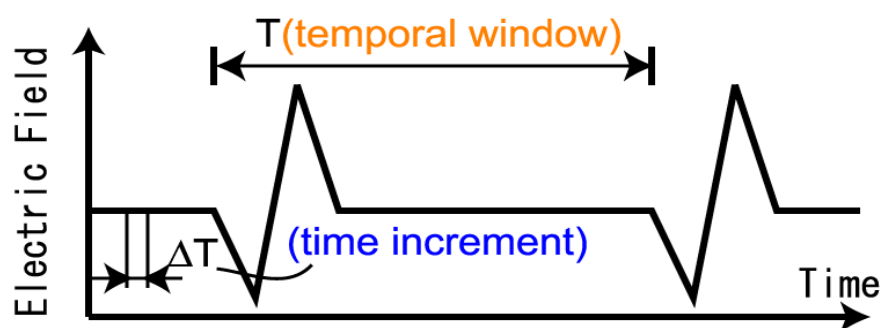
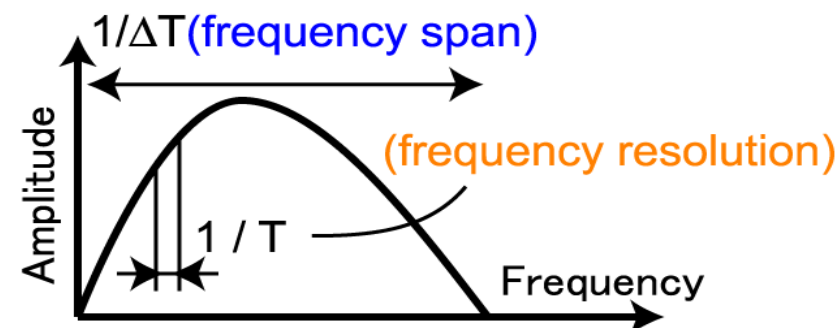
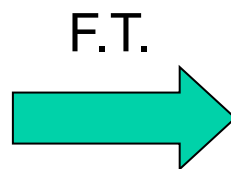


# THz time-domain spectroscopy



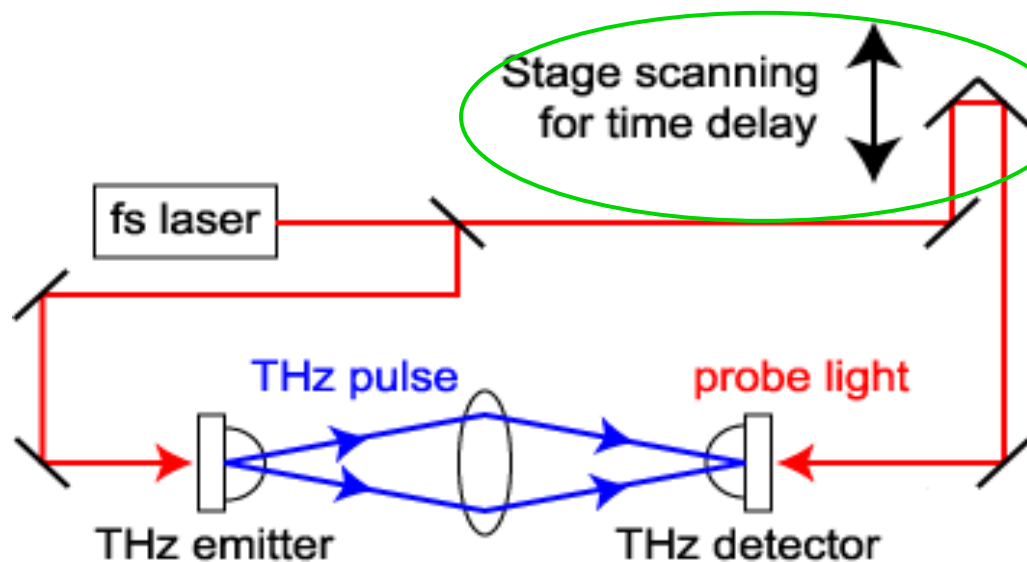
*Temporal waveform*

Spectral resolution  
= inverse of time window



*Fourier spectra*

Spectral accuracy  
= Precision of time delay



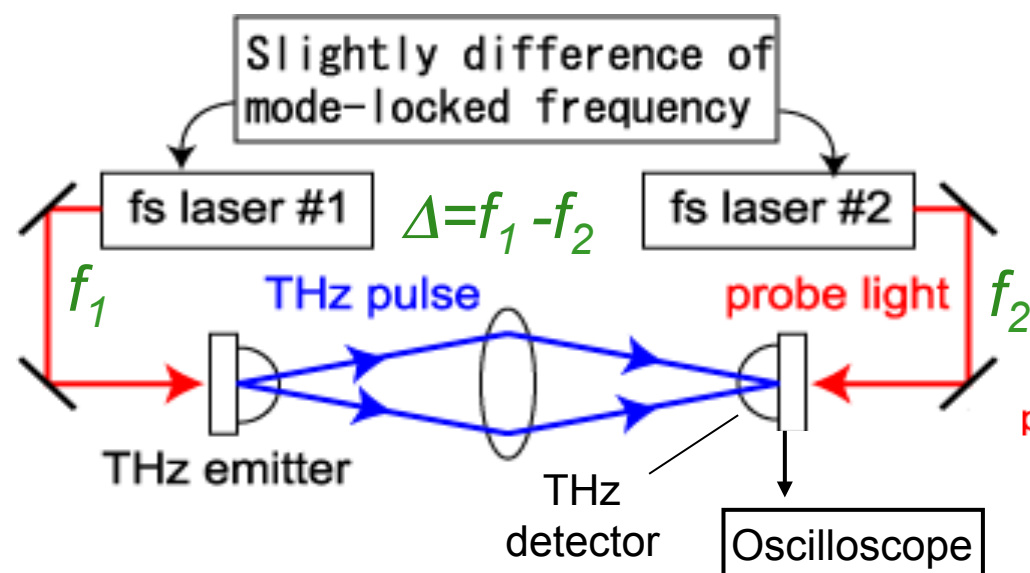
*Pump-probe experiment*

Marking of frequency scale is based on mechanical movement of stage

- Trade-off between spectral resolution and measurement time
- Spectral accuracy depends on positioning precision of stage

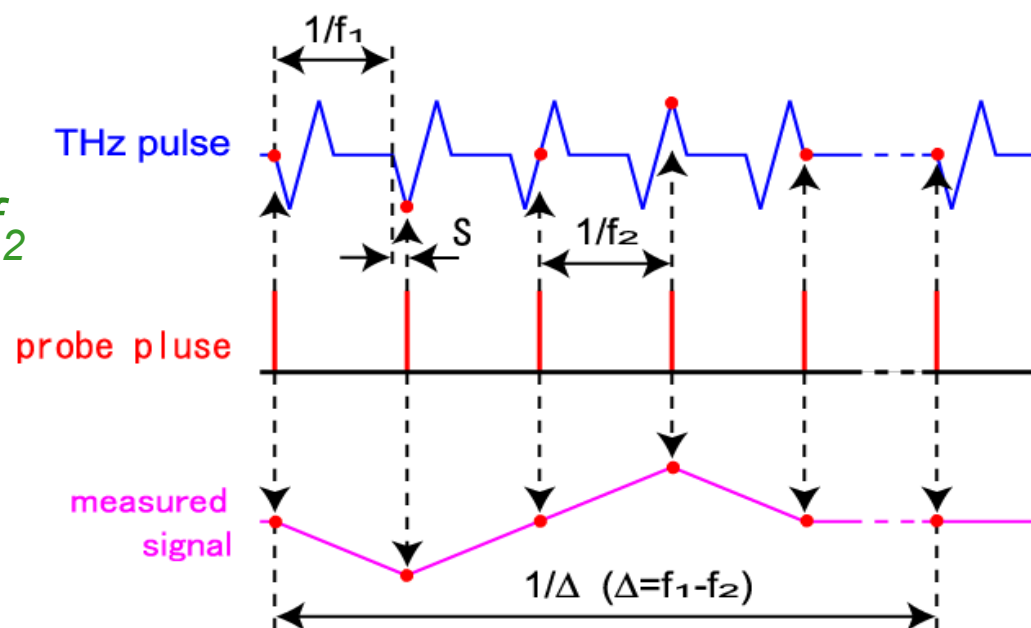
# Asynchronous-optical-sampling THz-TDS (AOS-TH-TDS)

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



**No need for mechanical stage**  
**No limitation for size of time window**

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

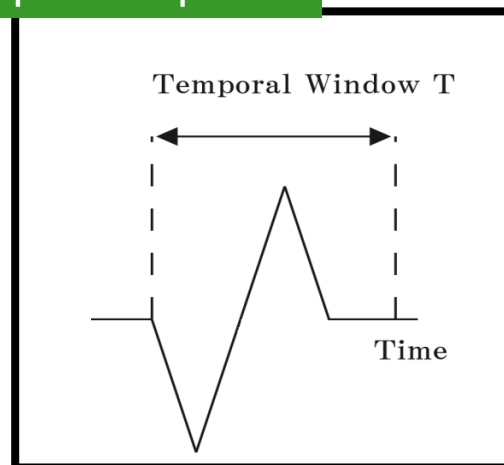
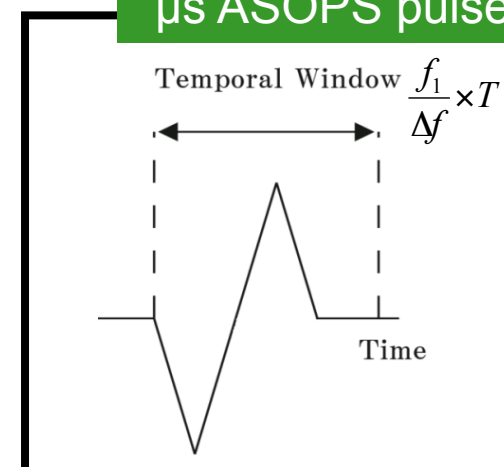


**Time scale of ps THz pulse is linearly expanded to μs order**

**High spectral resolution and accuracy can be achieved depending on laser stability**

# Conversion of frequency scale by ASOPS-THz-TDS

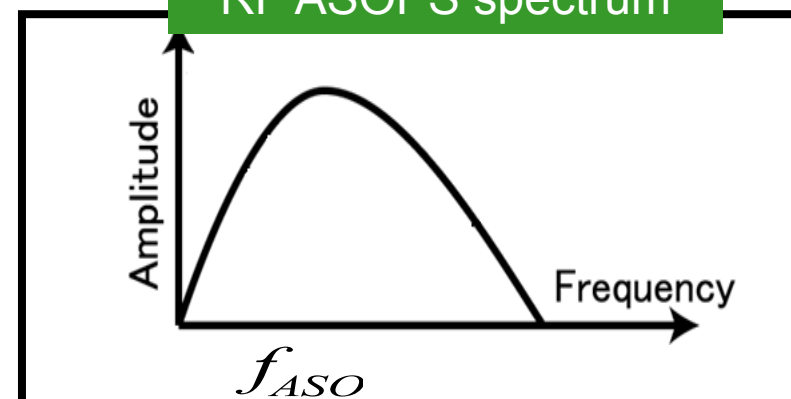
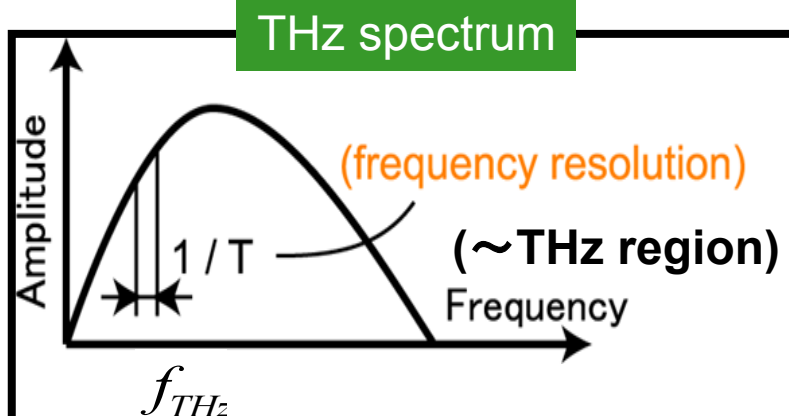
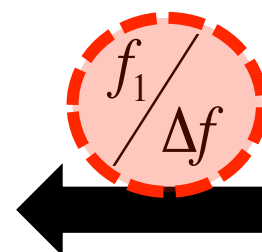
ps THz pulse

ASOPS  
(down-scaling) $\mu$ s ASOPS pulse

This factor is fluctuated  
by timing jitter of two  
lasers.

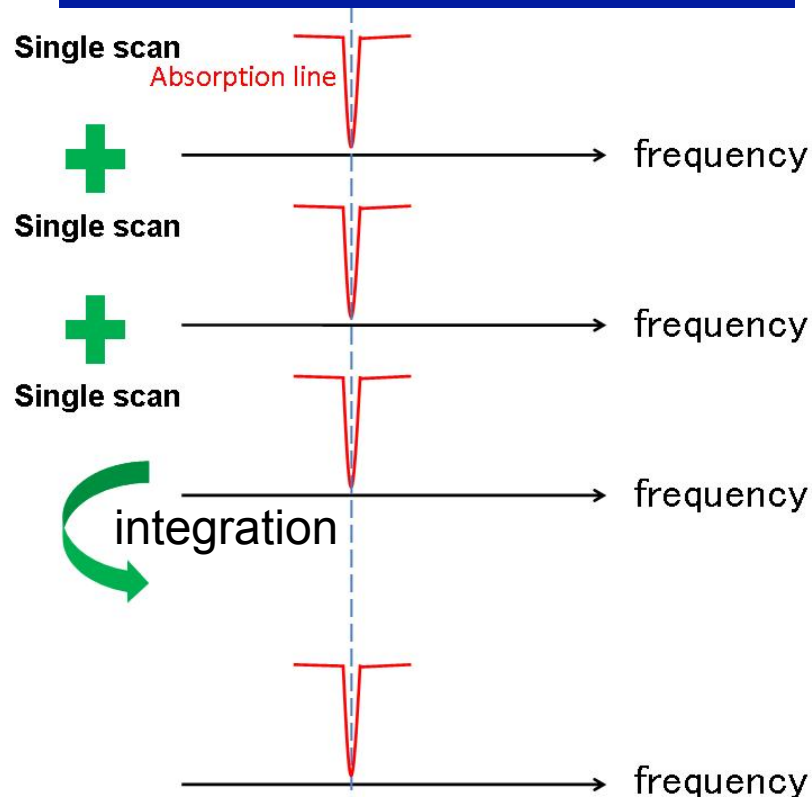
F.T.

RF ASOPS spectrum

Calibration  
(up-scaling)

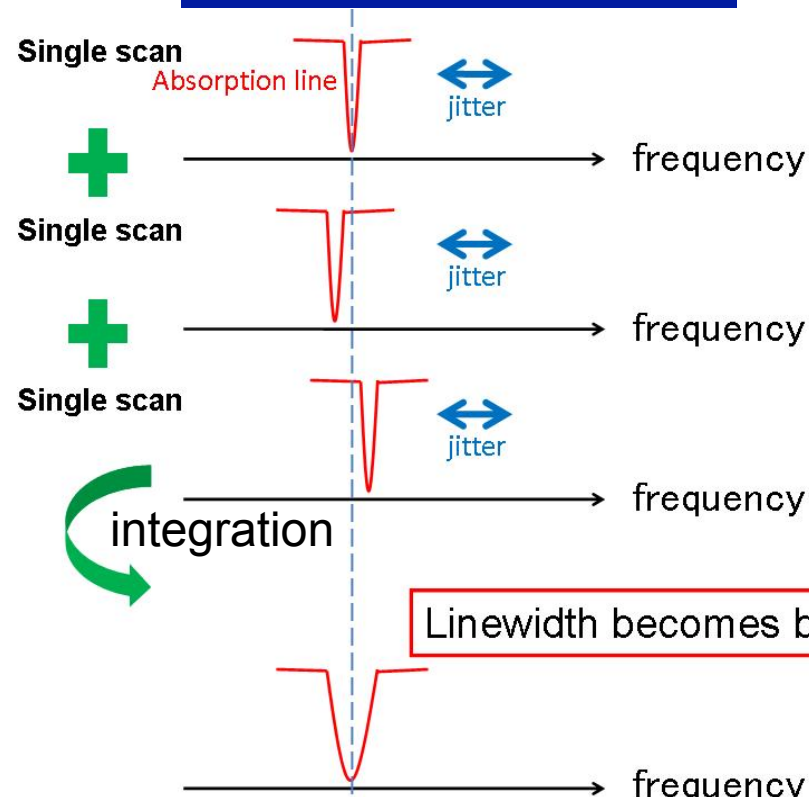
## Influence of timing jitter in absorption spectroscopy

## Conventional THz-TDS



spectral resolution  
= inverse of time window

## ASOPS-THz-TDS



Linewidth becomes broader!

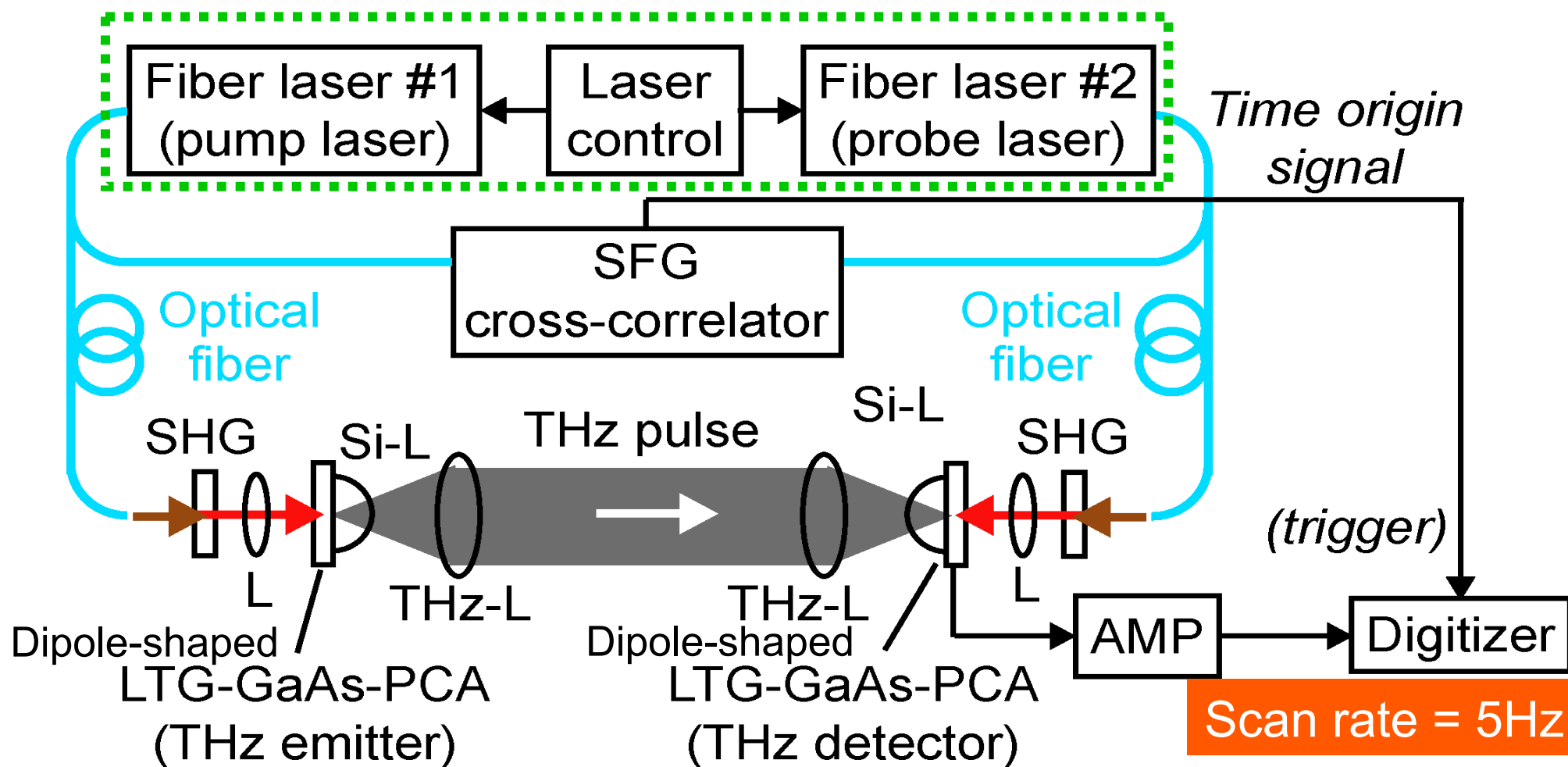
spectral resolution  
≥ inverse of time window

**Present  
talk**

Evaluate actual resolution and accuracy in ASOPS-THz-TDS using low-pressure gas spectroscopy

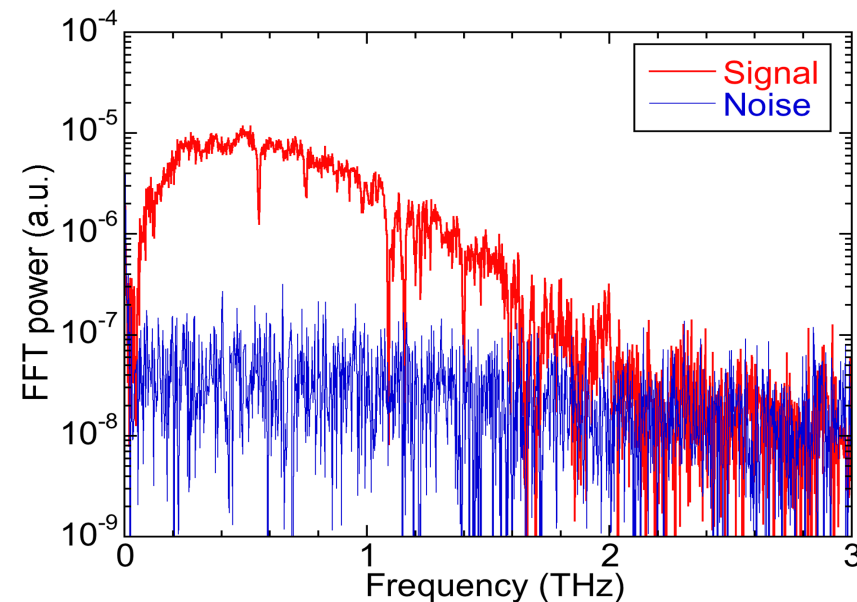
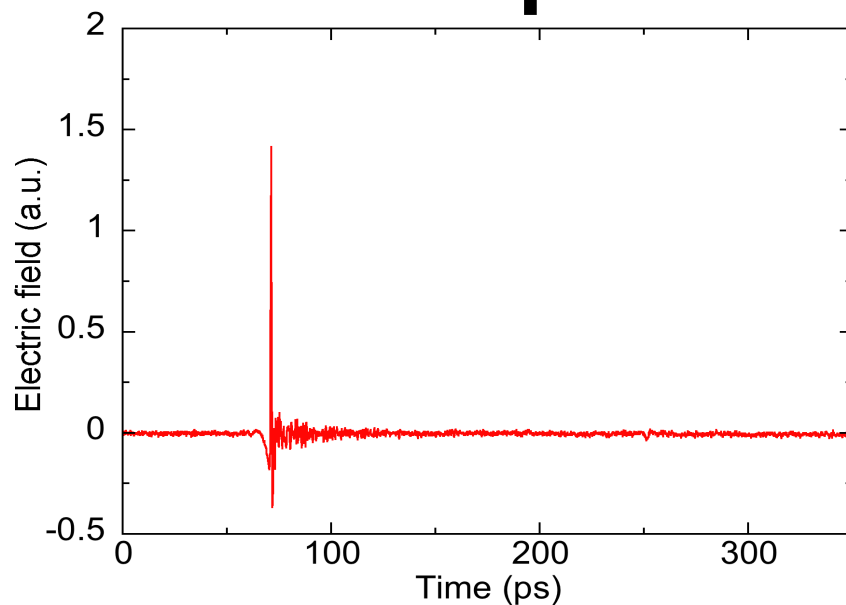
# Experimental setup

$\lambda_c = 1550\text{nm}$ ,  $\Delta\tau = 50\text{fs}$ ,  $P_{\text{avg}} = 100\text{mW}$ ,  
 $f_1 = 56.124\text{MHz}$ ,  $f_2 = 56.124\text{MHz} + 5\text{Hz}$ ,  $\Delta = f_2 - f_1 = 5\text{Hz}$ , Timing jitter  $< 300\text{fs}$

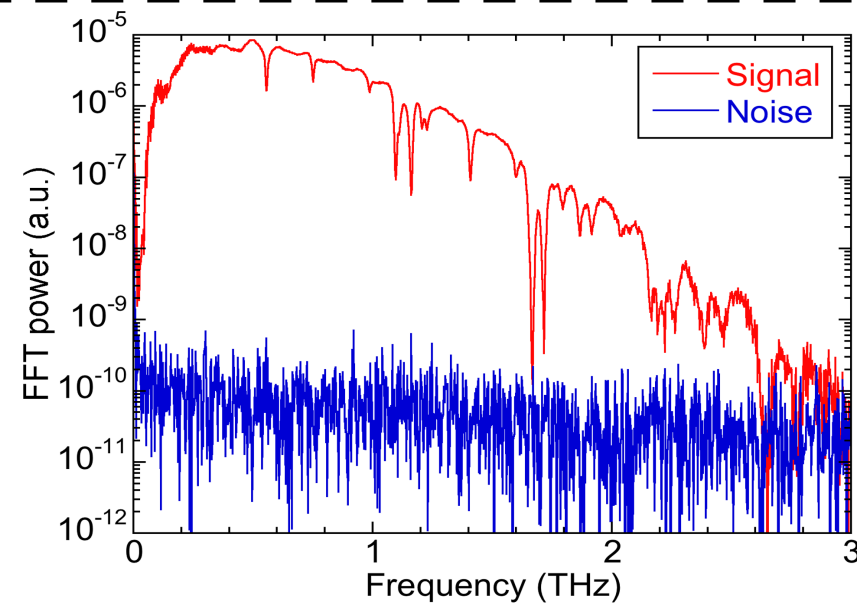
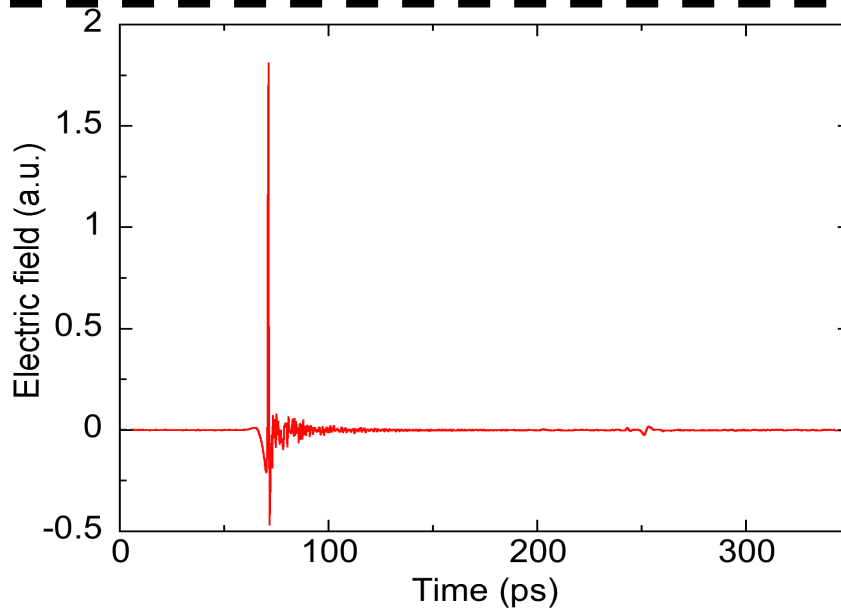


# Basic performance

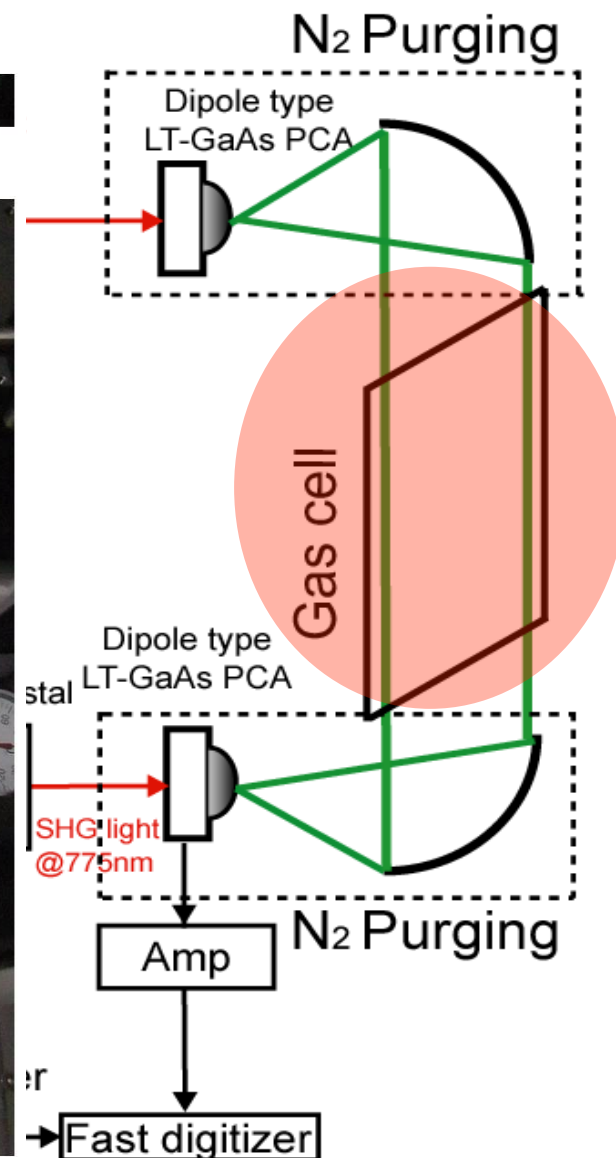
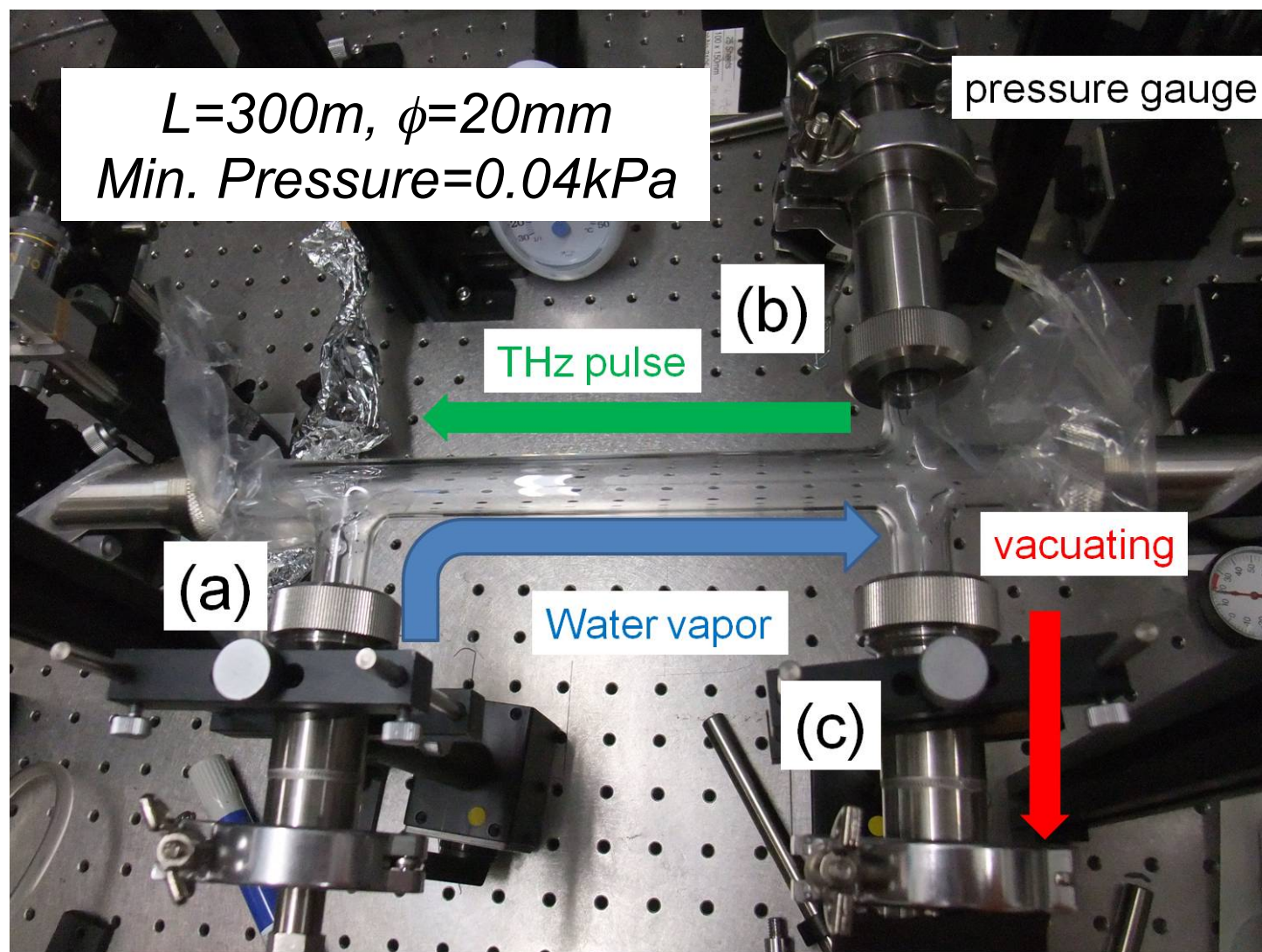
Single  
sweep  
(200ms)



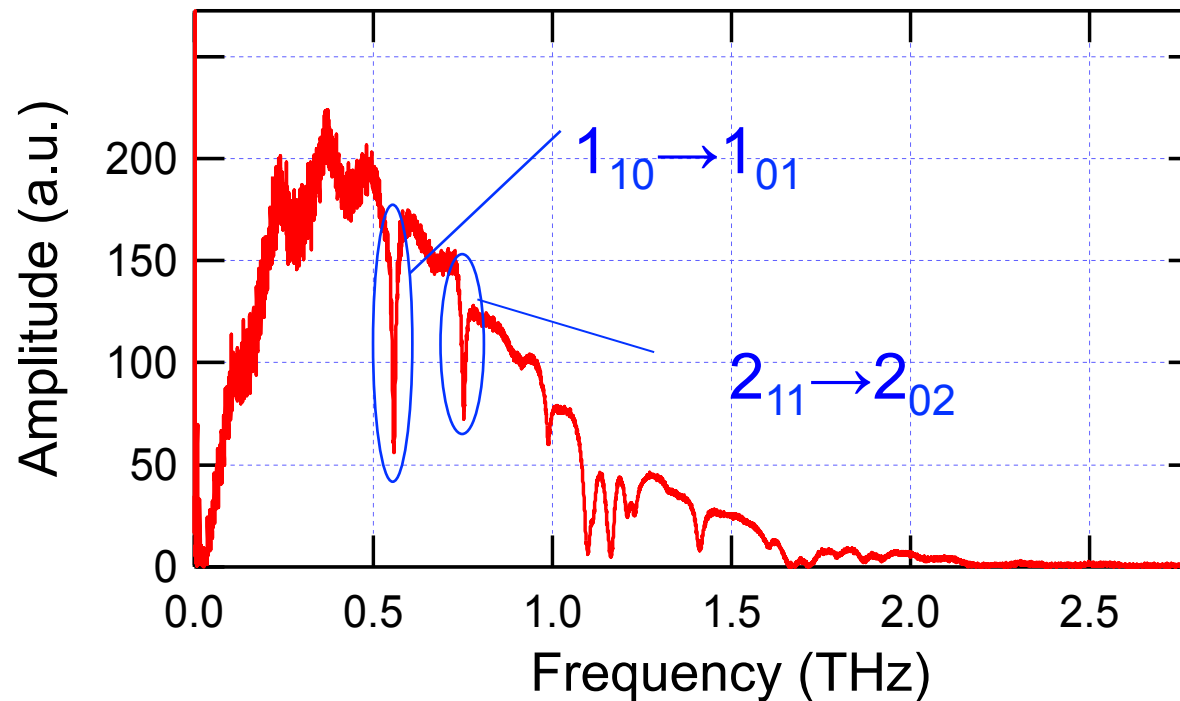
200  
sweeps  
(100s)



# Low-pressure gas cell



# Absorption lines of water vapor



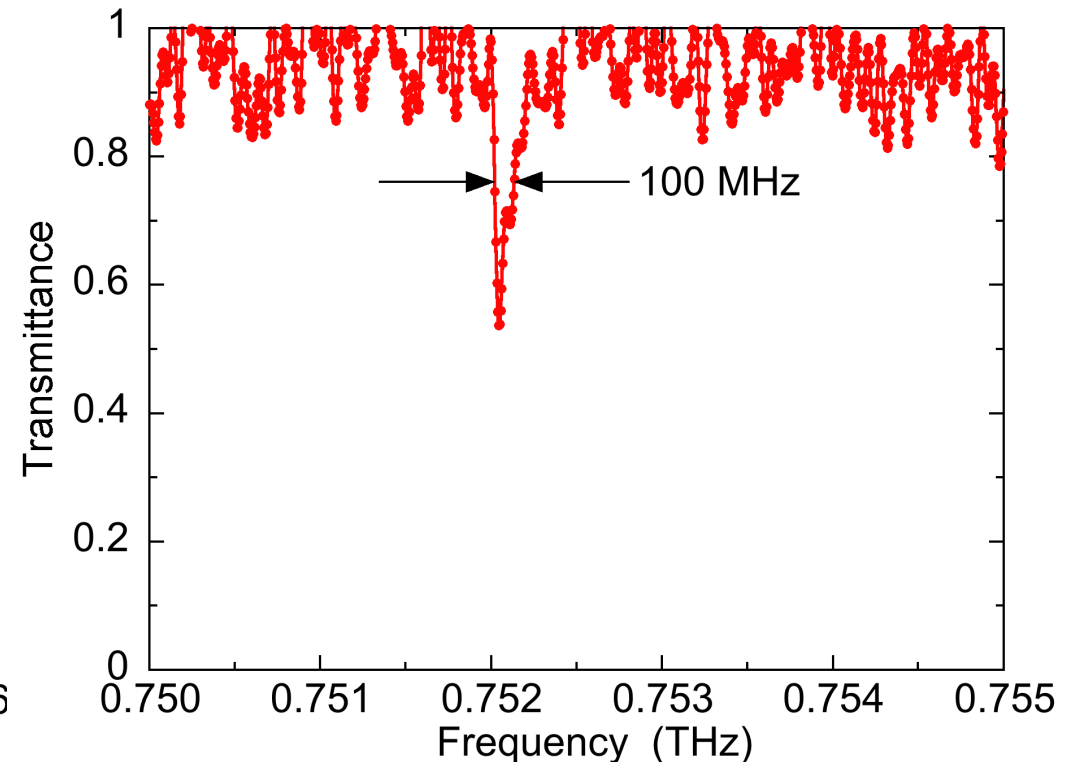
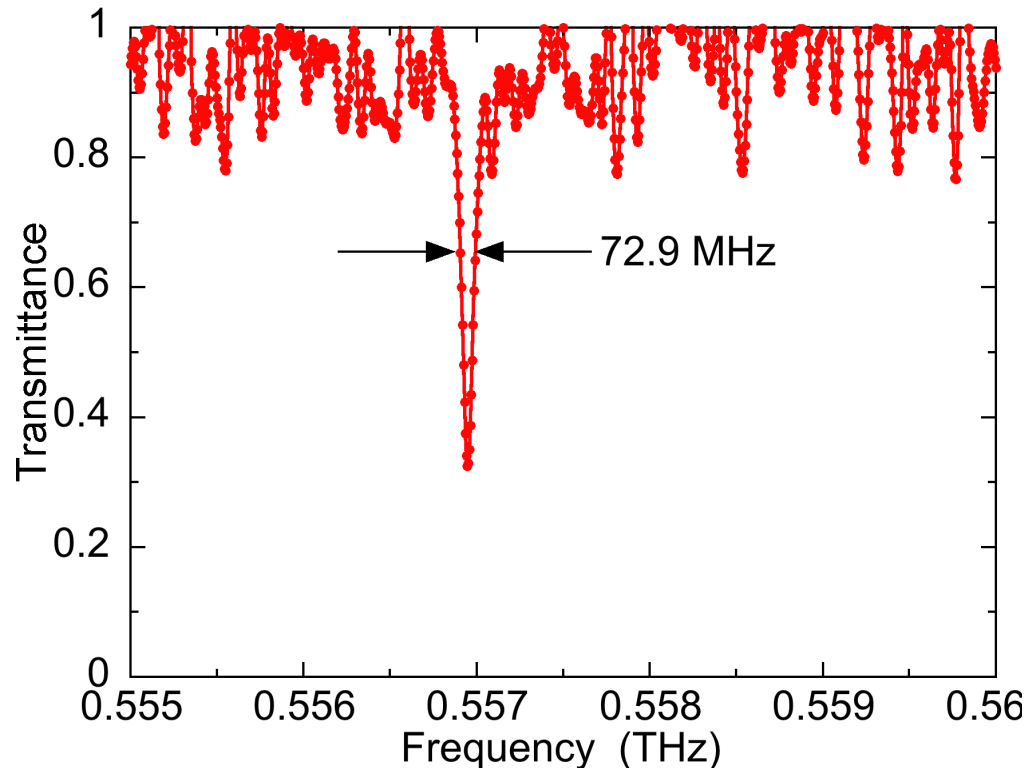
Transition	Absolute frequency (NASA database)	Pressure broadening at 1 kPa <sup>1)</sup>
$1_{10} \rightarrow 1_{01}$	0.556 936 0 THz	30.1 MHz
$2_{11} \rightarrow 2_{02}$	0.752 033 2 THz	29.7 MHz

<sup>1)</sup> T. Seta, et al., *J. Quantitative Spectroscopy & Radiative Transfer* **109**, 144-150 (2008).



# Evaluation of spectral resolution

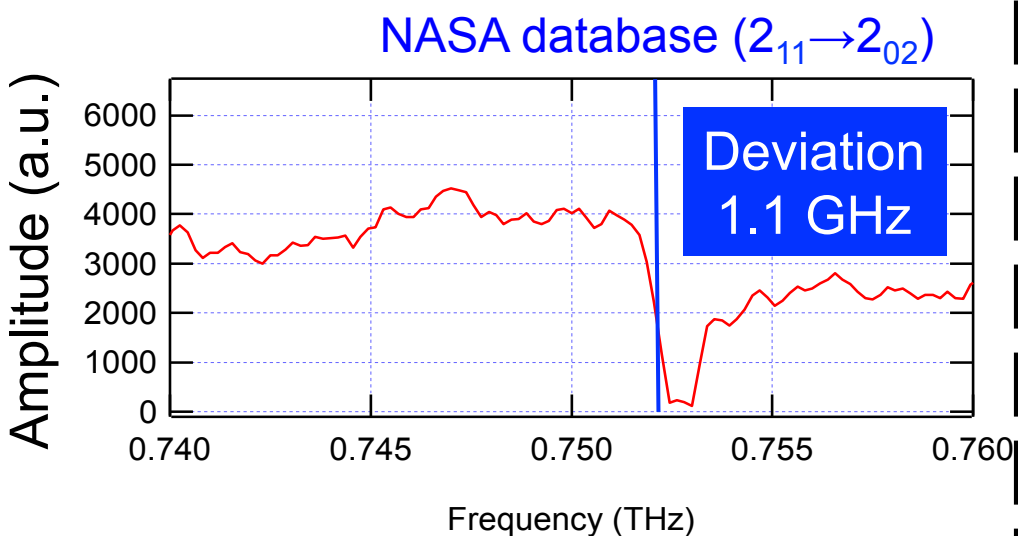
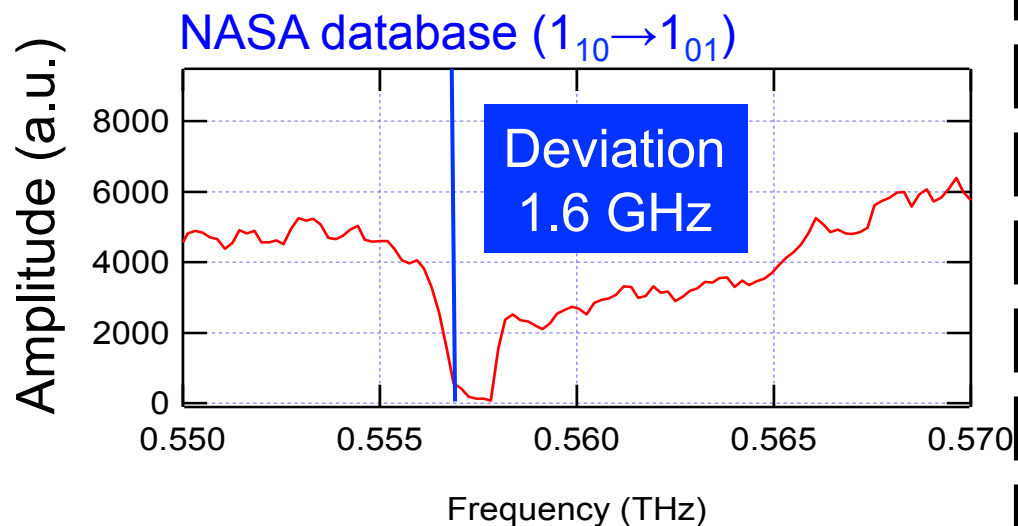
Time window = 17.9 ns (= one pulse period)  
Inverse of time window = 56 MHz



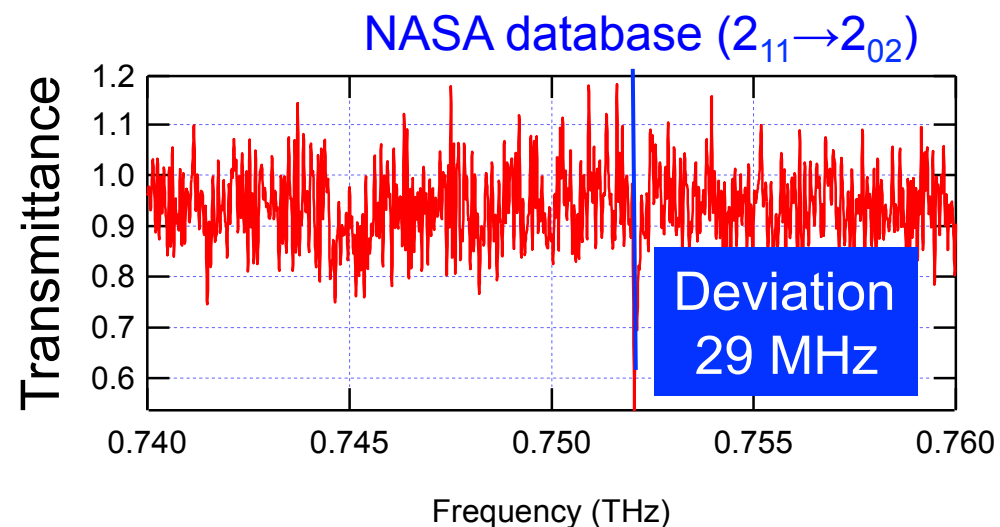
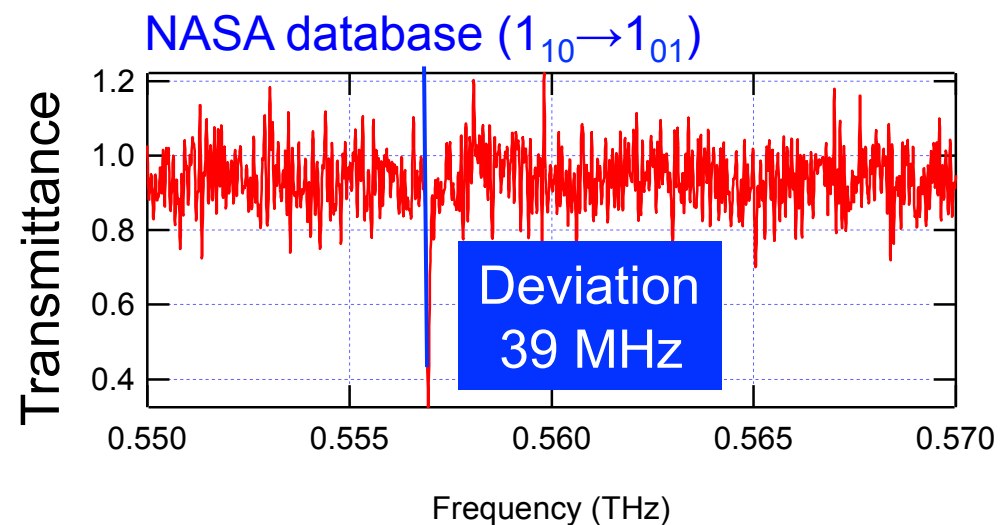
Actual spectral resolution  $\geq$  inverse of time window  
Decline of spectral resolution by timing jitter is small

# Frequency deviation from NASA database

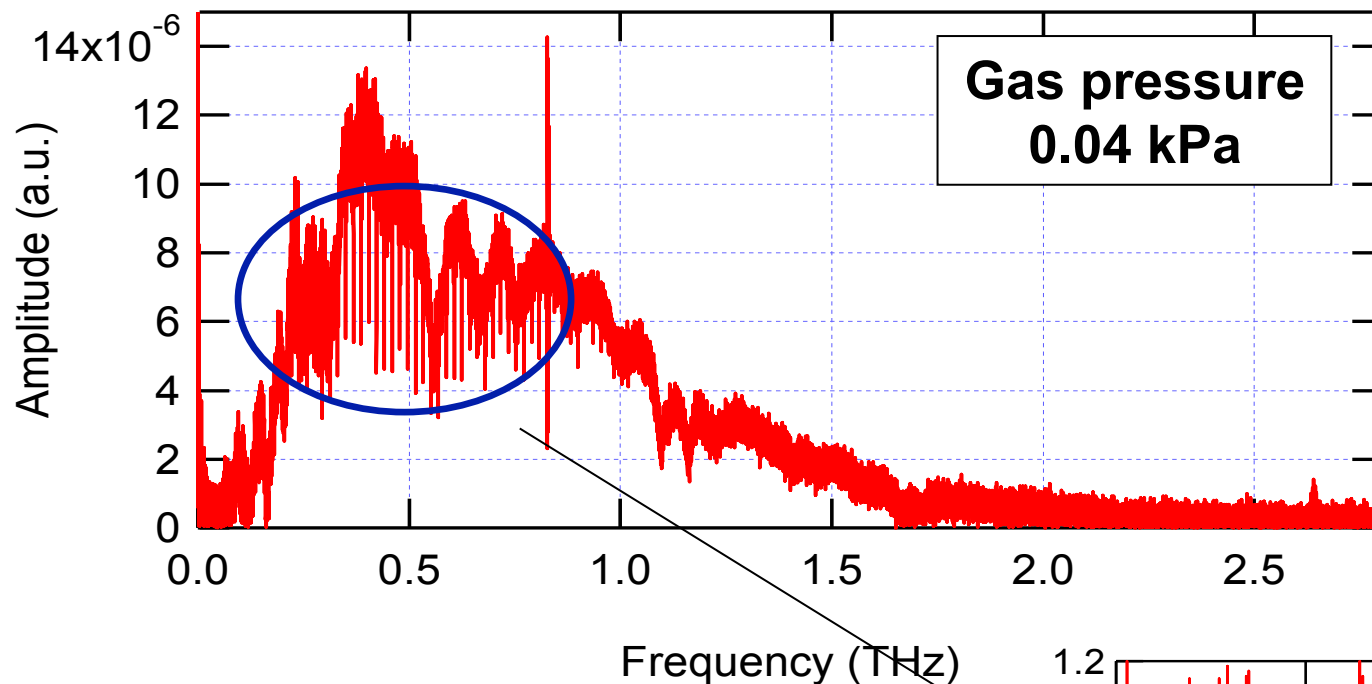
## Conventional THz-TDS



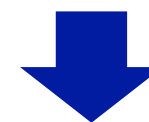
## ASOPS-THz-TDS



# Spectroscopy of acetonitrile gas

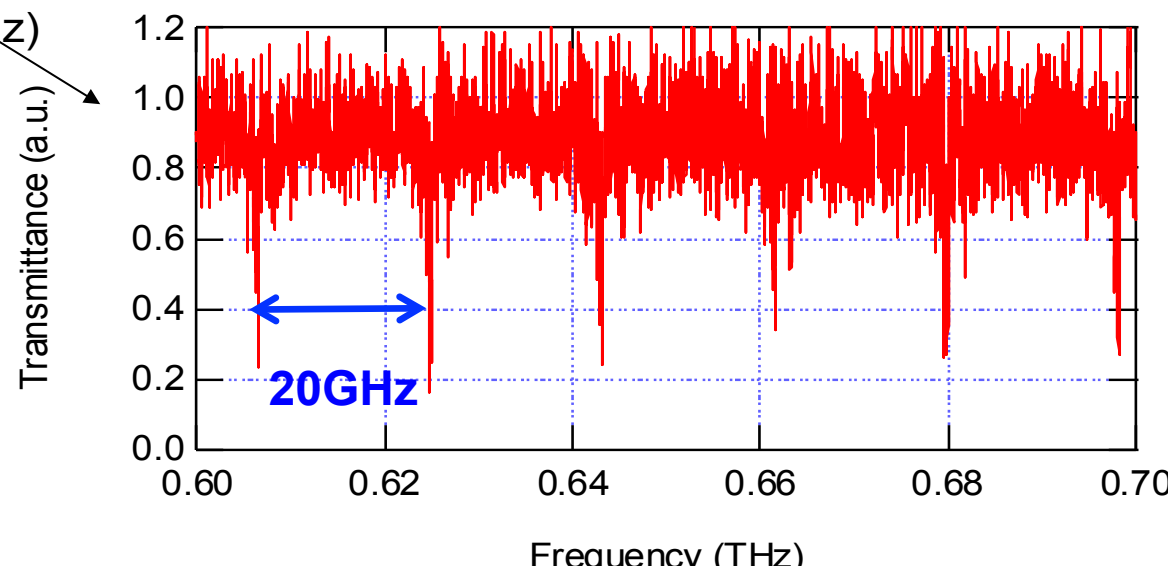


Interstellar molecule

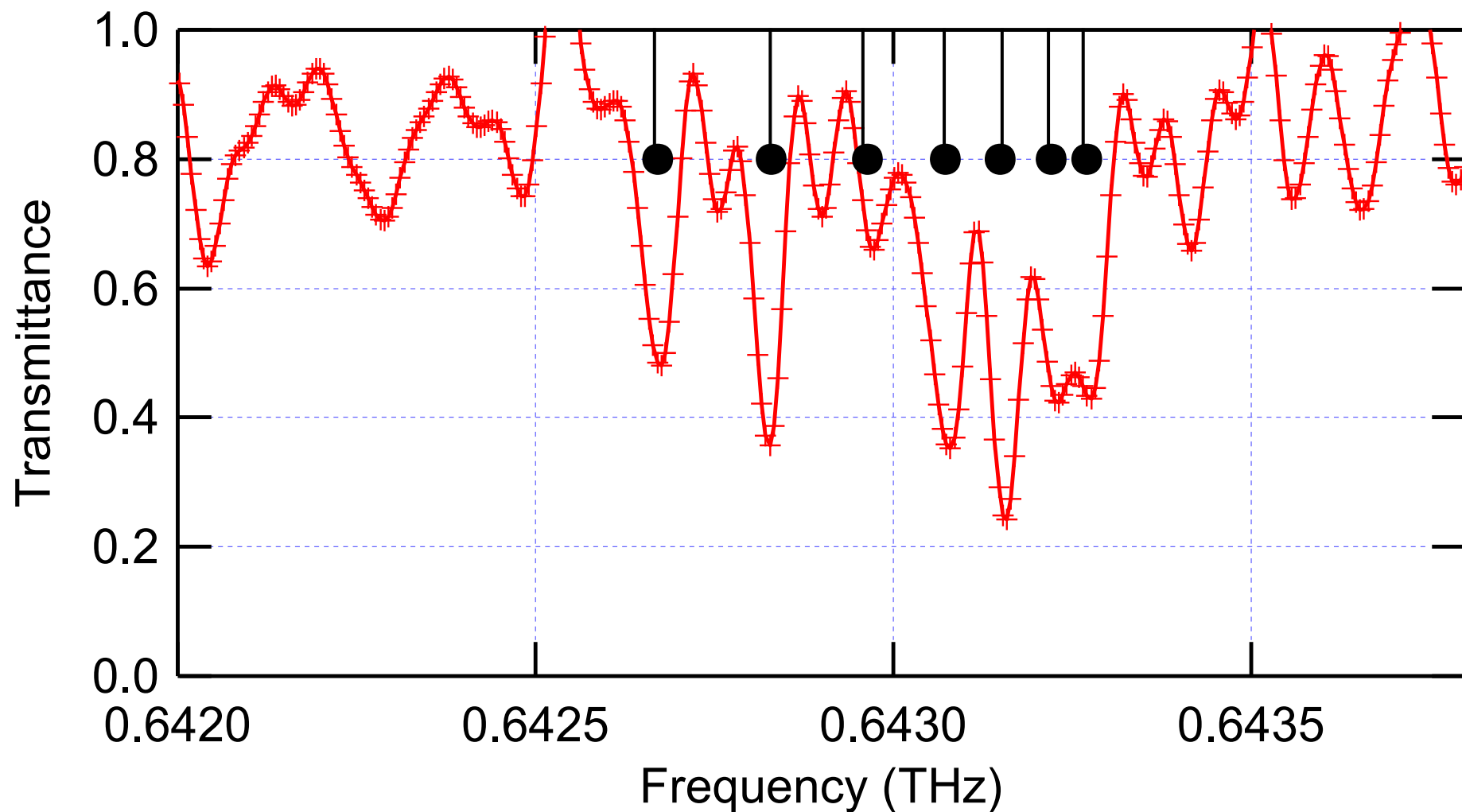


Important in the field  
of radio astronomy

Groups of absorption lines  
periodically appeared  
at an interval of 20 GHz



# Hyperfine structure of absorption lines in acetonitrile molecules



# Evaluation of spectral accuracy using absorption lines of acetonitrile

Experimental results [THz]	NASA database [THz]	Deviation[ MHz]
0.642 675 924 0	0.642 670 849 7	5.0743
0.642 827 458 8	0.642 829 579 3	2.1205
0.642 973 381 2	0.642 963 997 8	9.3834
0.643 080 016 7	0.643 074 051 0	5.9657
0.643 158 590 4	0.643 159 694 2	1.1038
0.643 231 551 6	0.643 220 892 8	10.6588
0.643 276 450 8	0.643 269 866 7	6.5841

**Mean accuracy =  $10^{-5}$**

# Discussion on spectral accuracy

Achieved accuracy =  $10^{-5}$   $\gg$  Rb atomic clock =  $10^{-12}$

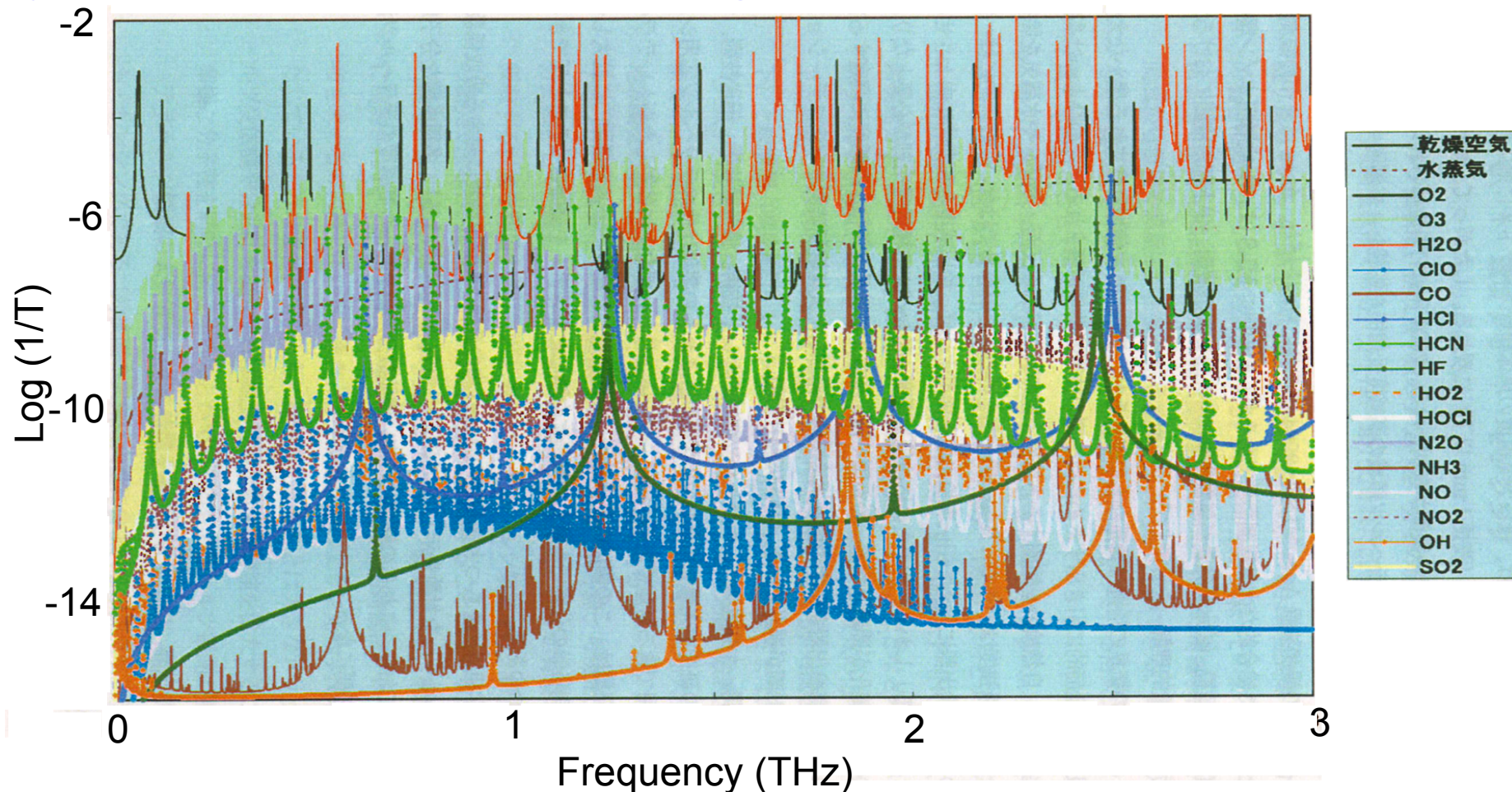

 Fluctuated by laser instability  
 $f_{THz}$   
 $\Delta f_{AS}$

	Frequency fluctuation	Mean frequency	Instability
$f_1$	several 10s mHz	56 MHz	$10^{-12}$
$\Delta f$		5 Hz	$10^{-5}$

**Limit the spectral accuracy**

# Future plan: Analysis of atmospheric gas molecules

Many absorption line of molecular gases are concentrated in THz region



*To identify each molecular gases correctly, high accuracy, high resolution, and broadband are required.*

# Conclusions

- (1) Evaluation of actual spectral resolution and accuracy in ASOPS-THz-TDS using low-pressure gas spectroscopy
  - resolution = 72.9 MHz, accuracy =  $10^{-5}$
  
- (2) Decline of spectral resolution caused by timing jitter is small.
  
- (3) Spectroscopy of acetonitrile molecular gas
  - periodical groups of absorption lines at every 20 GHz
  - hyperfine structure of individual absorption lines