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 (~1MHz)

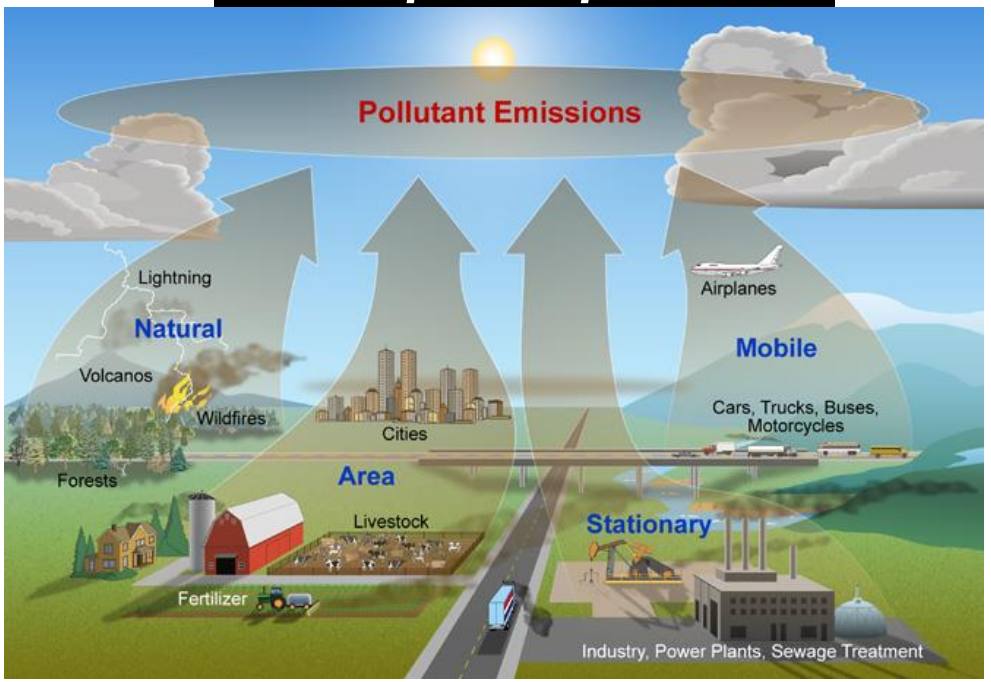
☞Multiple gas analysis

(2) Less scattering

- ✓ λ_{THz} >> particle diameter

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

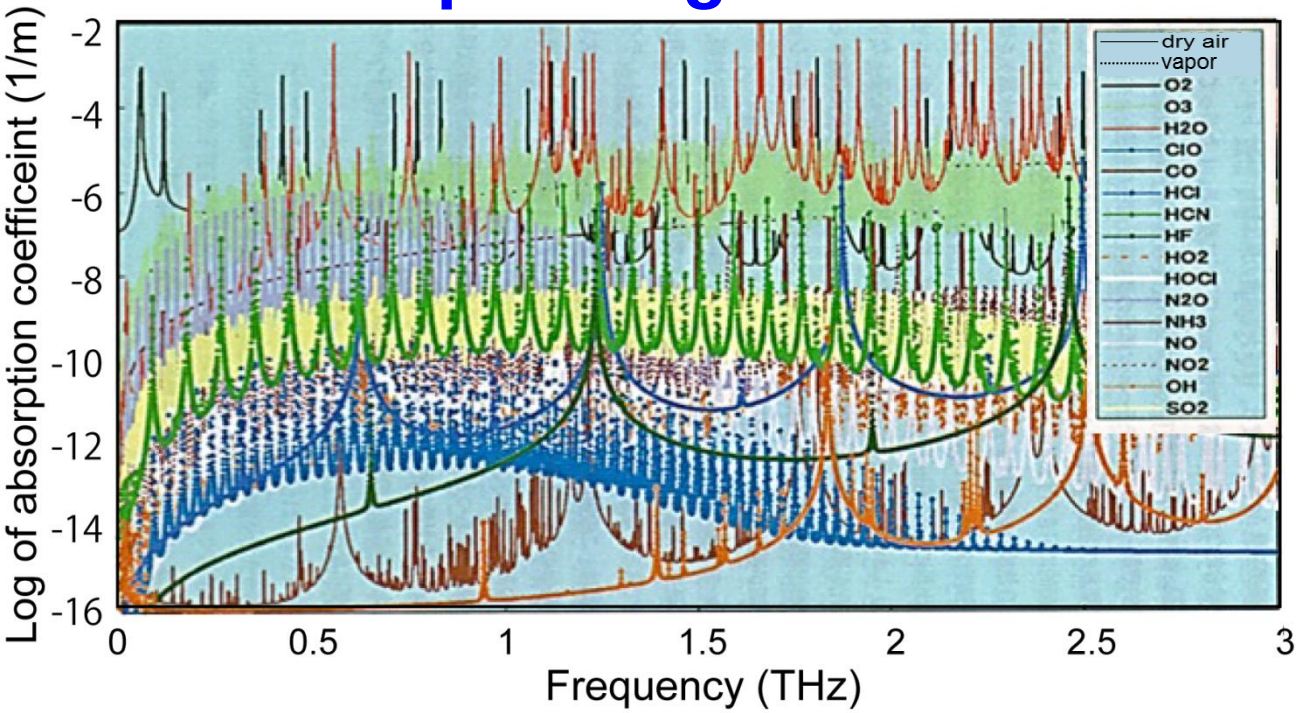
Atmospheric pollution



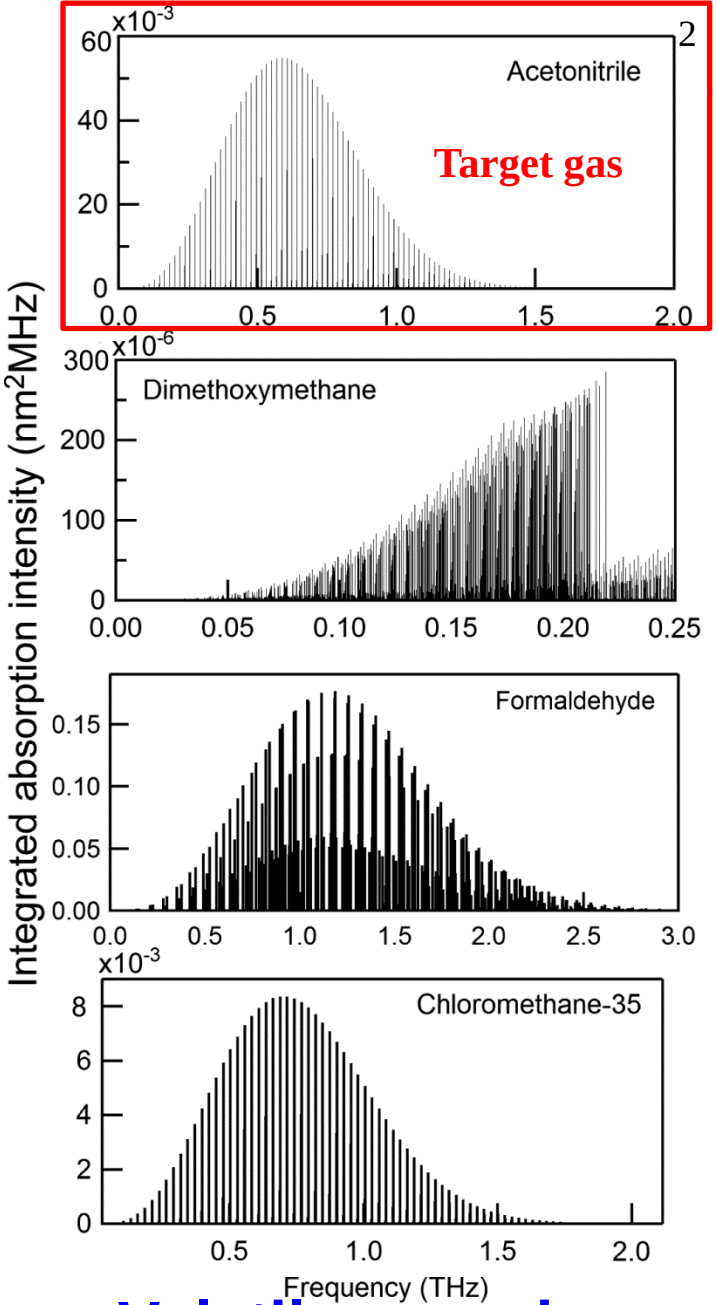
Ref)http://www.nature.nps.gov/air/aqbasics/sources.cfm

THz spectral fingerprints

Atmospheric gas molecule

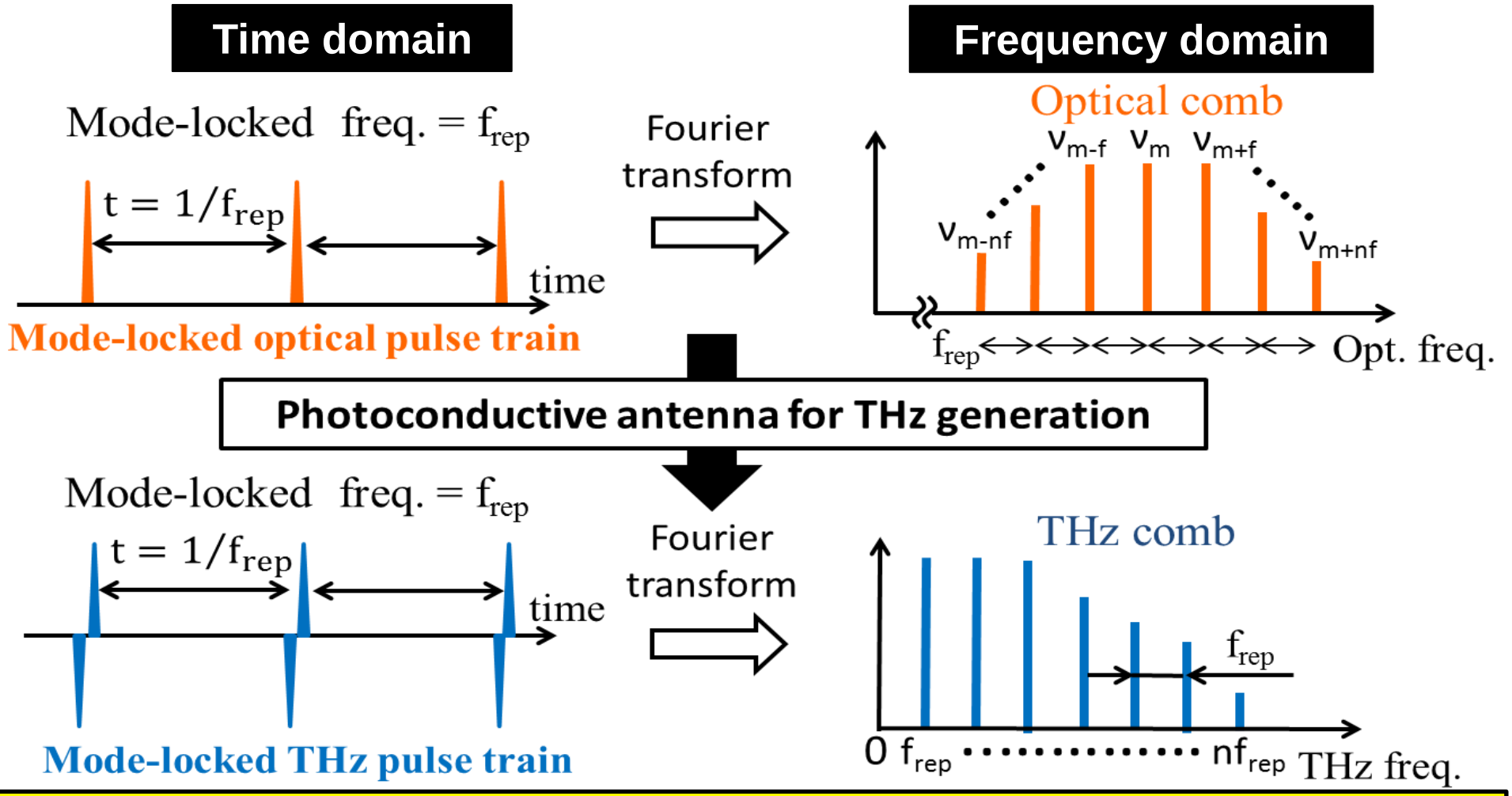


To discriminate the target gas correctly, high resolution, high accuracy, and broadband spectrum are required!!



Volatile organic compound (VOC) gas

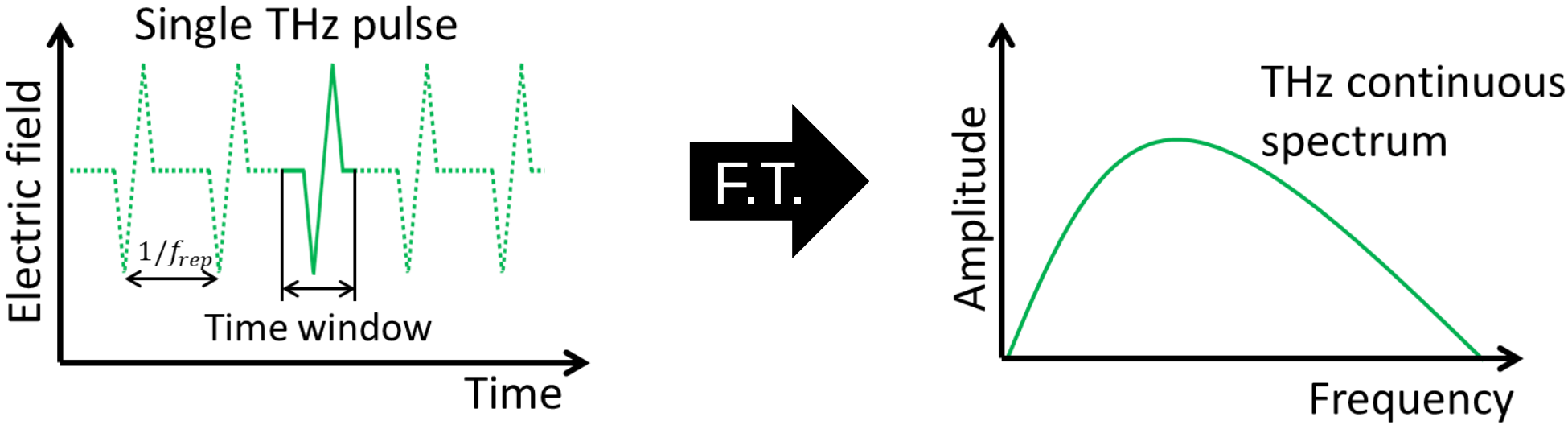
Optical comb & THz comb



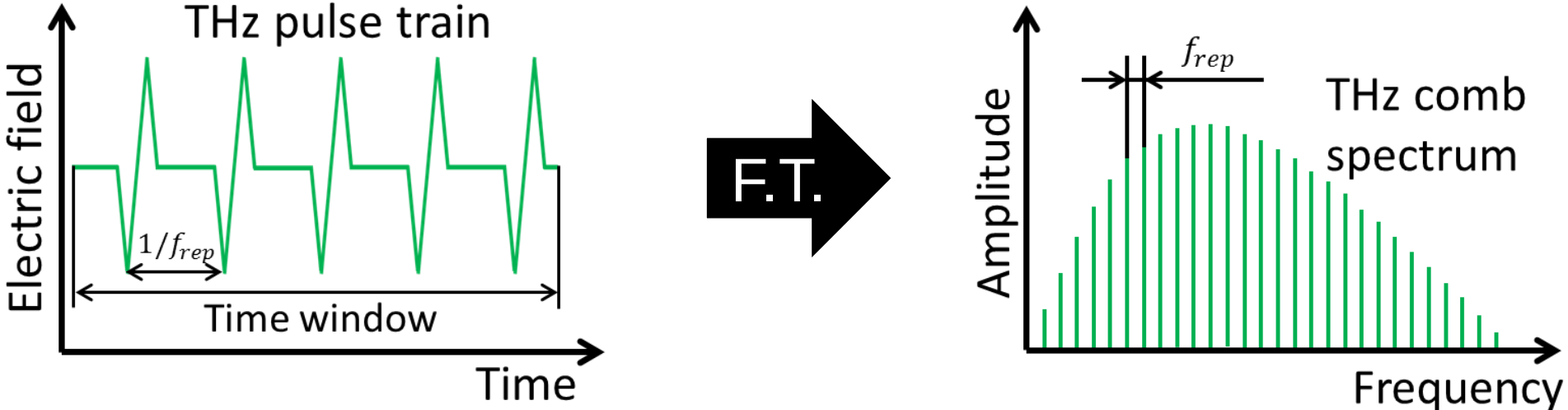
Simplicity, broadband selectivity, high spectral purity, and absolute frequency calibration

How to measure THz comb

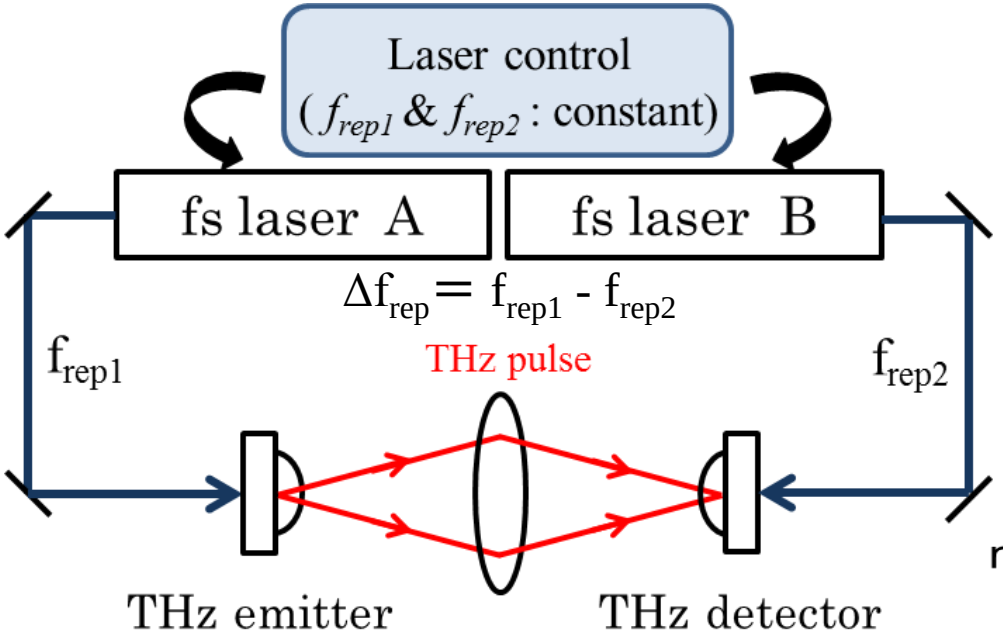
Traditional THz-TDS equipped with mechanical time-delay scanning



Time-window-extended THz-TDS

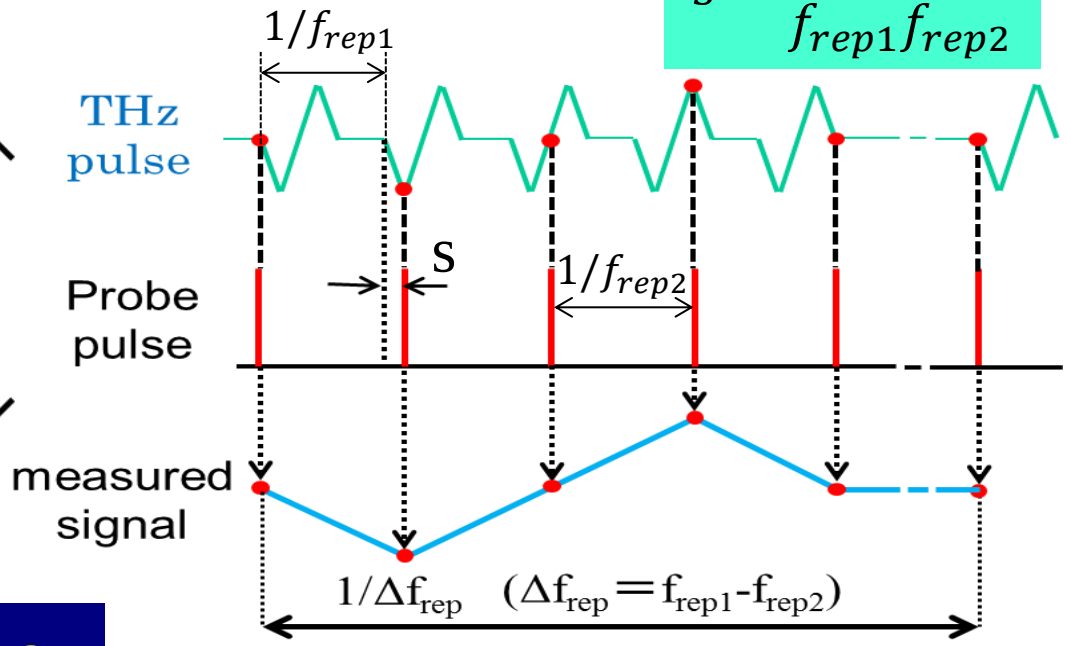


Asynchronous-optical-sampling THz-TDS (ASOPS-THz-TDS)



Overlap timing between THz and probe pulses automatically shifts for each pulse

$$s = \frac{\Delta f_{rep}}{f_{rep1} f_{rep2}}$$



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

Time scale of ps THz pulse is linearly expanded to μs order

Temporal magnification factor (TMF) = $\frac{f_{rep1}}{\Delta f_{rep}}$

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

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

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Expand the application fields of dual THz comb spectroscopy

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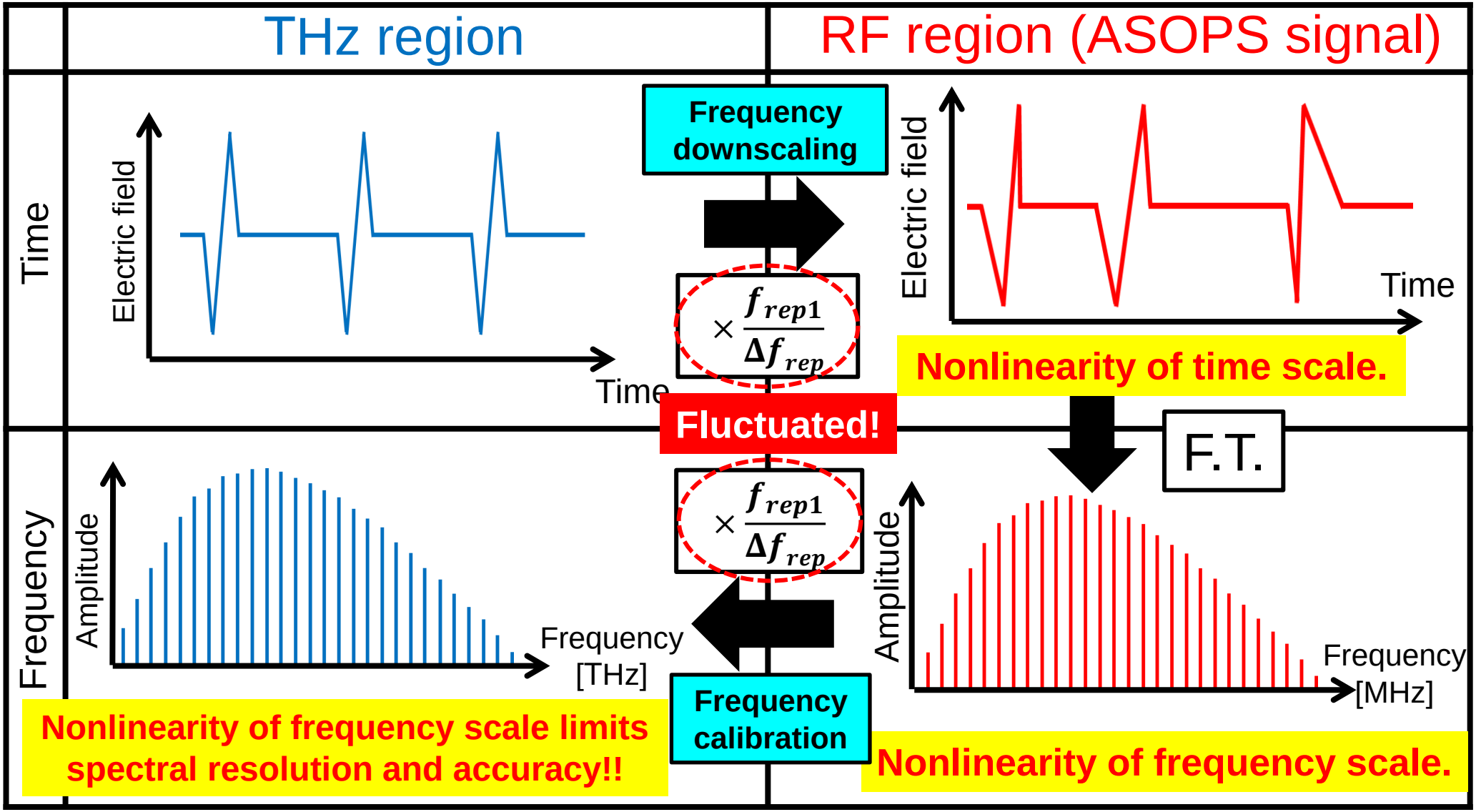
However, timing jitter between free-running dual fs lasers distort the linearity of time and frequency scales due to fluctuation of TMF!

Influence of timing jitter in ASOPS

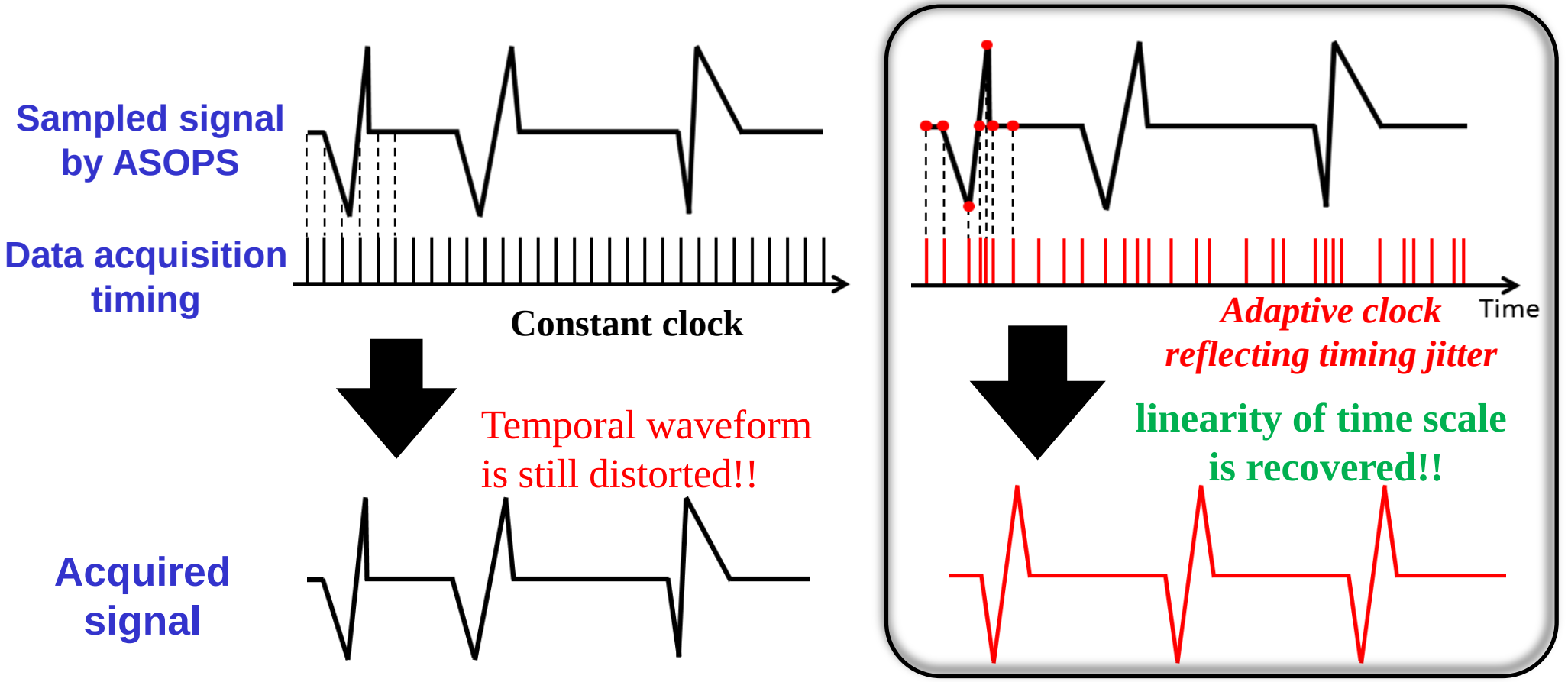


Adaptive sampling method

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

Conventional method (constant sampling)

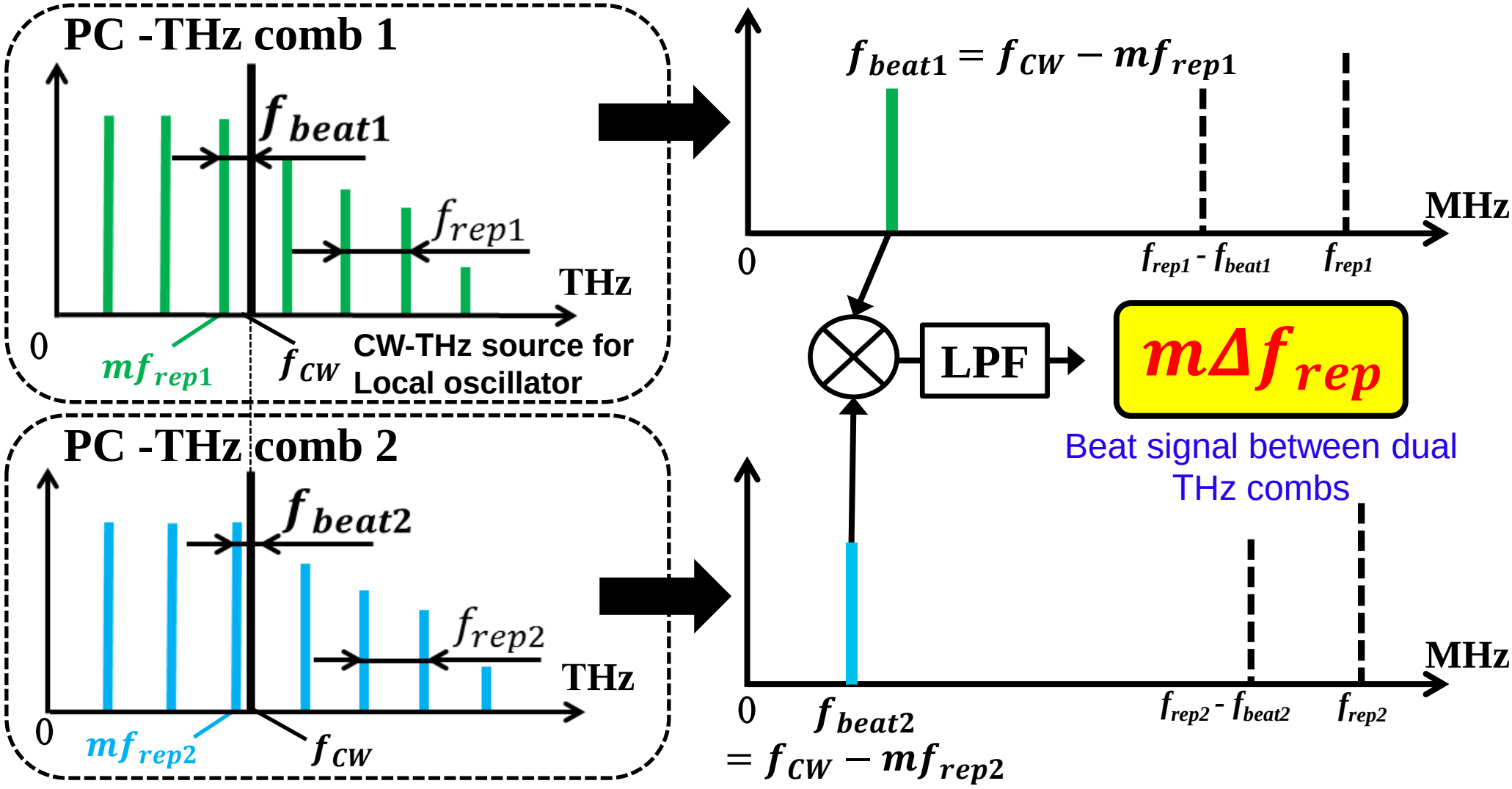
Proposed method (Adaptive sampling)



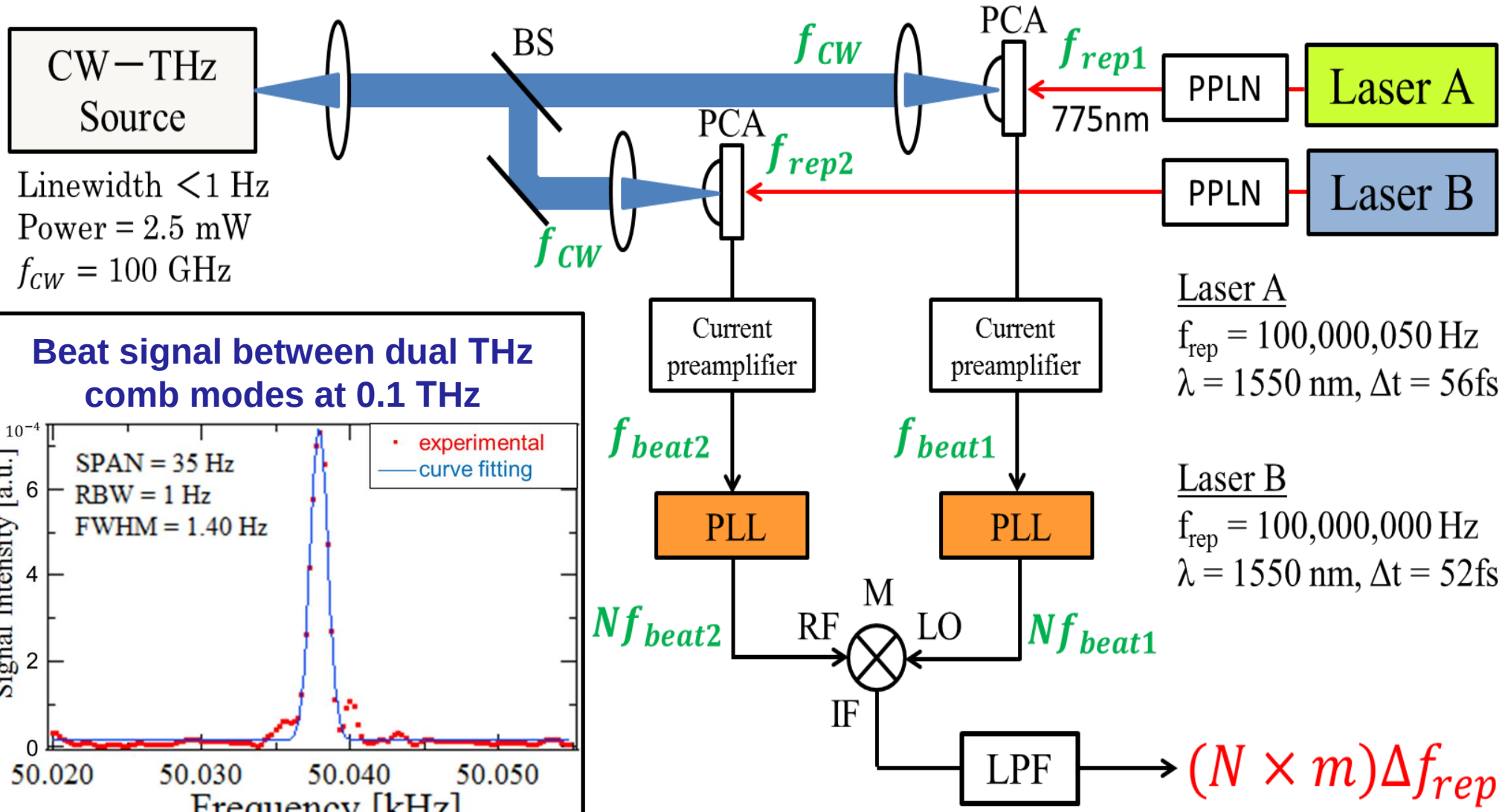
Adaptive clock can be generated by beat signal between dual THz combs!!

Beat signal between dual THz combs for adaptive sampling clock

ref) Shuko Yokoyama et al.,Optics Express, Vol. 16, Issue 17, pp. 13052-13061 (2008)

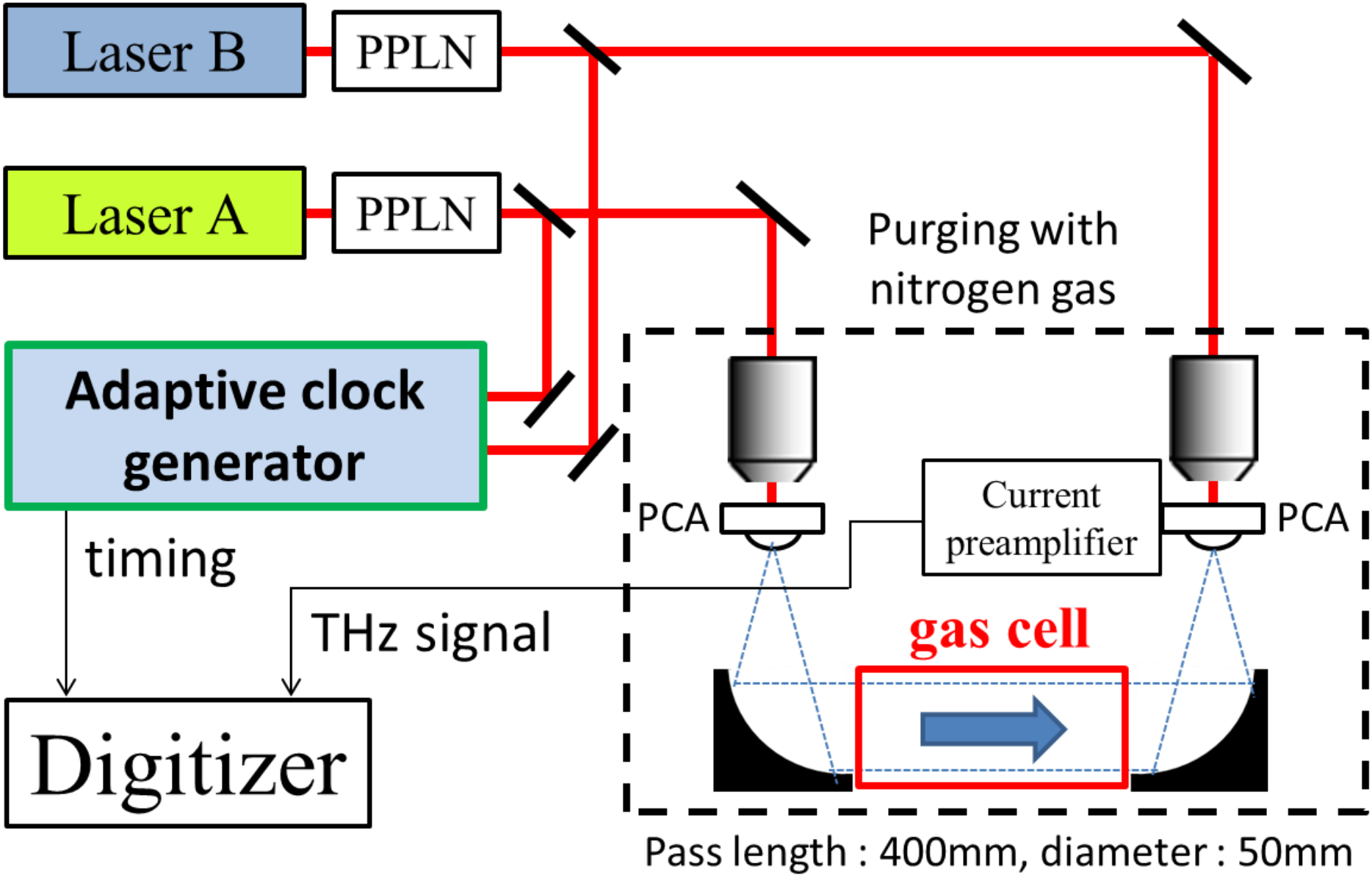


Experimental setup to extract the beat signal between dual THz combs



Beat signal between dual THz comb modes at 0.1 THz

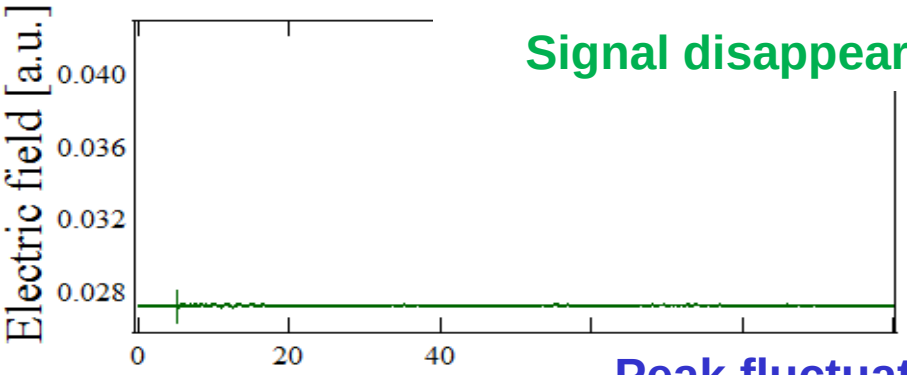
Setup for dual THz combs spectroscopy



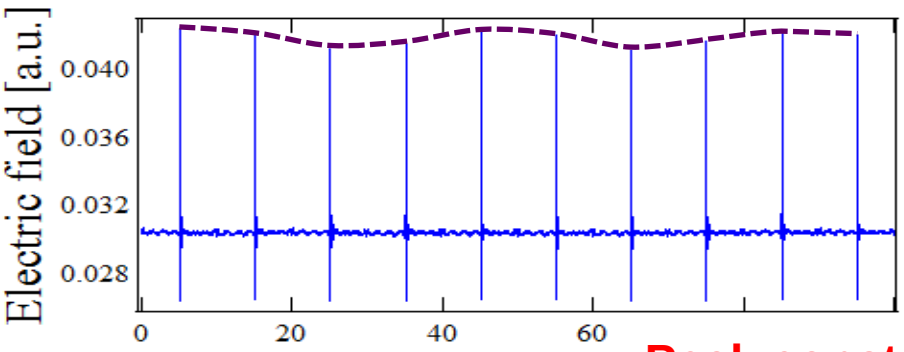
Integrated Temporal Waveform of 10 THz pulses

Integration number : 10000

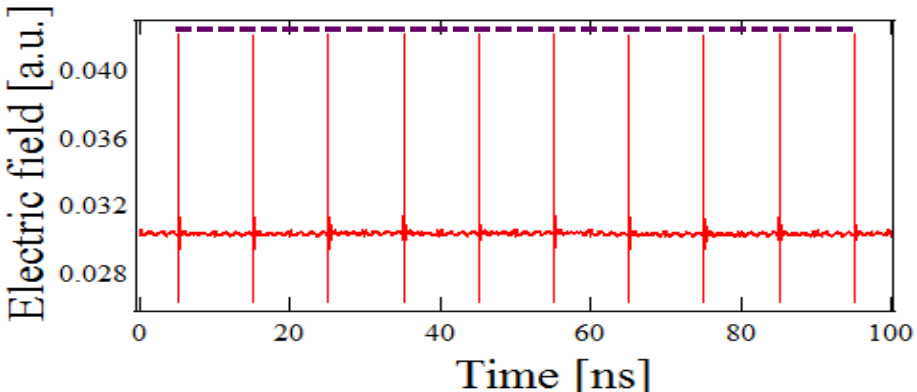
Constant clock
f_{rep1} & f_{rep2} free-running



Constant clock
f_{rep1} & f_{rep2} stabilized



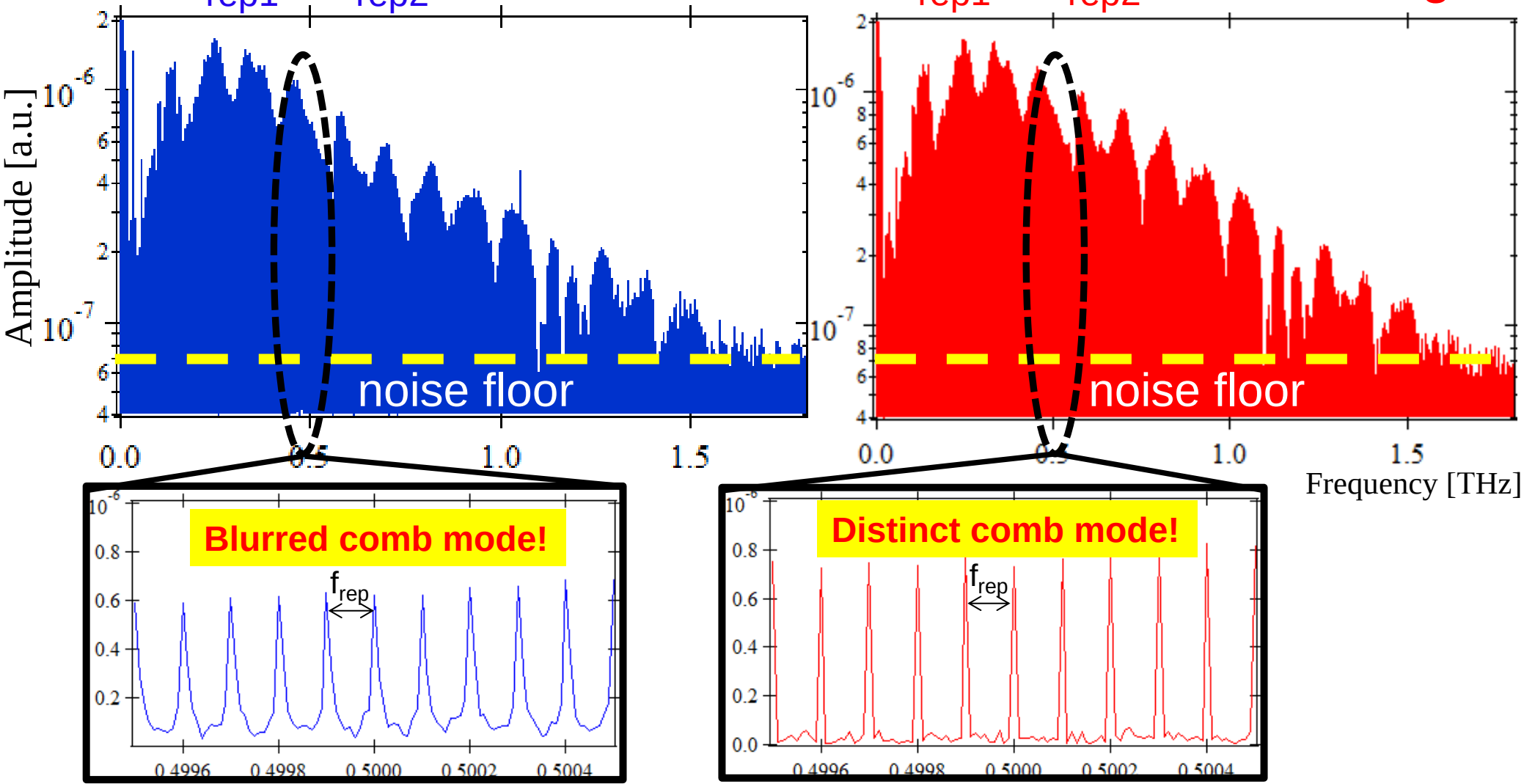
Adaptive clock
f_{rep1} & f_{rep2} free-running



THz comb spectrum

Constant clock
f_{rep1} & f_{rep2} stabilized

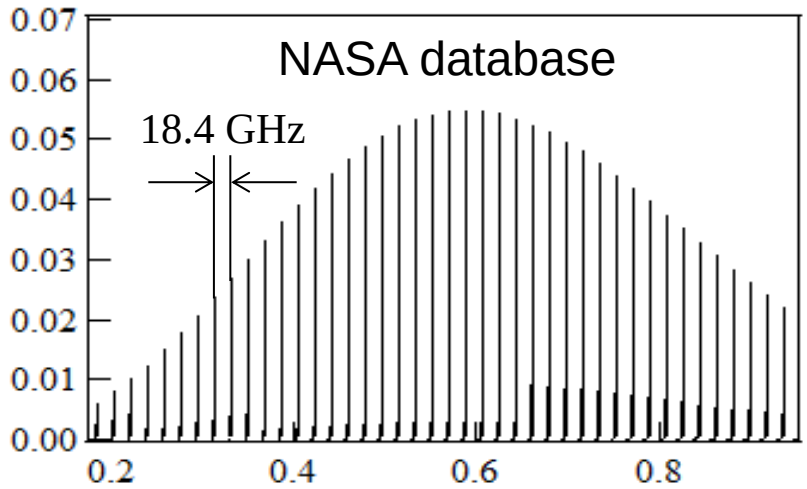
Adaptive clock
f_{rep1} & f_{rep2} free-running

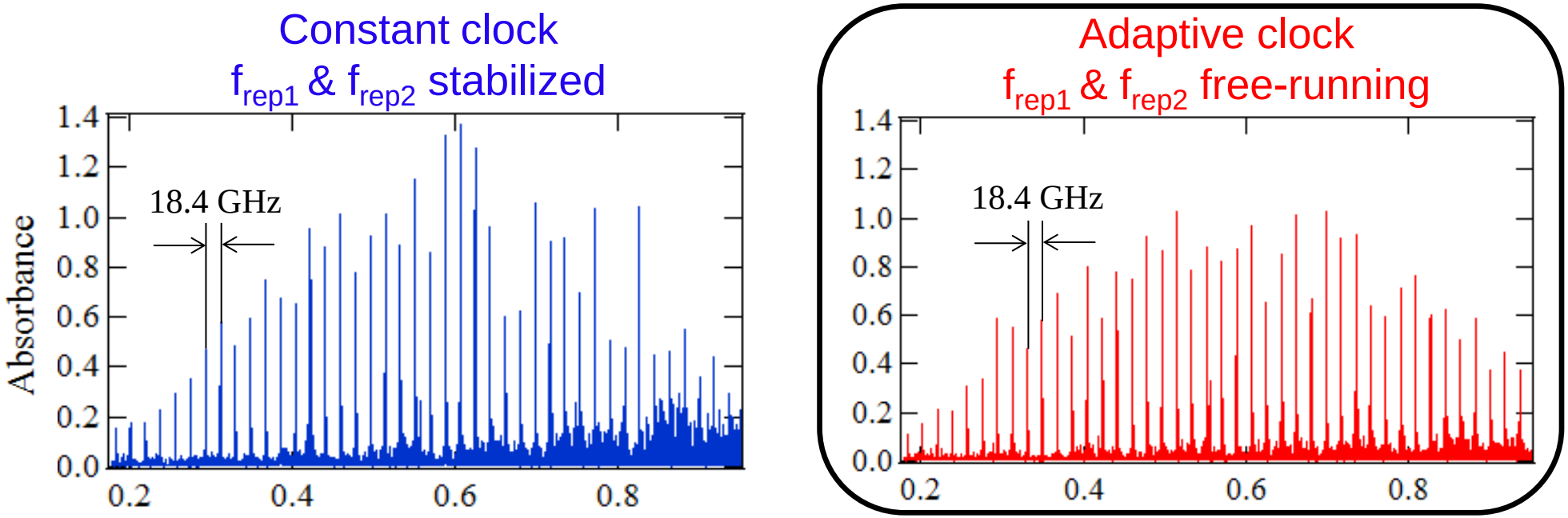


Acetonitrile gas at low pressure(1kPa)

Acetonitrile (CH_3CN)

- One of VOCs
- Very abundant species in interstellar medium





Summary

1. Dual THz comb spectroscopy was attained using free-running dual fs lasers.

2. Spectroscopic performance:
   Adaptive sampling with free-running dual lasers
   ≧*Constant sampling with stabilized dual lasers*

3. Possible to perform precision spectroscopy of low-pressure gas molecule using free-running dual fs lasers