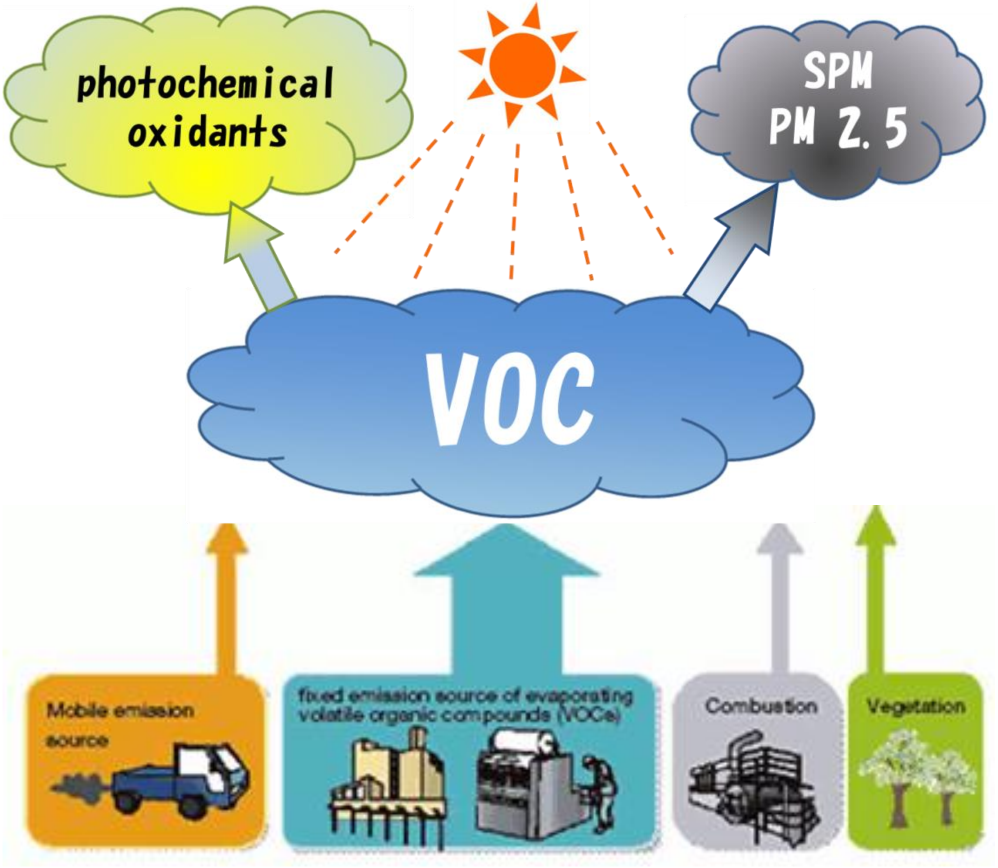
Adaptive Sampling Dual THz Comb Spectroscopy

8/4 M2 Ichikawa

Air pollution by VOCs



From : Environment of Tokyo

Primary air pollutants

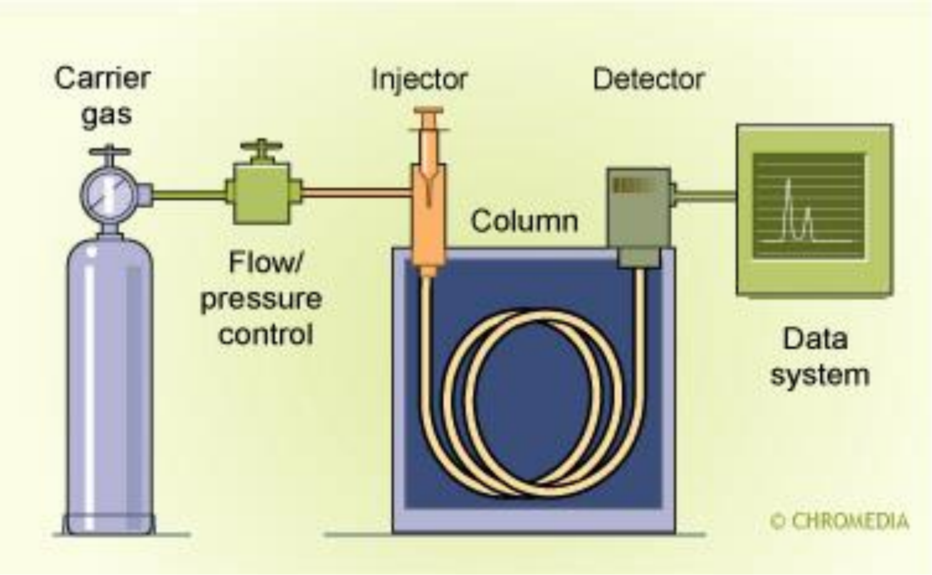
Nitrogen dioxide (NO_x)
Ozone (O_3)
Sulfur dioxide (SO_2)
Small particulate matter (SPM)
Carbon monoxide (CO)
Lead (Pb)
Volatile organic compounds(VOCs)

VOC is the general term of organic compounds that become gaseous in the atmosphere

The instrumental analysis of VOC gases is important to pollution control.

Conventional techniques

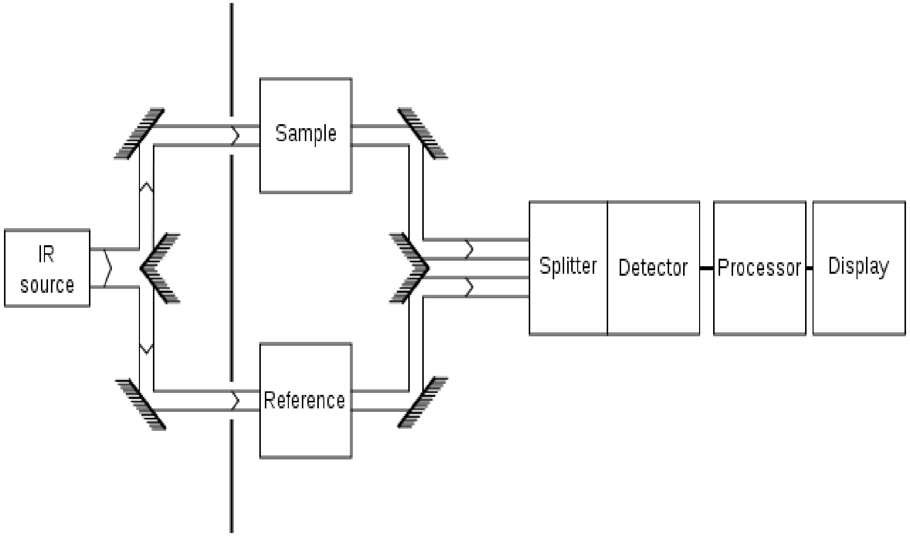
Gas chromatography



From : htto://chromedia.org/

Advantage : High resolution, sensitivity
Disadvantage : Need the skills for instrumental analysis,
Long measurement time, Sample preparation

Infrared spectroscopy



From : en.wikipedia.org

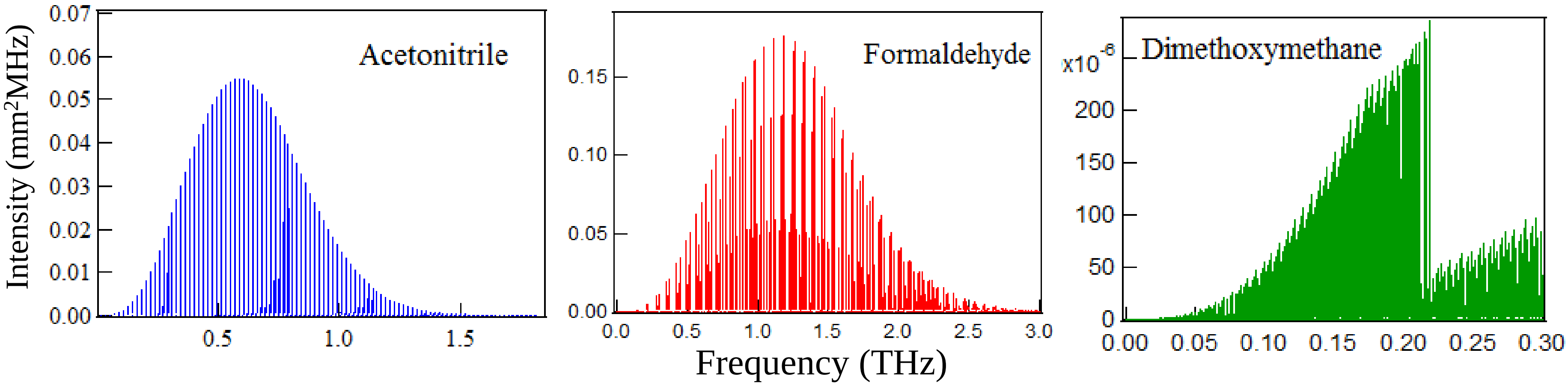
Advantage : High speed, Broadband spectrum
Disadvantage : Scattering by aerosol, Low sensitivity

With conventional techniques, it was difficult to analyze VOC gases directly without preparing samples in advance.

THz gas spectroscopy

(1)Rotational transition of polar molecules.

Rich spectral fingerprints, high discrimination, high sensitivity



THz Spectral fingerprints of VOC gases

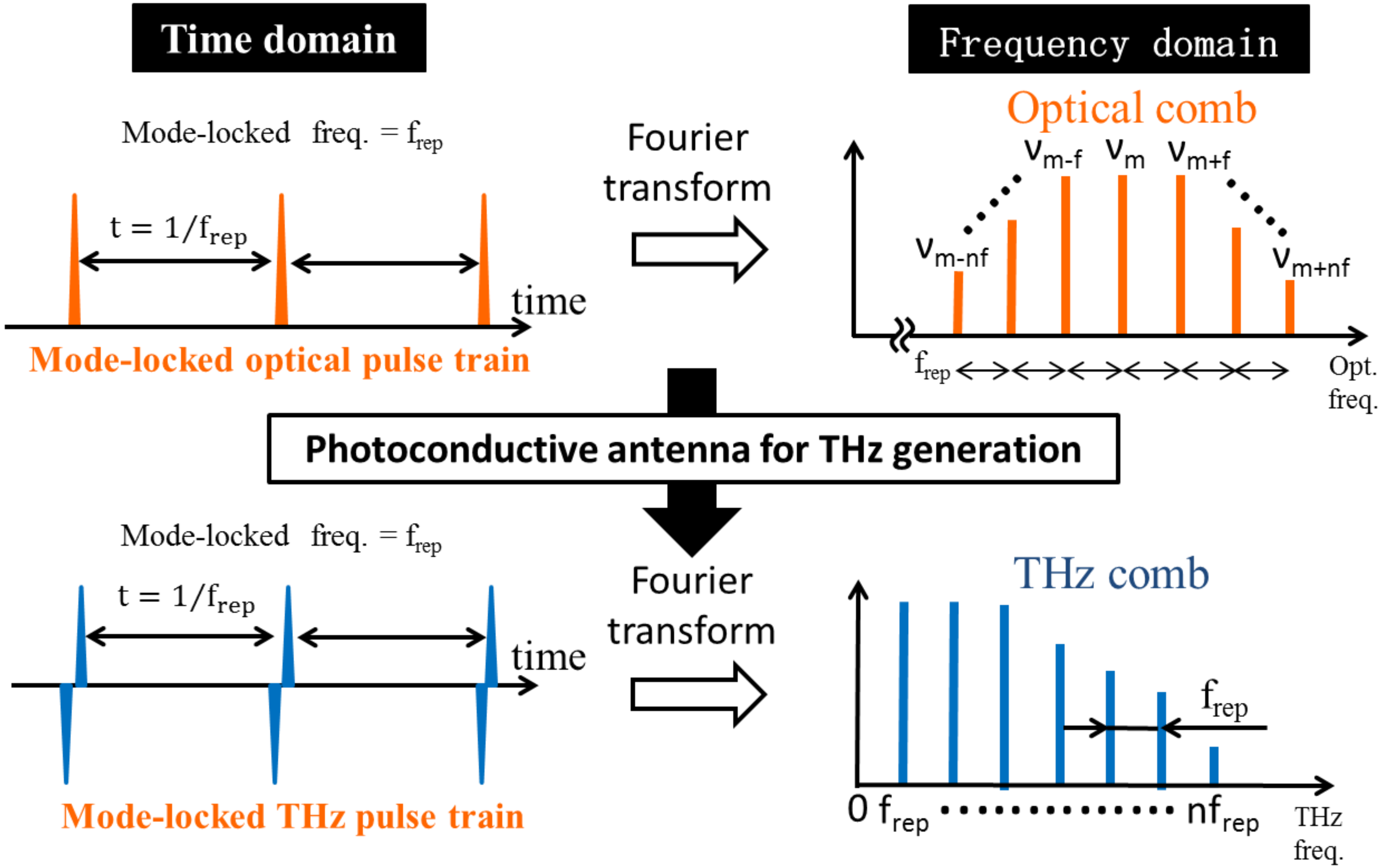
(2)Reduced scattering in small particles

$\lambda_{THz} >>$ particle diameter

possible to analyze gas molecules mixed with aerosols, fog, cloud, smoke, etc…

THz spectroscopy has high potential for analysis of VOC gas. However, spectral resolution, spectral accuracy and broadband spectral are required!!

Optical comb & THz comb



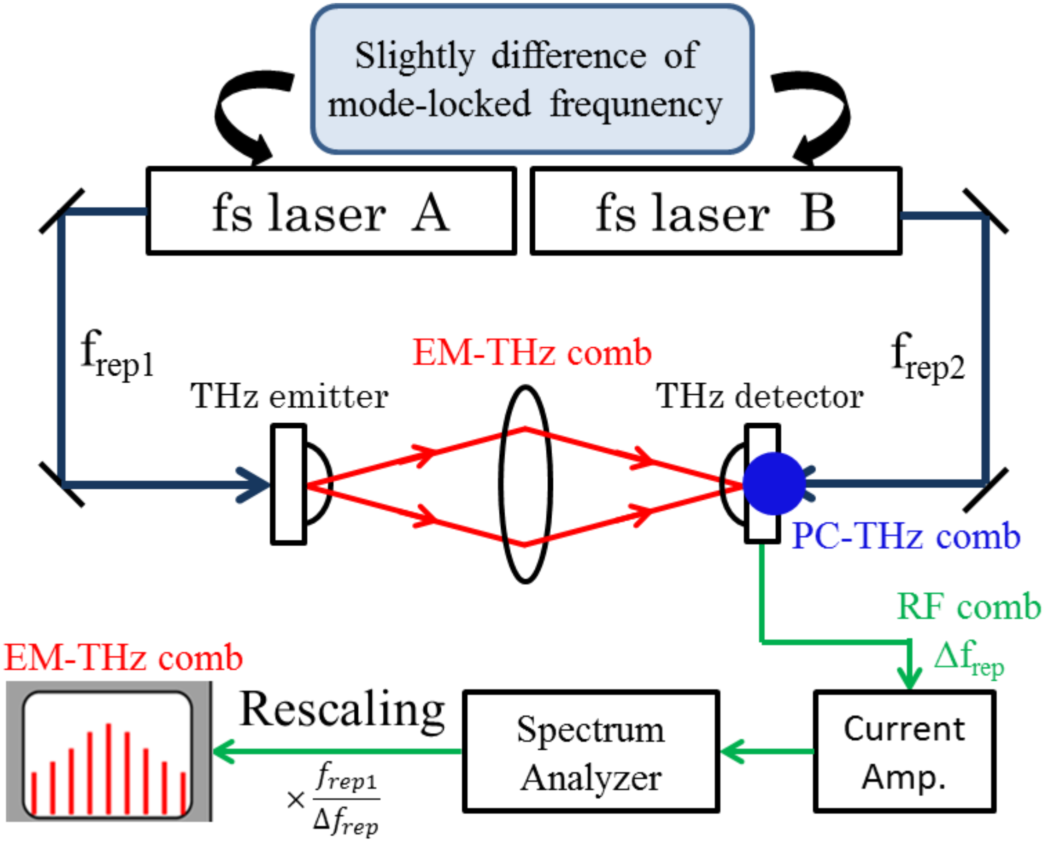
Time domain

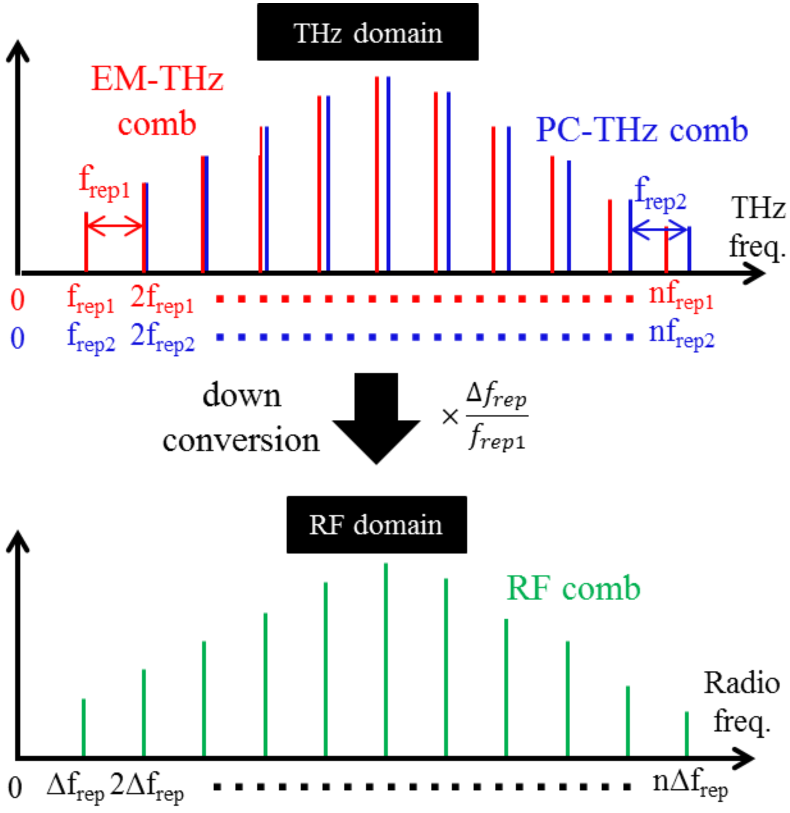
Frequency domain

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

Dual THz Comb Spectroscopy

Frequency Domain

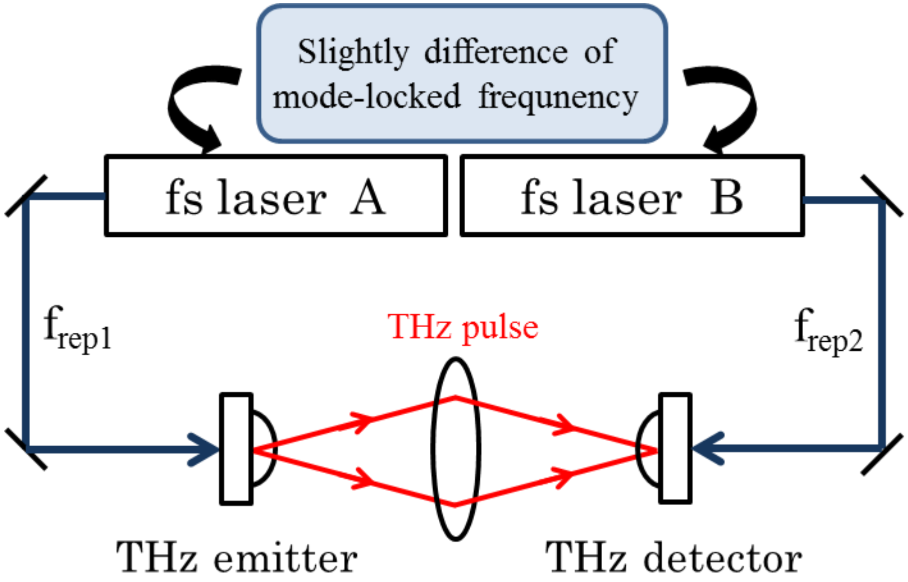




CW-THz波の狭線幅特性とTHzﾊﾟﾙｽの広帯域ｽﾍﾟｸﾄﾙ特性の融合
マイクロ波周波数標準へのトレーサビリティ

Dual THz Comb Spectroscopy

Time Domain (ASOPS-THz-TDS)



Slightly difference of mode-locked frequency

fs laser A

fs laser B

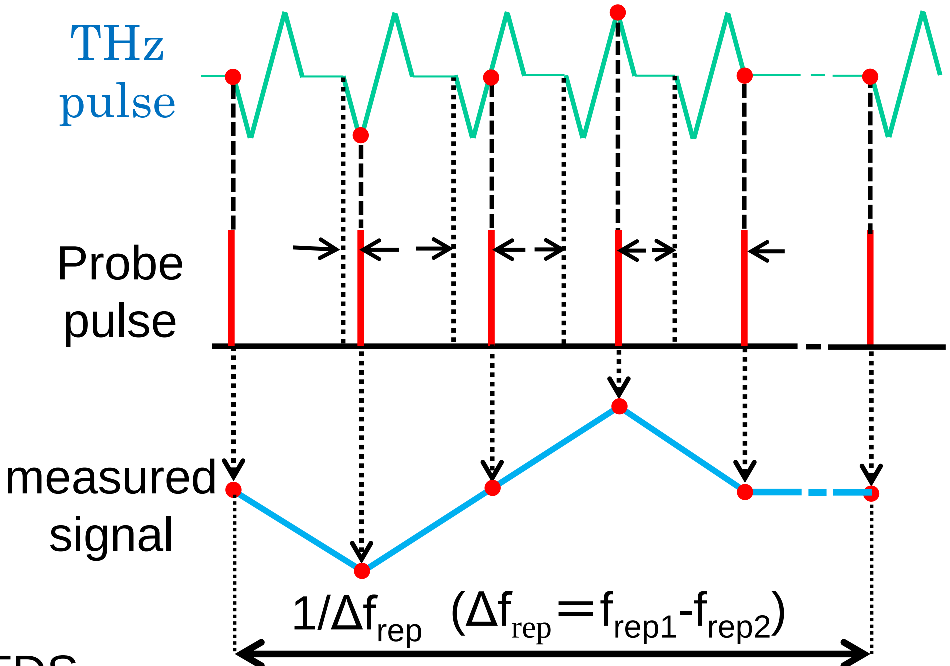
f_{rep1}

THz pulse

f_{rep2}

THz emitter

THz detector



THz pulse

Probe pulse

measured signal

$1/\Delta f_{rep}$ $(\Delta f_{rep} = f_{rep1} - f_{rep2})$

Comparison with conventional THz-TDS based on mechanical stage…

- No moving parts.
- High spectral resolution.
- Measurement time is very fast.

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

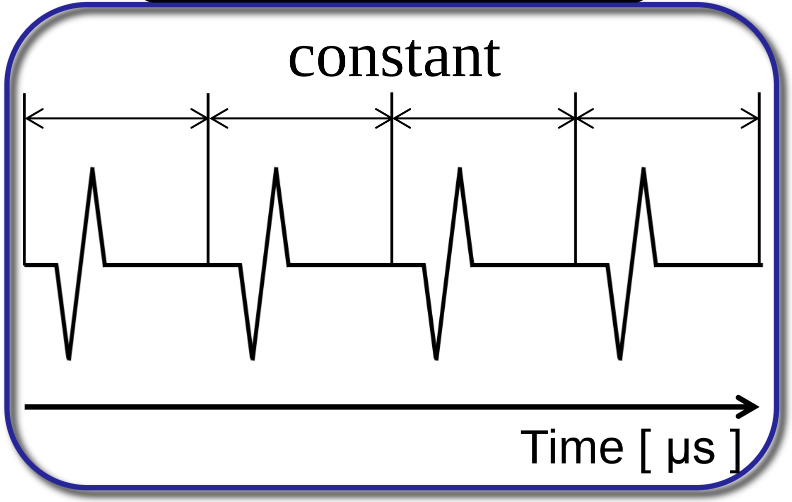
Conversion factor $\frac{f_{rep1}}{\Delta f_{rep}}$

temporal waveform is expanded to the micro second order.

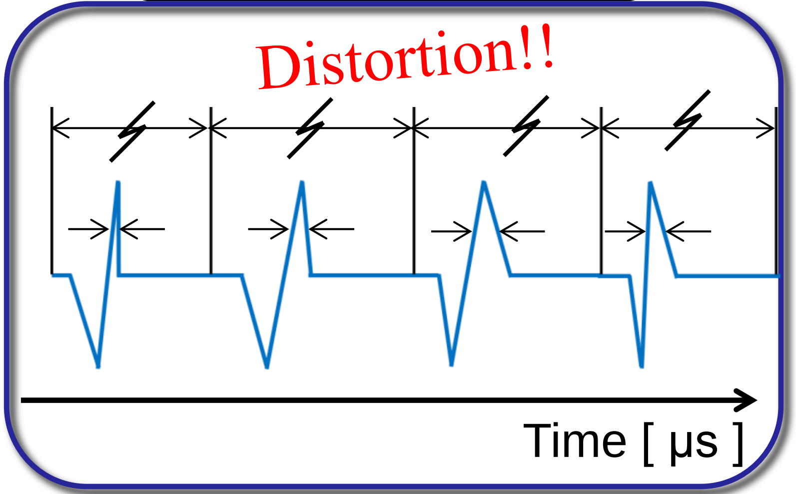
Adaptive sampling

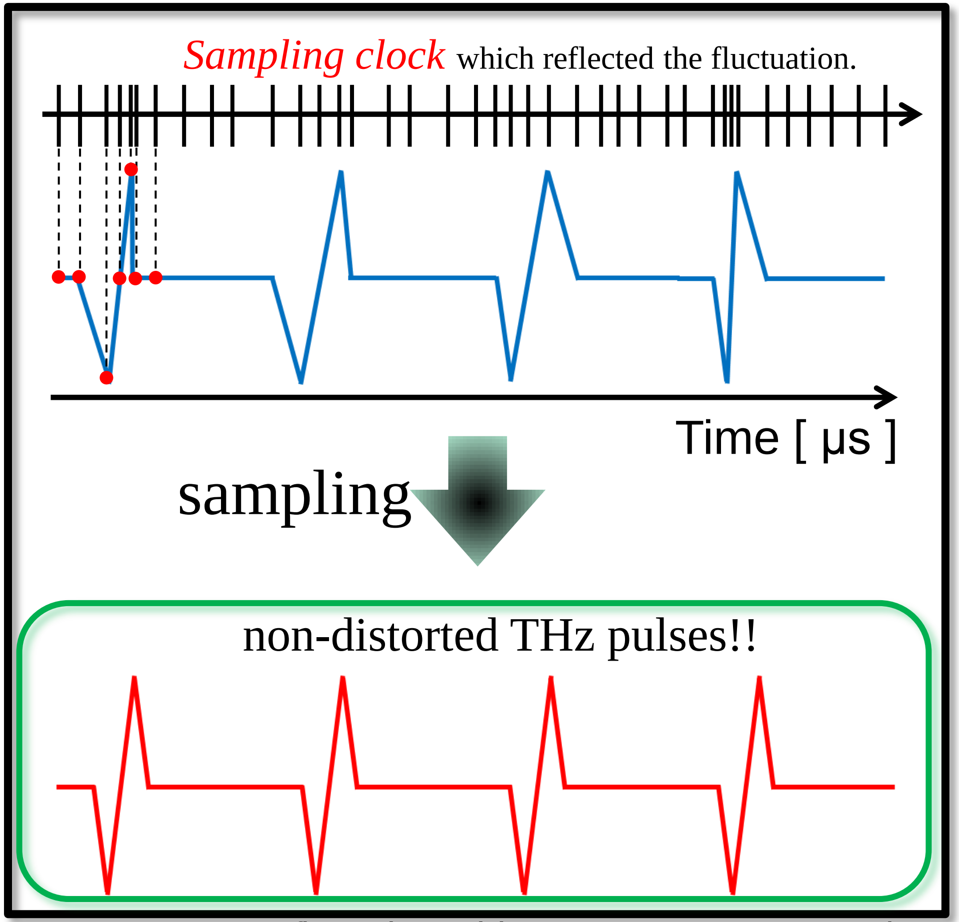
By, generate the sampling clock which reflected the fluctuation, linearity of temporal axes can be kept.

f_{rep1}, f_{rep2} locked



$freerun$

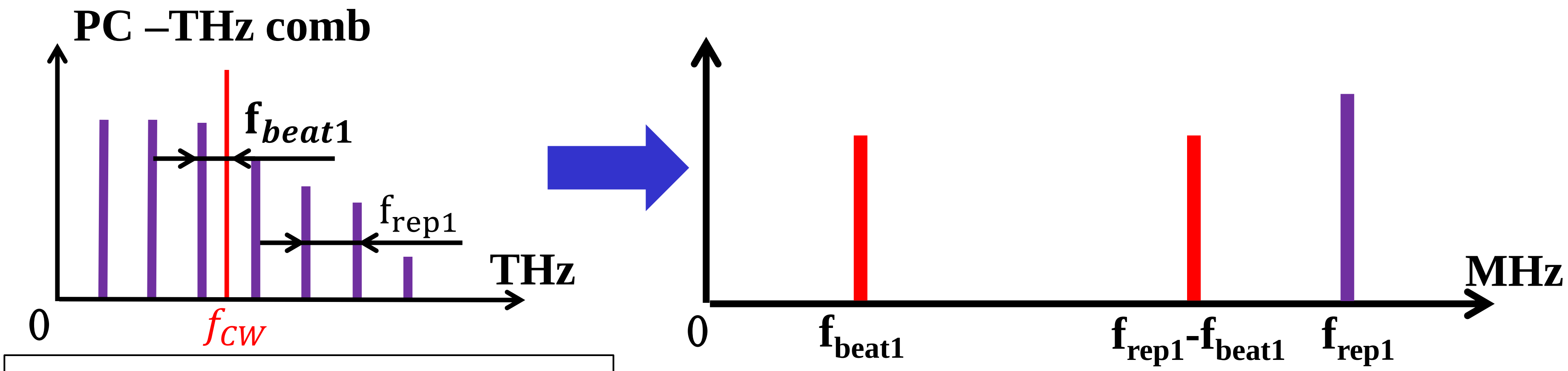




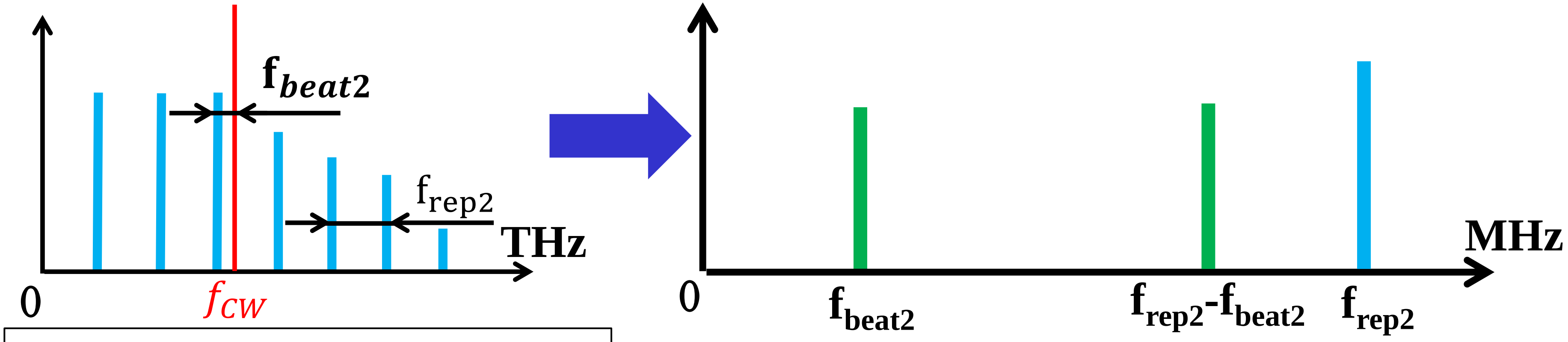
Ref) T. Ideguchi, Nat. Comm., **5**, 3375 (2014).

Extraction of beat signal between dual THz combs

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

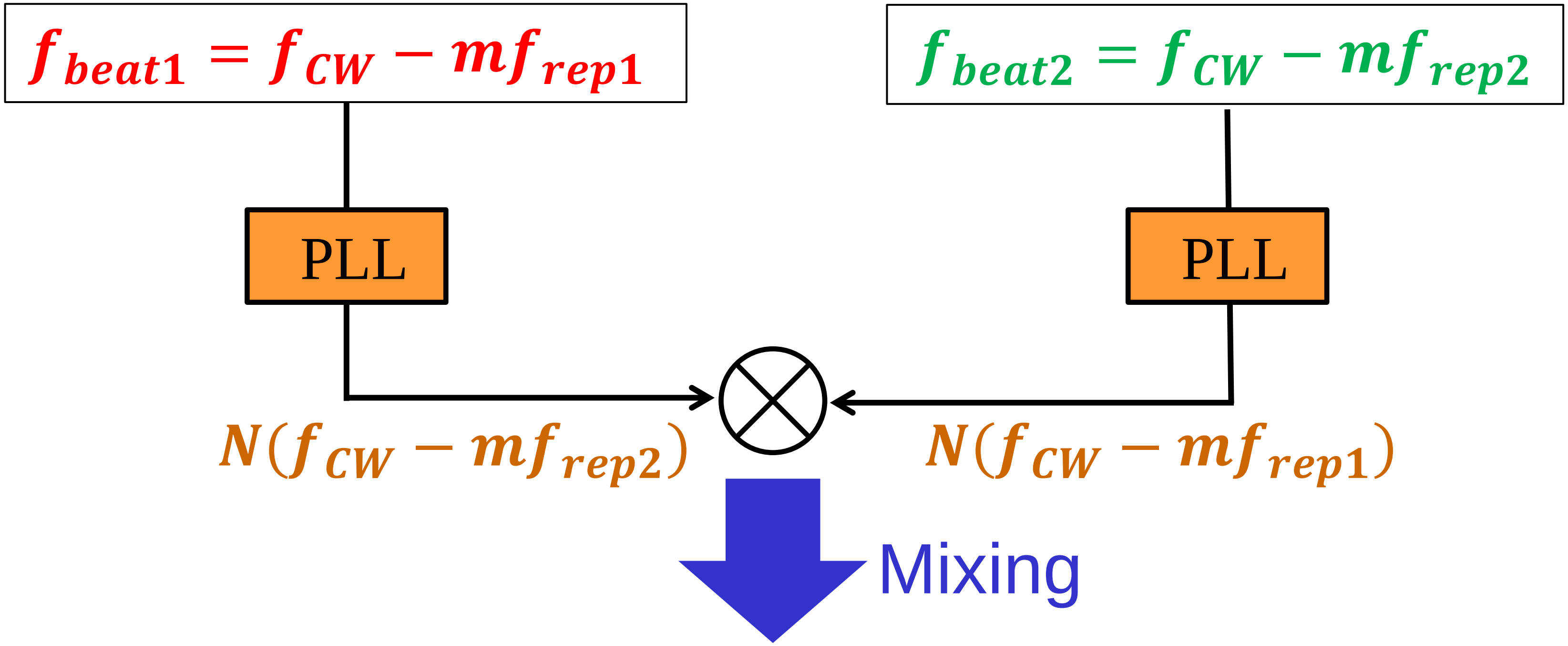


$$f_{beat1} = f_{CW} - mf_{rep1}$$



$$f_{beat2} = f_{CW} - mf_{rep2}$$

Extraction of beat signal between dual THz combs



$$f_{beat1} = f_{CW} - m f_{rep1}$$

$$f_{beat2} = f_{CW} - m f_{rep2}$$

$$N(f_{CW} - m f_{rep2})$$

$$N(f_{CW} - m f_{rep1})$$

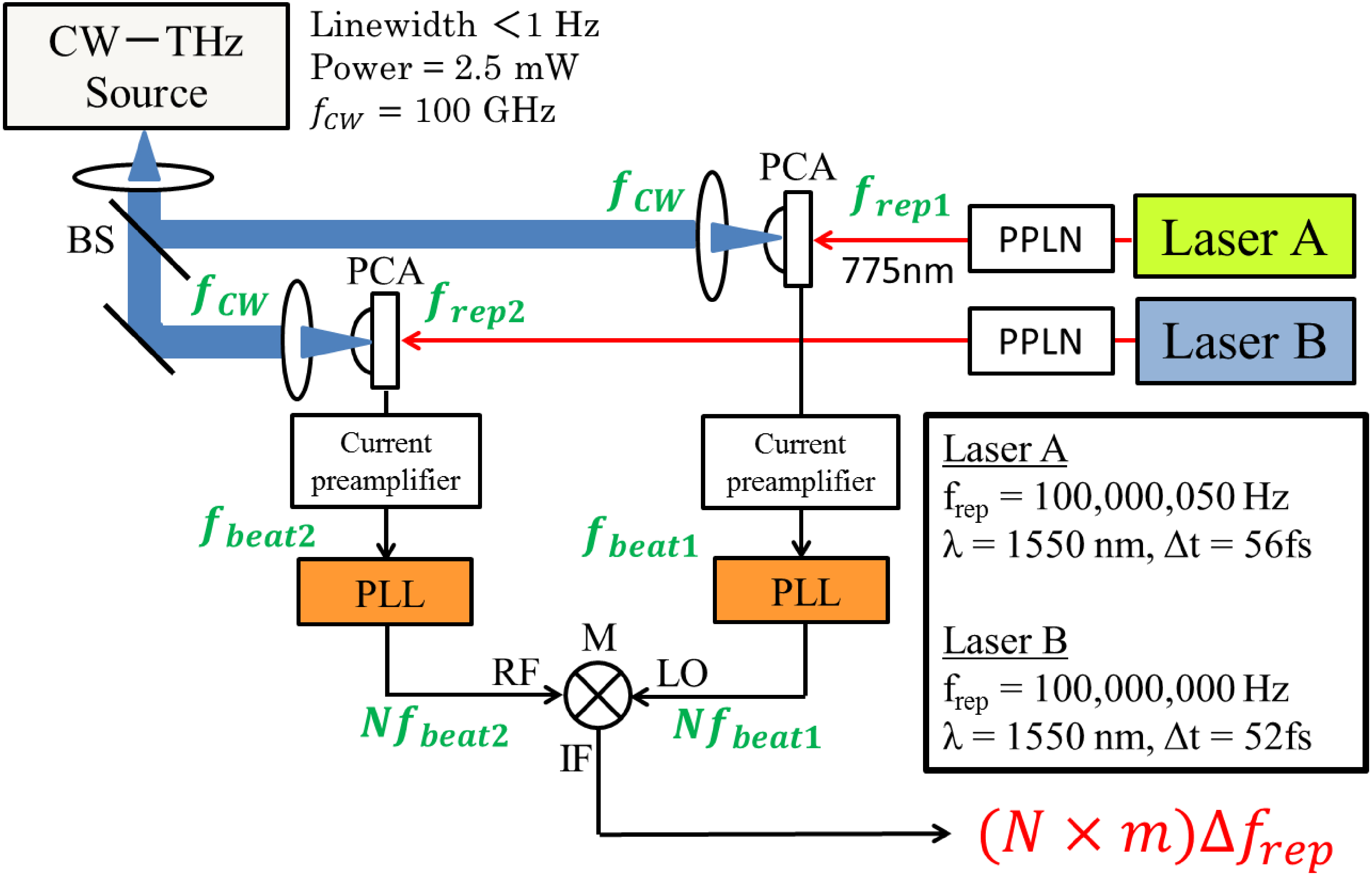
Mixing

$$\underline{f_{beat1} - f_{beat2} = N \times m\left(f_{rep2} - f_{rep1}\right) = N \times m\Delta f_{rep}}$$

Adaptive clock

Experimental Setup for Adaptive Clock

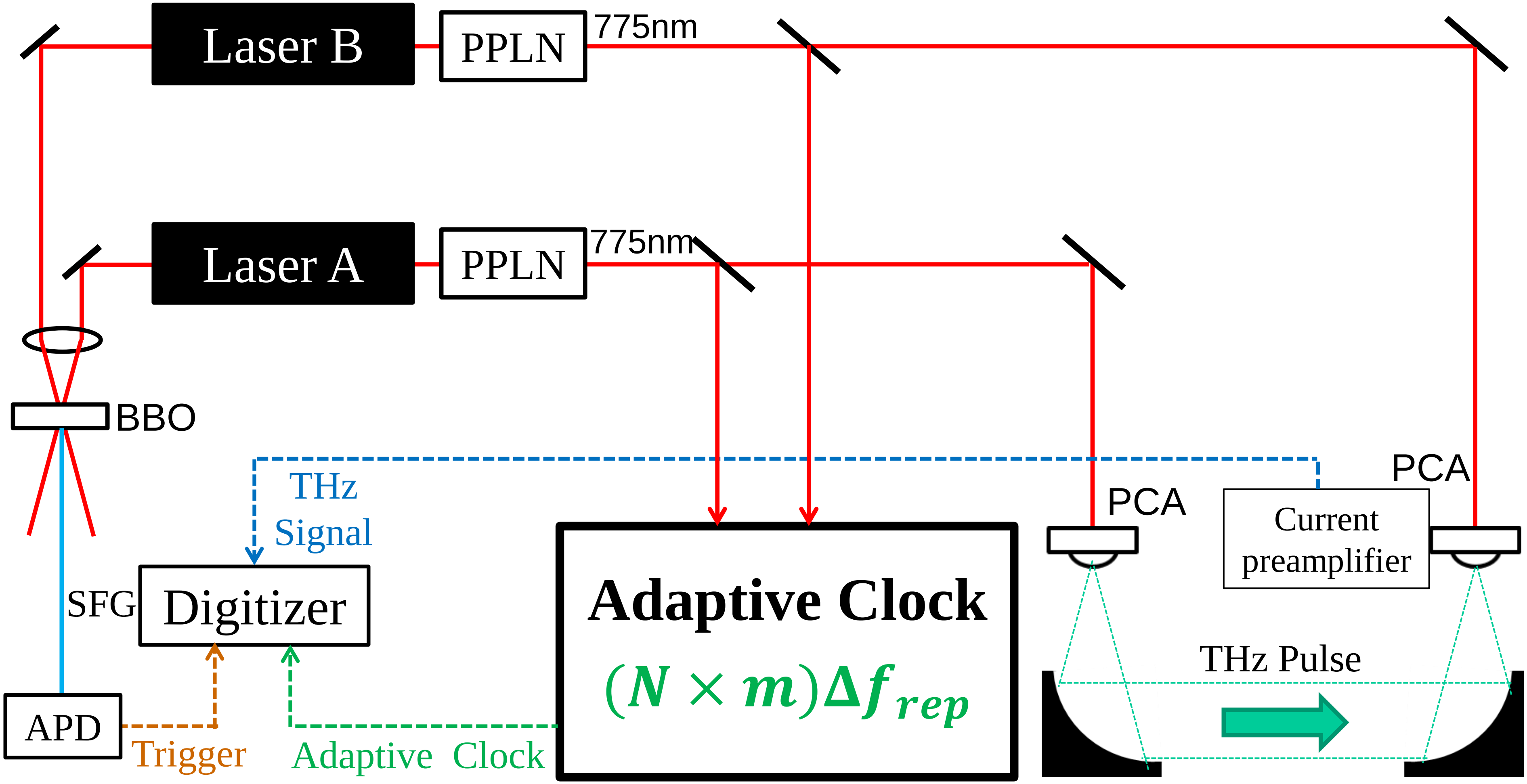
Linewidth <1 Hz
Power = 2.5 mW
f_{CW} = 100 GHz



Laser A
f_{rep} = 100,000,050 Hz
λ = 1550 nm, Δt = 56fs

Laser B
f_{rep} = 100,000,000 Hz
λ = 1550 nm, Δt = 52fs

Experimental Setup



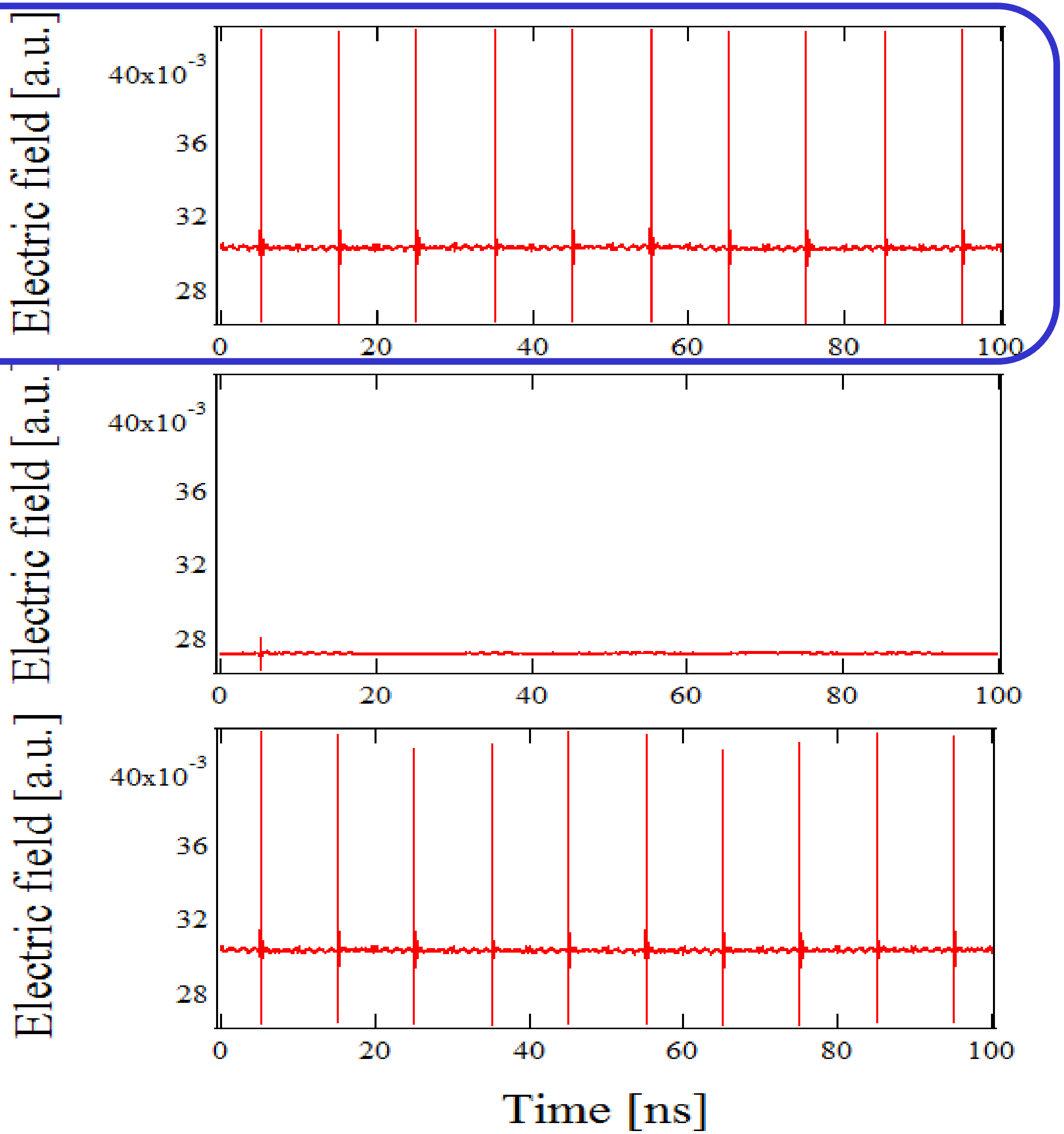
Result（Temporal Waveform）

Integration Number10000, Sampling rate2 MHz

free-run
Adaptive clock

free-run
Constant clock

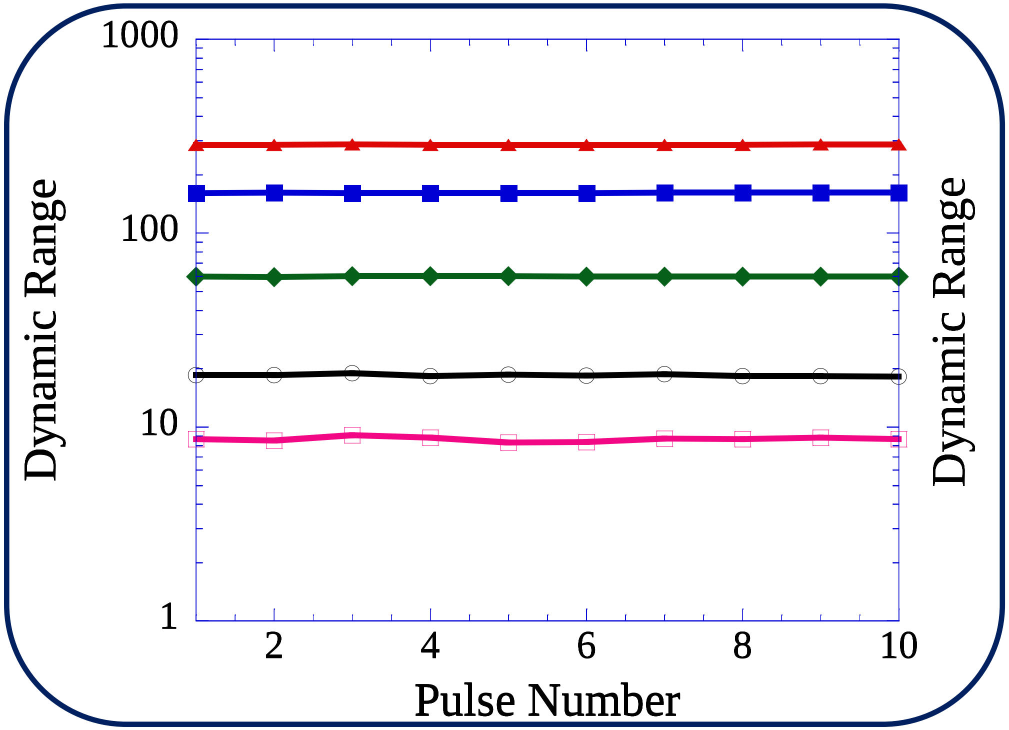
f_{rep} locked
Constant clock



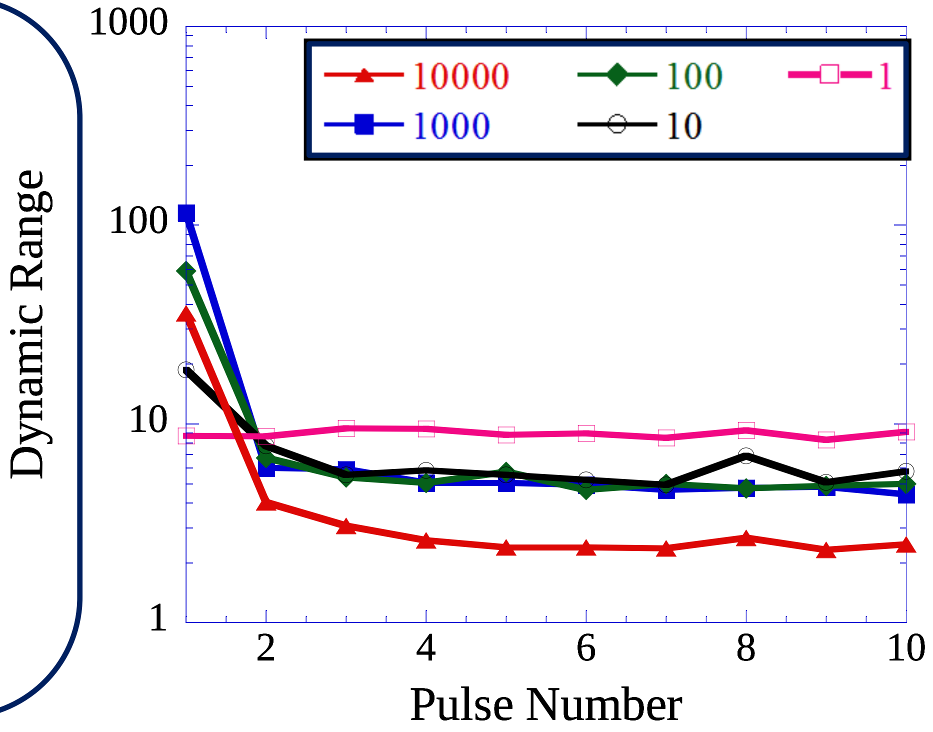
Comparison of dynamic range

Integration : 1, 10, 100, 1000, 10000, Sampling Rate 2 MHz

FRL · Adaptive Clock



FRL · Constant Clock



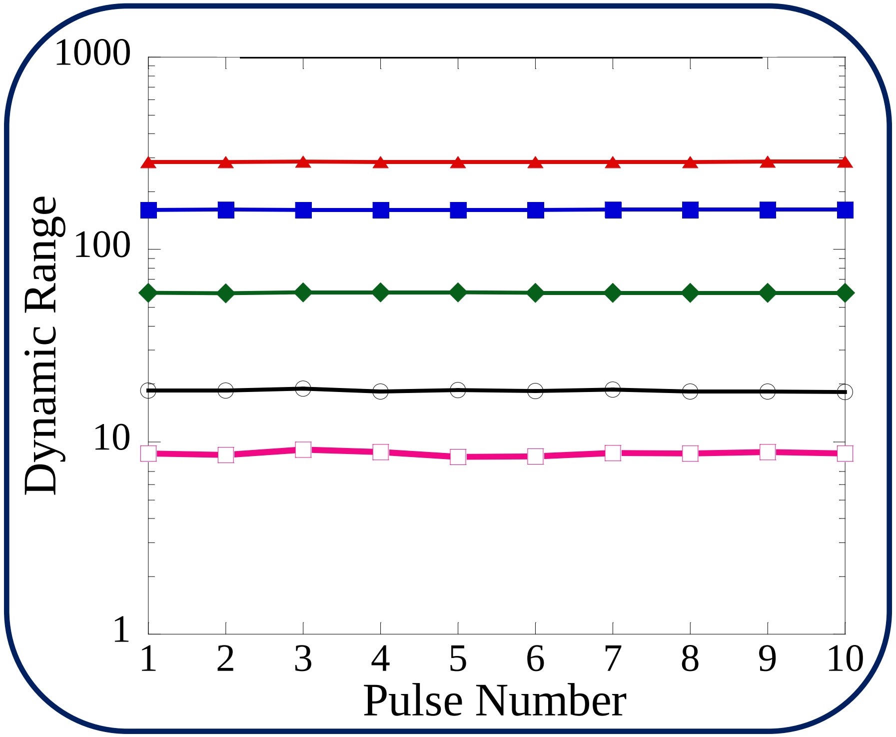
Effect of timing jitter is not observed.

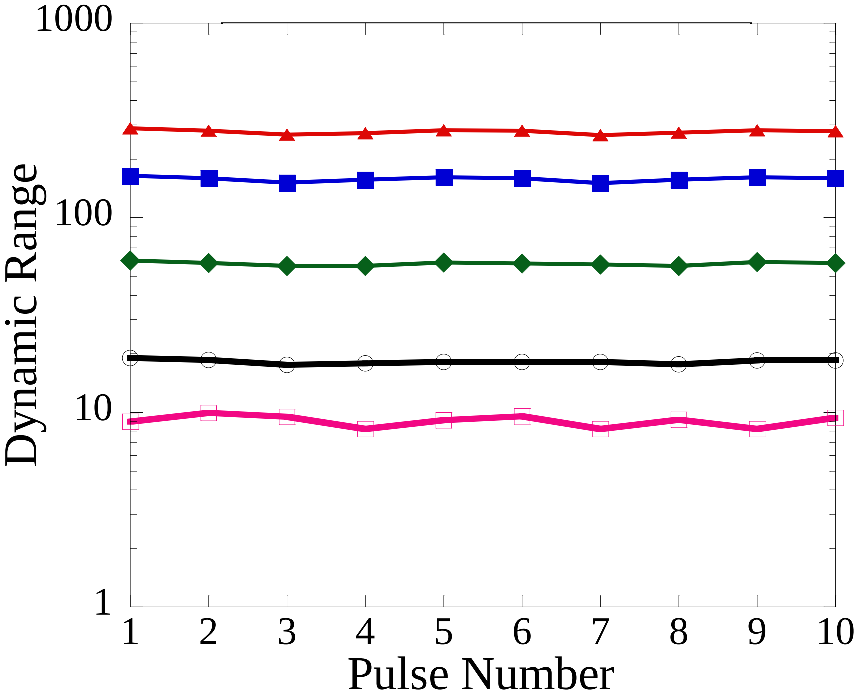
Comparison of dynamic range

Integration : 1, 10, 100, 1000, 10000, Sampling Rate 2 MHz

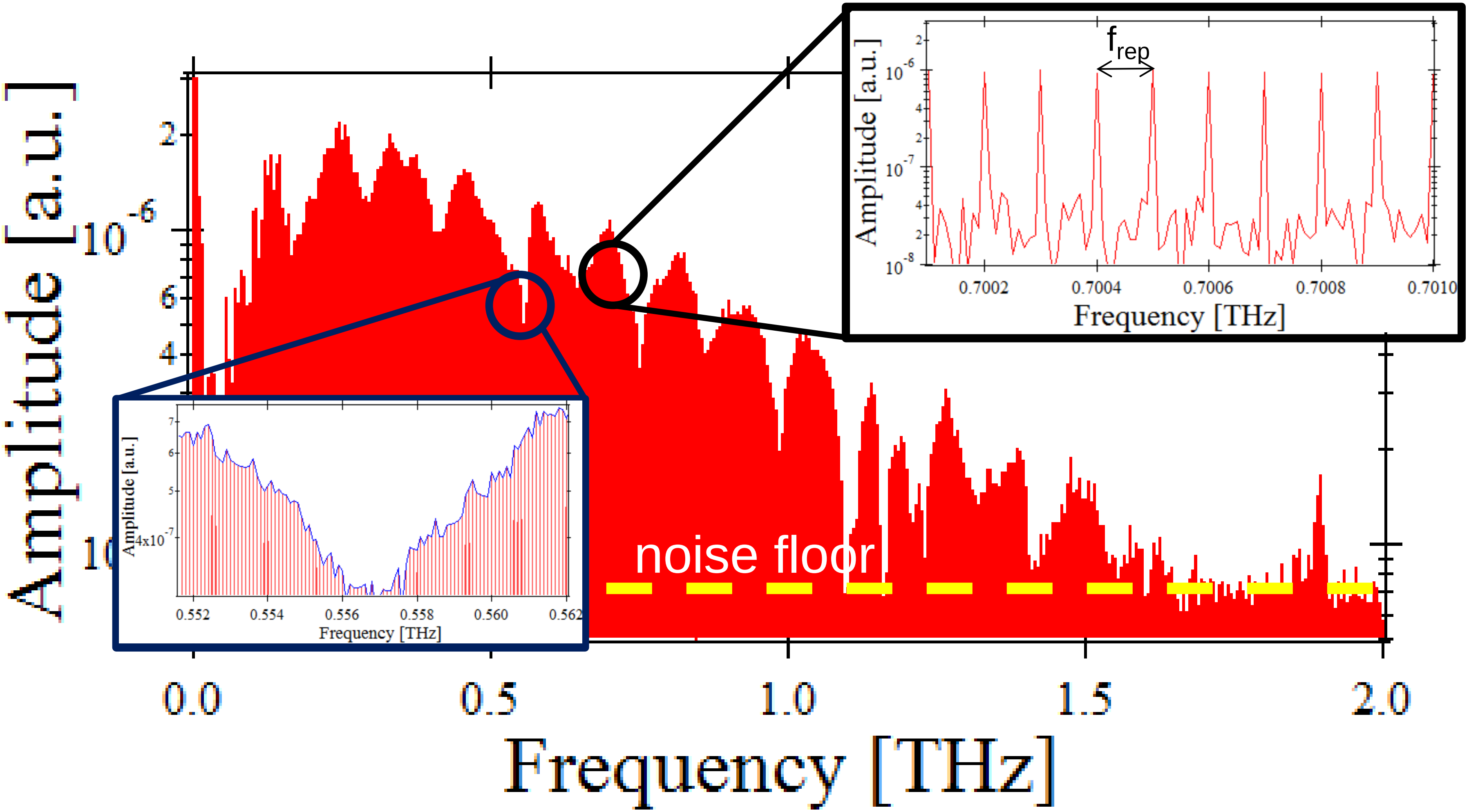
FRL・Adaptive Clock

f_{rep} locked・Constant Clock





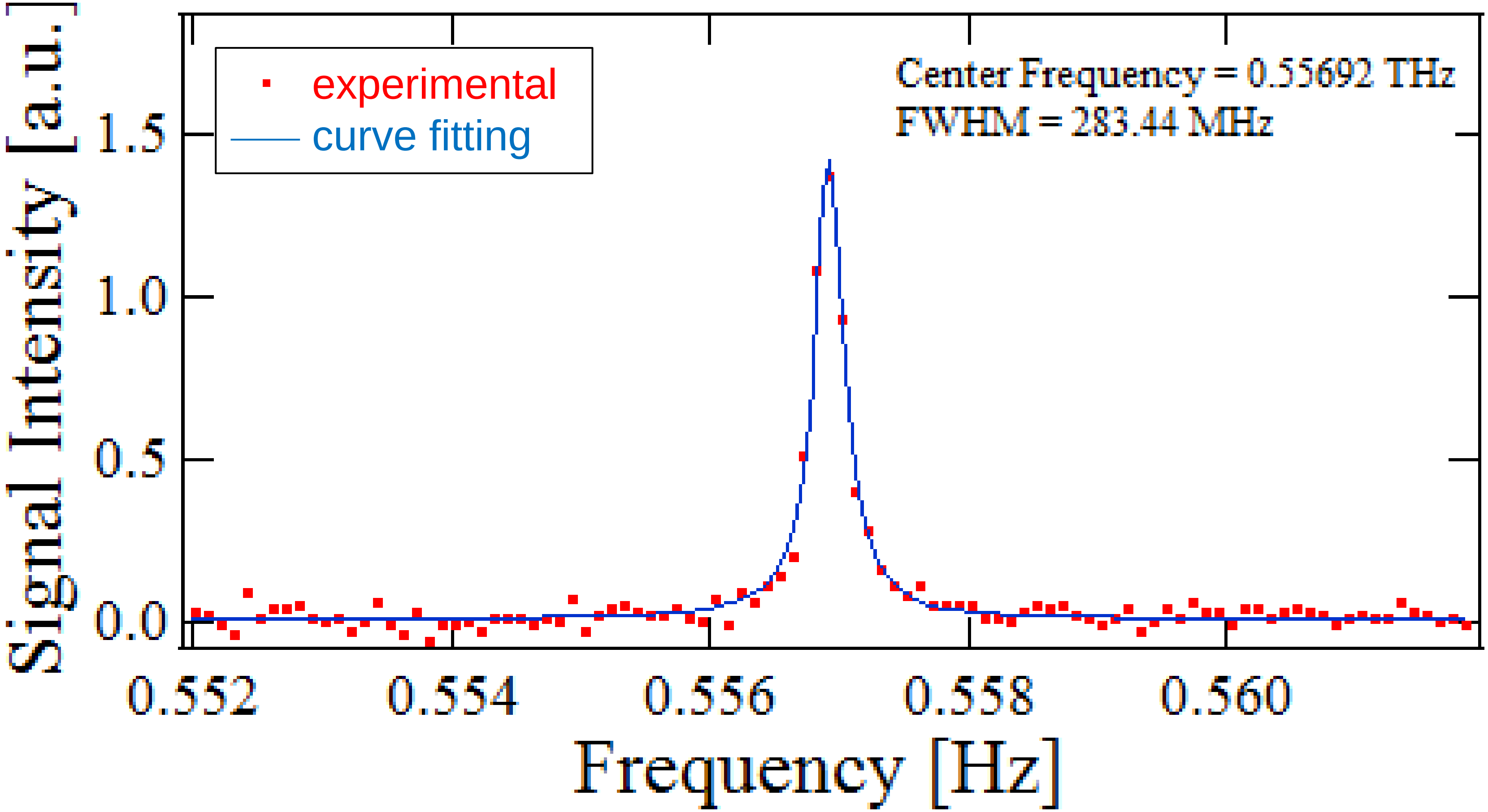
THz Comb Spectrum



Dual THz comb spectroscopy using FRL is attained.

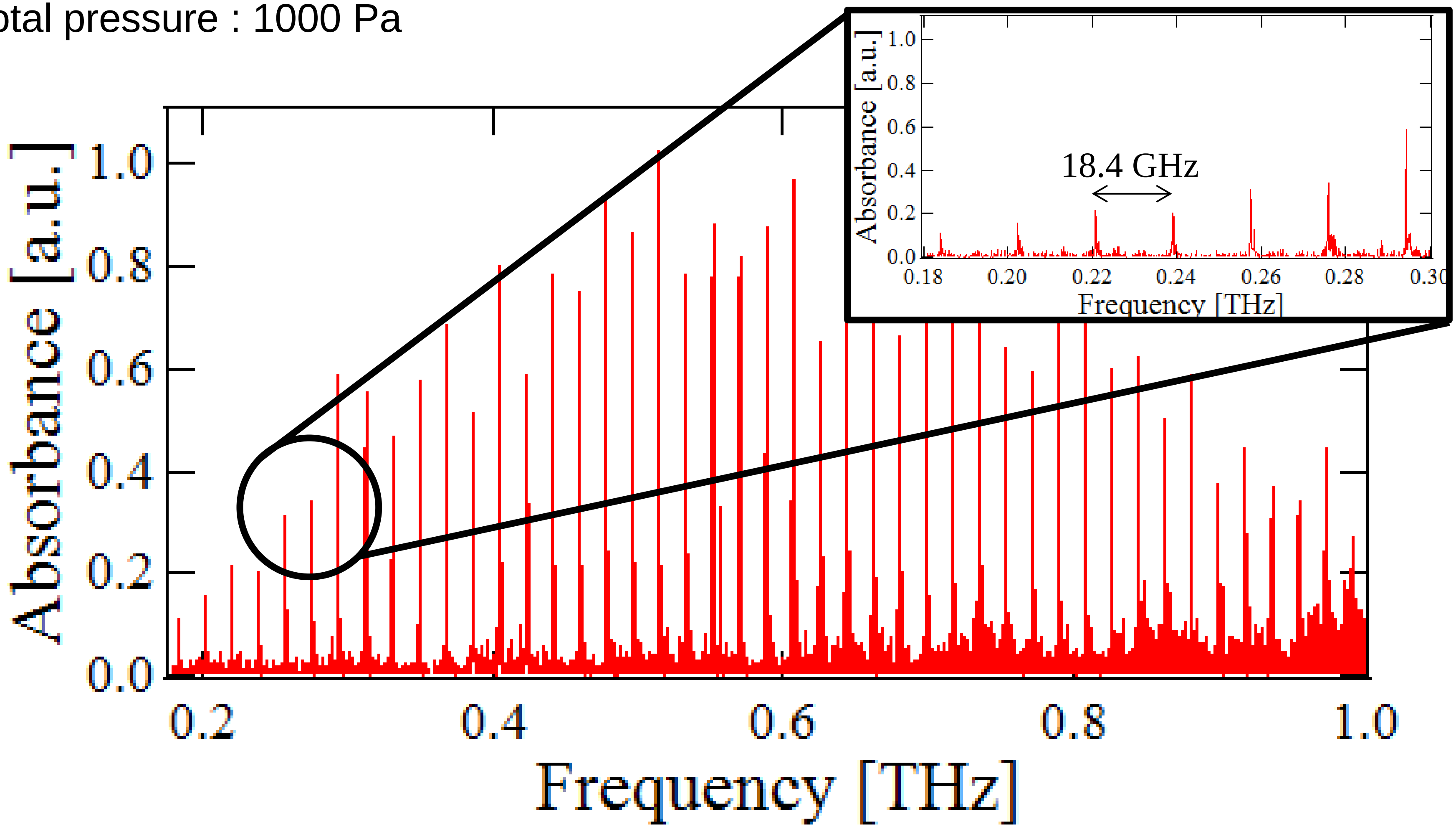
Spectroscopy of low-pressure water vapor

Rotational transition $1_{10} \leftarrow 1_{01}$: 0.5569 THz @ NASA database
Nitrogen : 2400 Pa
Water Vapor : 170 Pa
Theoretical linewidth : 200 MHz



Absorbance spectrum of acetonitrile gas

- Acetonitrile (CH_3CN)
  Symmetric top molecule, rotational constant *B* = 9.2 GHz
  Manifold of rotational transitions regularly spaced by 2*B*
- Total pressure : 1000 Pa



Summary

Dual THz comb spectroscopy using free-running laser is attained.

- Effect of timing jitter is not observed.
- The result is equal to f_{rep} locked・constant clock.

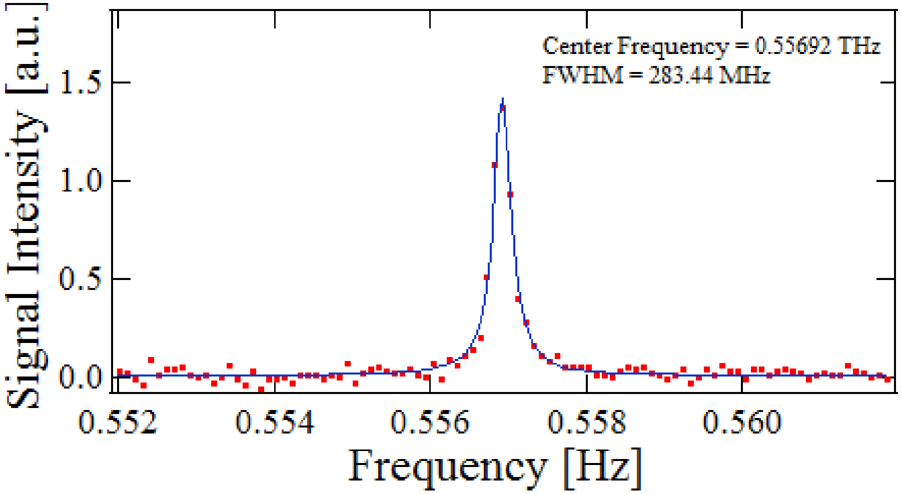
Future work

Adaptive sampling Dual optical comb spectroscopy.

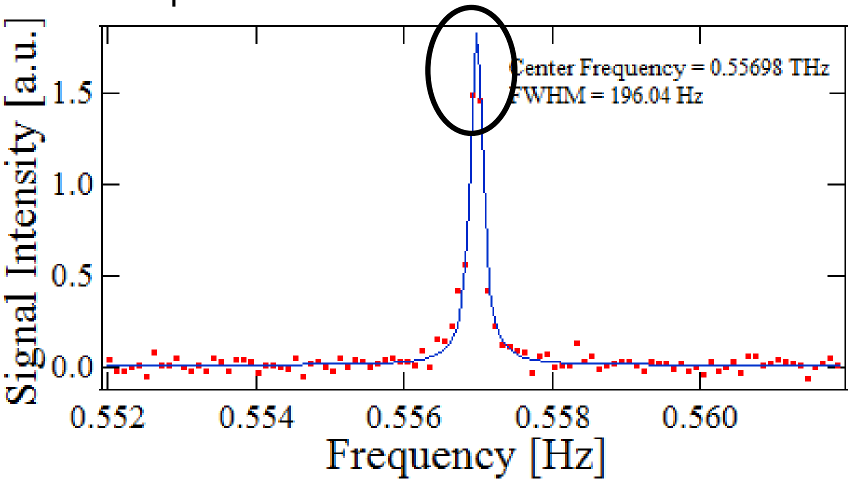
Spectroscopy of low-pressure water vapor

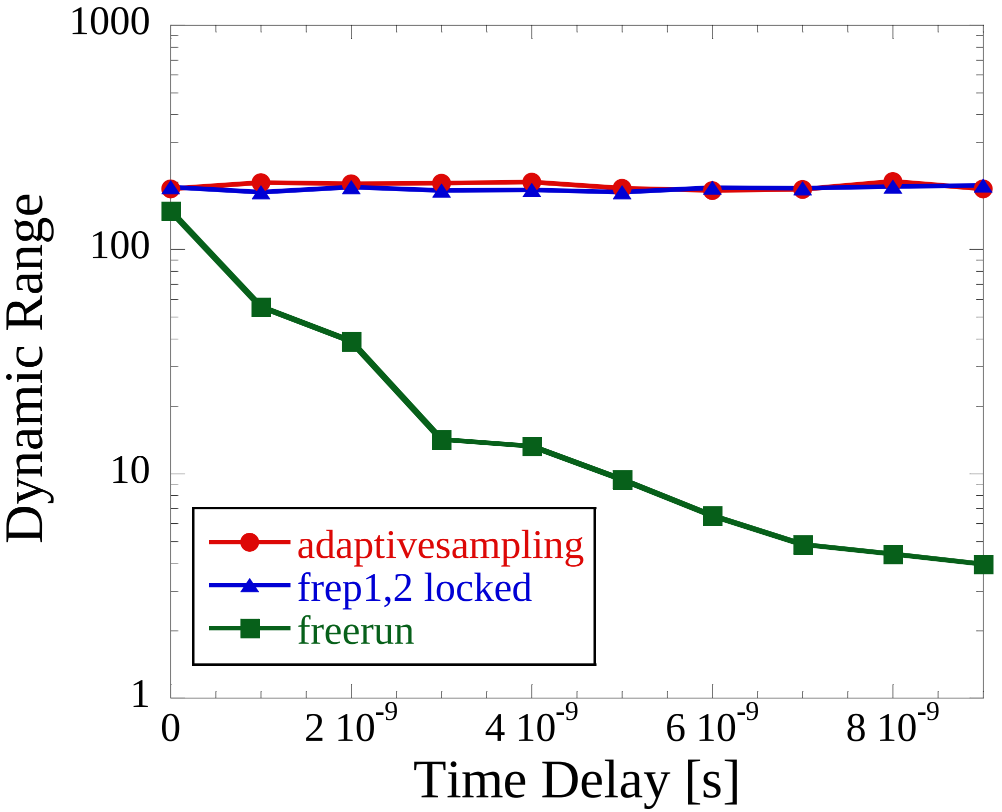
Nitrogen : 2400 Pa
Water Vapor : 170 Pa
Theoretical linewidth : 200 MHz

FRL・Adaptive Clock



f_{rep} locked・Constant Clock



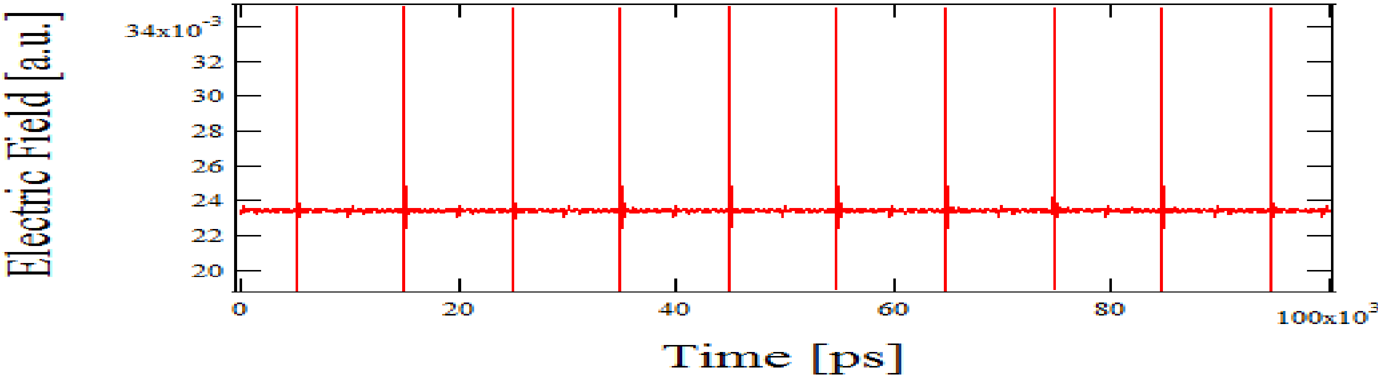

1000
100
10
1
Dynamic Range
0
2 10-9
4 10-9
6 10-9
8 10-9
Time Delay [s]
adaptivesampling
frep1,2 locked
freerun

10連THzパルス列の計測

freerun

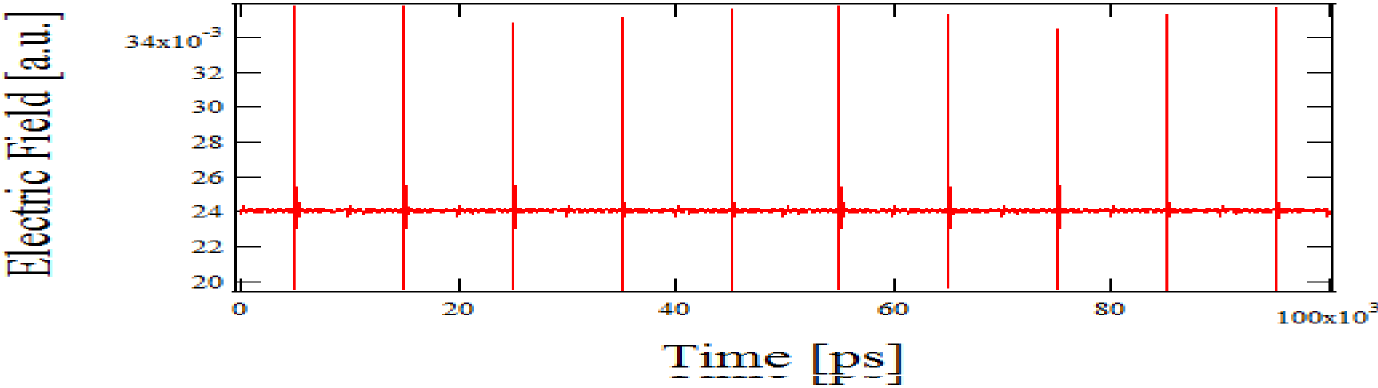
Adaptive clock

ジッターの影響をキャンセル



f_{rep1}, f_{rep2} locked

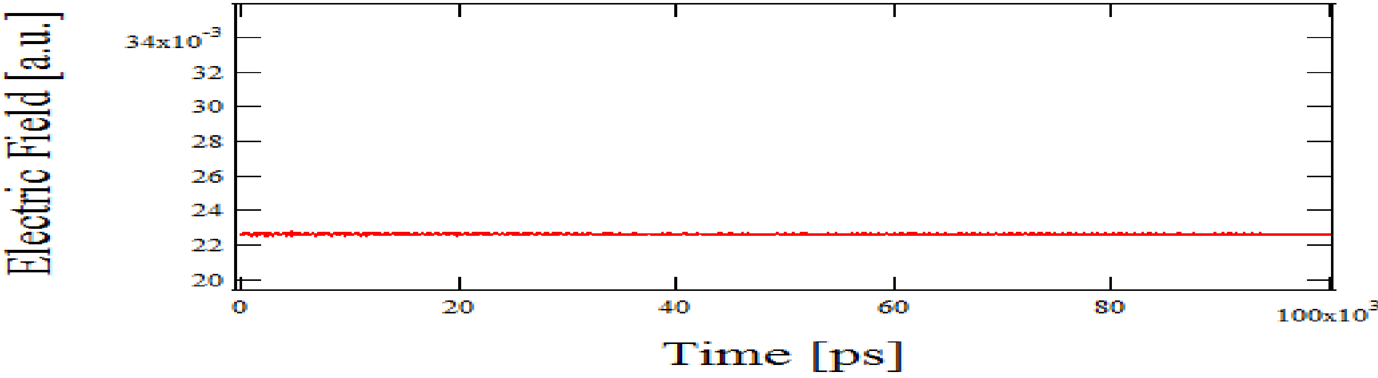
Constant clock



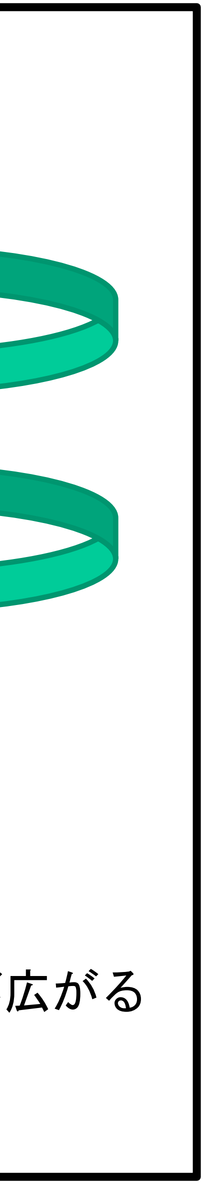
freerun

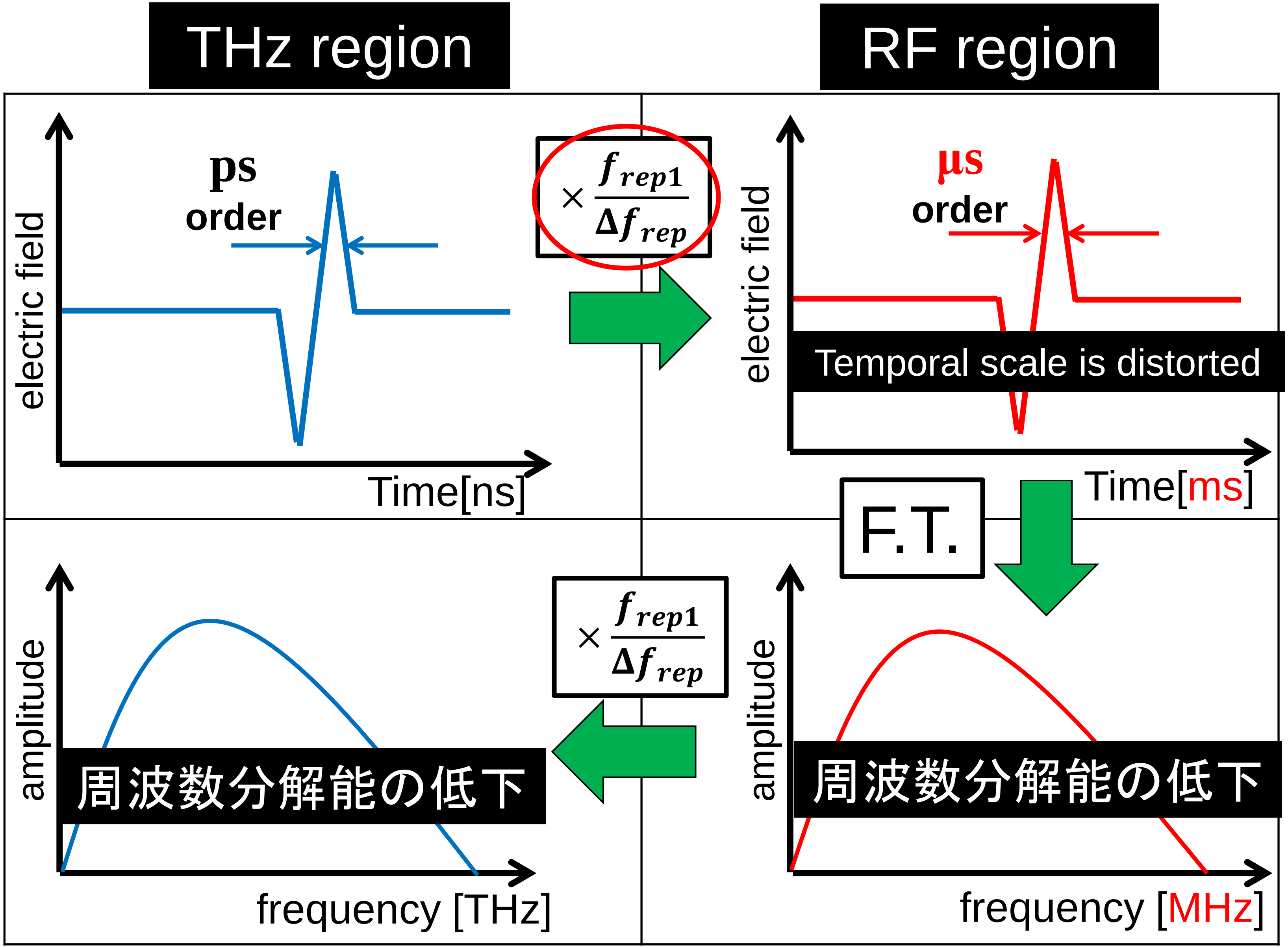
Constant clock

ジッターの影響で積算できない



ASOPS-THz-TDSにおける信号の流れ





$\times \frac{f_{rep1}}{\Delta f_{rep}}$

Temporal scale is distorted

$\times \frac{f_{rep1}}{\Delta f_{rep}}$

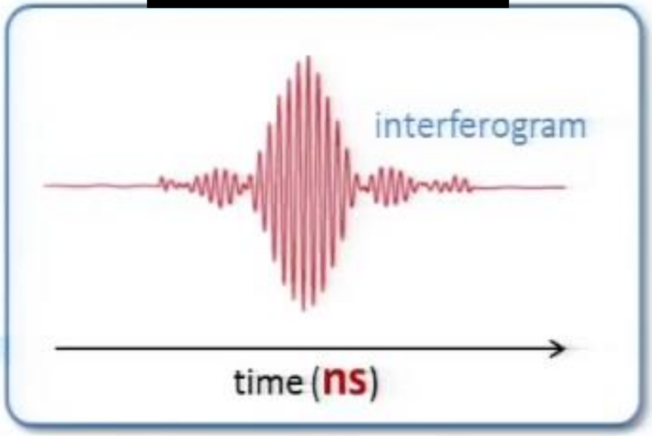
周波数分解能の低下

周波数分解能の低下

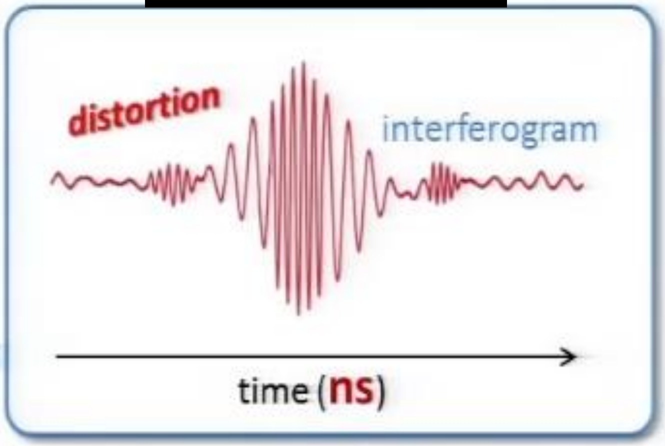
Adaptive sampling

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

安定化制御

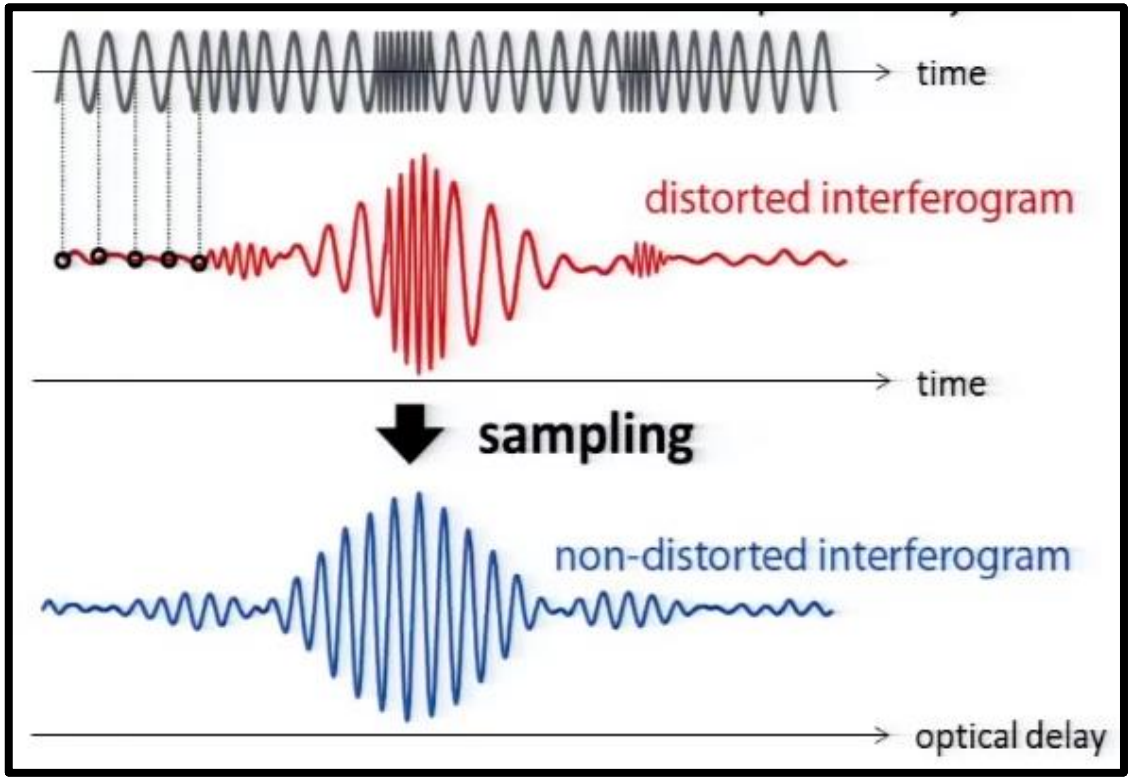


free-running



時間軸の線形性が保たれない！

モード同期周波数の揺らぎを反映したサンプリング・クロックを生成すれば、時間軸の線形性を保てる！



揺らぎ＝ Δf_{rep} ＝コム間ビート