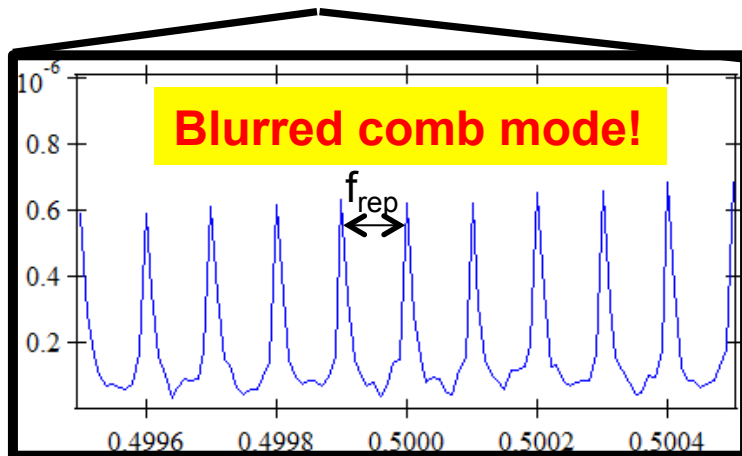
Mode-resolved THz comb spectrum

Constant clock
with stabilized lasers

Adaptive clock
with free-running lasers



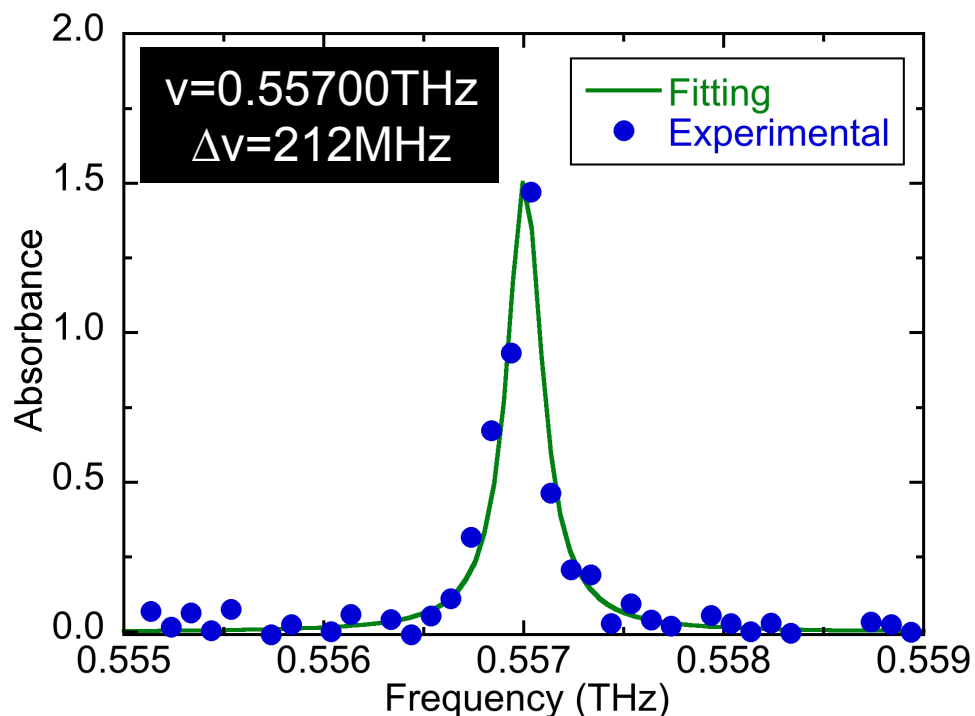
Frequency [THz]



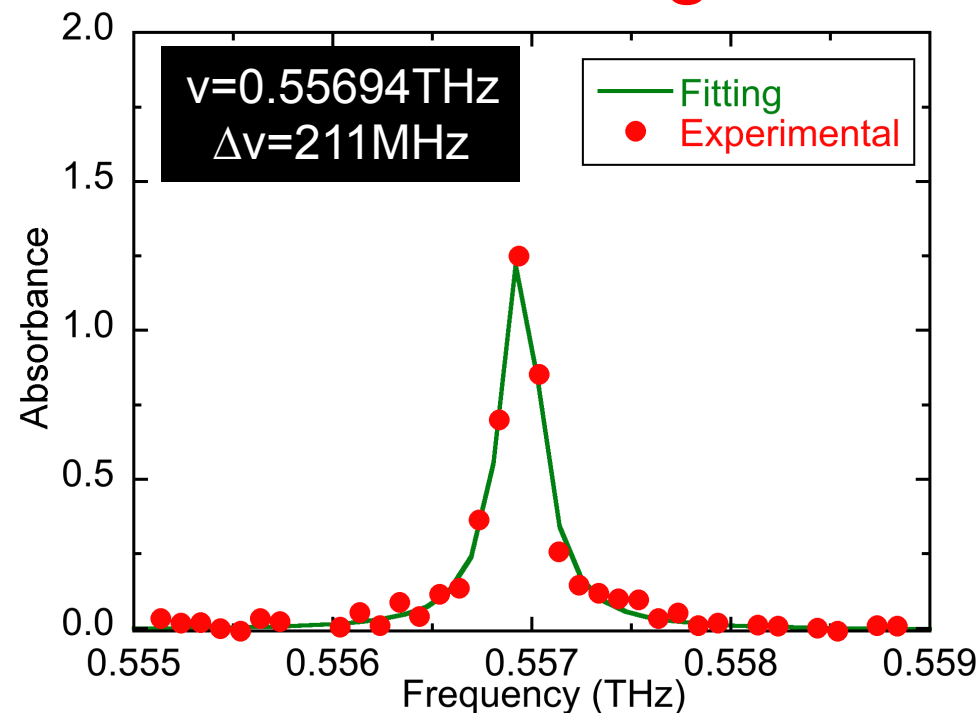
Water vapor at low pressure

Rotational transition $I_{10} \leftarrow I_{01}$ @ 0.556936THz
Pressure broadening linewidth = 200MHz

Constant clock
with stabilized lasers



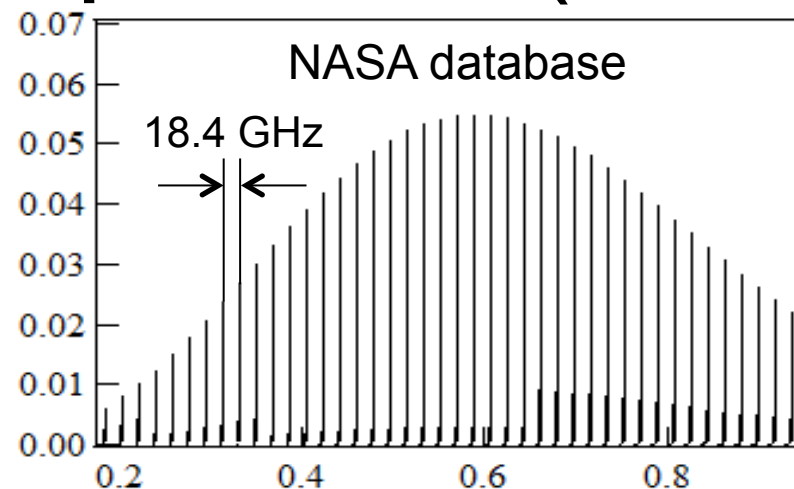
Adaptive clock
with free-running lasers



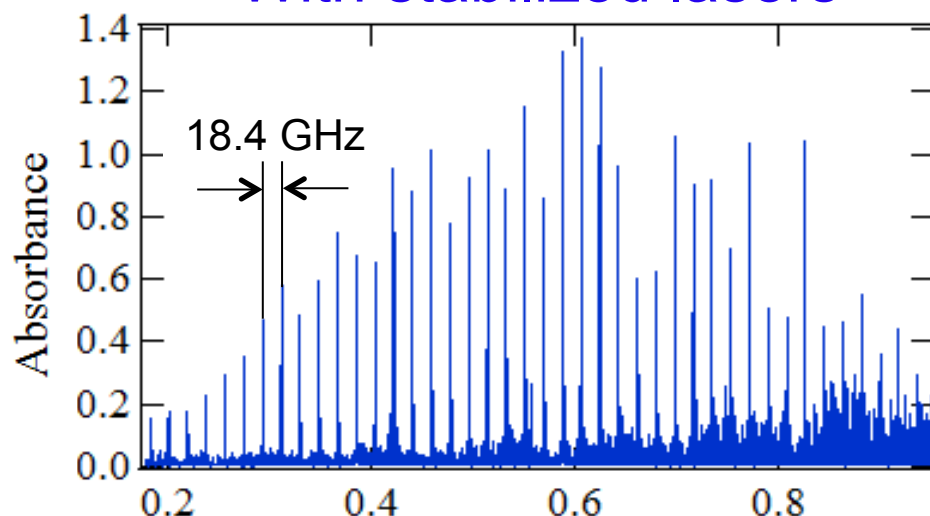
Acetonitrile gas at low pressure(1kPa)

Acetonitrile (CH_3CN)

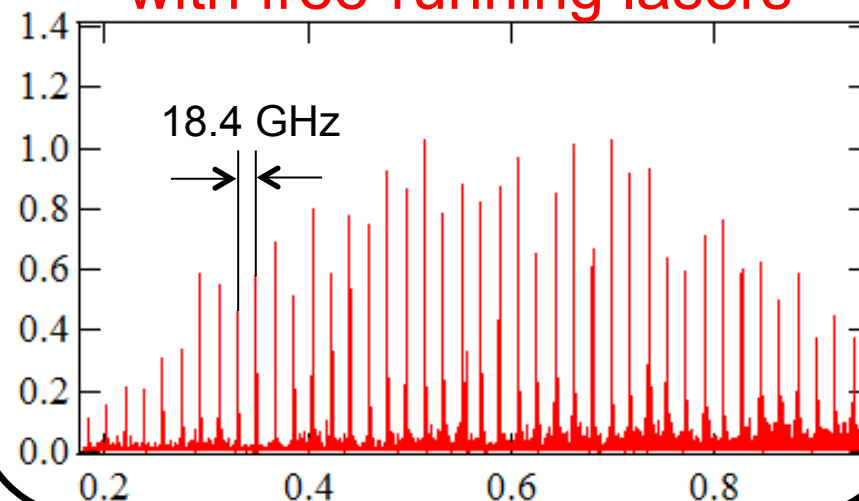
- One of VOCs
- Very abundant species in interstellar medium



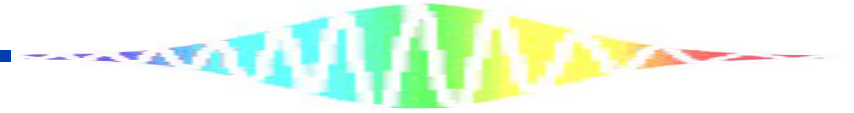
Constant clock
With stabilized lasers



Adaptive clock
with free-running lasers



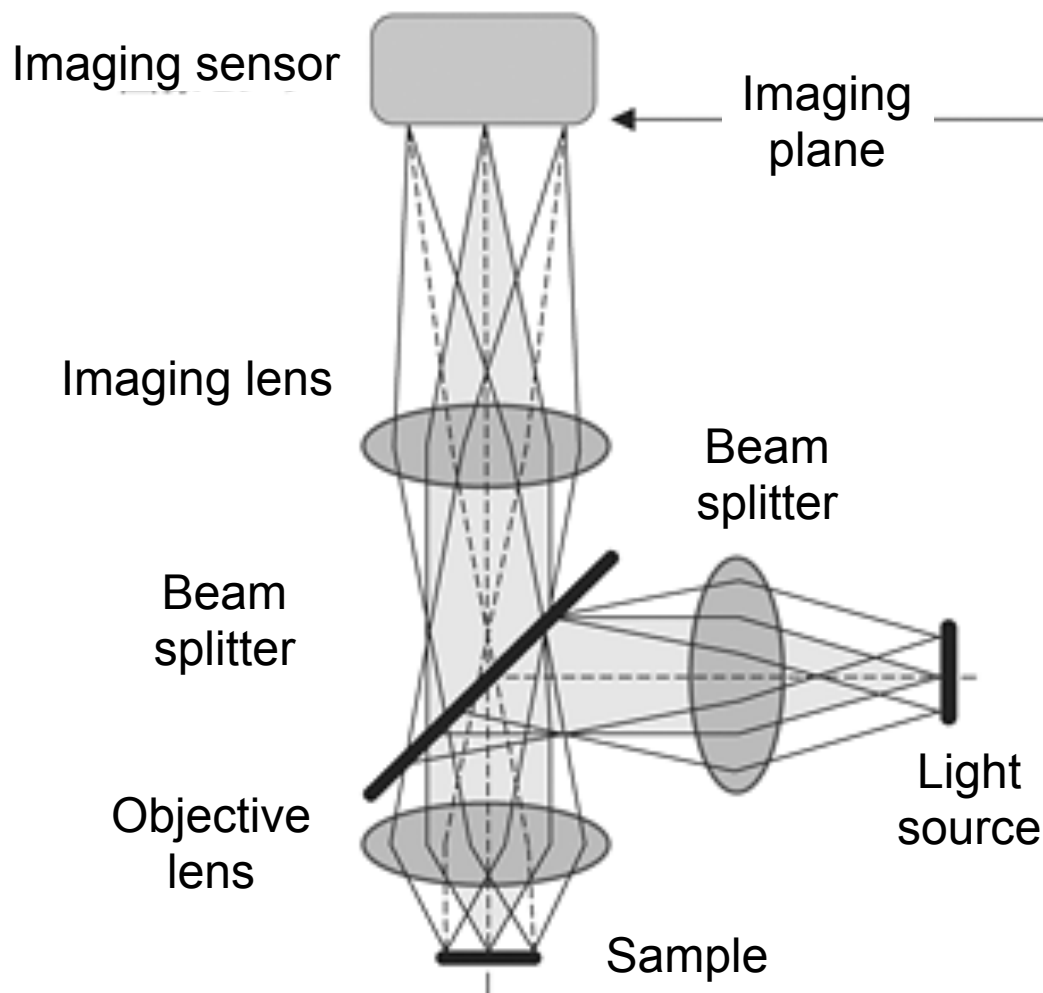
Frequency [THz]



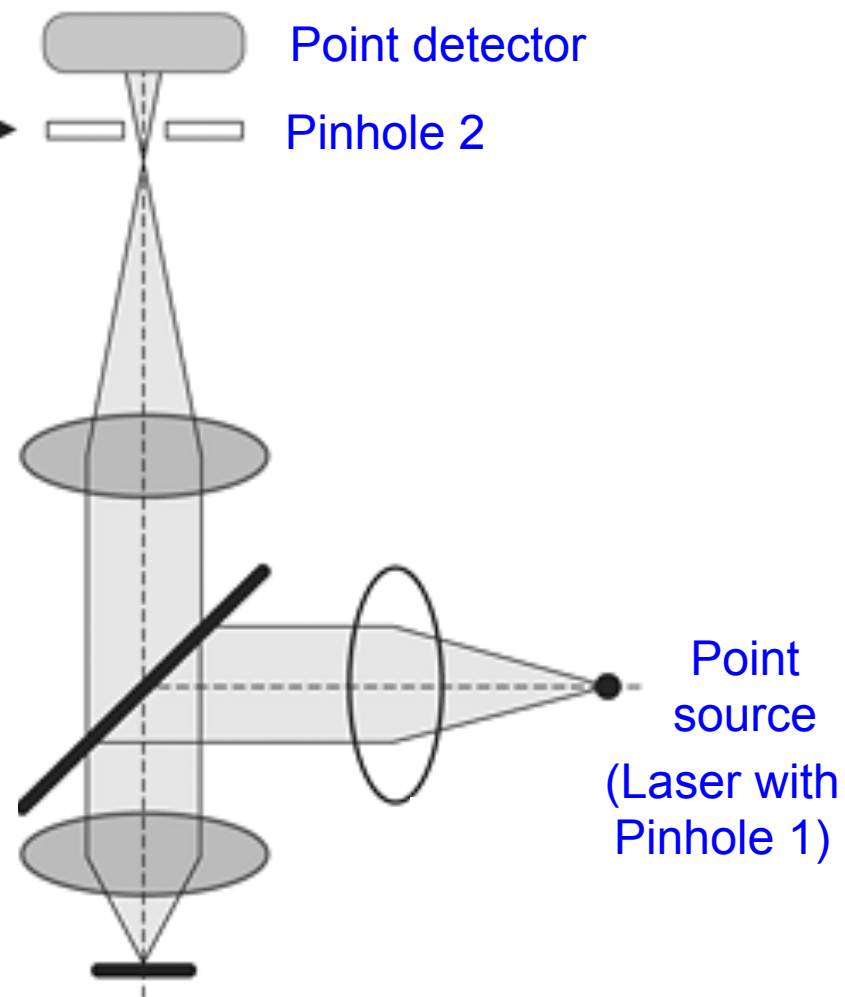
(2) Full-field confocal optical-comb microscopy

Optical microscopy

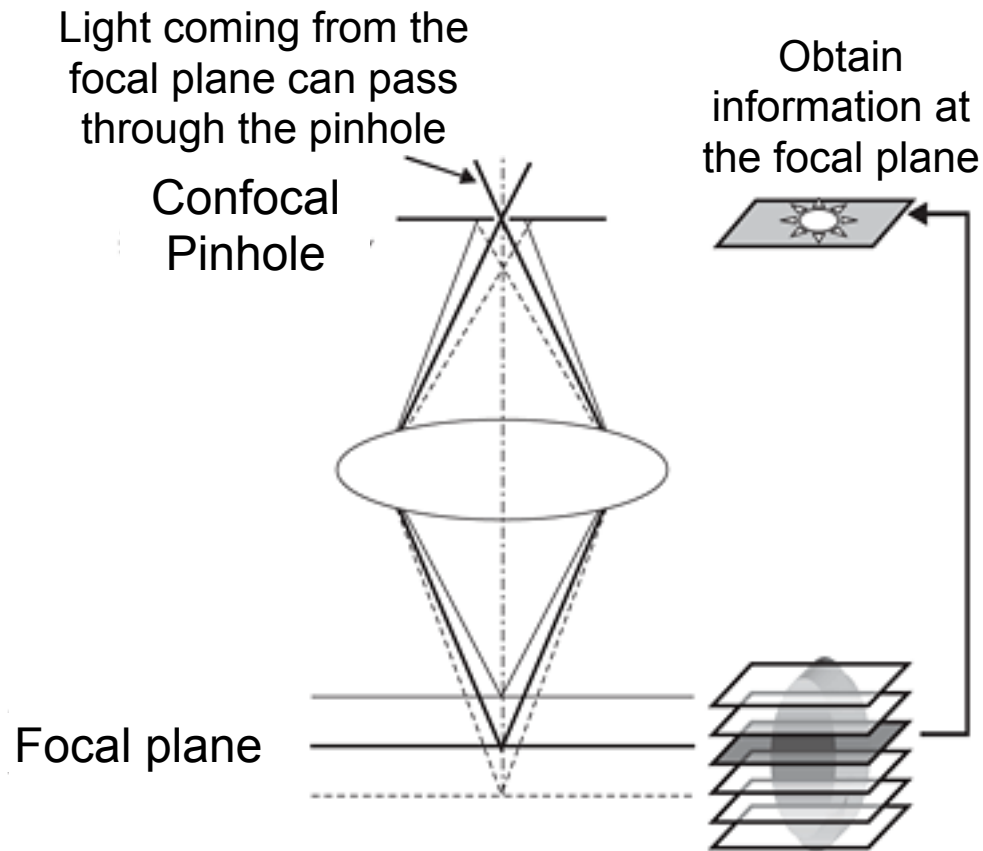
Normal microscopy



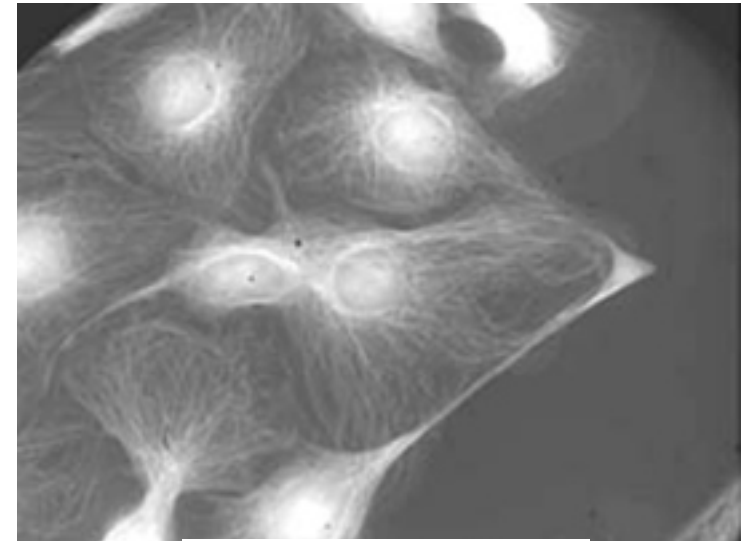
Confocal microscopy



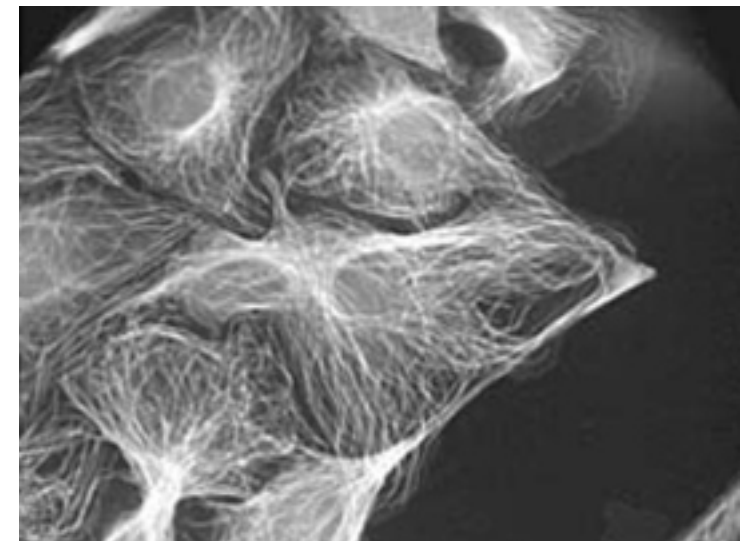
Confocal optical microscopy



- Detect signal at focal plane
- No stray light
 - Depth resolution
 - Applicable for thick sample

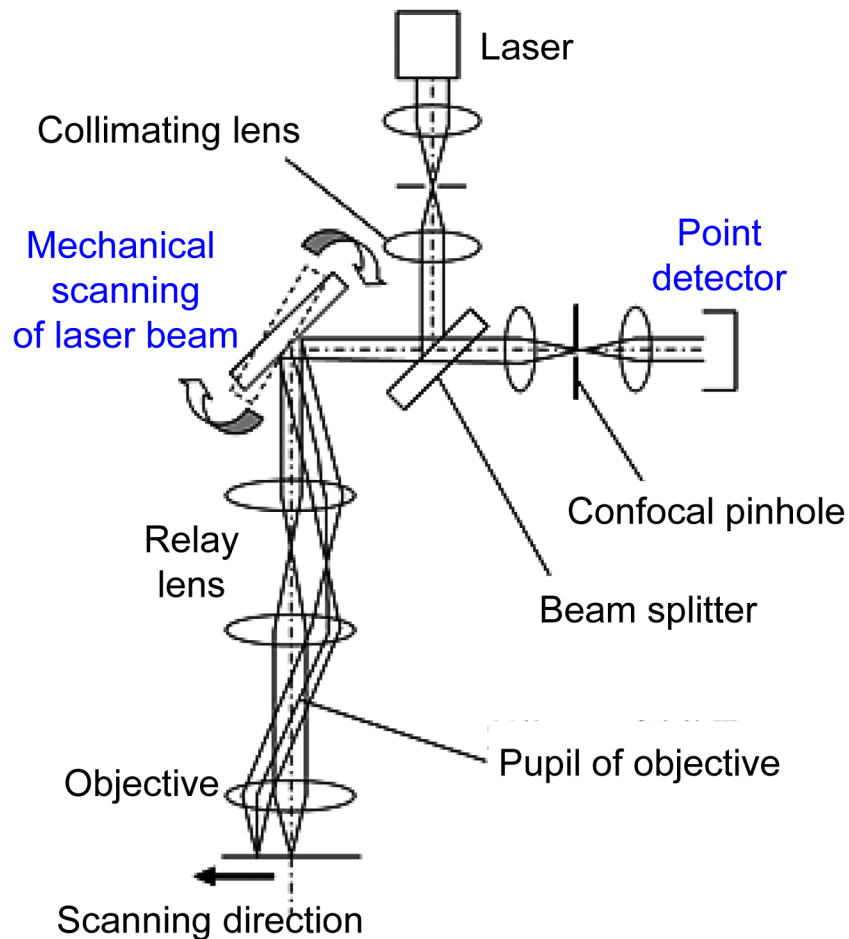


Normal microscopy



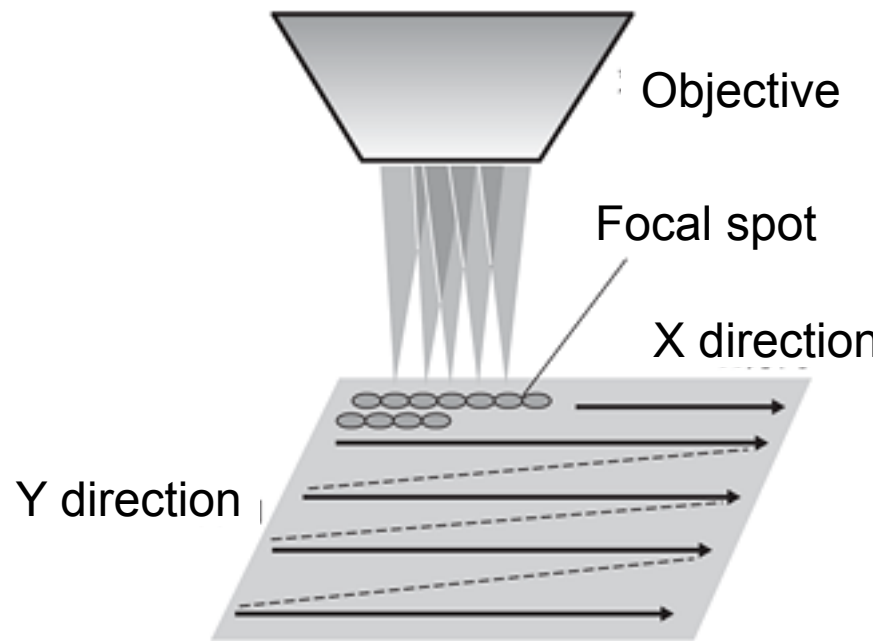
Confocal microscopy

Confocal laser microscopy



Optical configuration

http://bioimaging.jp/learn/023/index_2.html



Raster scanning of a focused laser spot

<http://bioimaging.jp/learn/023/>

Faster image acquisition ! Higher spatial resolution!

Fast confocal microscopy

Conventional

Serial imaging based on mechanical scanning



Optical comb

Proposal

One-shot imaging based on parallel data acquisition

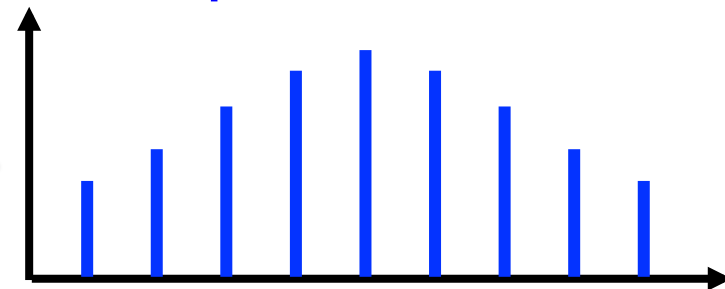


Dimension-converting comb

Temporal
Spatial
Polarization
etc

Conversion to
wavelength

Optical comb



Information is superimposed
on each comb mode

Dual comb spectroscopy

Number of comb mode

☞ Tremendous channel number

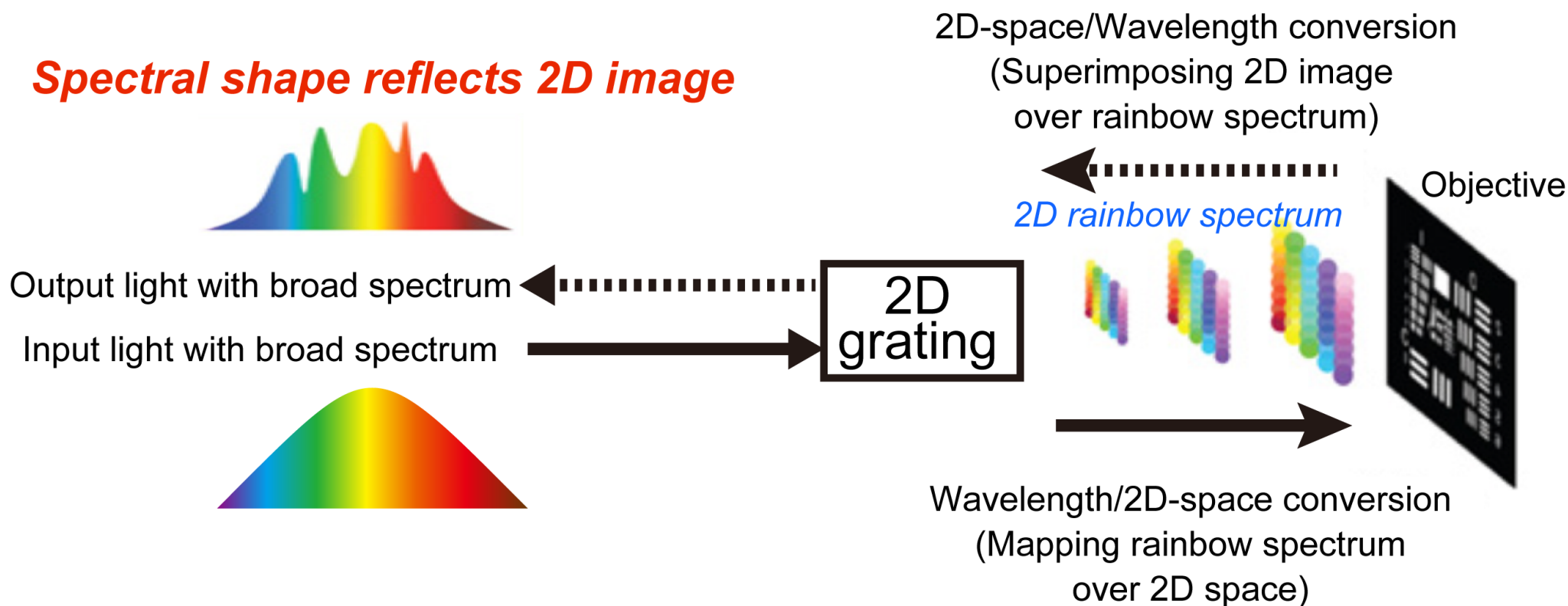
Linewidth of comb mode

☞ Infinitesimal sampling interval

Mode-resolved
comb spectrum

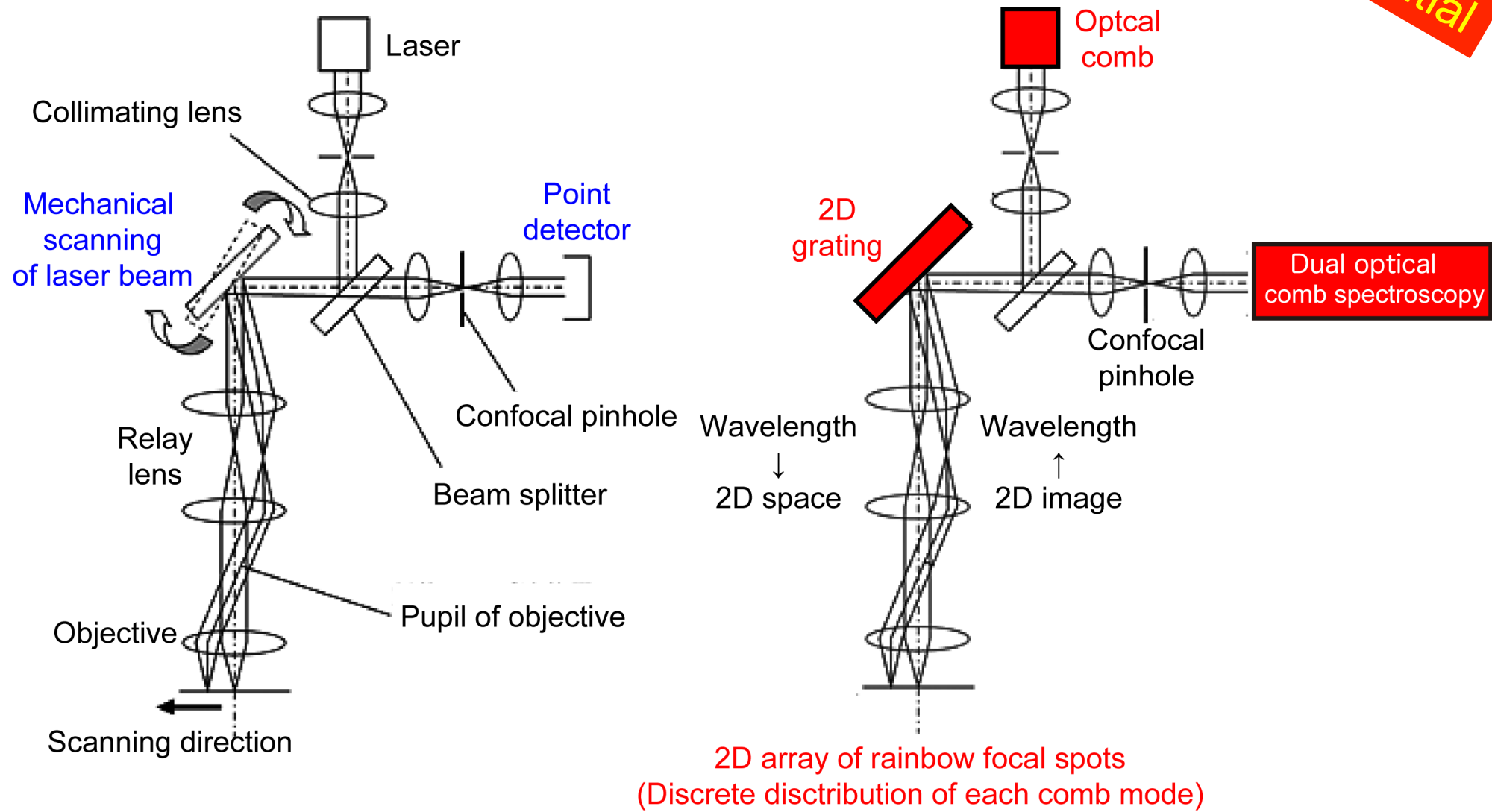
Single-shot imaging based on bidirectional conversion between wavelength and 2D space

Spectral shape reflects 2D image



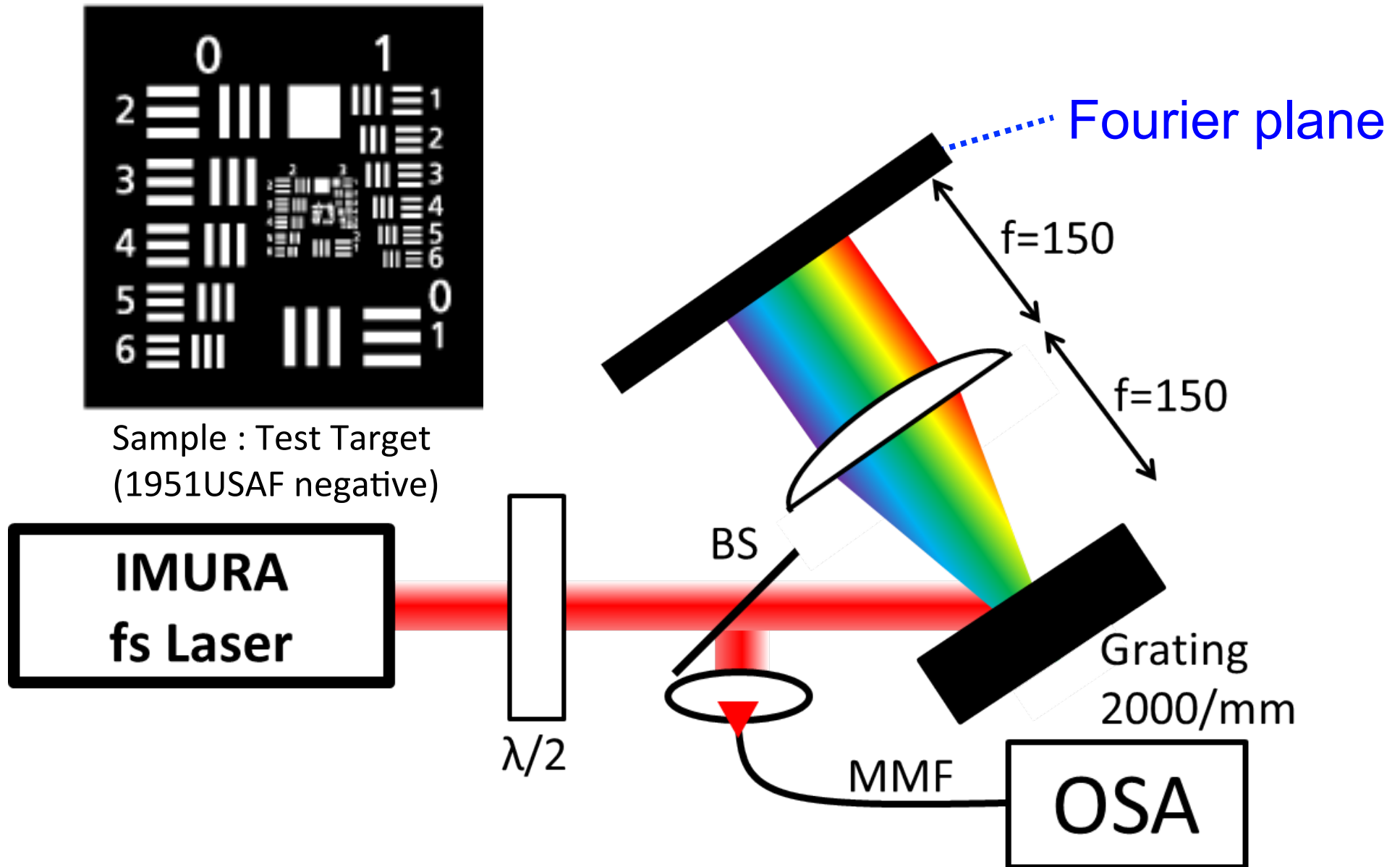
Optical-comb confocal microscopy

Confidential

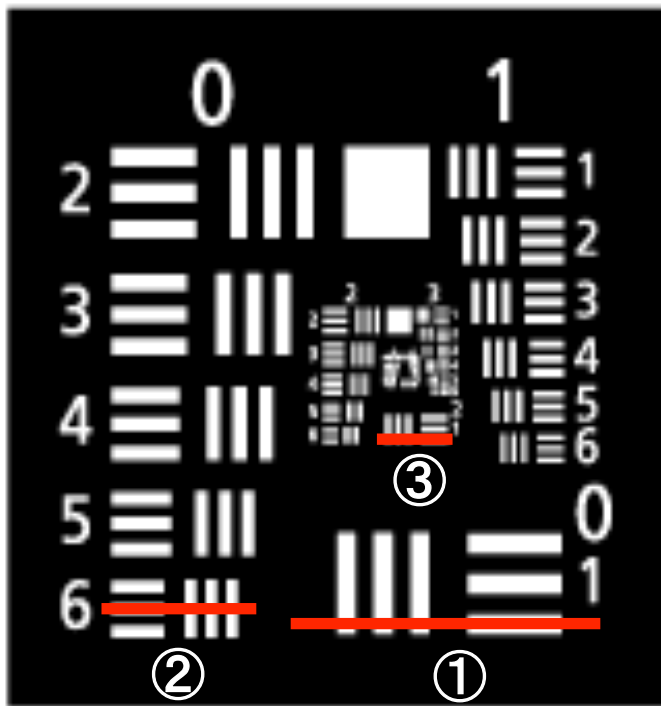


No mechanical scanning, single-shot imaging, super resolution

Optical setup for line imaging with OSA

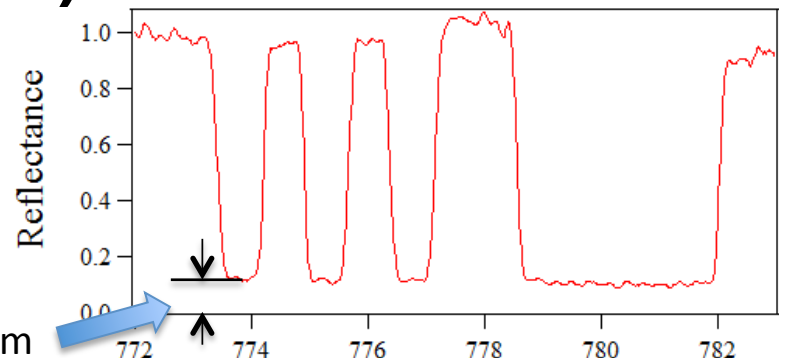


Results (1)

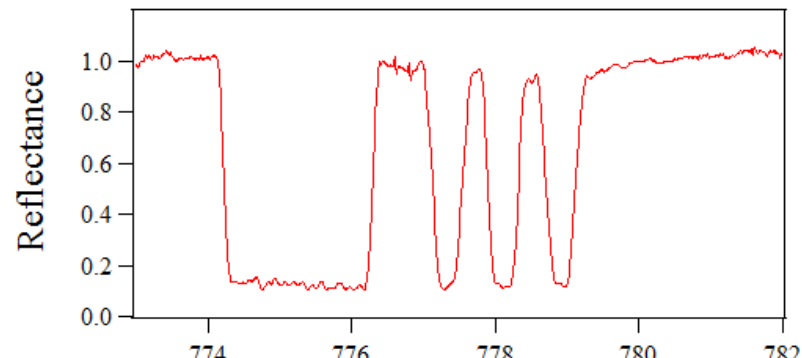


要素番号	0	1	2
1	1.00	2.00	4.00
2	1.12	2.24	4.49
3	1.26	2.52	5.04
4	1.41	2.83	5.66
5	1.59	3.17	6.35
6	1.78	3.56	7.13

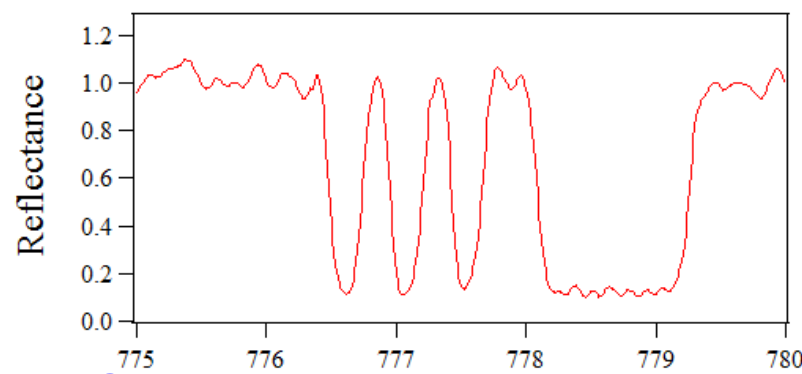
Reflection from glass



① Spec. res=0.1 nm, span=11 nm



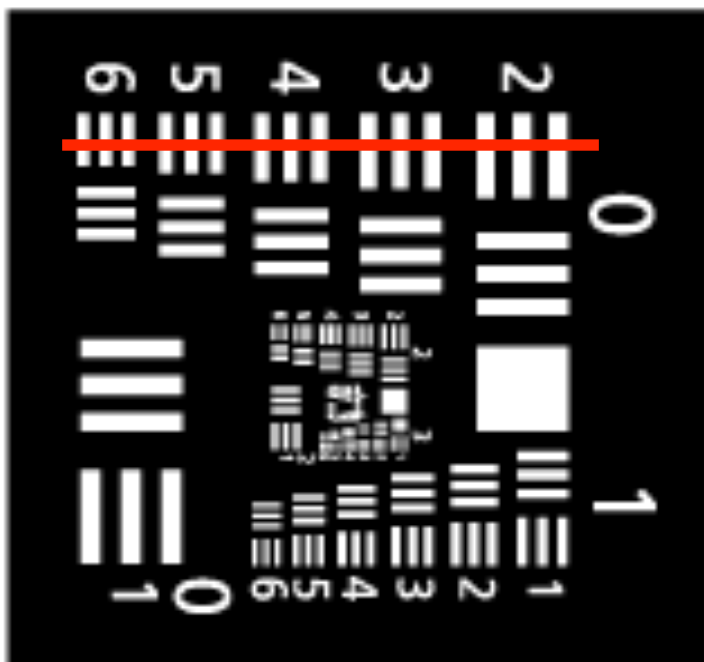
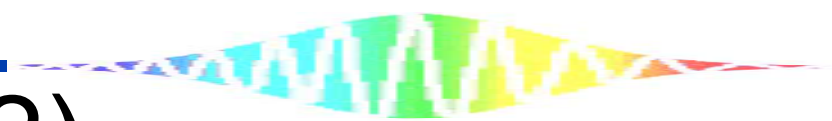
② Spec. res=0.05 nm, span=9 nm



③ Spec. res=0.05 nm, span=5 nm

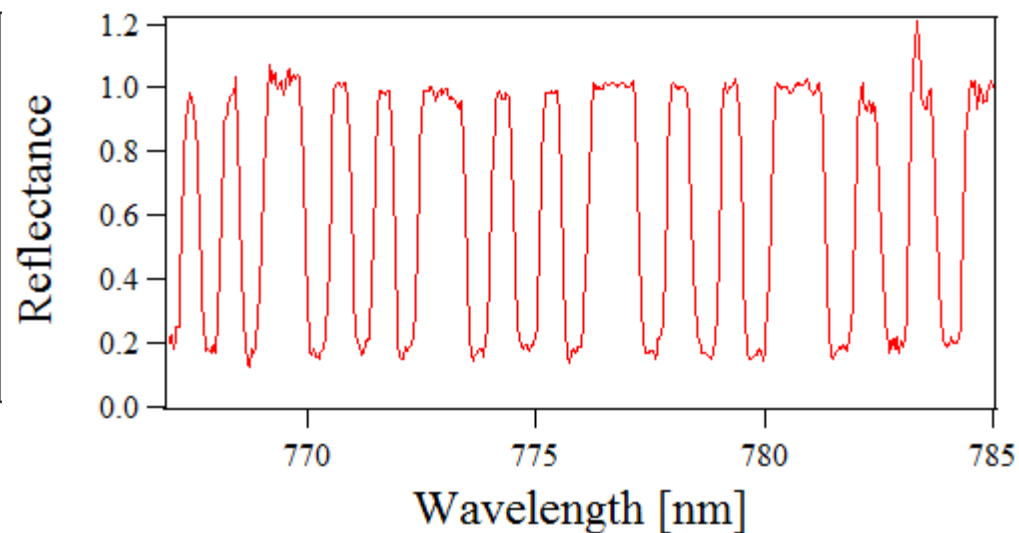
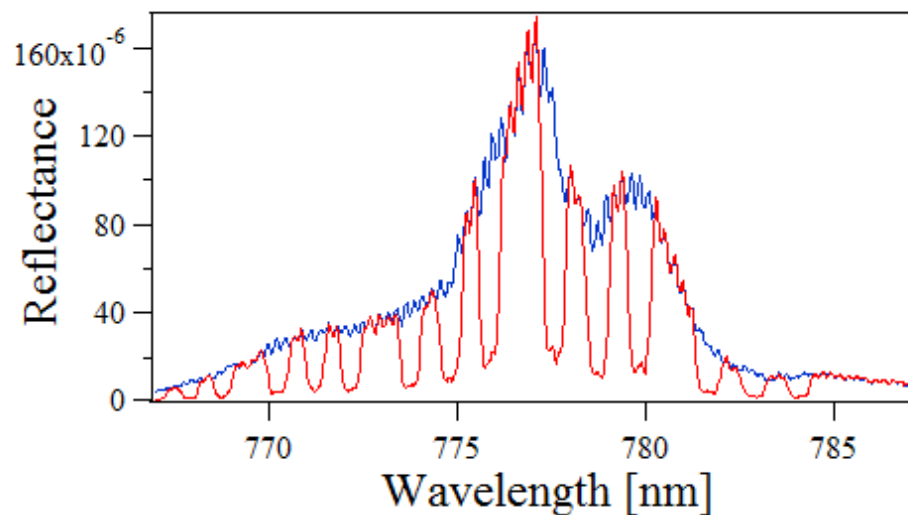
Can not resolved

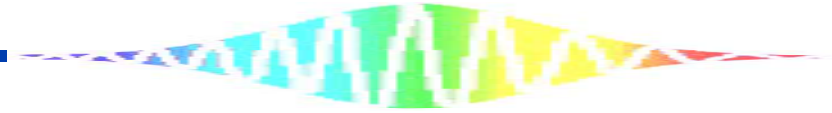
Results (2)



1 mmあたりのラインペアの数 (IP/mm)

要素番号	グループ			
	0	1	2	3
1	1.00	2.00	4.00	8.00
2	1.12	2.24	4.49	8.98
3	1.26	2.52	5.04	10.10
4	1.41	2.83	5.66	11.30
5	1.59	3.17	6.35	12.70
6	1.78	3.56	7.13	14.30



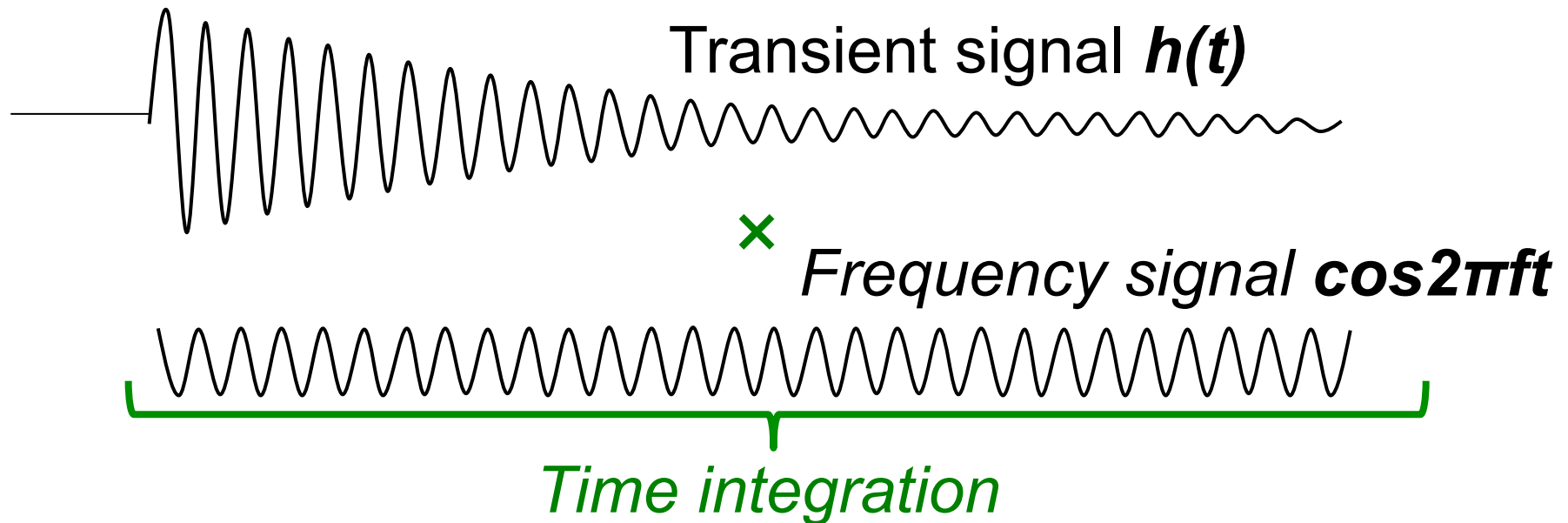


(3) Discrete Fourier transform infrared spectroscopy (in collaboration with AIST)

Fourier transform of transient signal $h(t)$

*Formula
of FT*

$$H(f) = \int_{-\infty}^{+\infty} h(t) \cdot \exp(-2\pi ift) dt$$



Actual spectral resolution is determined by observed time window.

**Pulse train of
THz electric field**

Upper limit of time window
= repetition period of THz pulse

FT of transient signals induced by THz pulse train

Relaxation time
of transient signal

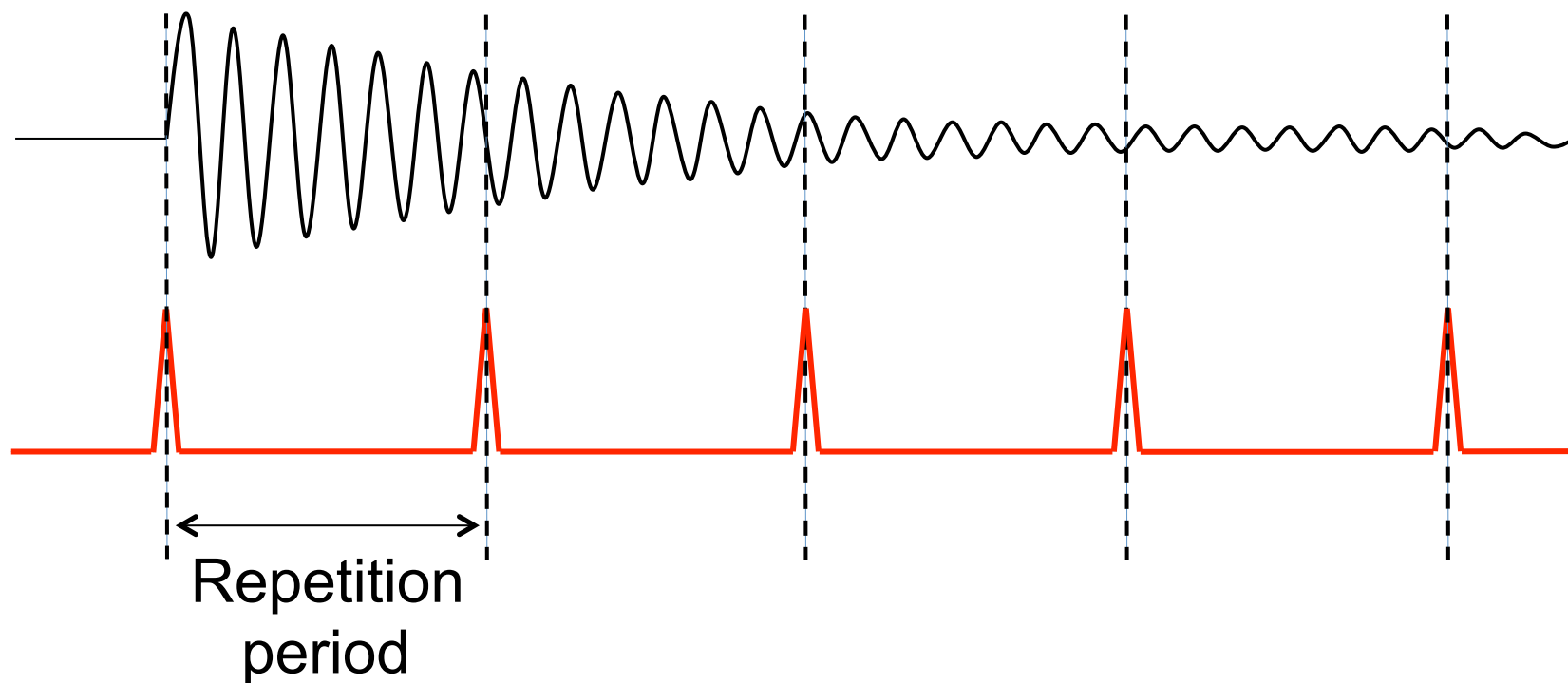
>

Repetition period of
THz pulse train

e.g. FID signal

Transient signal $h(t)$

Periodic
excitation

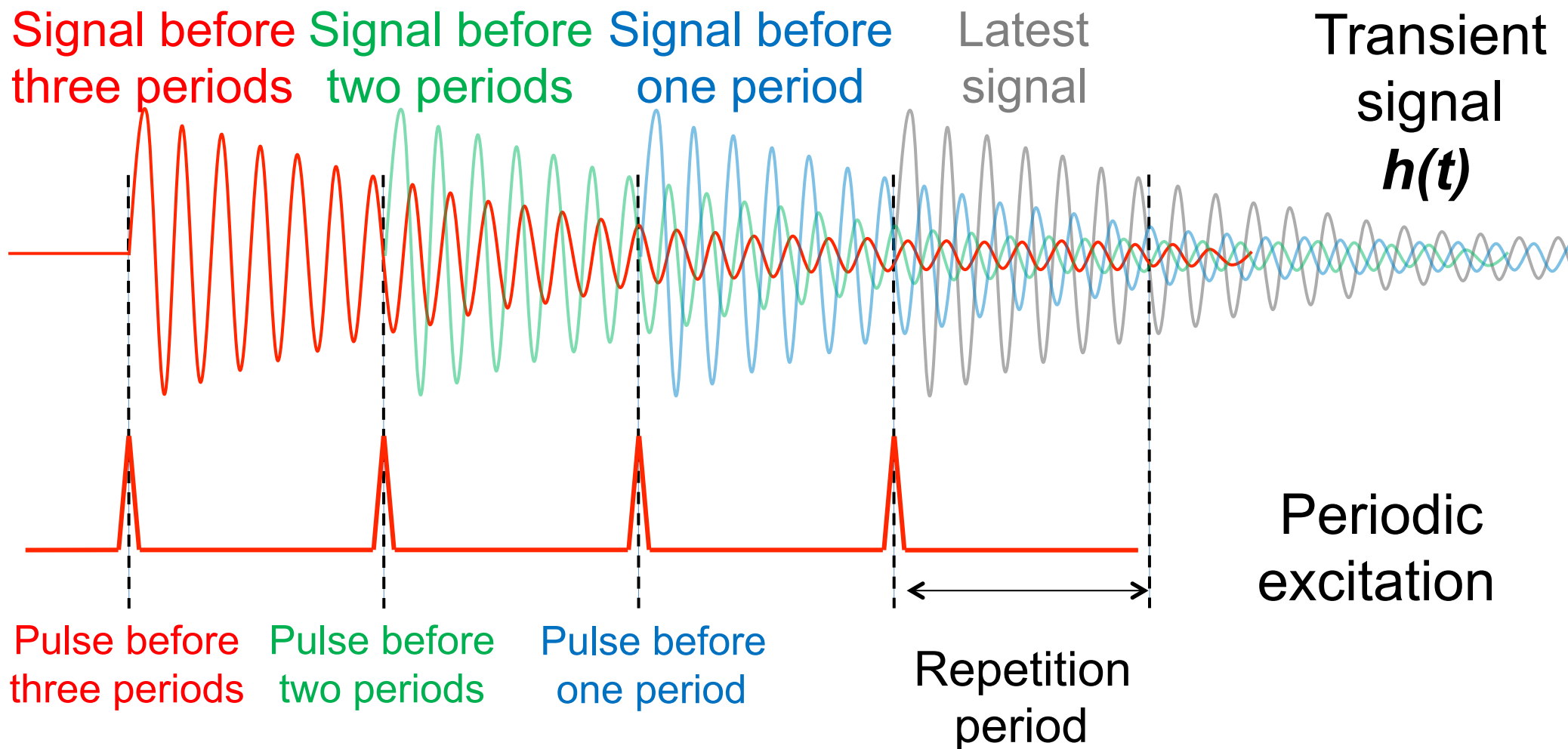


FT of transient signals induced by THz pulse train

Relaxation time
of transient signal

>

Repetition period of
THz pulse train

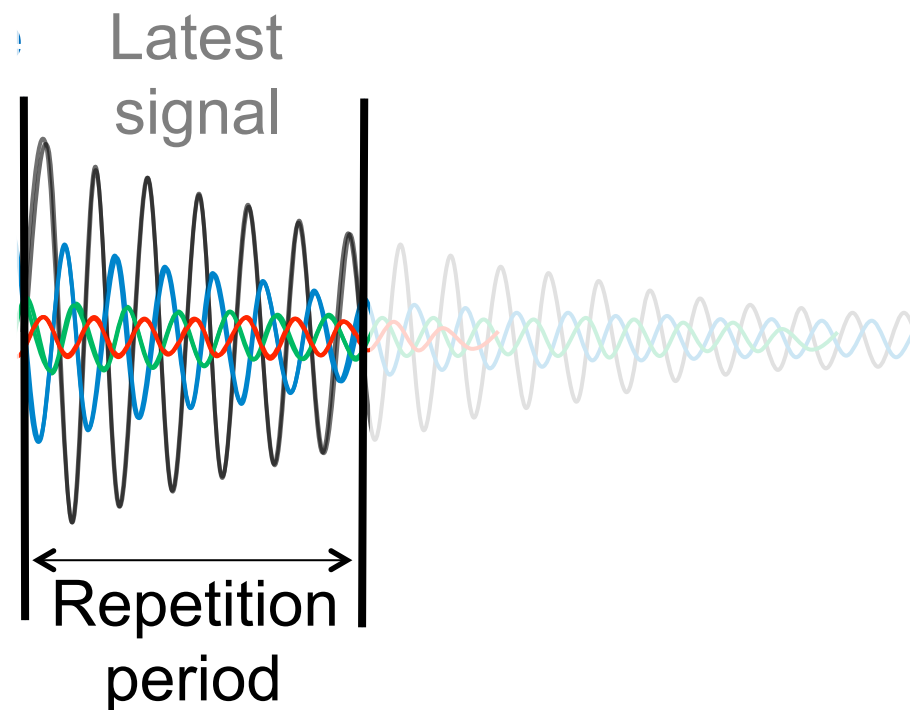


FT of transient signals induced by THz pulse train

Relaxation time
of transient signal

>

Repetition period of
THz pulse train

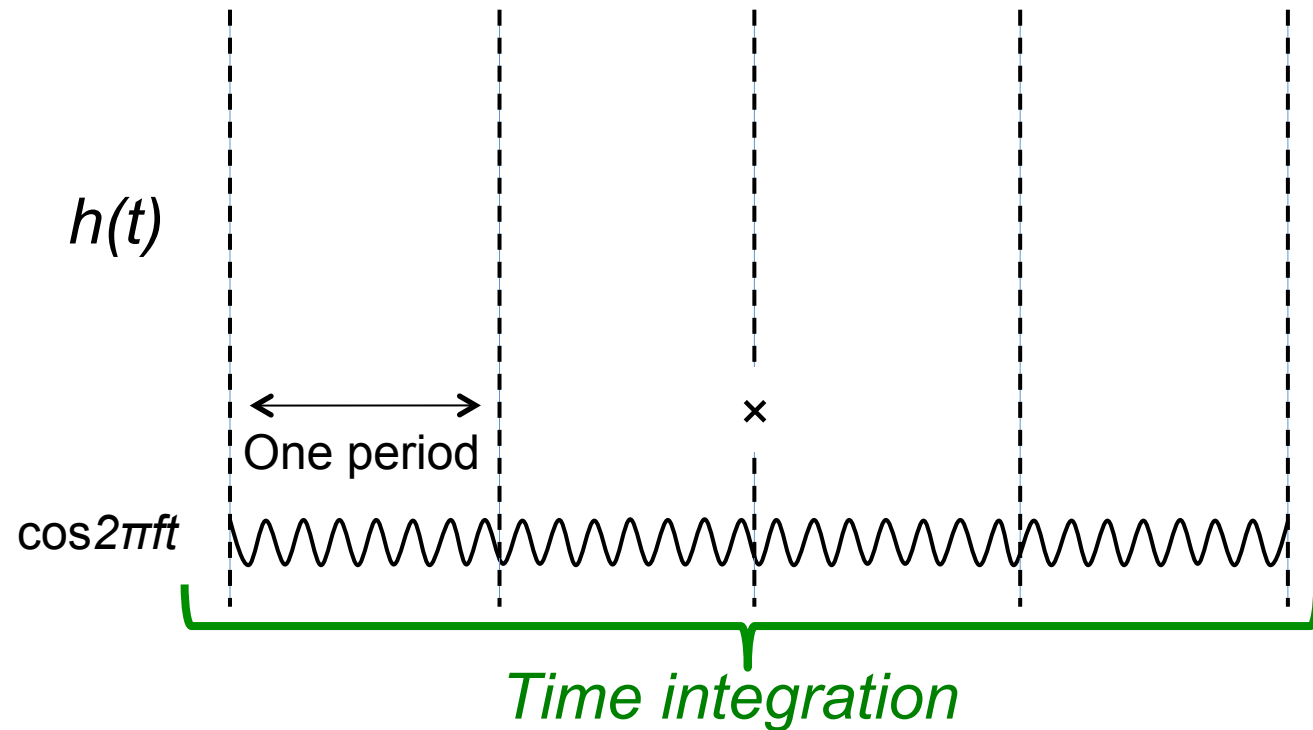
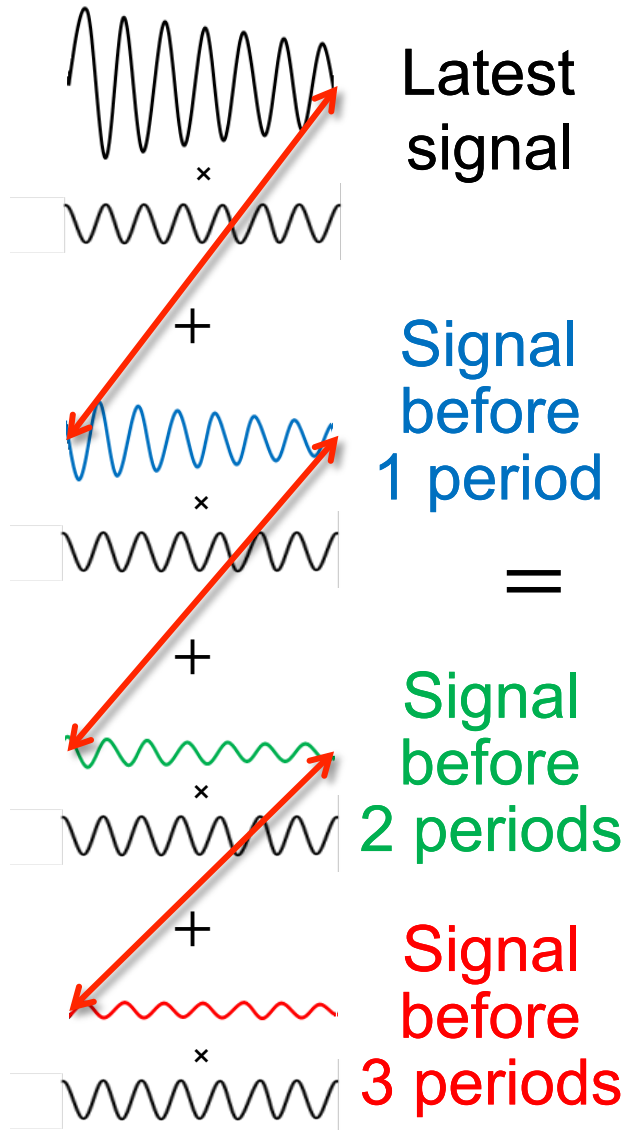


Focus on pieces of different transient signals included in the same one period

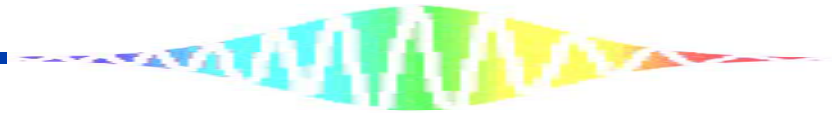
**Observed time window
= One repetition period**

Fields induced by THz pulse train

Each piece can be temporally connected!



**Equivalent to integration of $h(t) \cdot \cos 2\pi ft$
within infinite time window!**

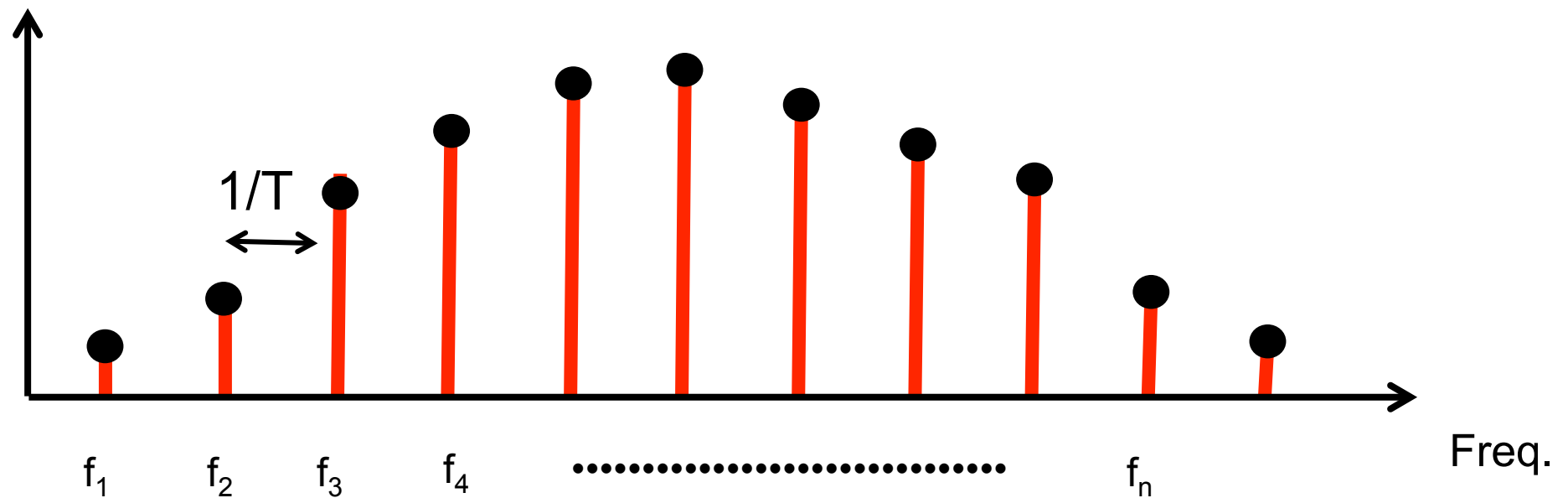


Discrete Fourier transform spectrum

Frequency of each plot

$$f_n = \frac{n}{T}$$

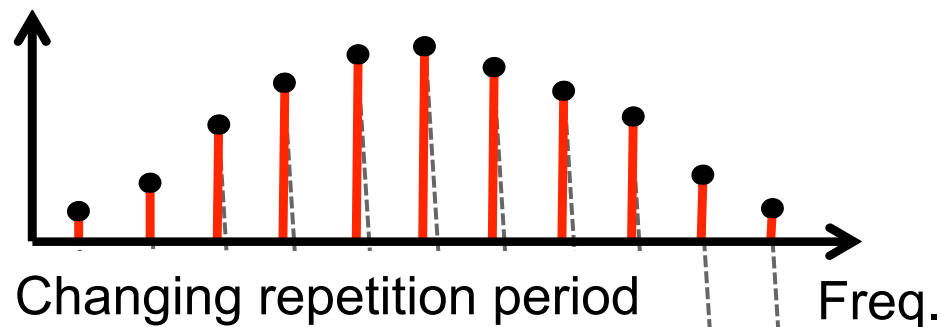
n : number of data plot
 T : repetition period



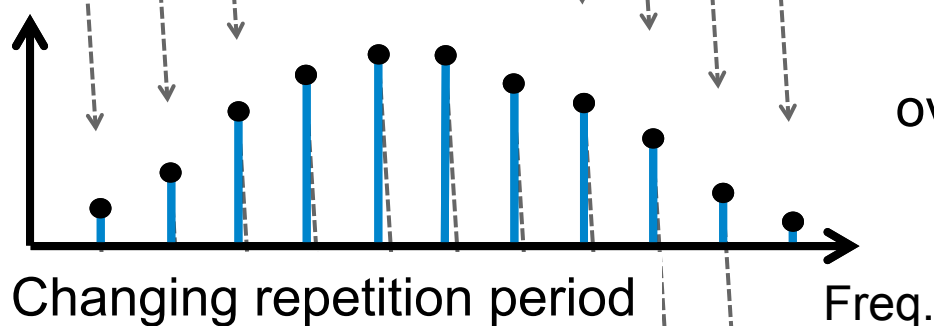
Each plot has infinite spectral resolution due to infinite time window. However, discrete distribution limits spectral resolution to plot interval.



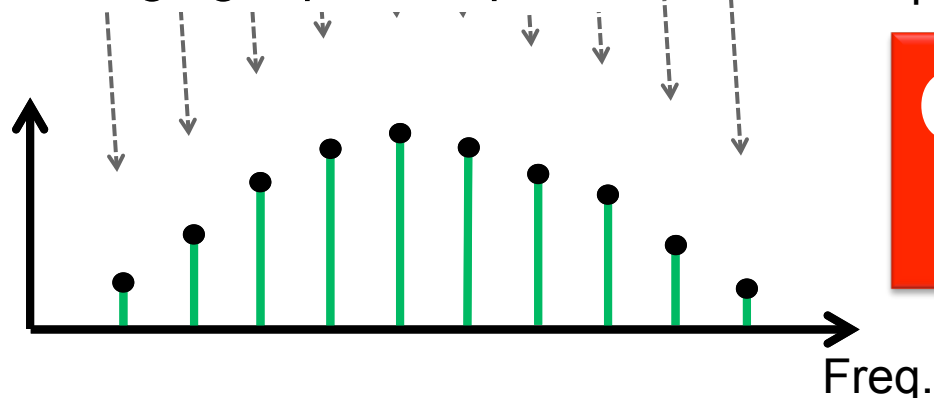
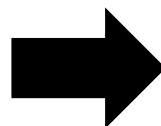
Sweeping of discrete FT spectrum



Spectrally interleaving spectrum

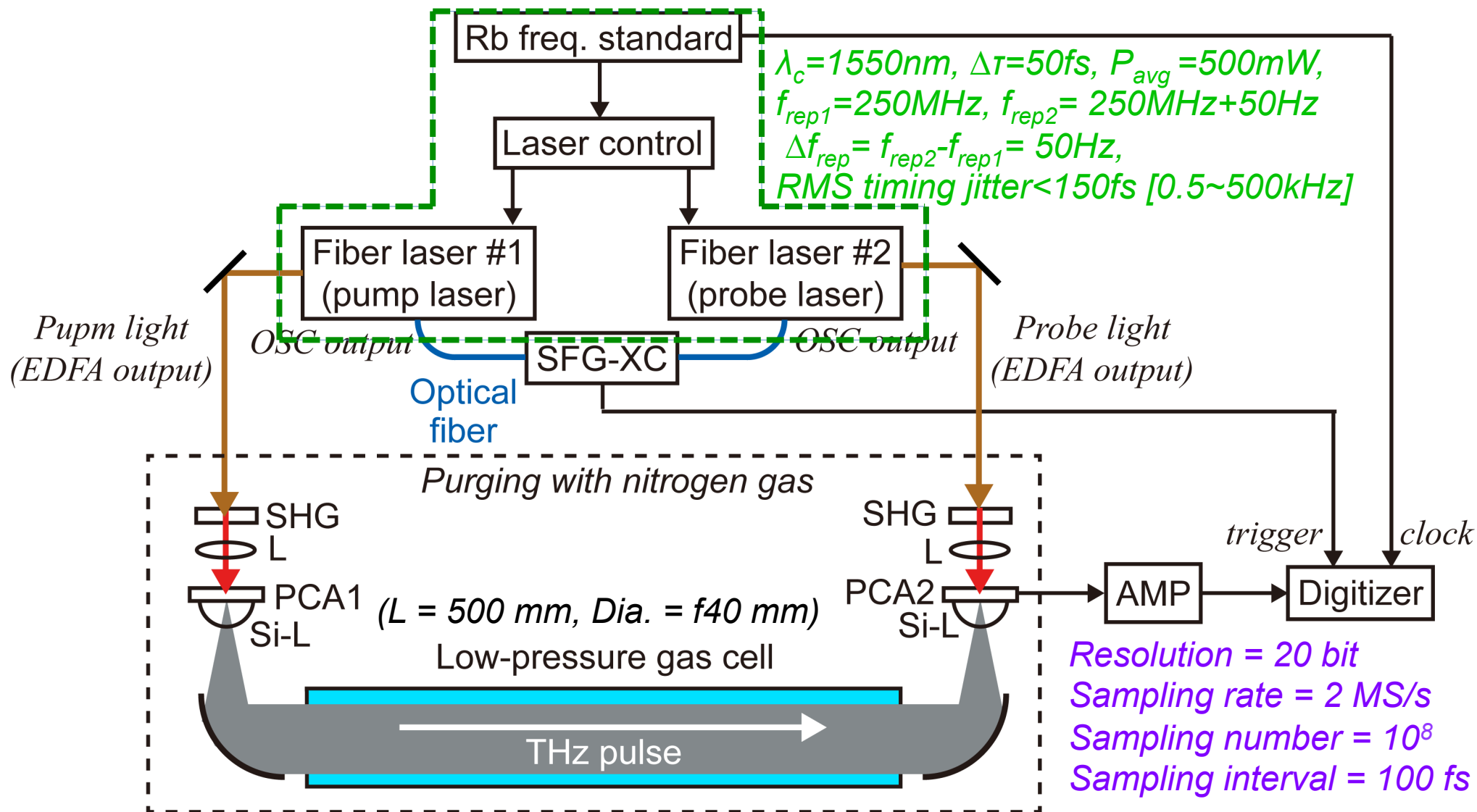


Spectral overlapping



Gaps between data plots can be filled in

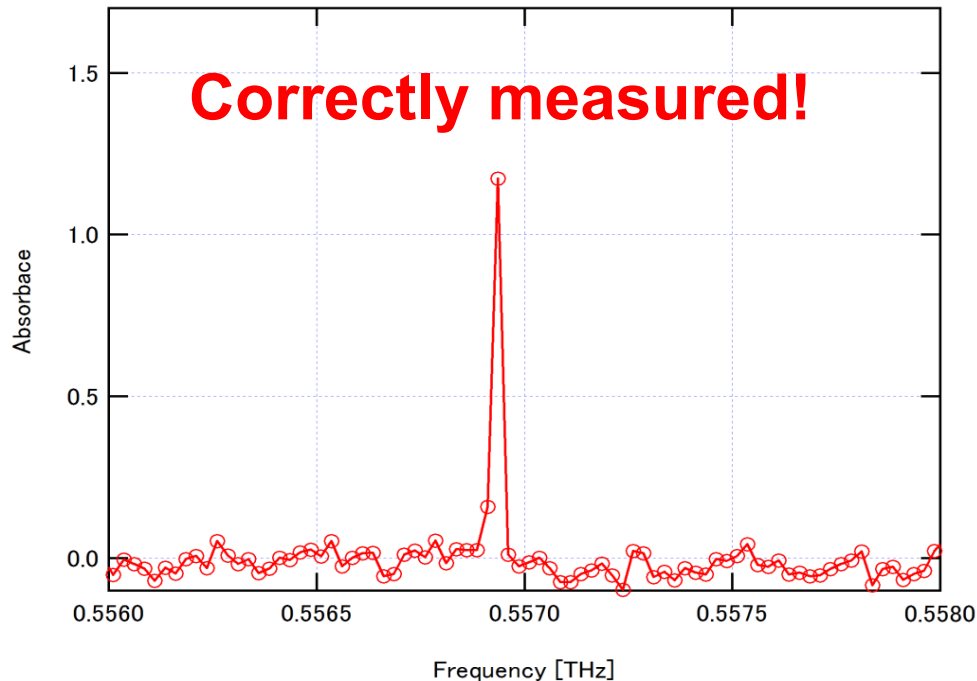
Experimental setup



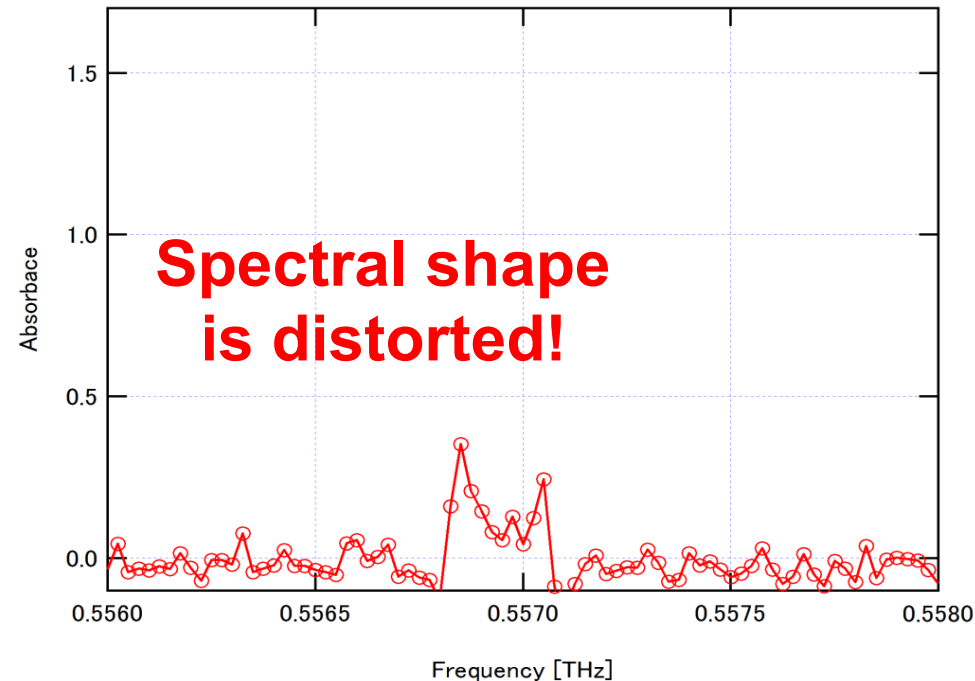
Relation between time window and repetition period (Relaxation time > repetition period)

Rotational transition of H₂O at 0.557 THz (H₂O@6 Pa & N₂@140 Pa)
Expected pressure broadening linewidth = 10.6 MHz
Relaxation time of absorption = 94 ns (25 periods)

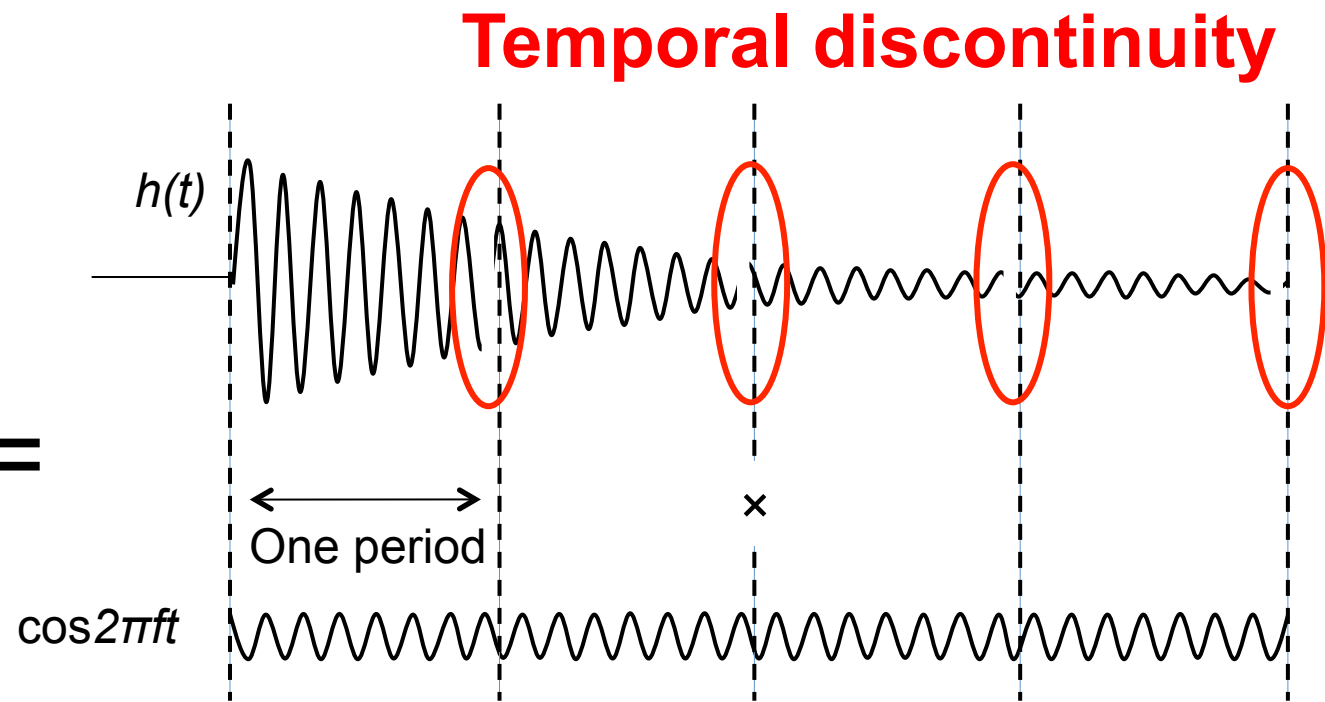
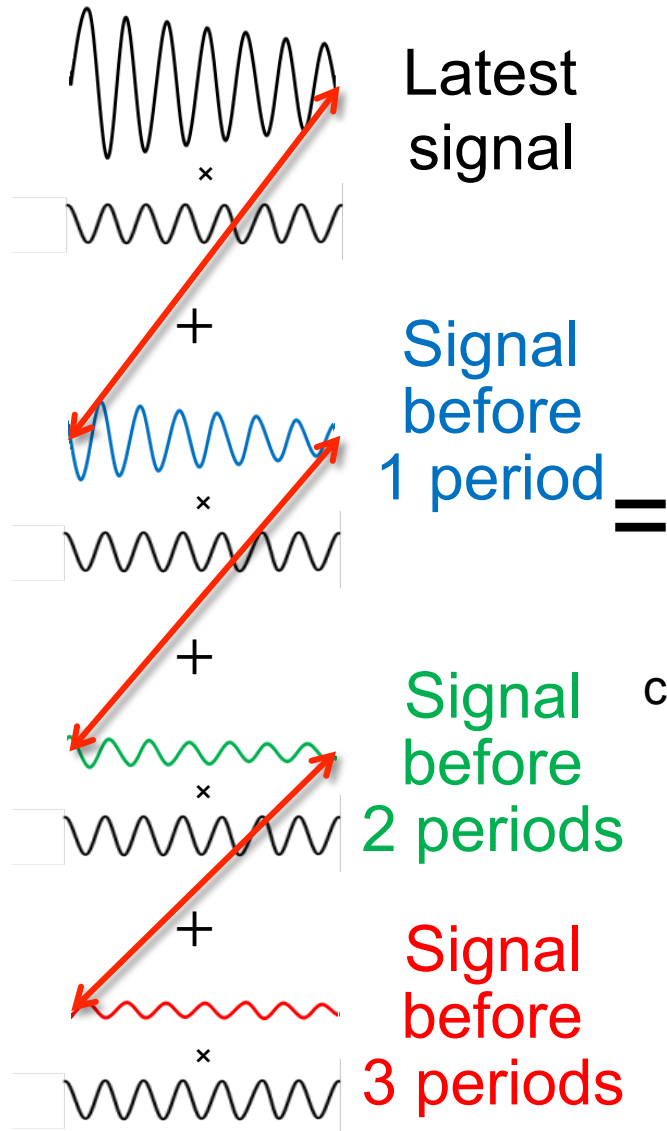
Time window = 1 period



Time window = 0.9995 period



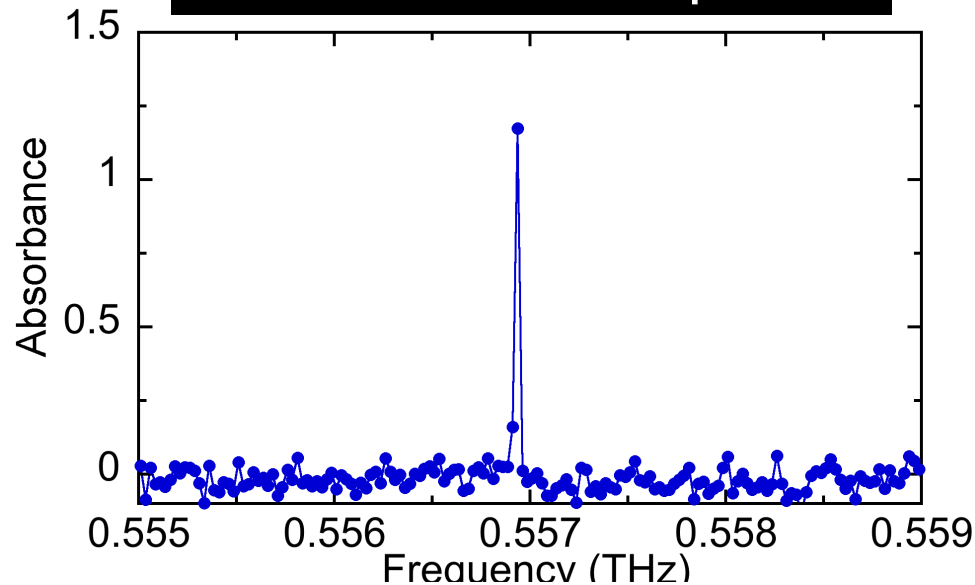
Relation between time window and repetition period (Relaxation time > repetition period)



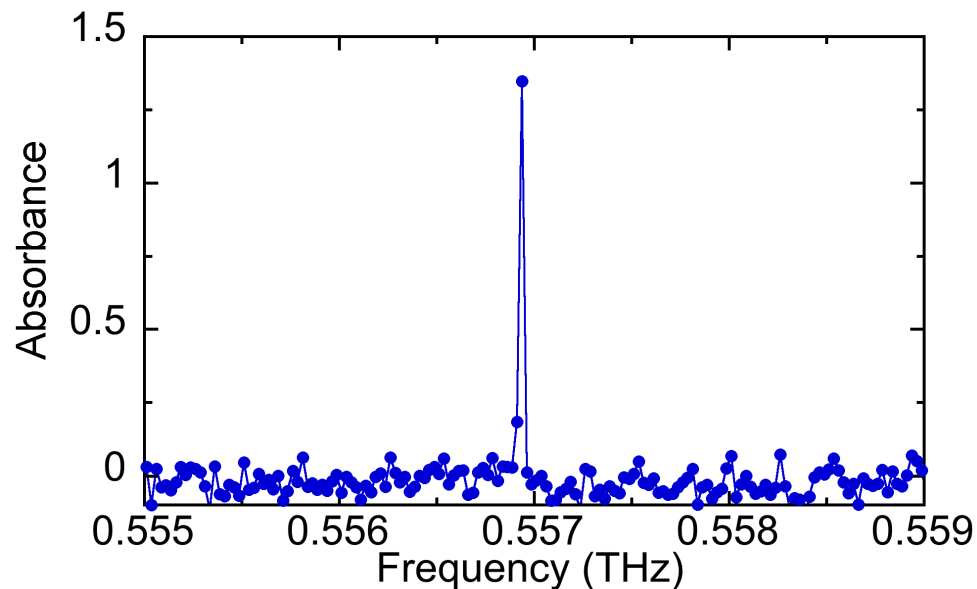
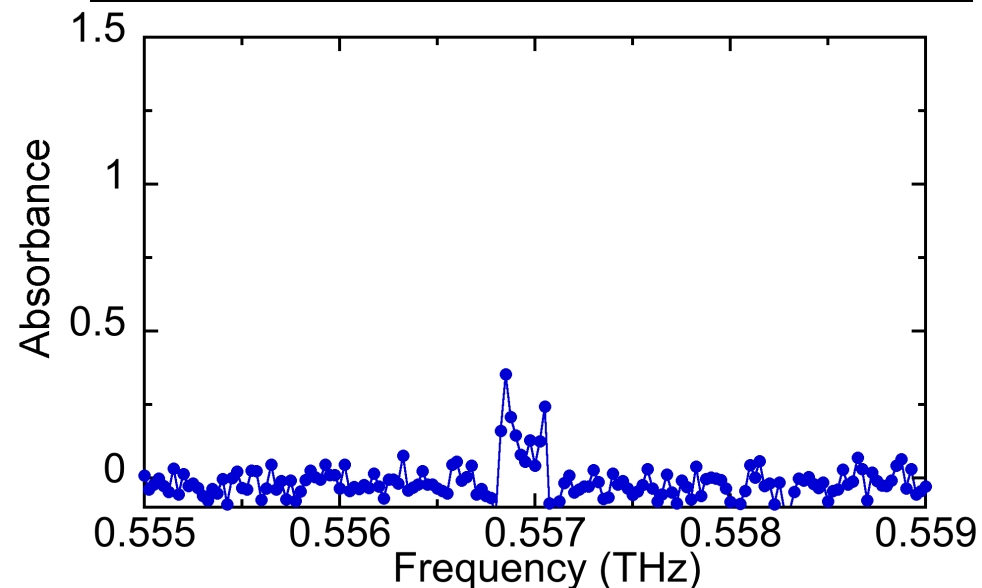
Each piece of different transient signals can not be connected!

Relation between time window and repetition period

Time window = 1 period



Time window = 0.9995 period



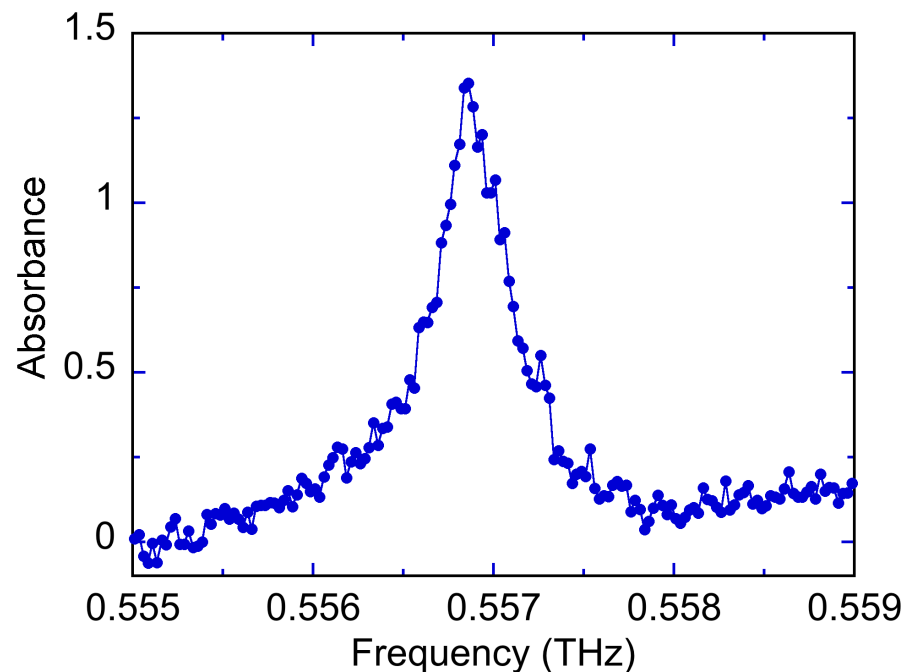
Time window = 1 period

Data for 0.9995period
+
Null for 0.0005period

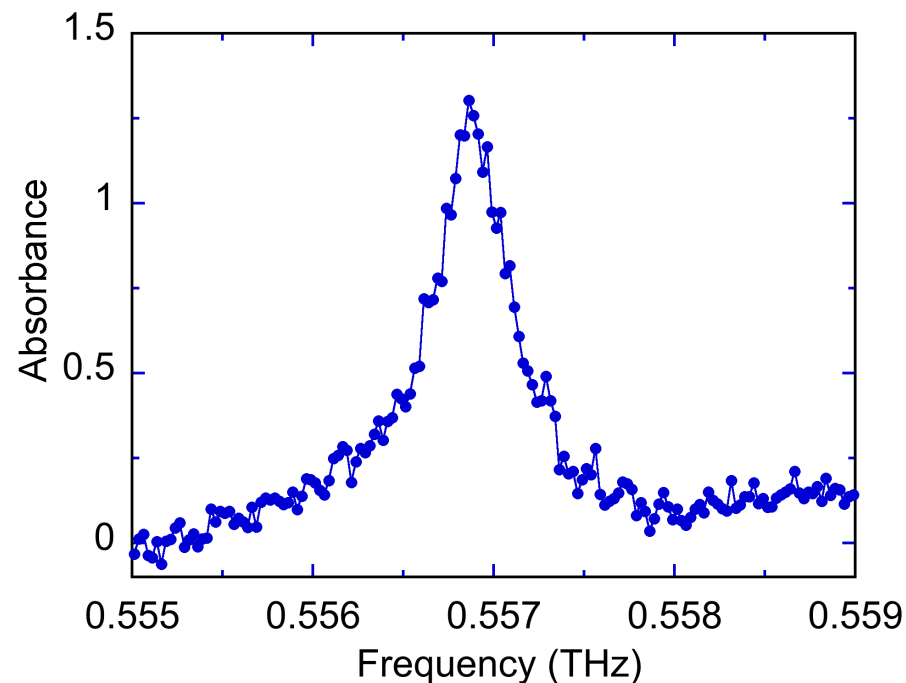
Relation between time window and repetition period (Relaxation time < repetition period)

Rotational transition of H₂O at 0.557 THz (H₂O@1kPa & N₂@3.5kPa)
Expected pressure broadening linewidth = 500 MHz
Relaxation time of absorption = 2 ns (0.5 periods)

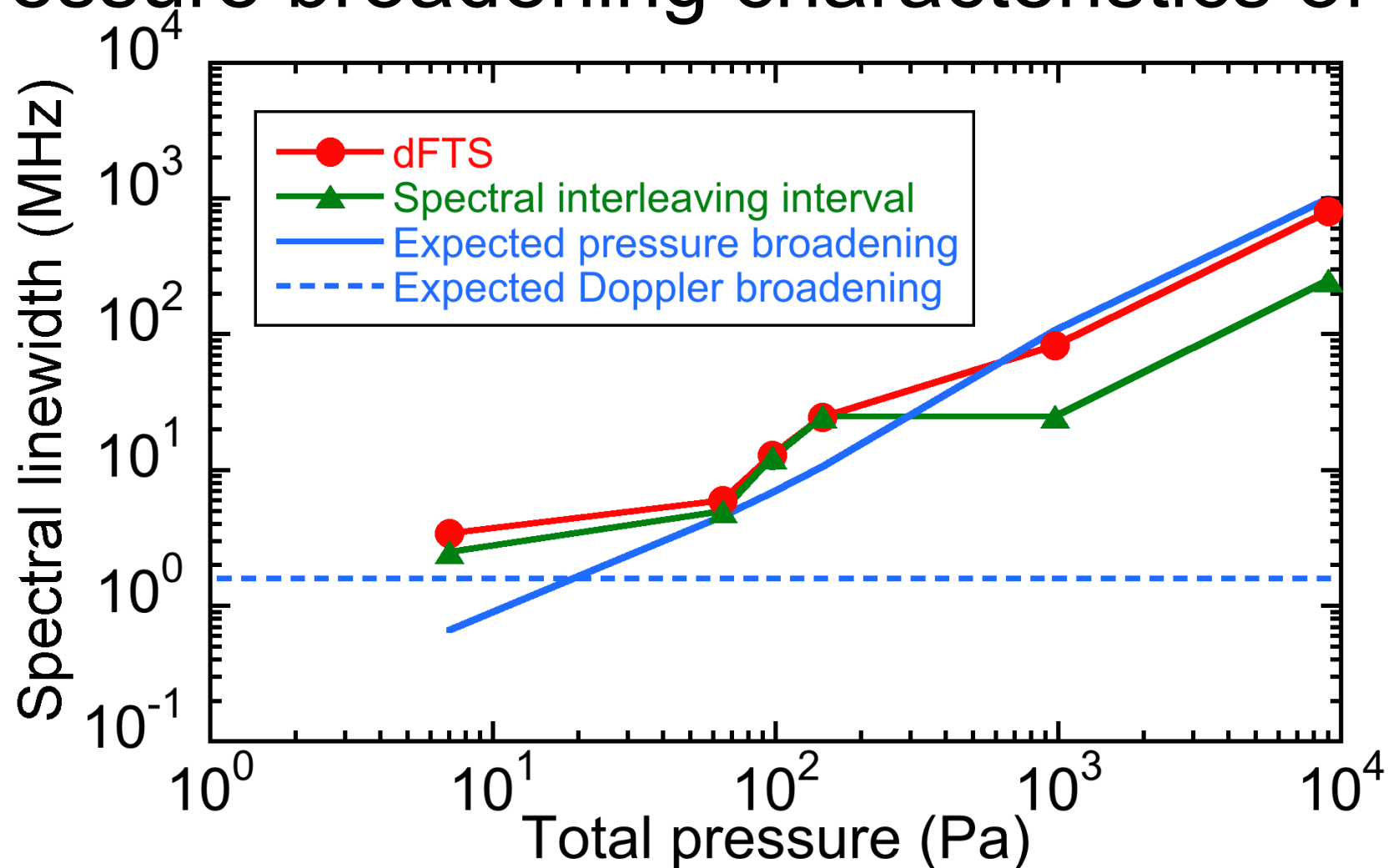
Time window = 1 period



Time window = 0.9995 period



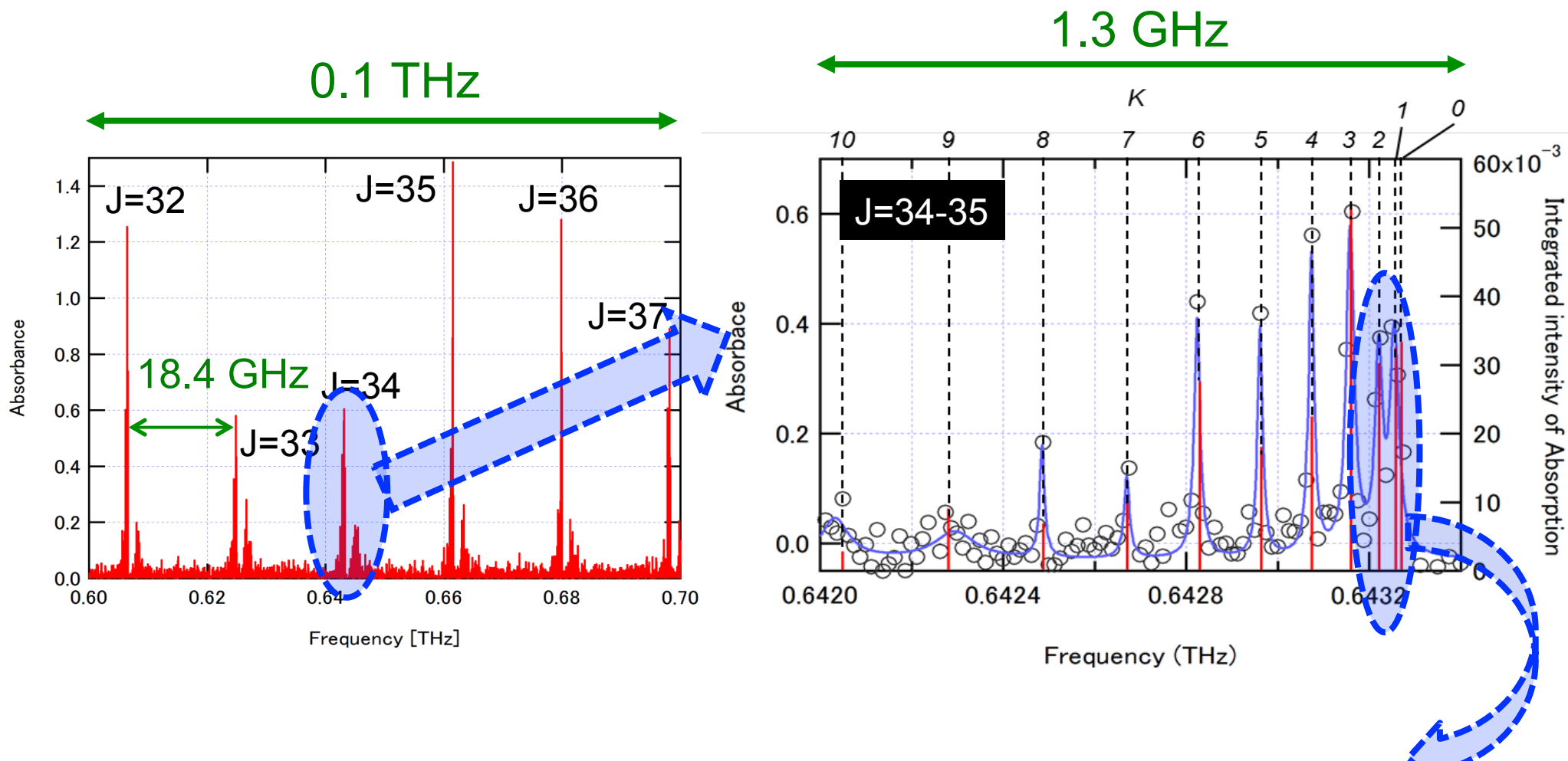
Pressure broadening characteristics of H₂O



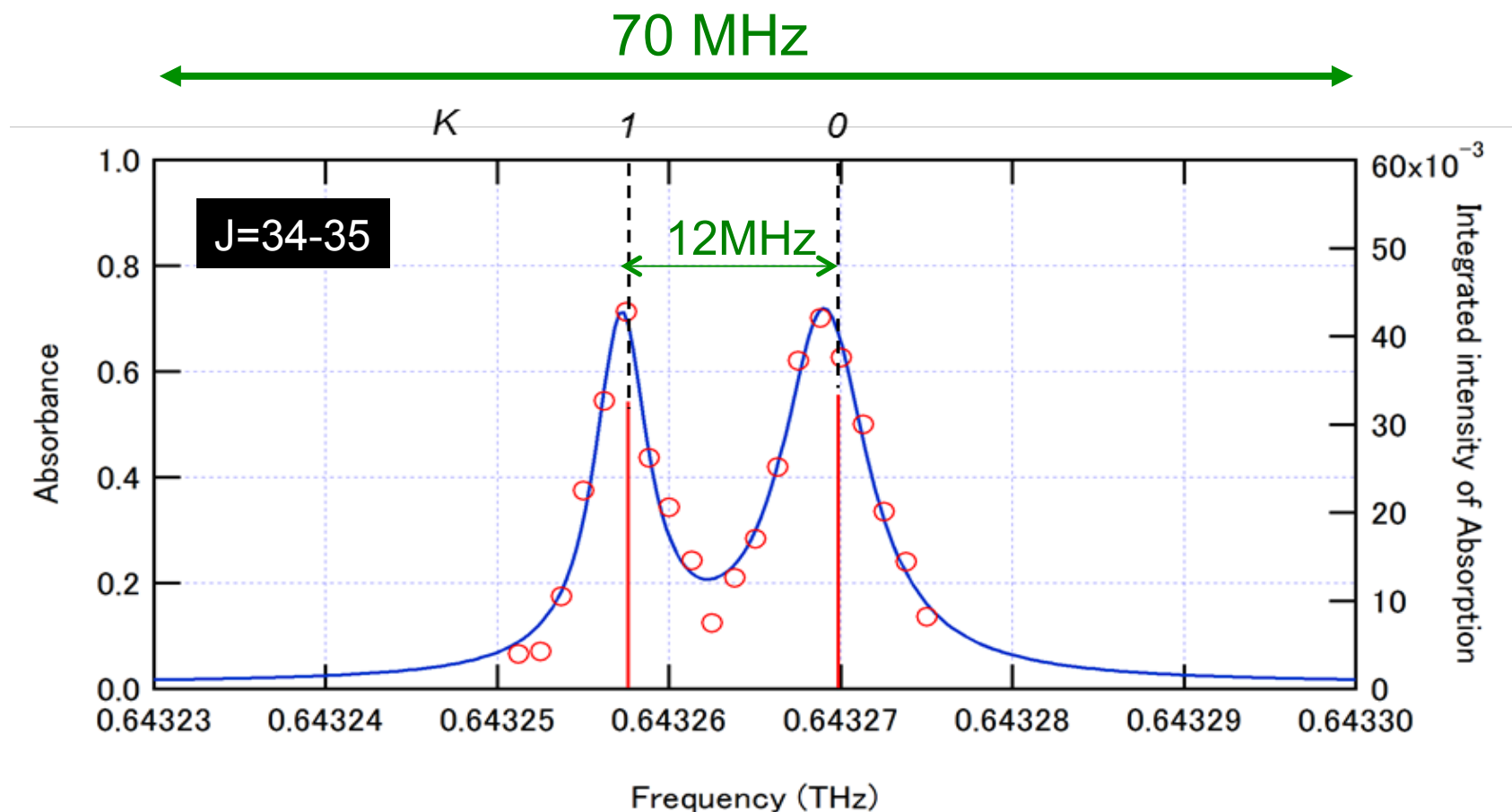
Spectral interleaving interval limits spectral resolution independent of time window size

Spectroscopy of multiple absorption lines

Sample gas: CH₃CN@30Pa
 Symmetric top molecule with rotational constant B of 9.2GHz



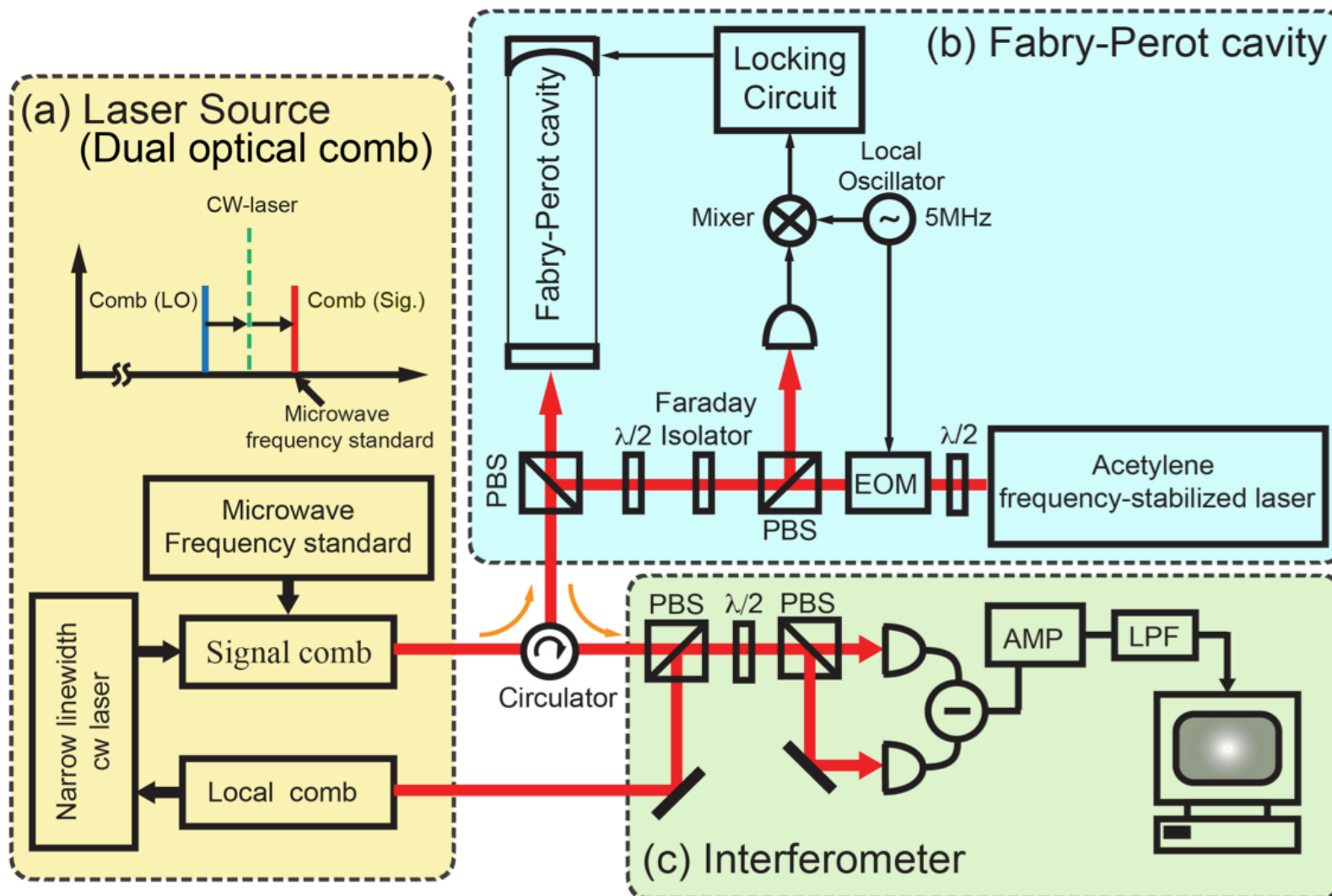
Spectroscopy of multiple absorption lines



K	Literature value (THz)	Experimental value (THz)	Discrepancy (MHz)
1	0.643 257 622 0	0.643 257 498 7	0.12
0	0.643 269 866 7	0.643 268 764 0	1.10

Experimental setup

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$$f_{rep_LO} = 48,350,631 \text{ Hz}$$

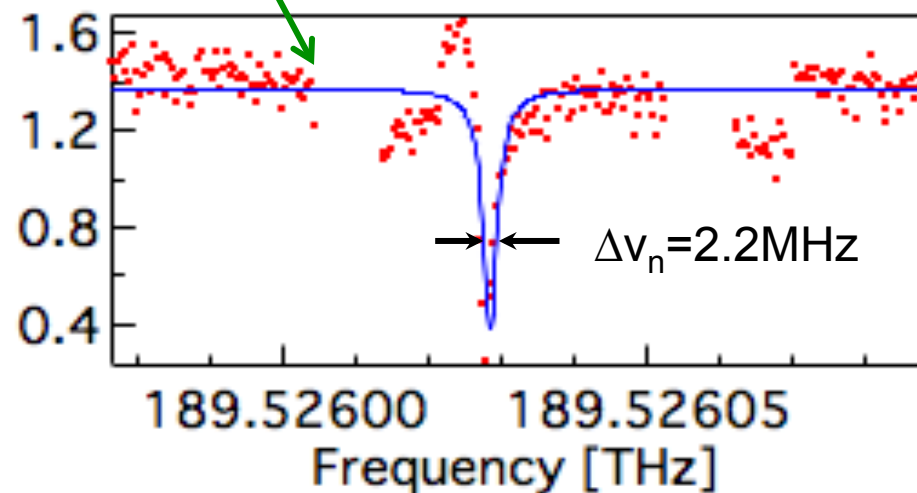
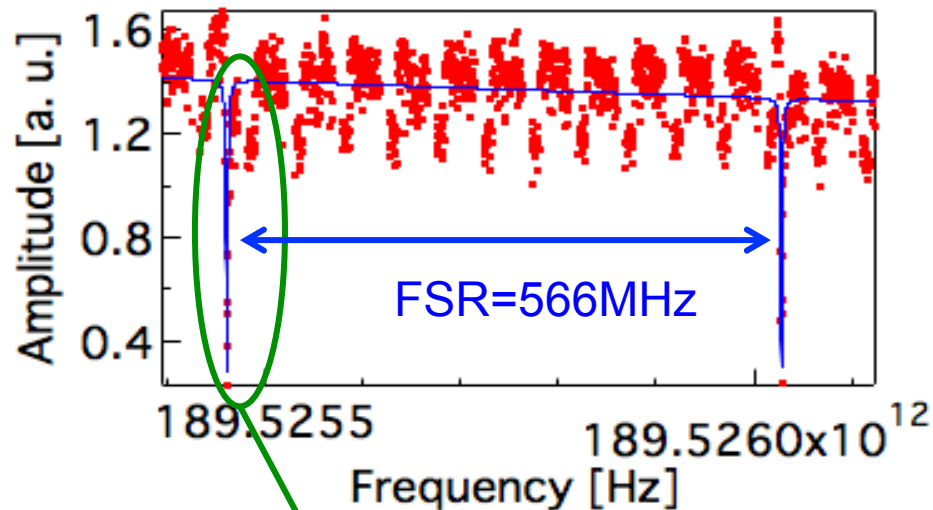
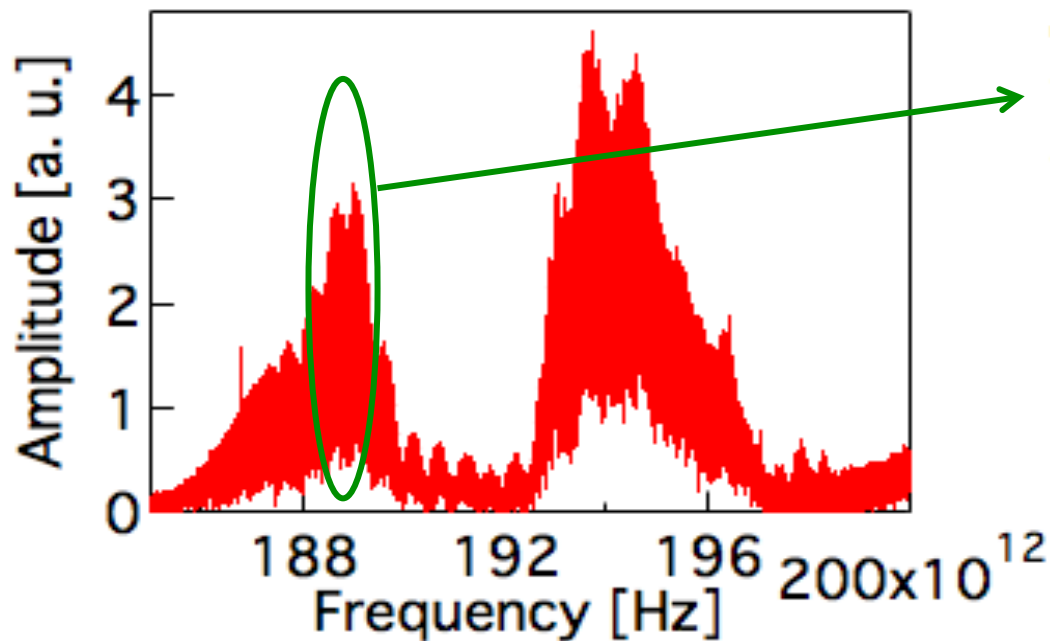
$$f_{rep_S} = 48,350,676 \text{ Hz}$$

$$\Delta f_{rep} = f_{rep_S} - f_{rep_LO} = 45 \text{ Hz}$$

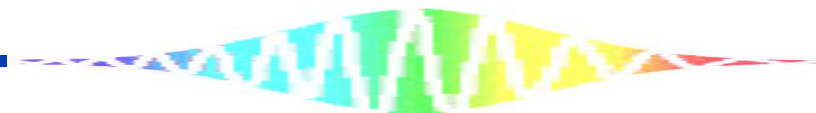
Result

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- $f_{rep, S} = 48.350676\text{MHz} \rightarrow 48.350685\text{MHz}$, 100 times sweep
- $\Delta f = 44.99513\text{Hz}$



The resonance mode in Fabry-Perot cavity



Summary

(1) Intelligent THz instrumentation and application

- Adaptive sampling dual THz comb spectroscopy (CLEO, to be submitted in this year)
- THz-comb-referenced spectrum analyzer (CLEO)

(2) Novel optical comb measurement and application

- Full-field confocal optical-comb microscopy (Patent preparation)
- Discrete Fourier transform infrared spectroscopy (CLEO)