Le couplage entre optique ultra-rapide et microscopie à effet tunnel (le THz-STM)

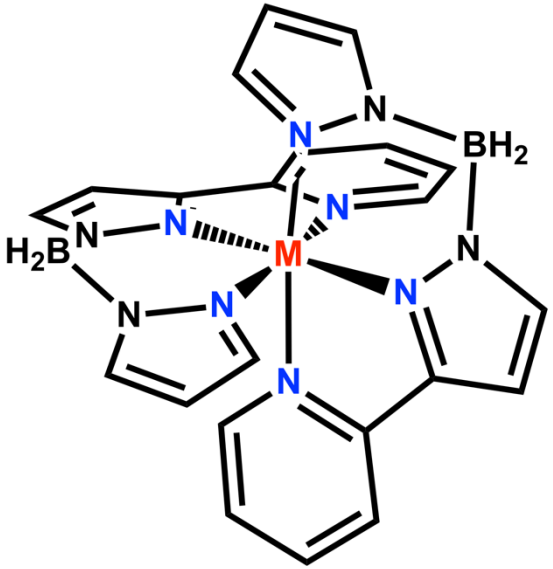
Manuel Gruber

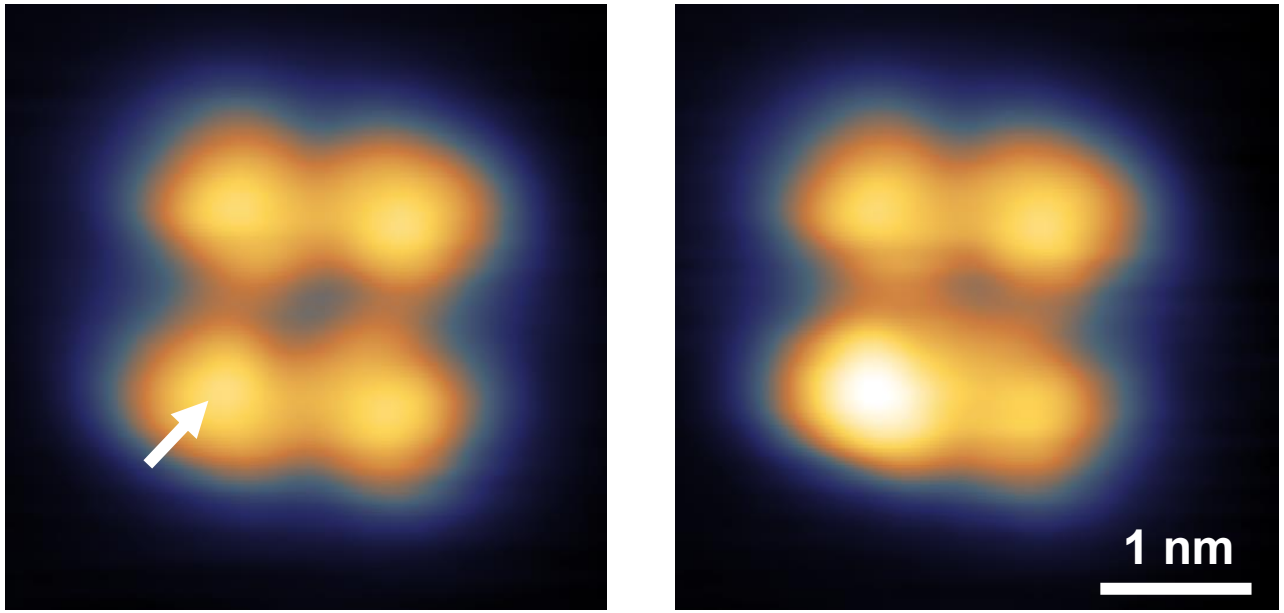
Couplage de la microscopie à sonde locale et la nanophotonique

UNIVERSITÄT
DUISBURG
ESSEN

Offen im Denken

Why temporal resolution? Example of spin crossover





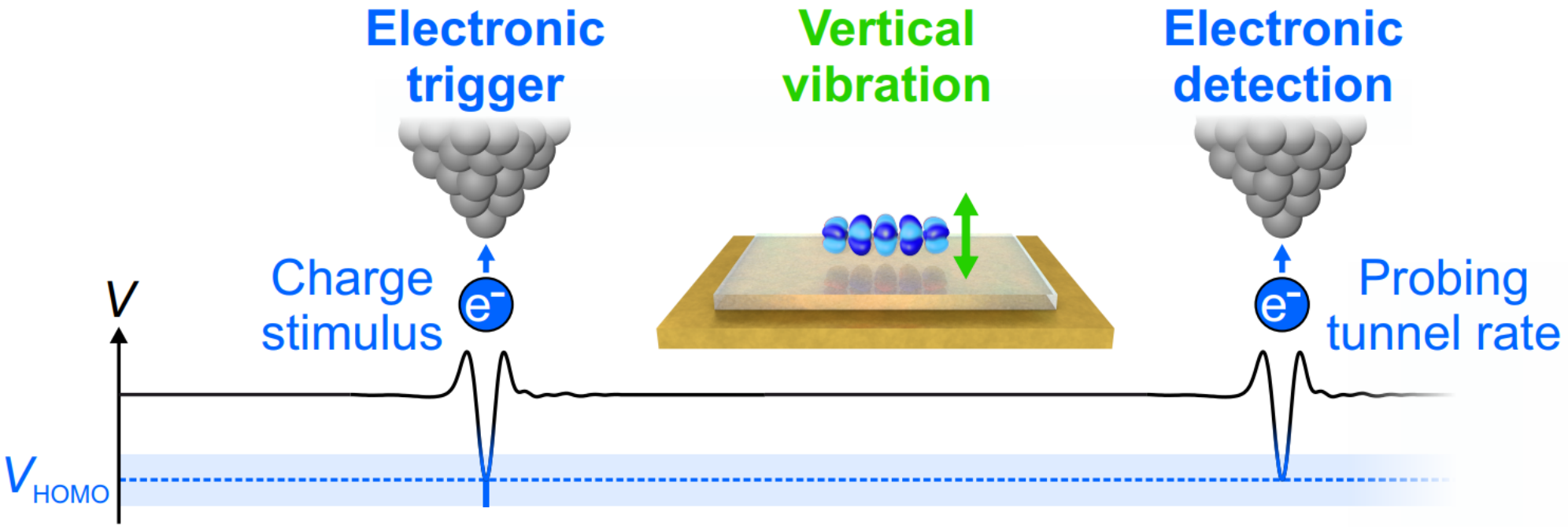
- Spin-state switching of Fe(II) complexes at the single-molecule level
- Switching triggered by tunnel electrons, involve transient occupation of orbital, and most likely excitation of vibrational modes
- We miss all the dynamics with conventional STM; only see the result

Time scales

- Transient charging: fs to ps
- Vibrations: 100 fs to 100 ps
- Chemical reactions: ps to ns

Johannsen, … MG,ACS Nano **15**, 11770 (2021)

Ultrafast dynamics of a single molecule

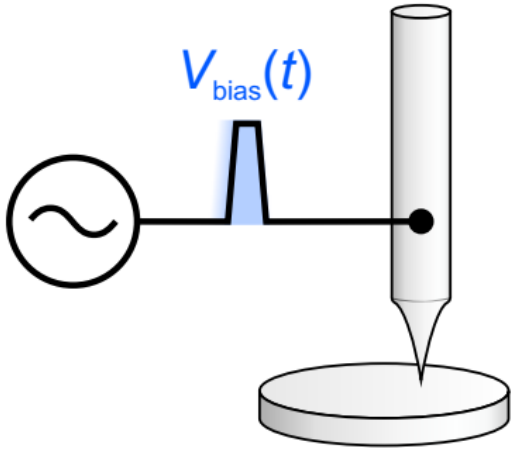


Cocker et al., Nature 539, 263 (2016)

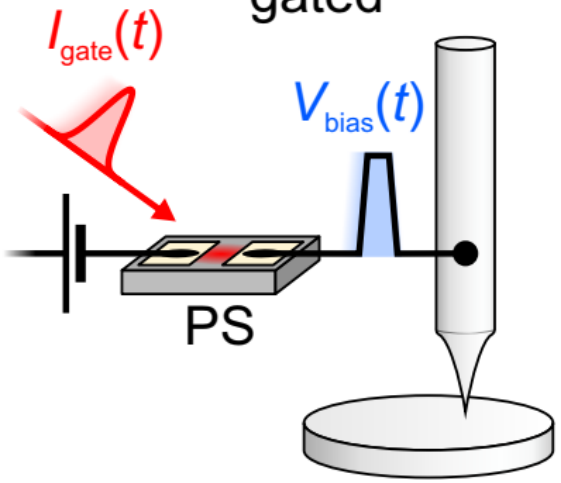
Peller, PhD thesis (2020)

How to achieve short voltage/current pulses?

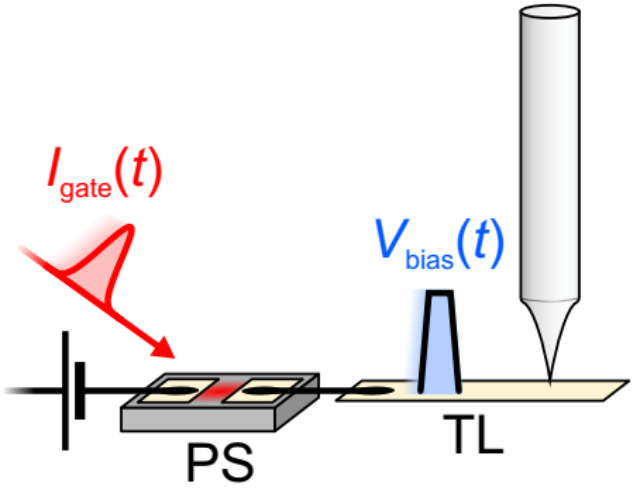
All-electronic



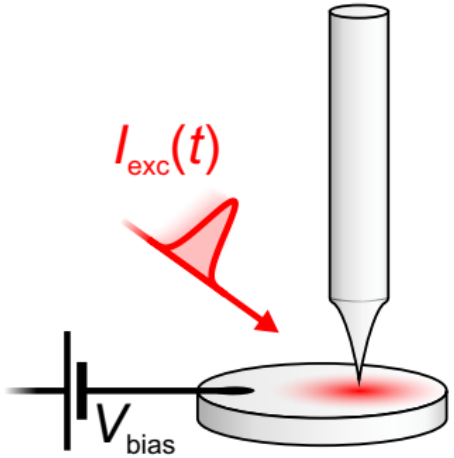
Photoconductively gated



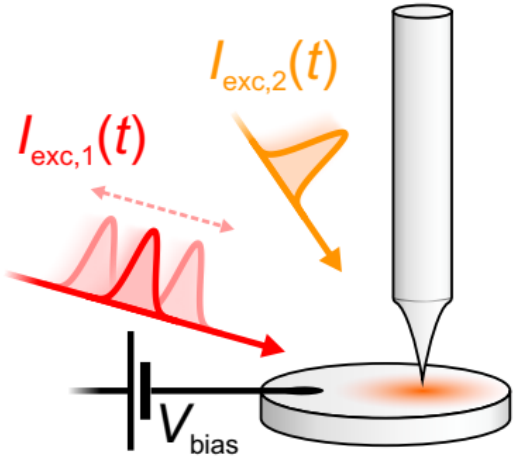
Junction-mixing



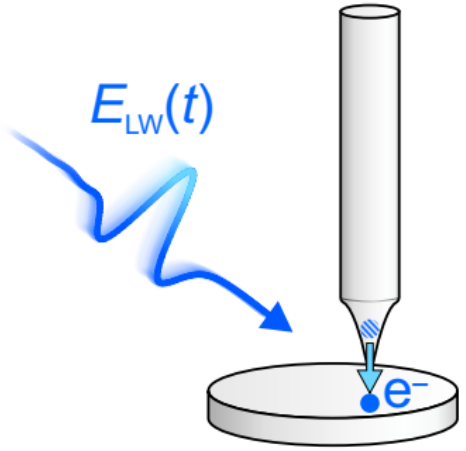
Surface photovoltage



Shaken-pulse-pair excited

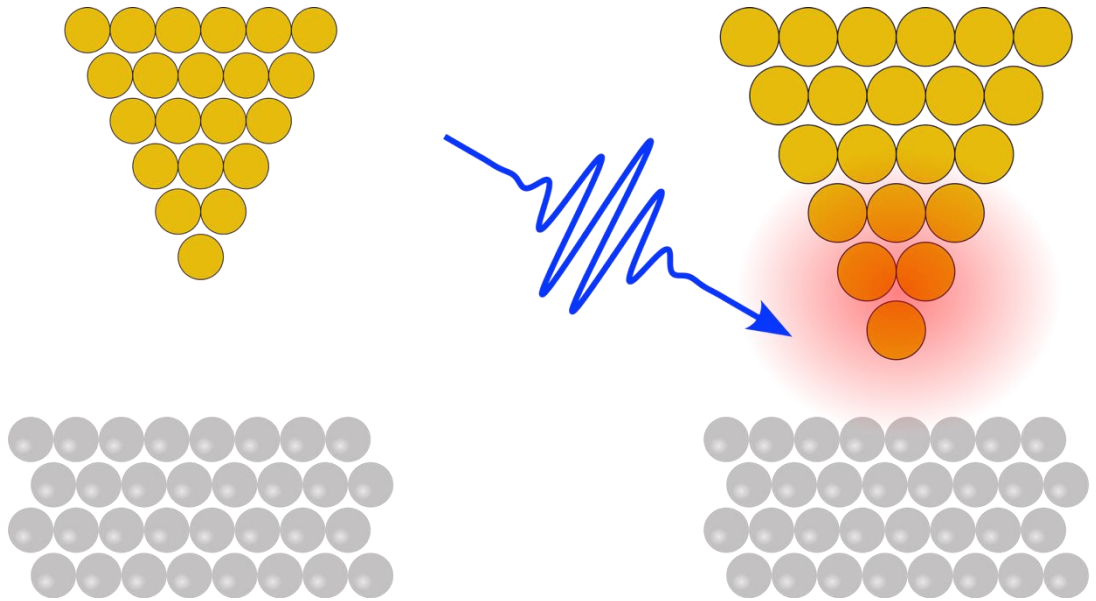


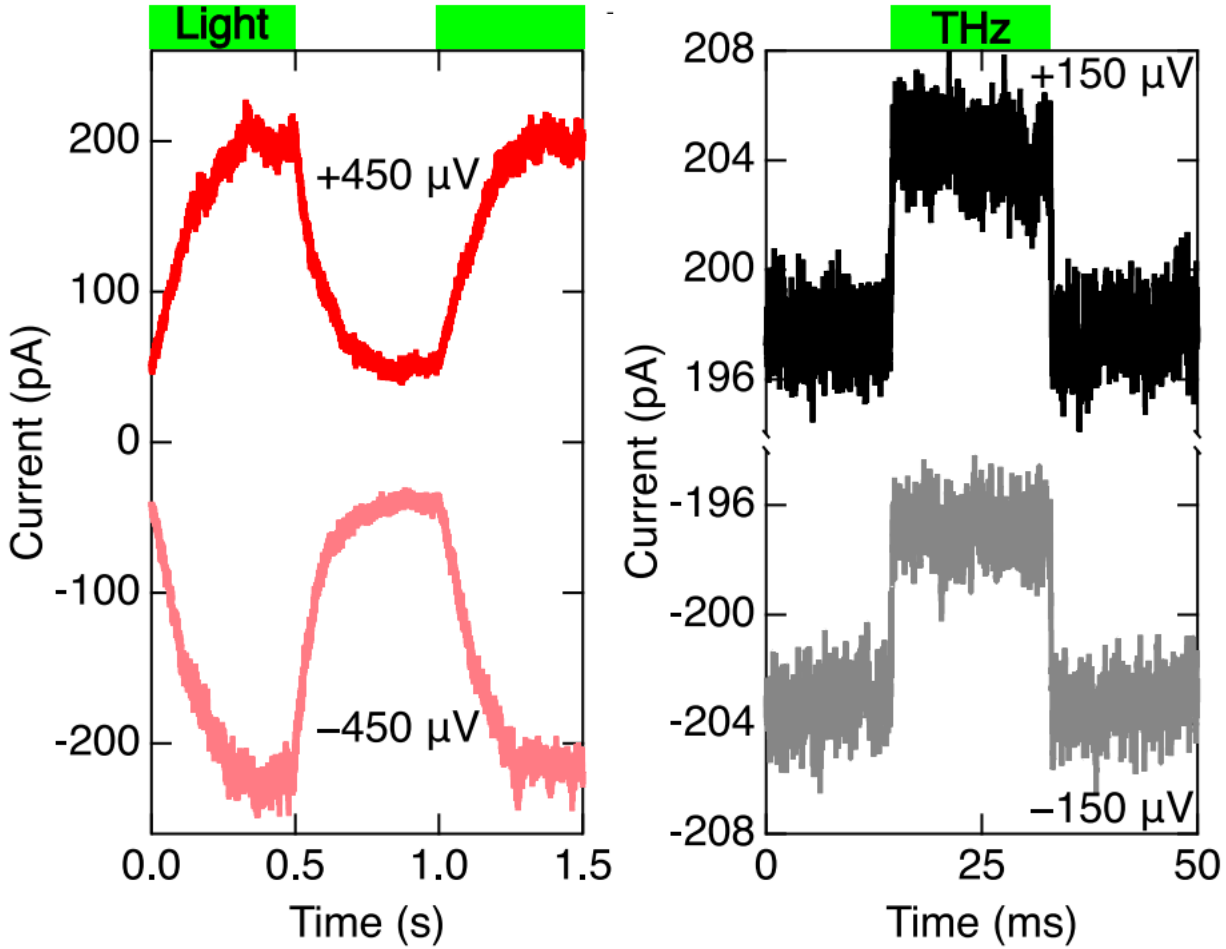
Lightwave-driven



Peller, PhD thesis (2020)

Thermal effects at the junction under illumination





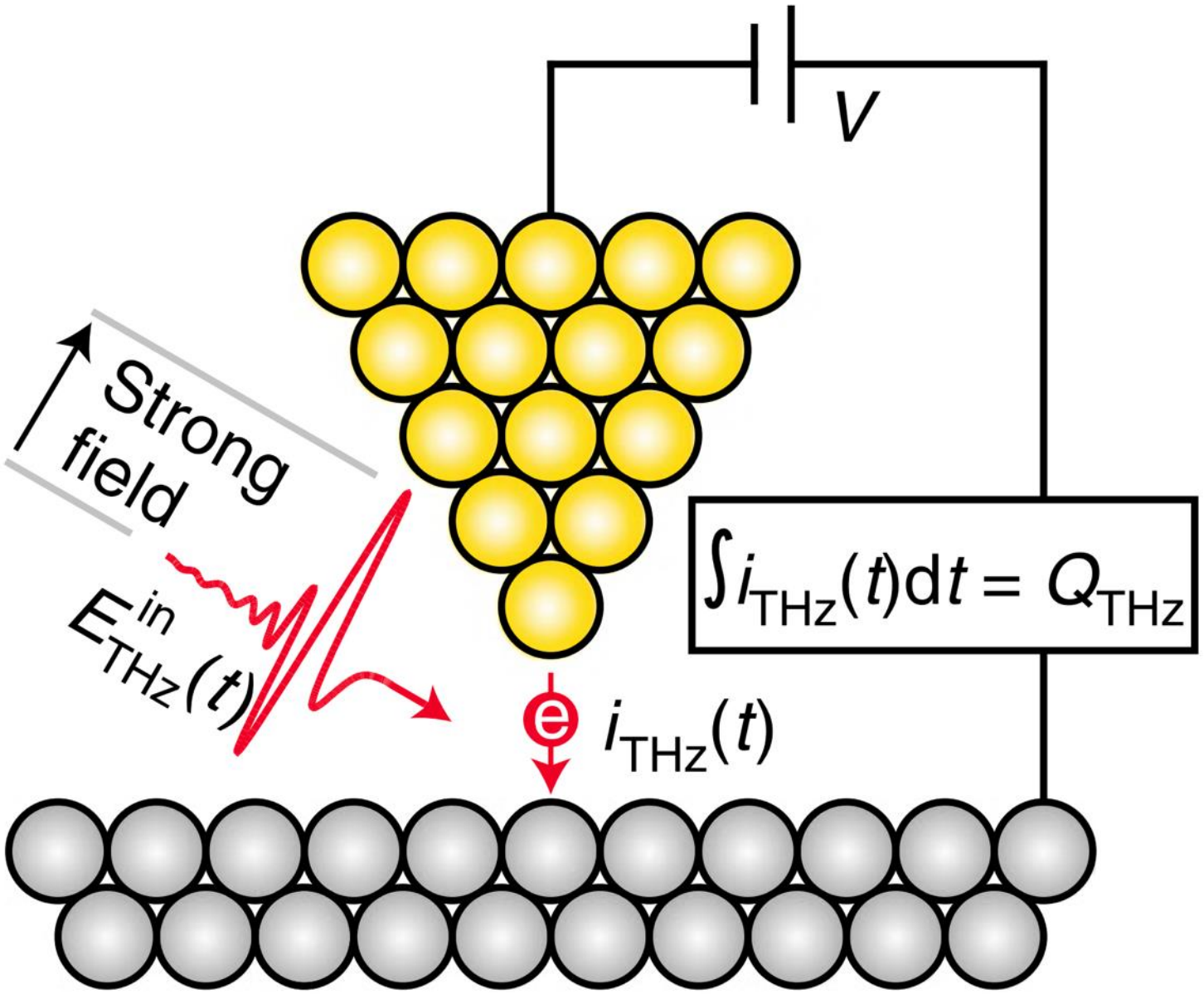
- Typically, mW visible light for photo-induced current (versus µW for THz)
- Energy absorbed, and lead to thermal expansion (ms dynamics)
- The problem is NOT the thermal expansion but its variation over time (chopping)
- Practical tricks such as shaken-pulse-pair

Grafström *et al.*, J. Vac. Sci. Technol. B **9**, 568 (1991)
Azazoglu, …, MG, Surf. Sci. 743, 122465 (2024)

Lightwave-driven scanning tunnelling microscopy

What is it?

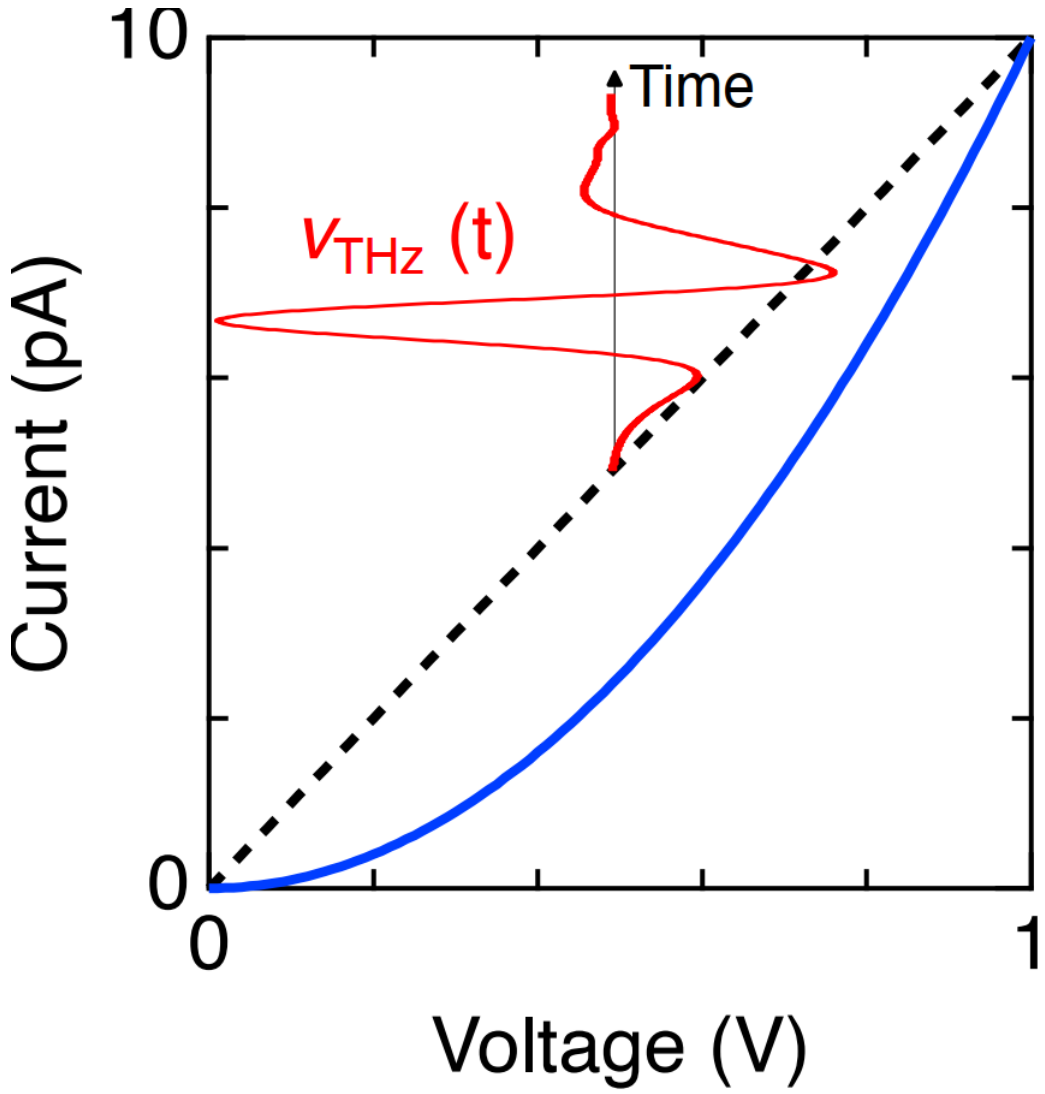
Lightwave-driven STM

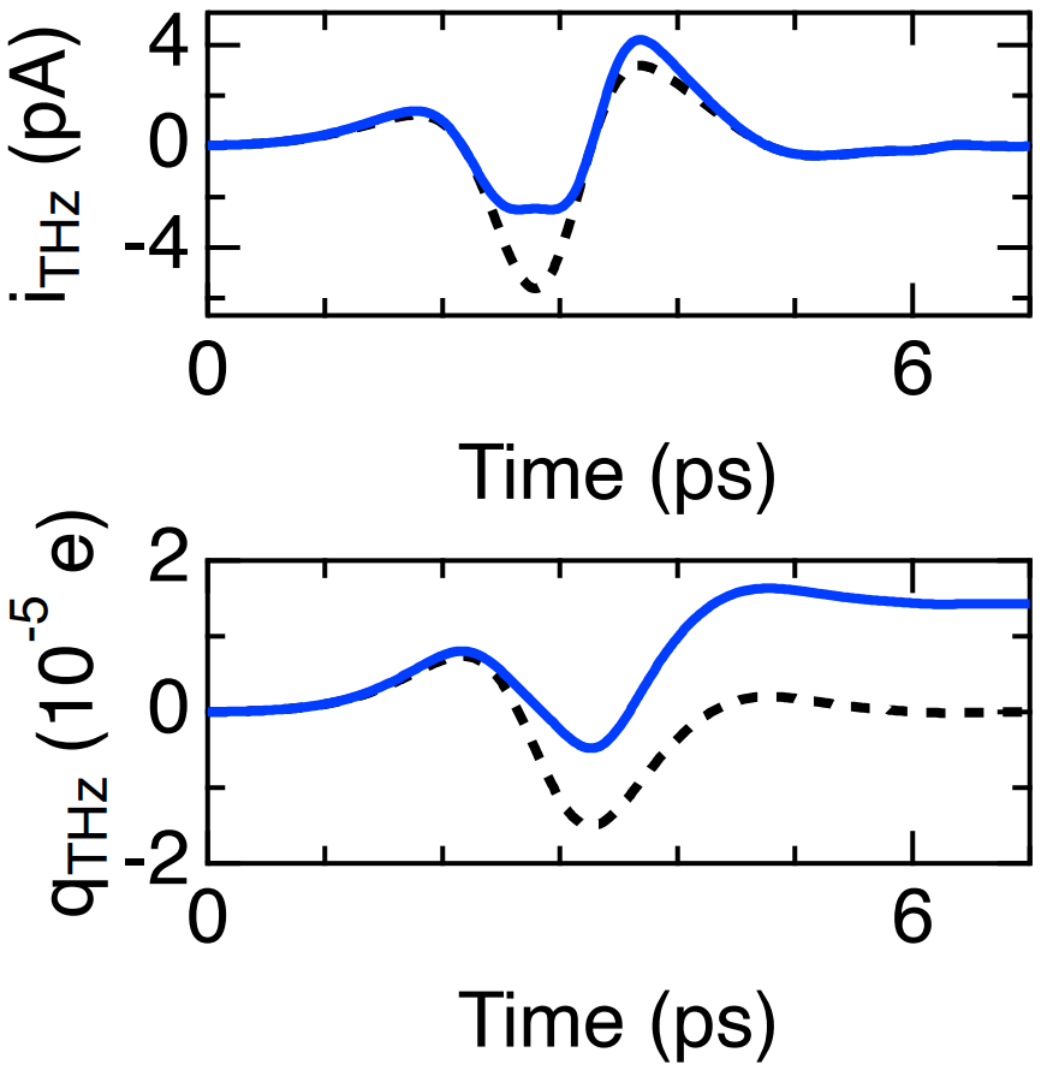


- Far-field THz couples to STM junction
- Field enhancement by factor ~10^5 at junction (near field, antenna effect)
- Electric field between metal electrodes equivalent to transient voltage
- Transient tunnelling current $i_{THz}(t)$, whose integral leads to a net transfer of charge
- Most common scheme for time resolution
  - Tunnel e^- triggers the dynamics
  - Change of junction's conductance is probed

Cocker *et al.*, Nat. Photonics **15**, 558 (2021)

Rectification of the THz pulse





- Time-integral of far-field THz pulse is zero
- Net transfer of charge if and only if *I-V* characteristic is non-linear

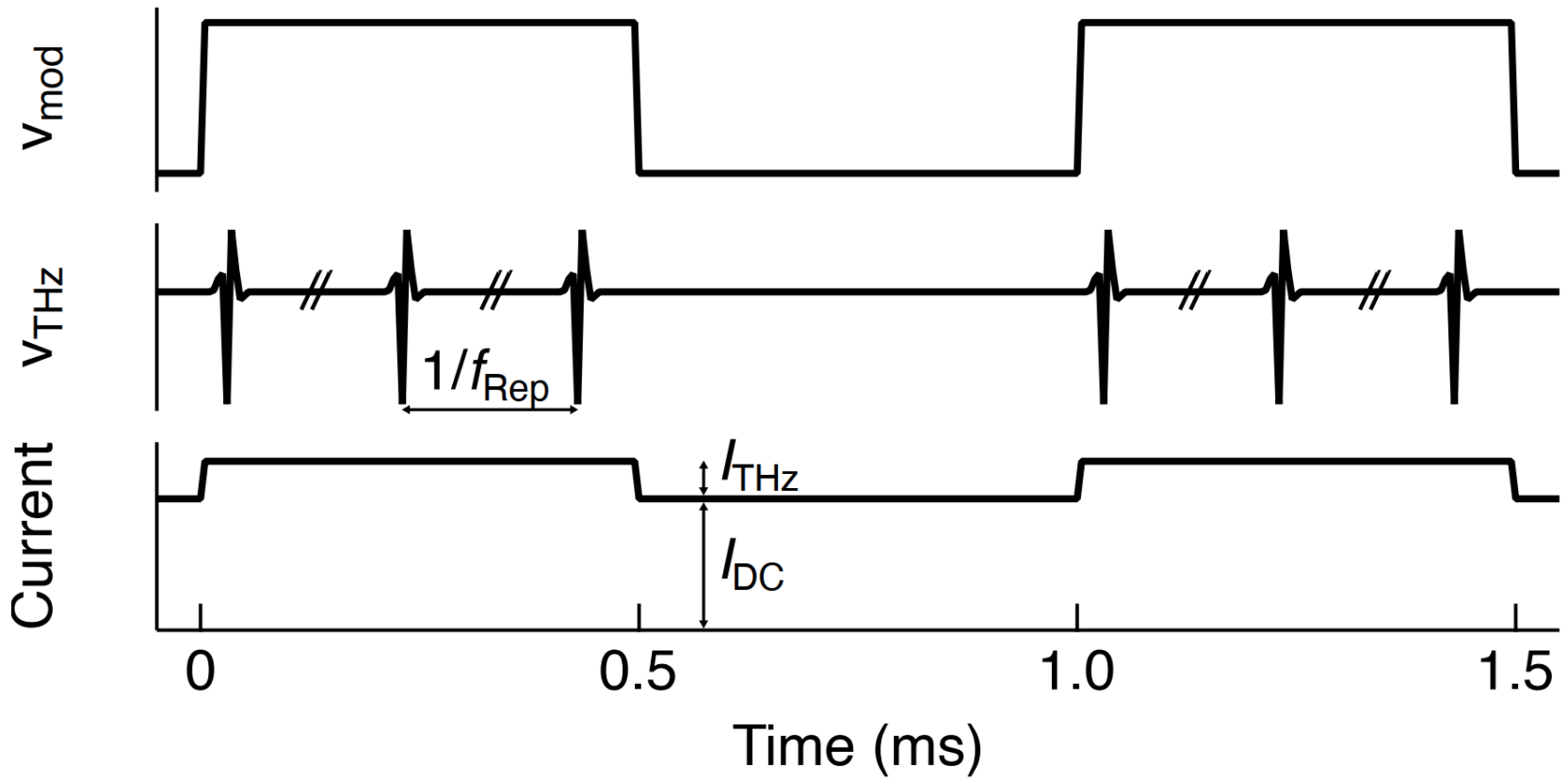
Current detection via lock-in

What current to expect?

- Assume
  - 1 e^- transferred per pulse
  - 1 MHz repetition rate (high)
- 200 fA of THz-induced current vs. ~10 pA DC tunnel current

Lock-in detection

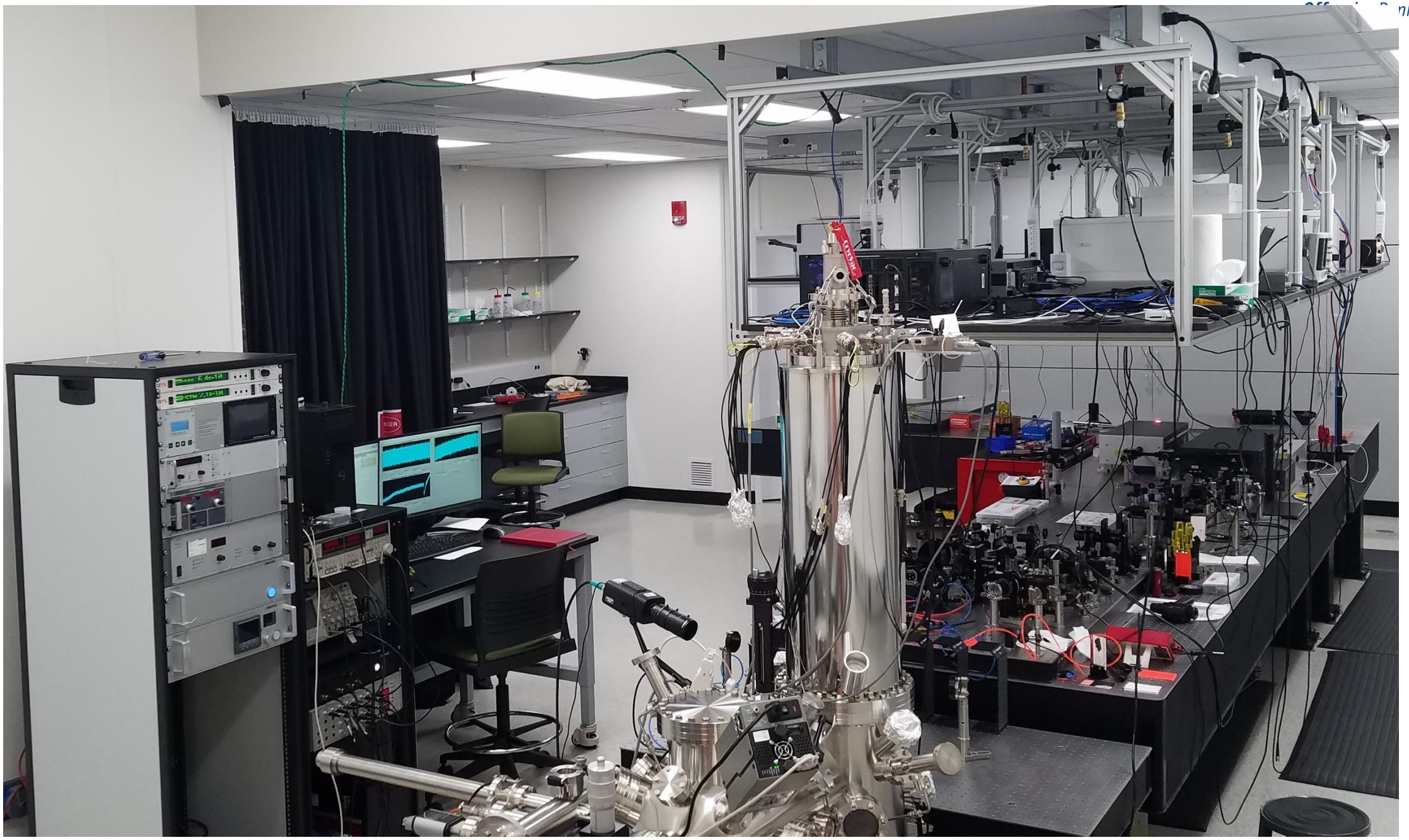
- THz pulses are chopped with $f_{Rep} \approx 1$ kHz
- Lock-in extract f_{Rep} component from the tunnelling current
- Convert lock-in output into I_{THz} , the average THz-induced current



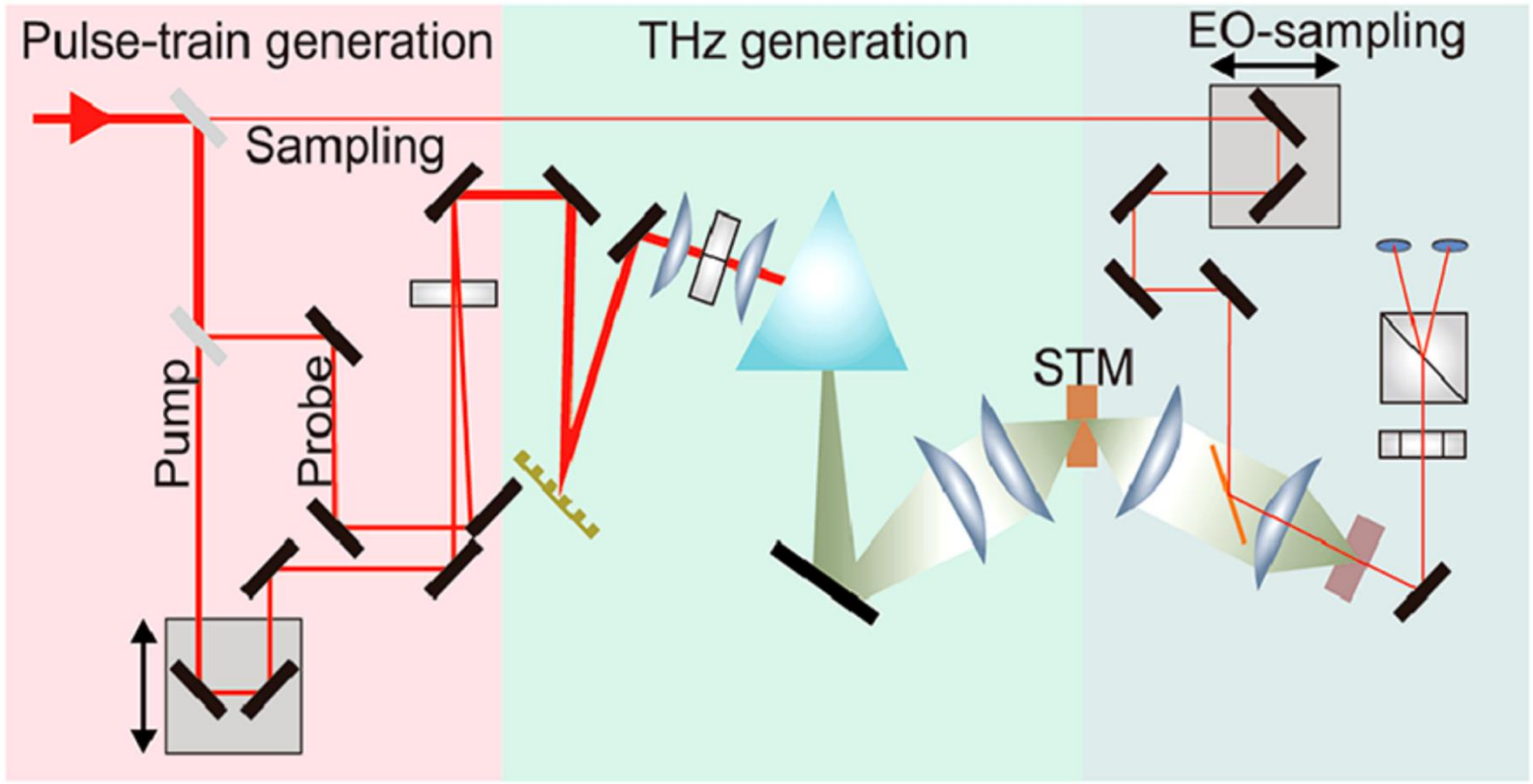
Lightwave-driven scanning tunnelling microscopy

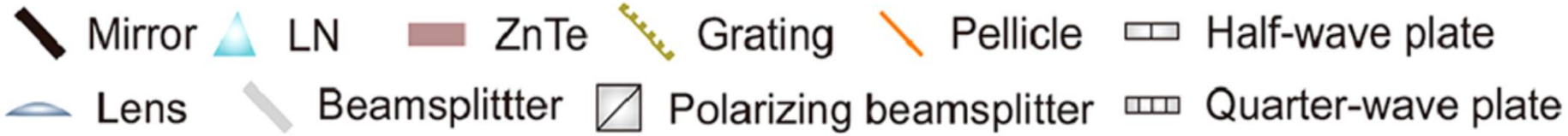
How to implement it?

Tyler Cocker's lab



Another example from the Loth's group (Stuttgart)



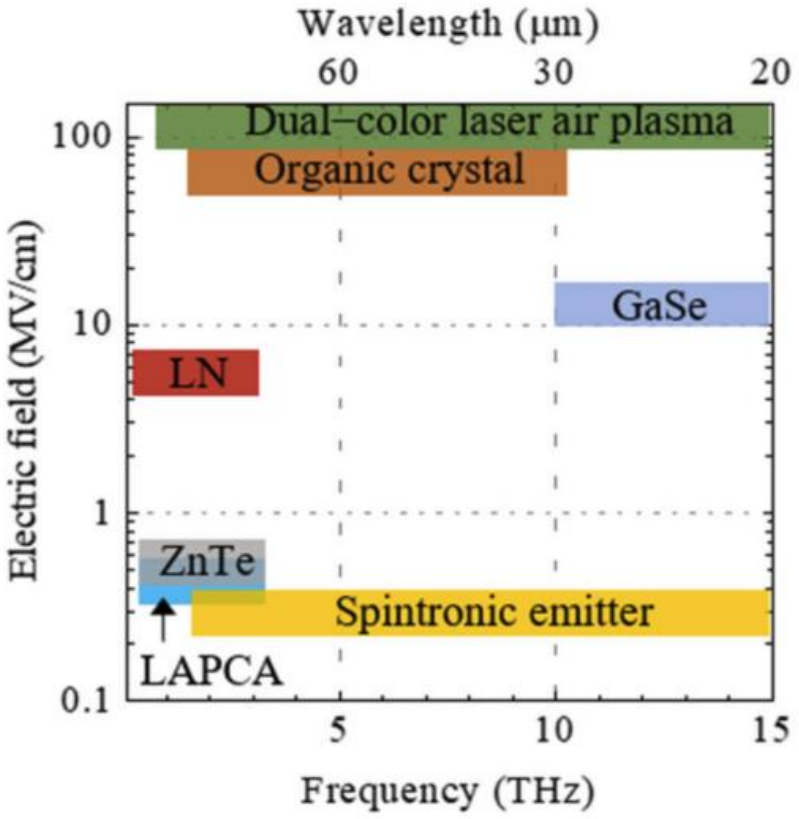


Key steps

- Pulse-train generation
- THz generation
- THz Focusing
- Survey of far-field THz waveform

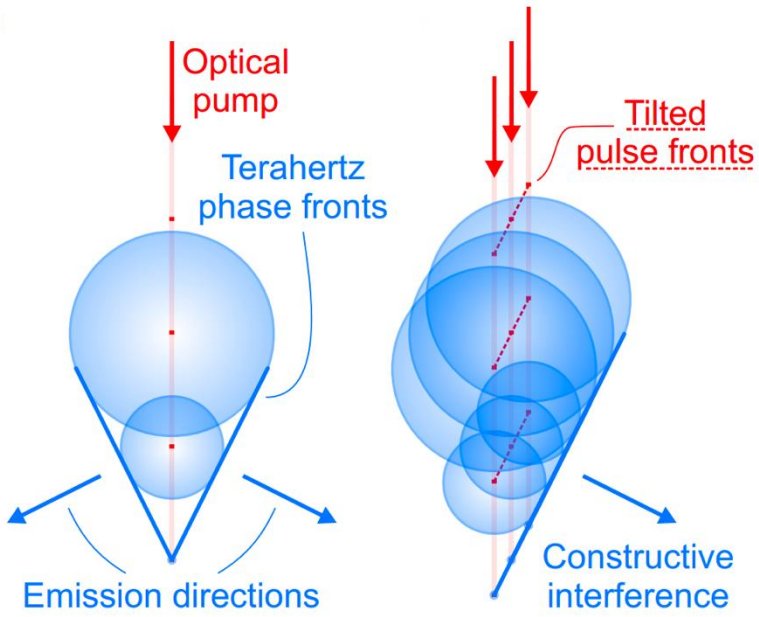
Abdo *et al.*, ACS Photonics **8**, 702 (2021)

Generation of THz pulses



Zhu *et al.*, Photonics **8**, 183 (2021)

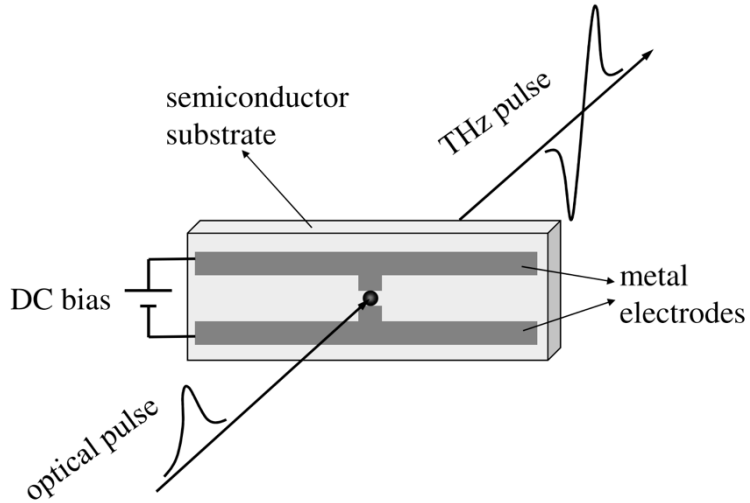
Tilted pulse front rectification (LN)



Peller, PhD thesis (2020)

- Method of reference
- Produce large THz fields
- Require energetic optical pump

Photoconductive antenna



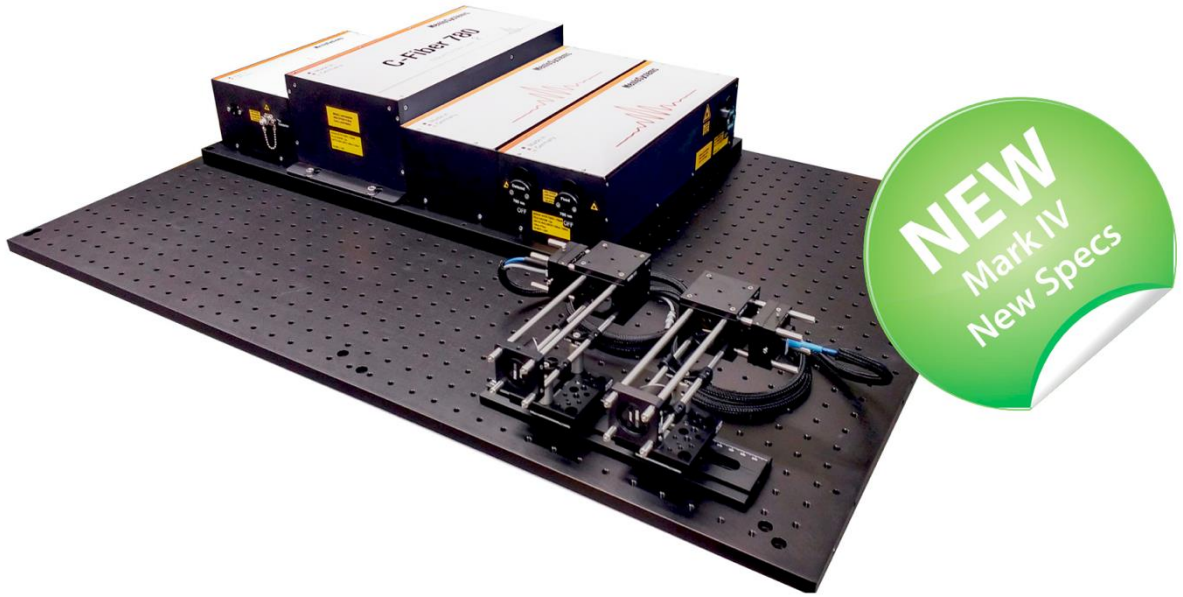
Lee, Principle of THz Science and Technology (2009)

- Commercial turn-key solution for time-domain THz spectroscopy
- Produce low-power THz pulses (improving)

Commercial, turn-key solution for THz generation

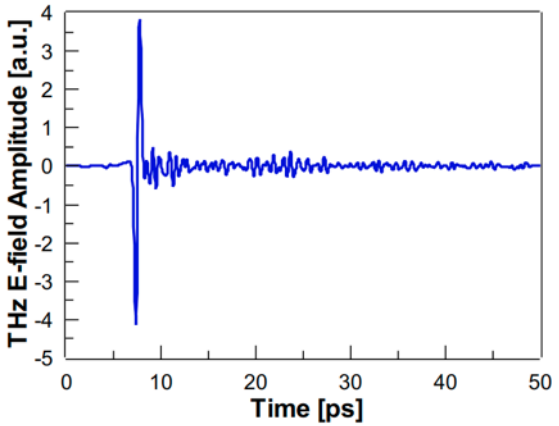
TERA K15

Versatile, All-fiber THz Time-Domain Platform
Based on 1560 nm Femtosecond Fiber Laser

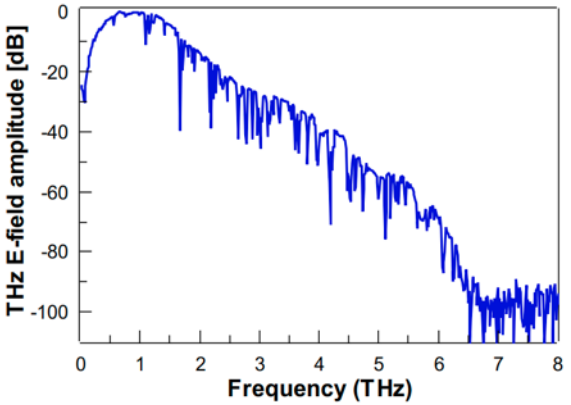


MenloSystems

THz pulse measured in ambient air*



THz spectrum showing absorption lines of atmospheric water vapor

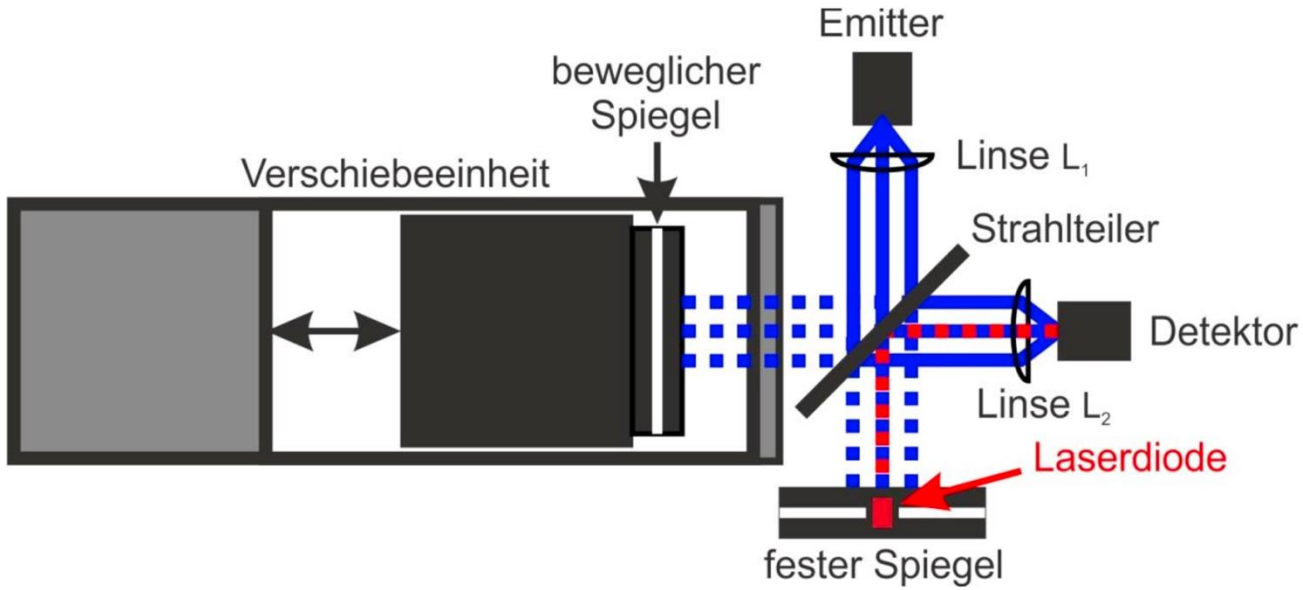


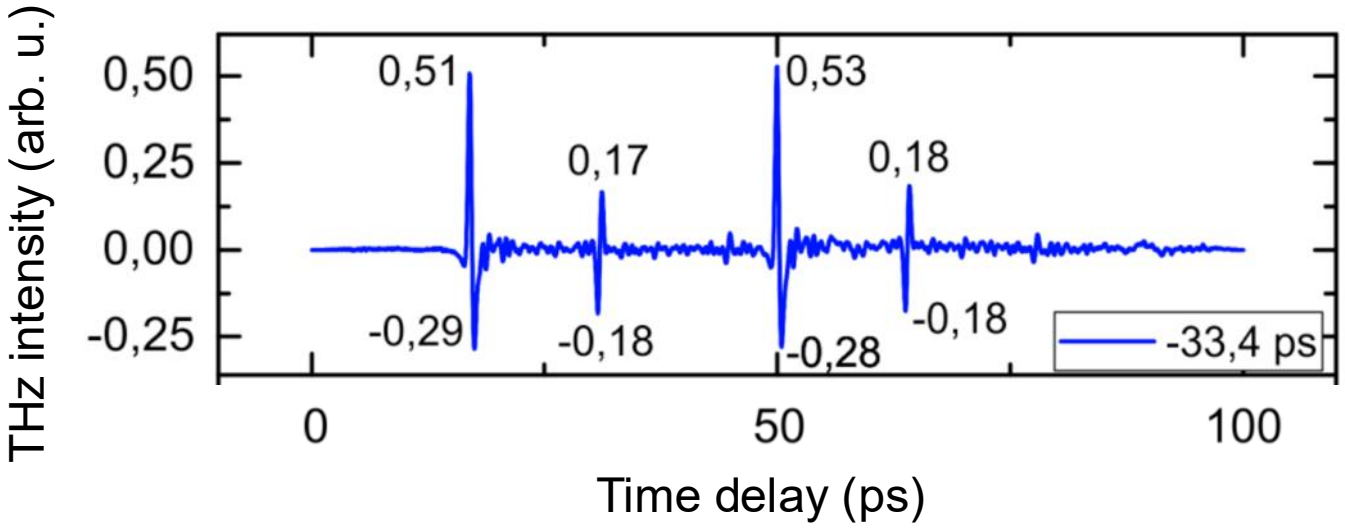
- Turn-key system
- Light transmitted via fiber
- Includes photoconducting-antenna emitter & detector

Need to

- Split & delay the THz pulse (pump-probe)
- Focus on the THz beam on the junction

Splitting of THz beam



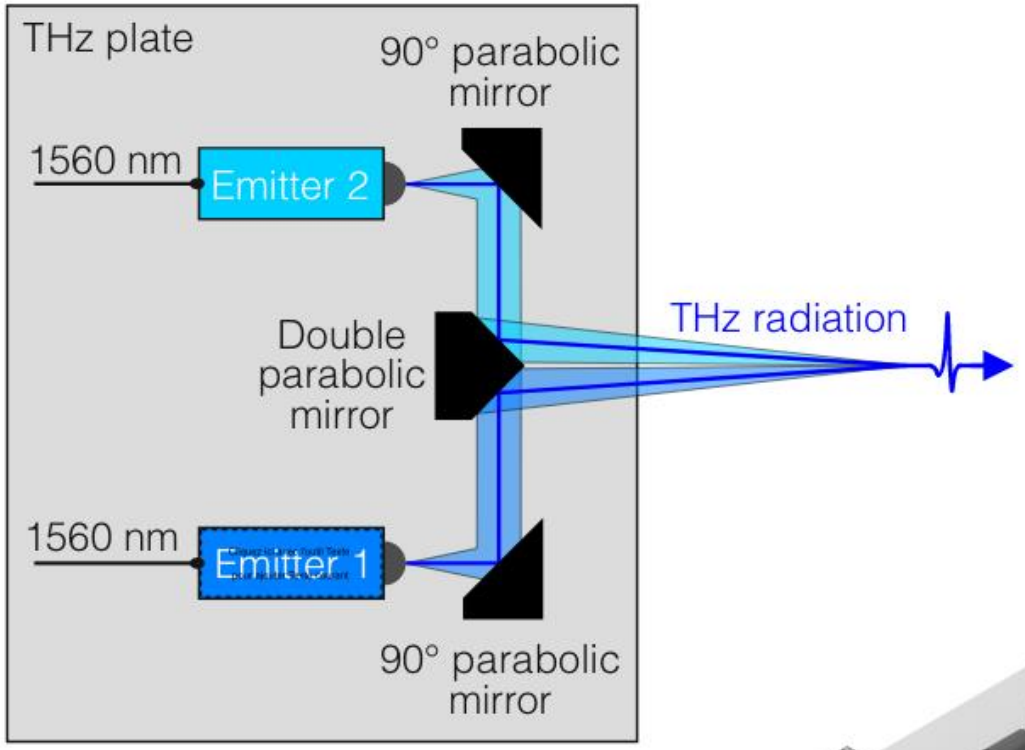


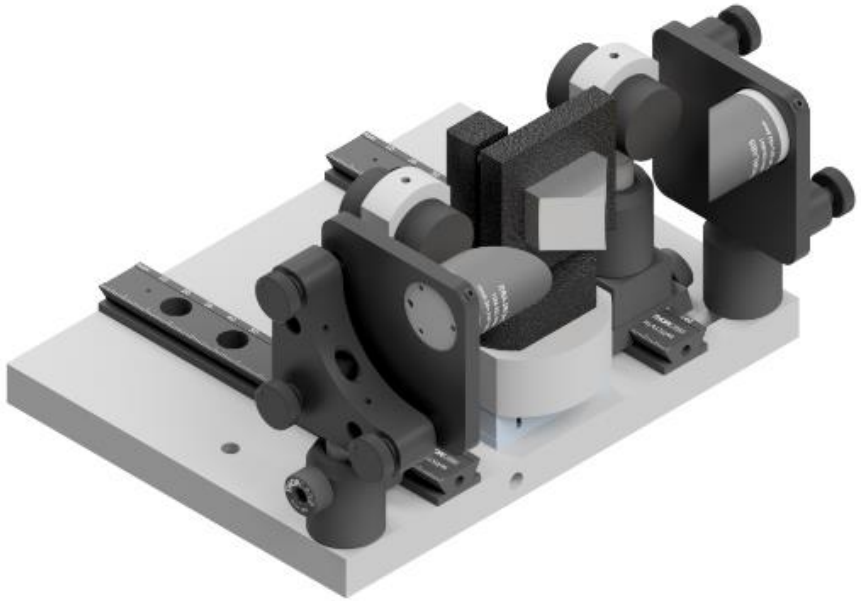
- Used high-resistive Si as splitter
- Internal reflections produce echos, that can be damped with metal coating
- Metal coating is leading to absorption
- Overall: Beam-splitting is challenging if beam intensity is low!

Our strategy

- Split the laser excitation and use two emitters
- Then focus the two beams to the STM junction

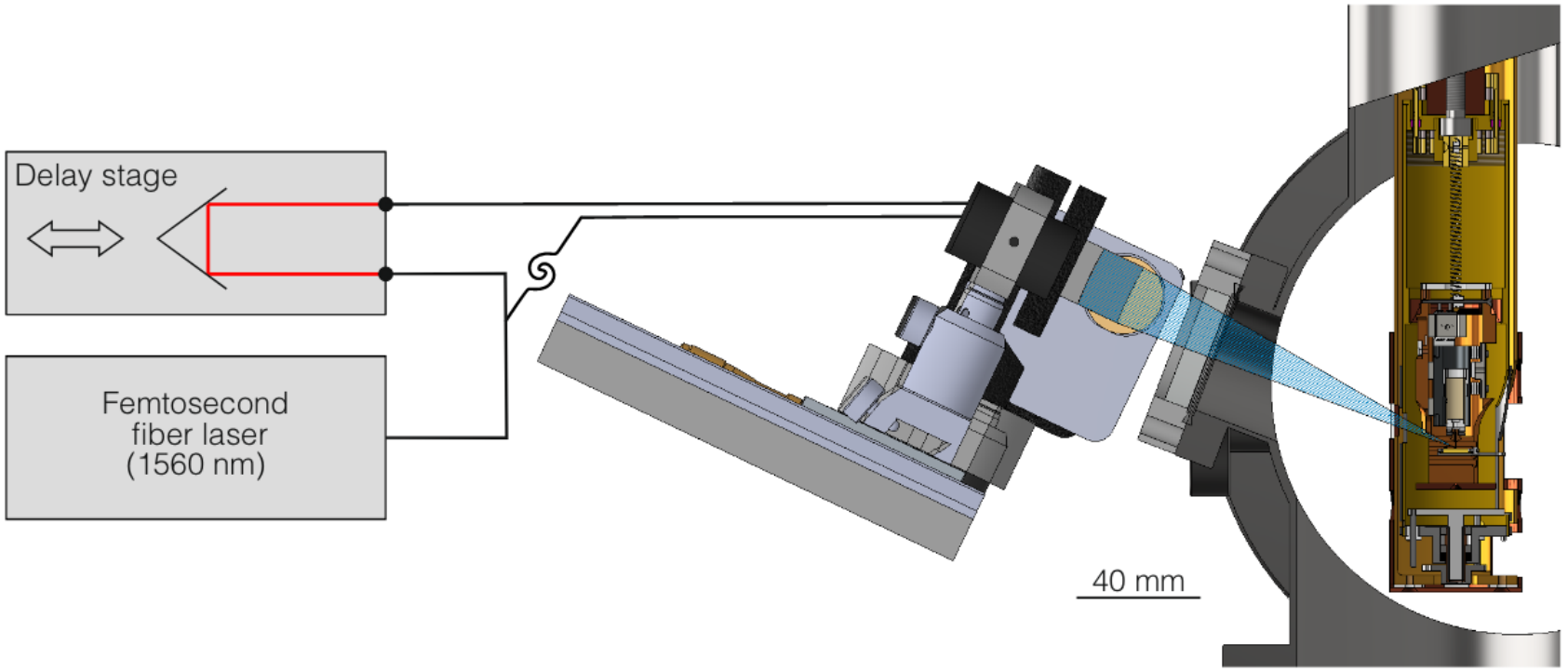
Portative plate for THz generation & focusing

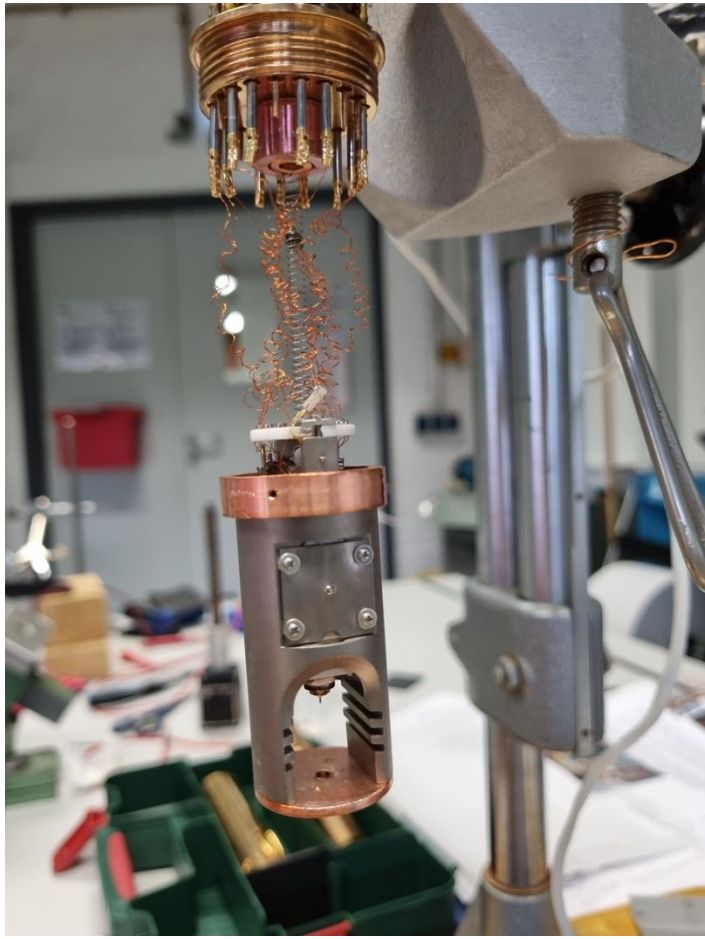




- THz free-space path is short (not much water absorption)
- Plate mounted on the UHV system of the STM

THz plate mounted on home-made variable-T STM



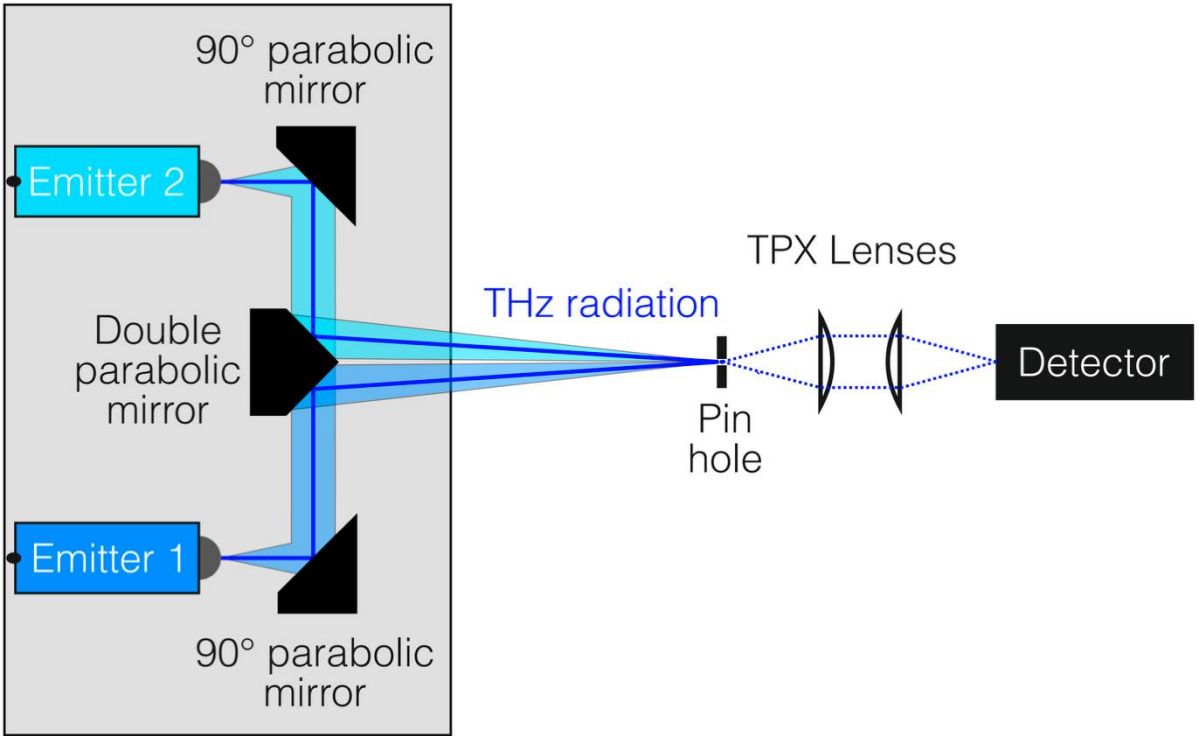


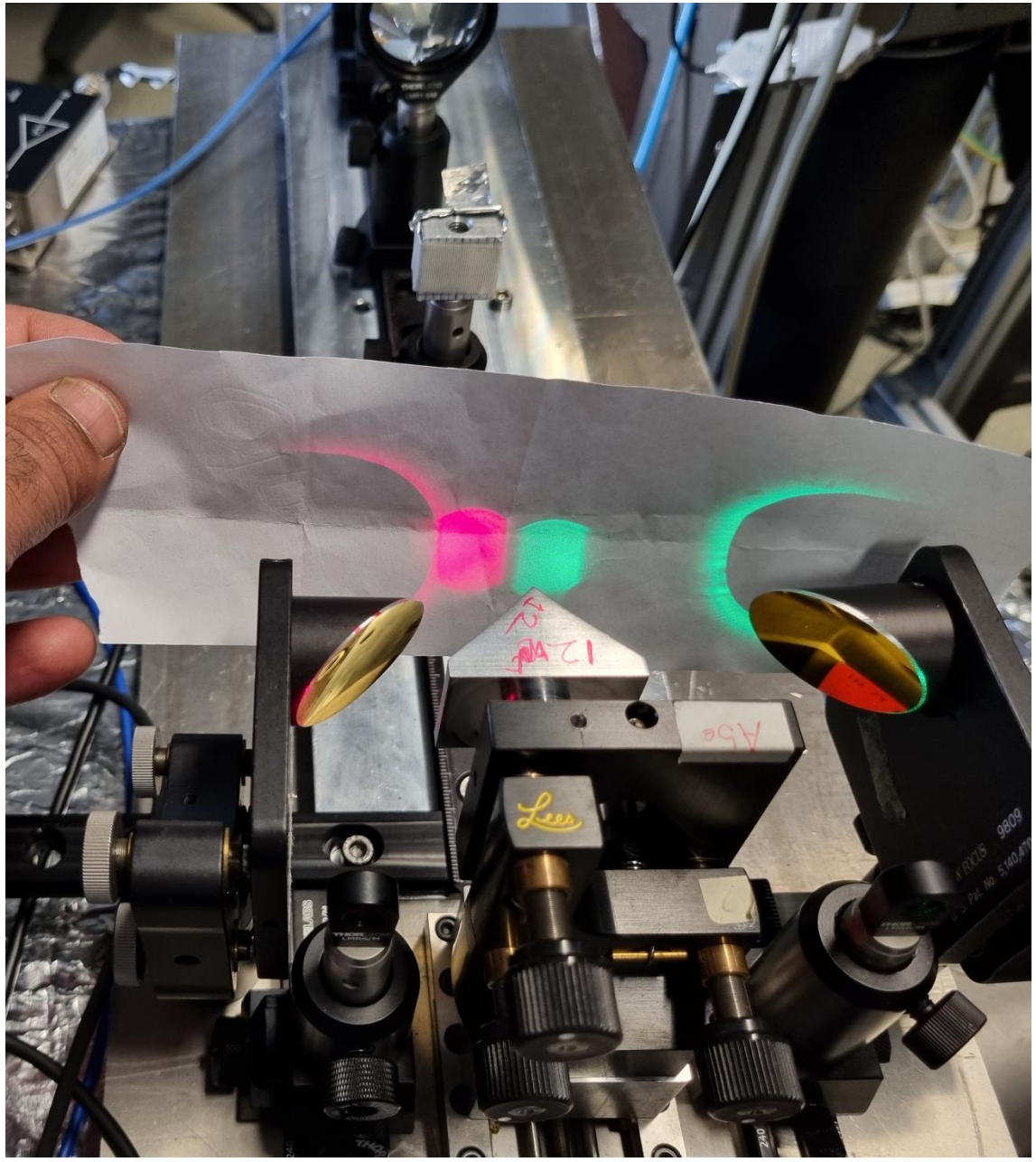
- Focusing realized “outside” (no mirror in UHV)
- Require high-quality quartz z-cut windows for large transmission

Azazoglu, …, MG, Rev. Sci. Instrum. **95**, 023703 (2024)

Alignment of THz beams

- Optical elements on plate are aligned on an optical table
- Pre-alignment with lasers





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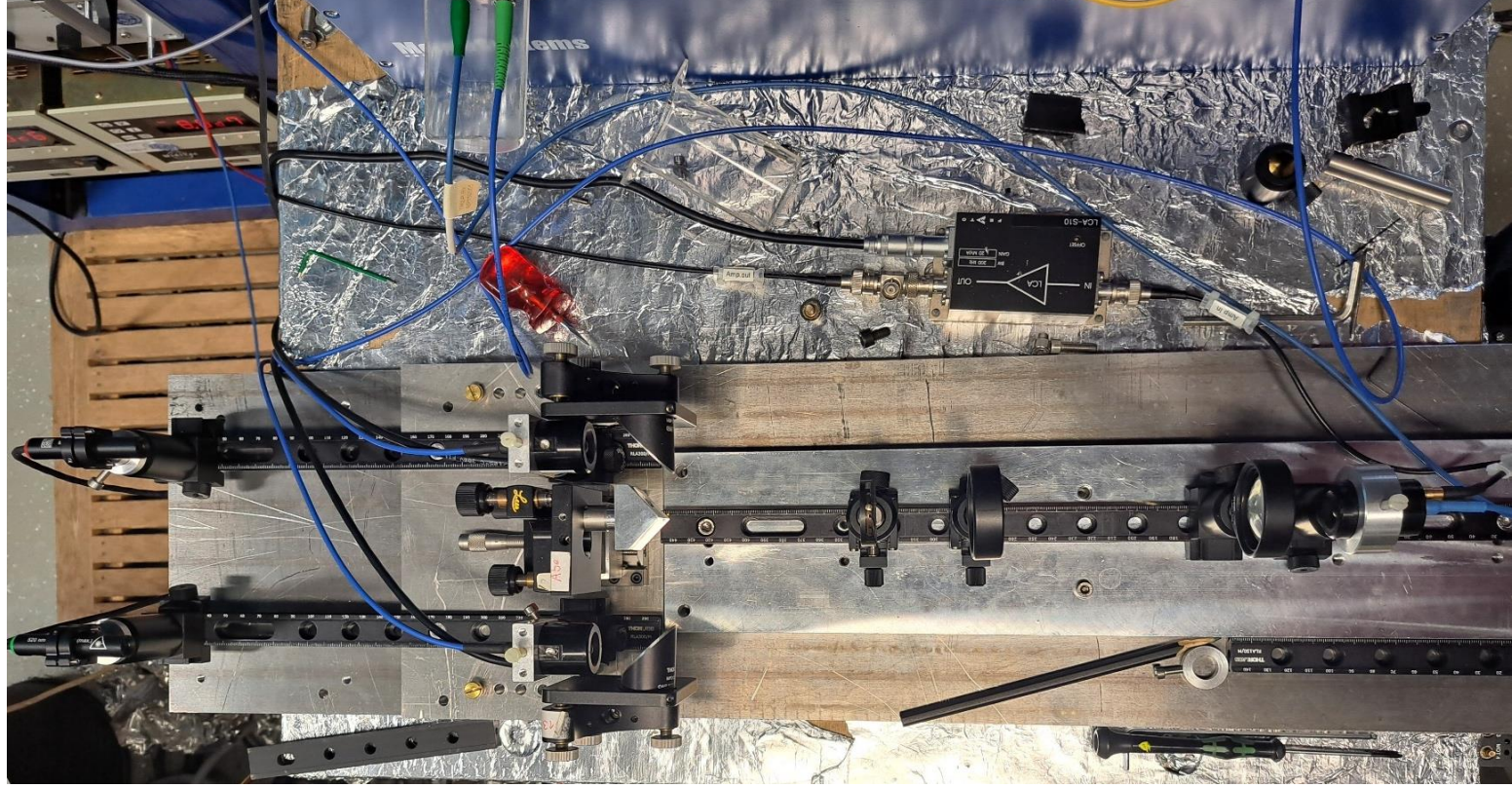
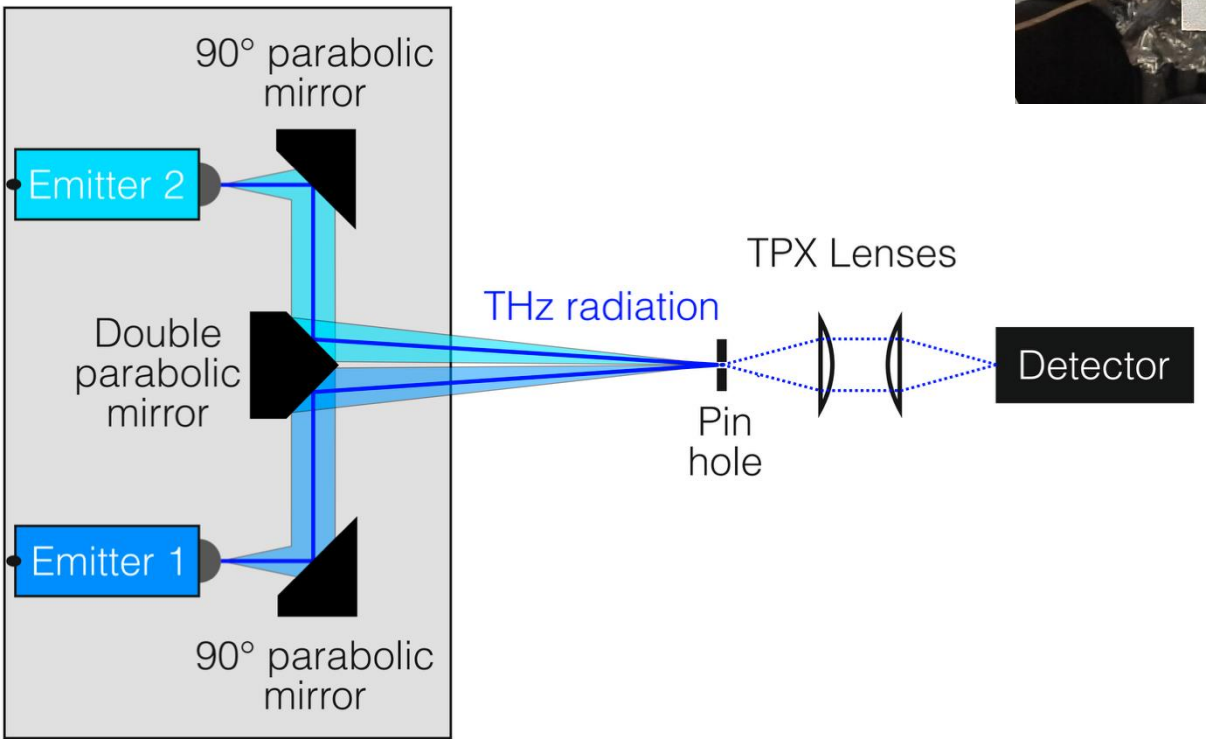
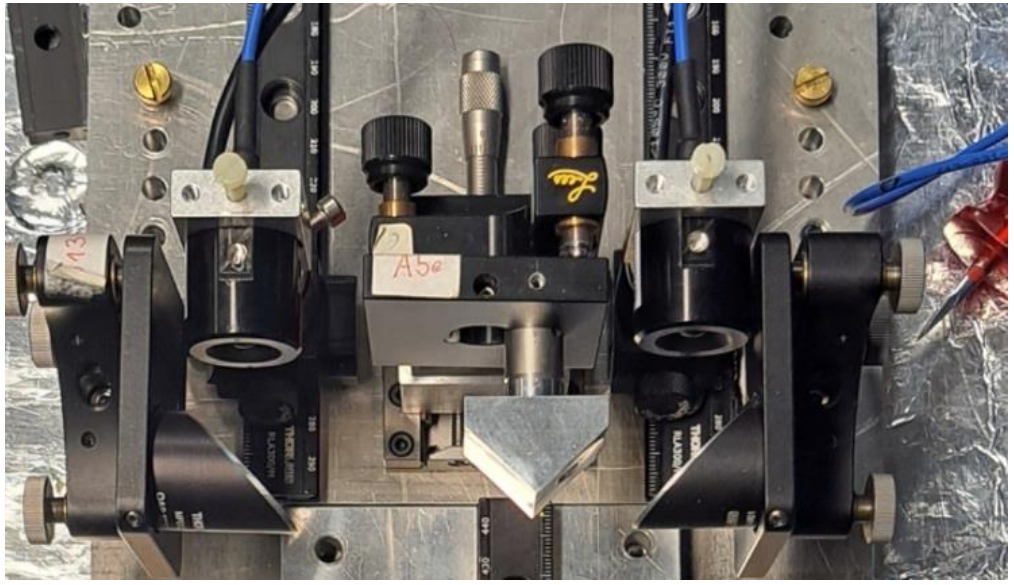
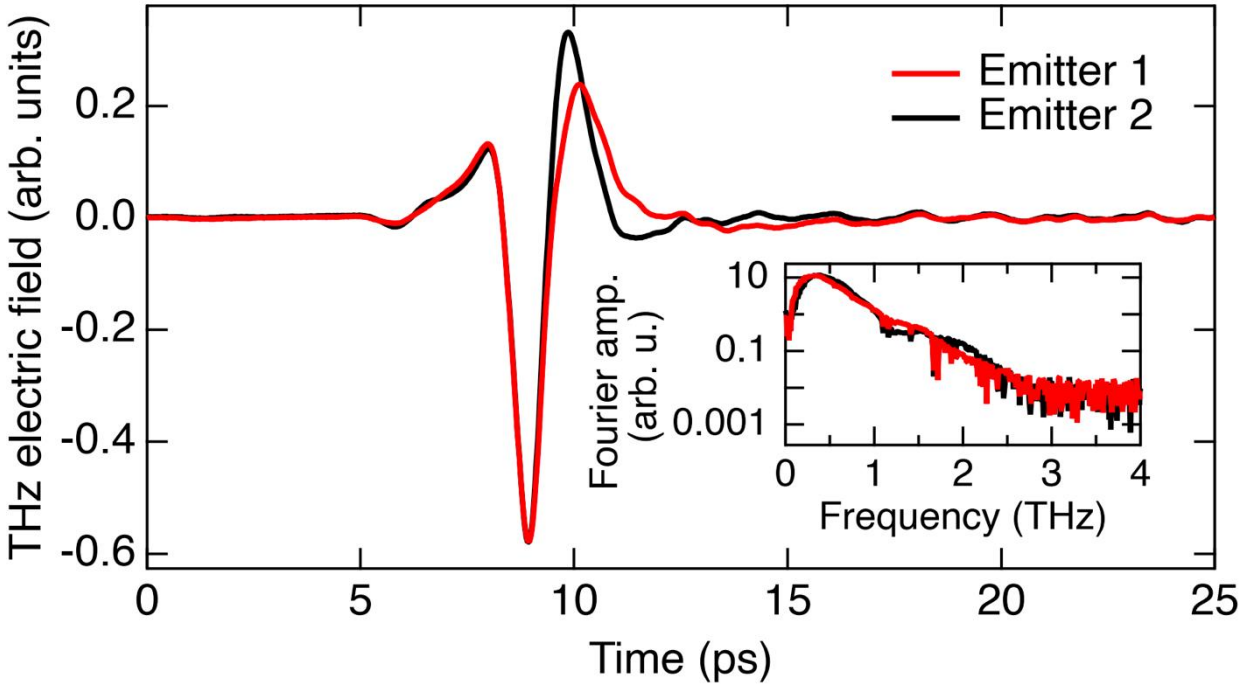
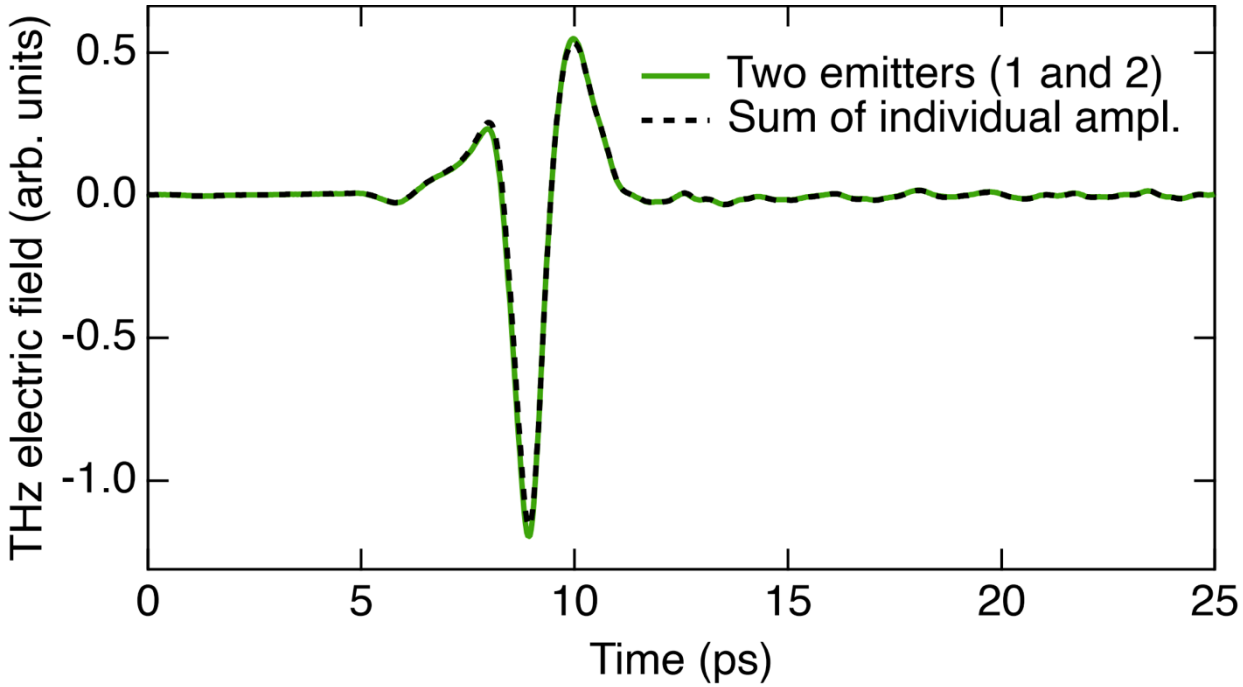






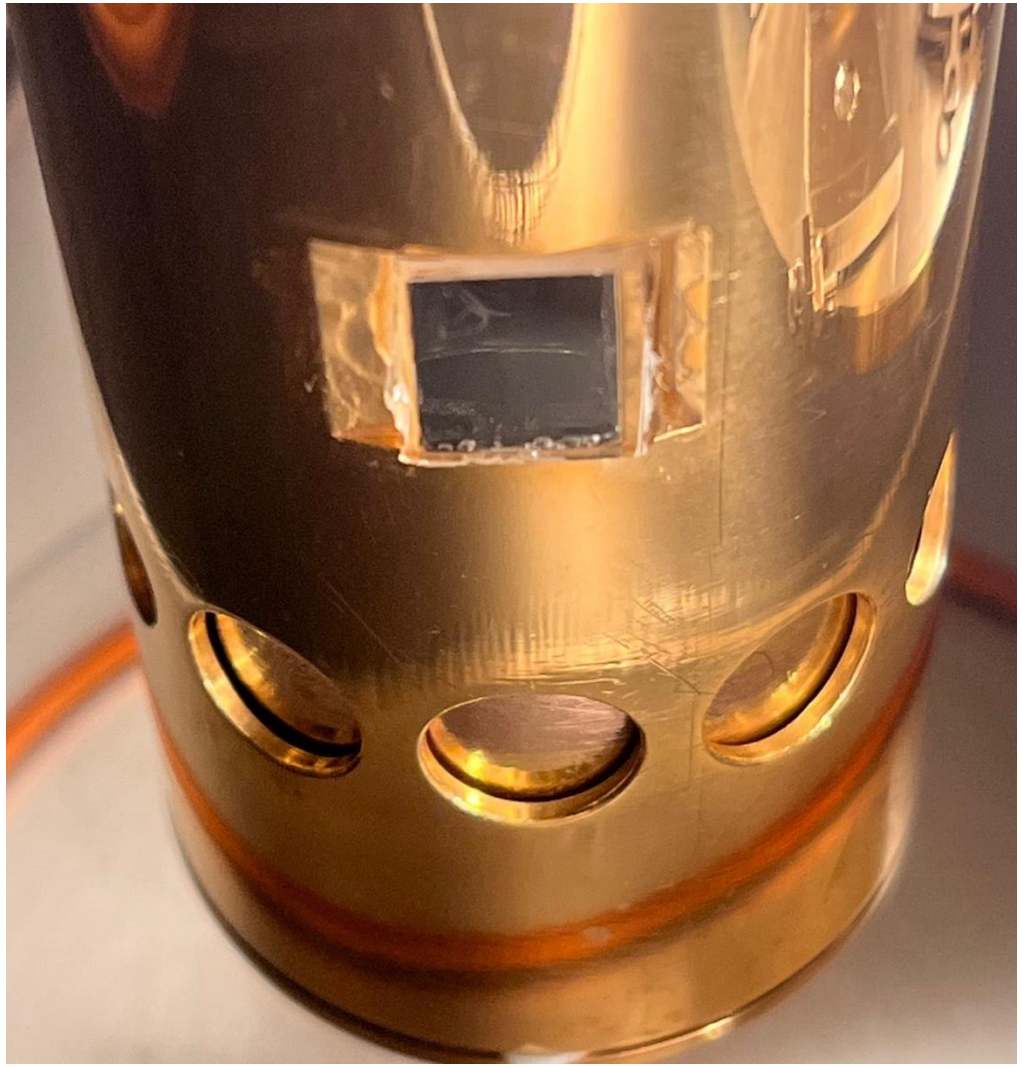
Verification of the alignment & waveform

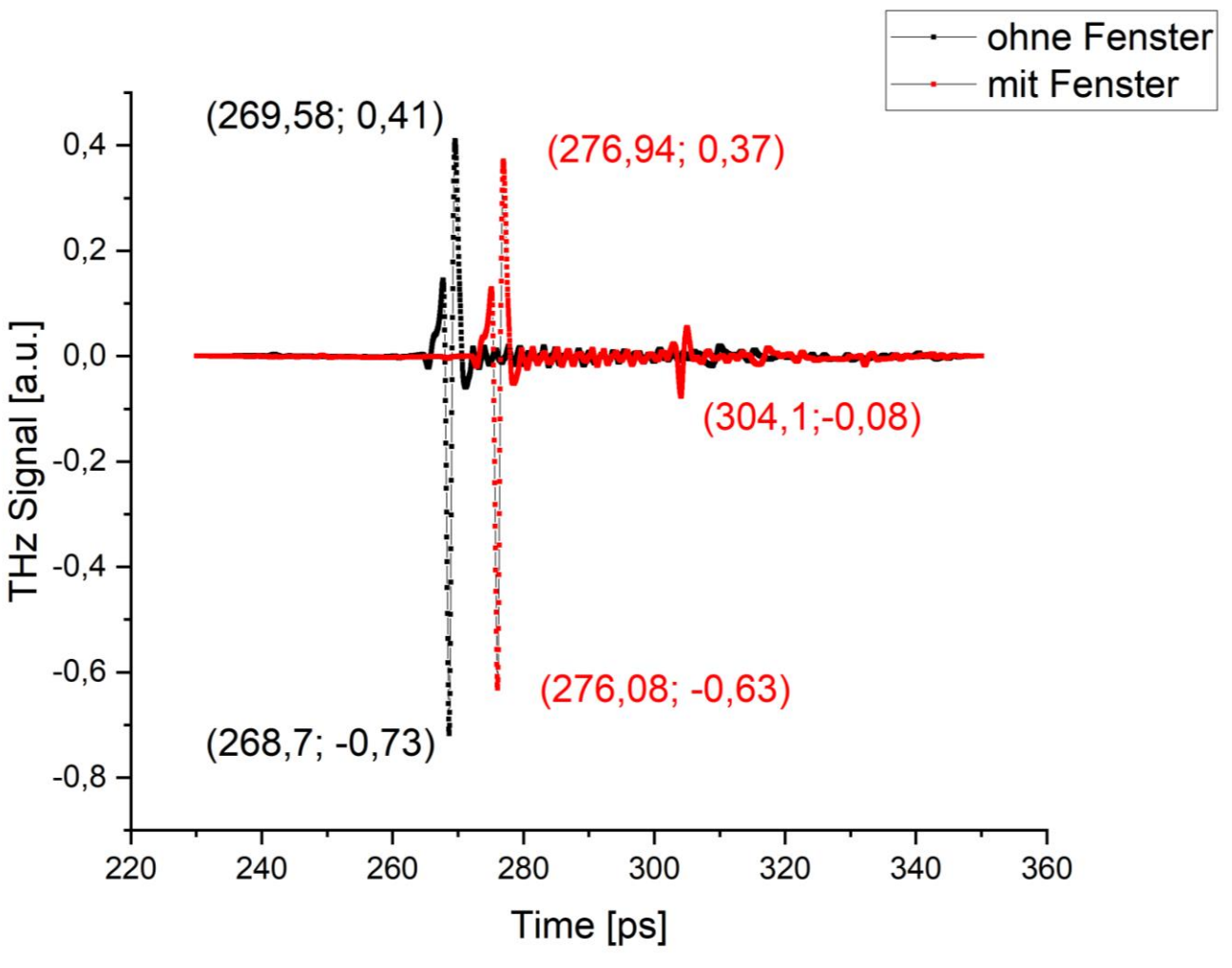




- Two emitters have similar waveforms
- Frequencies up to 3 THz
- Note: waveform is expected to change upon coupling to junction

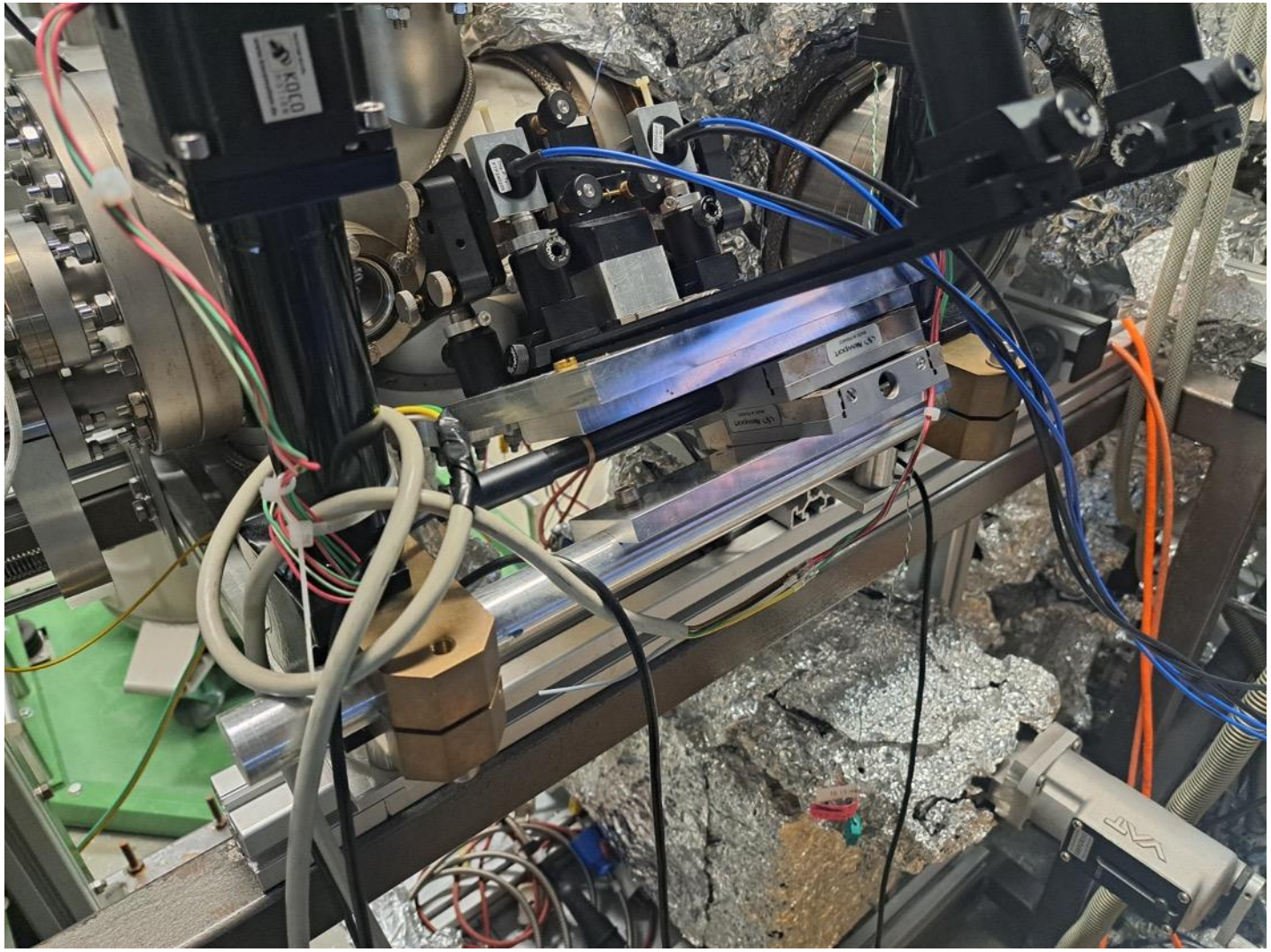
Characterization of the windows

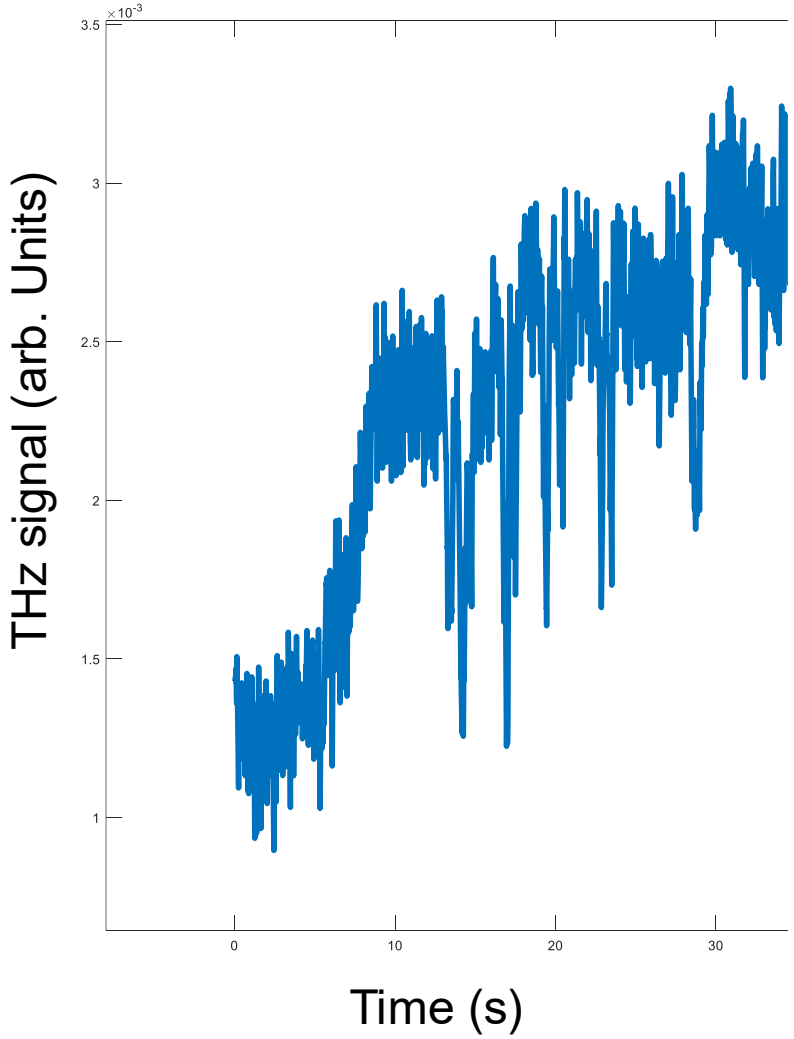




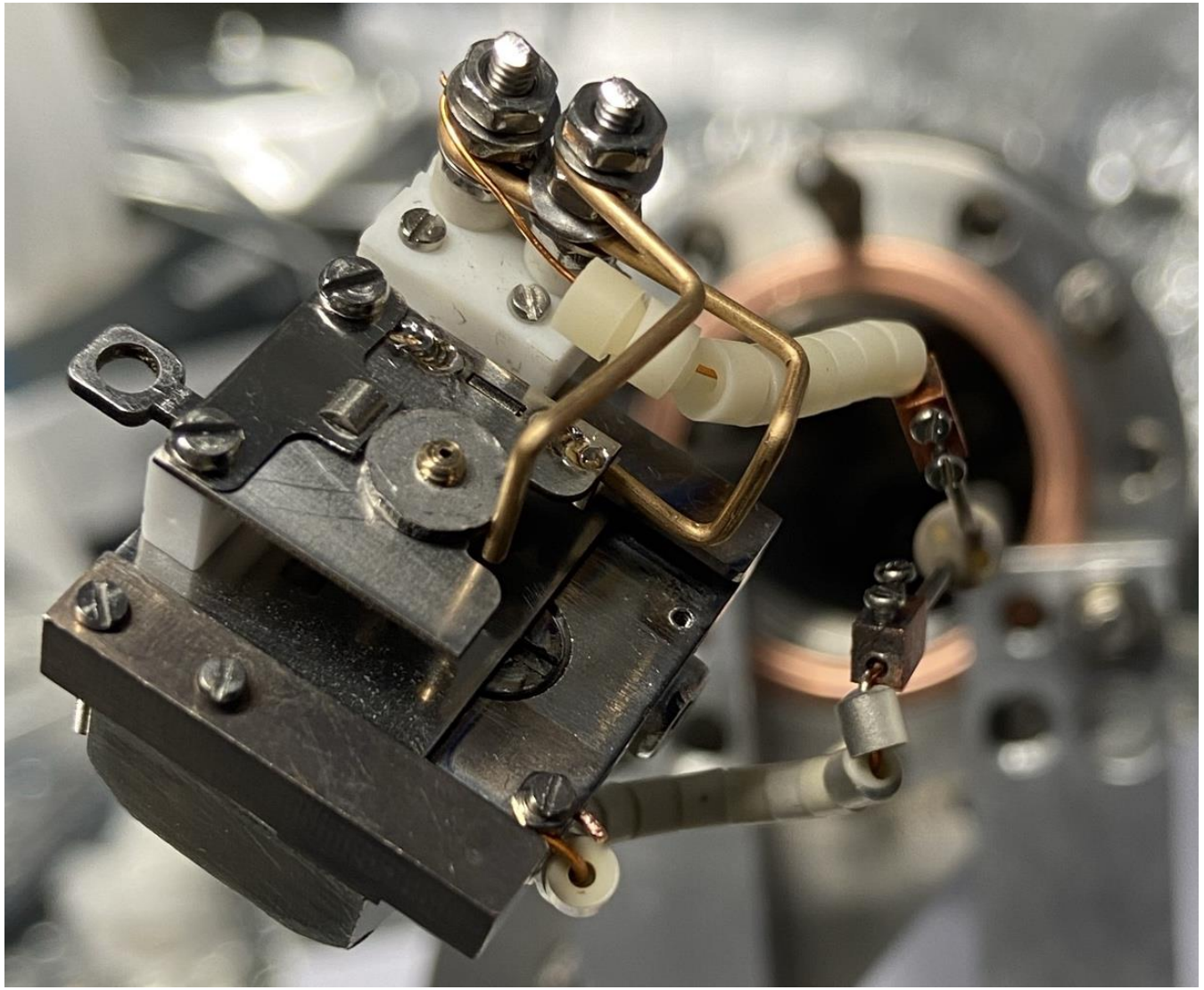
- 2 mm thick quartz *z*-cut (sold in wafer)
- Transmission of 85 % (with 7 ps delay)
- Echo after 28 ps due to internal reflections

THz-plate mounted on the STM





THz-induced current strongly depends on tip



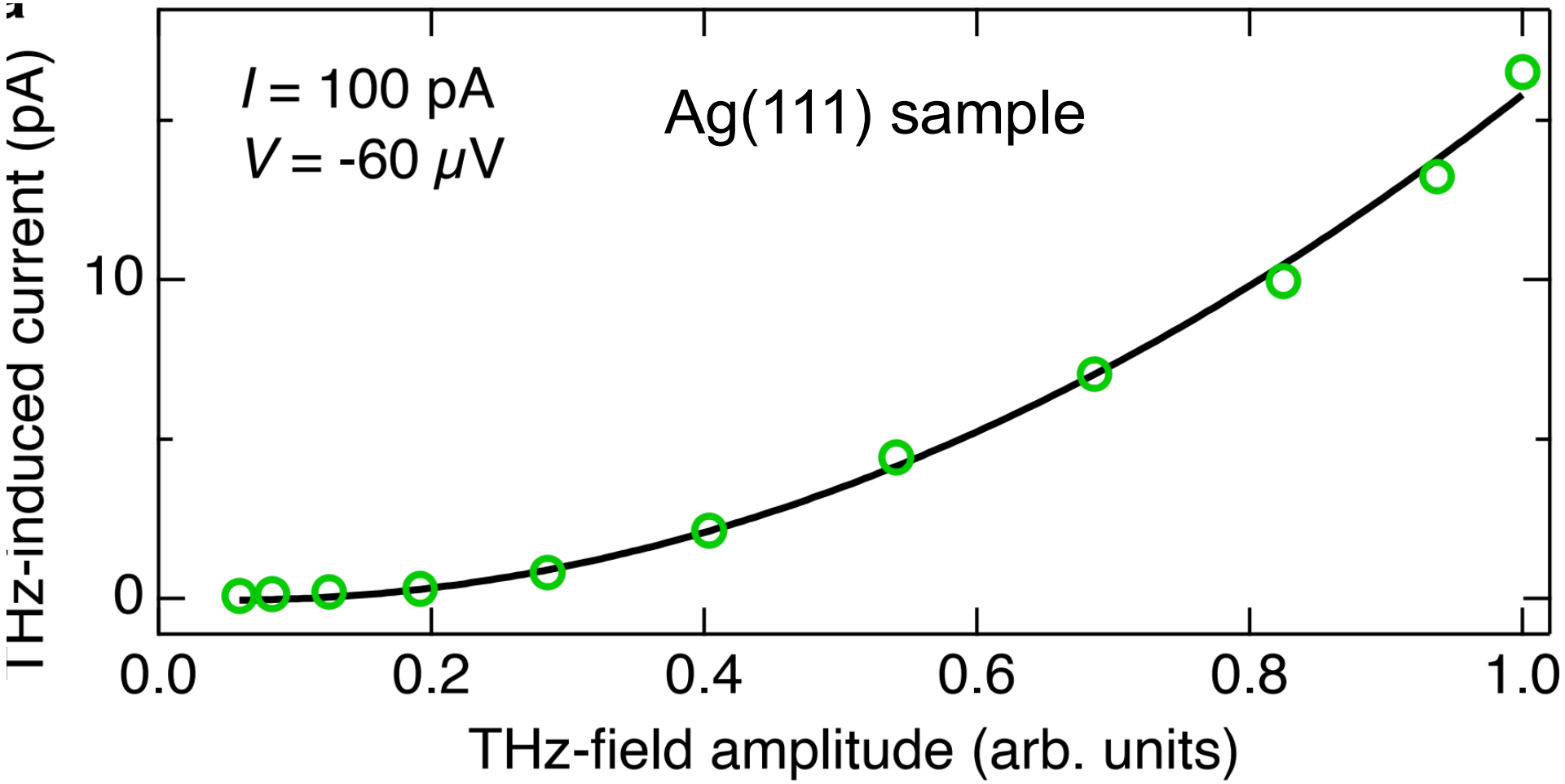
- Electrochemical etching of W tip
- Self-sputtering in Ne atmosphere (10^{-5} mbar)

Ernst, PhD thesis (2006)

Lightwave-driven scanning tunnelling microscopy

How to be sure it is THz-induced current?

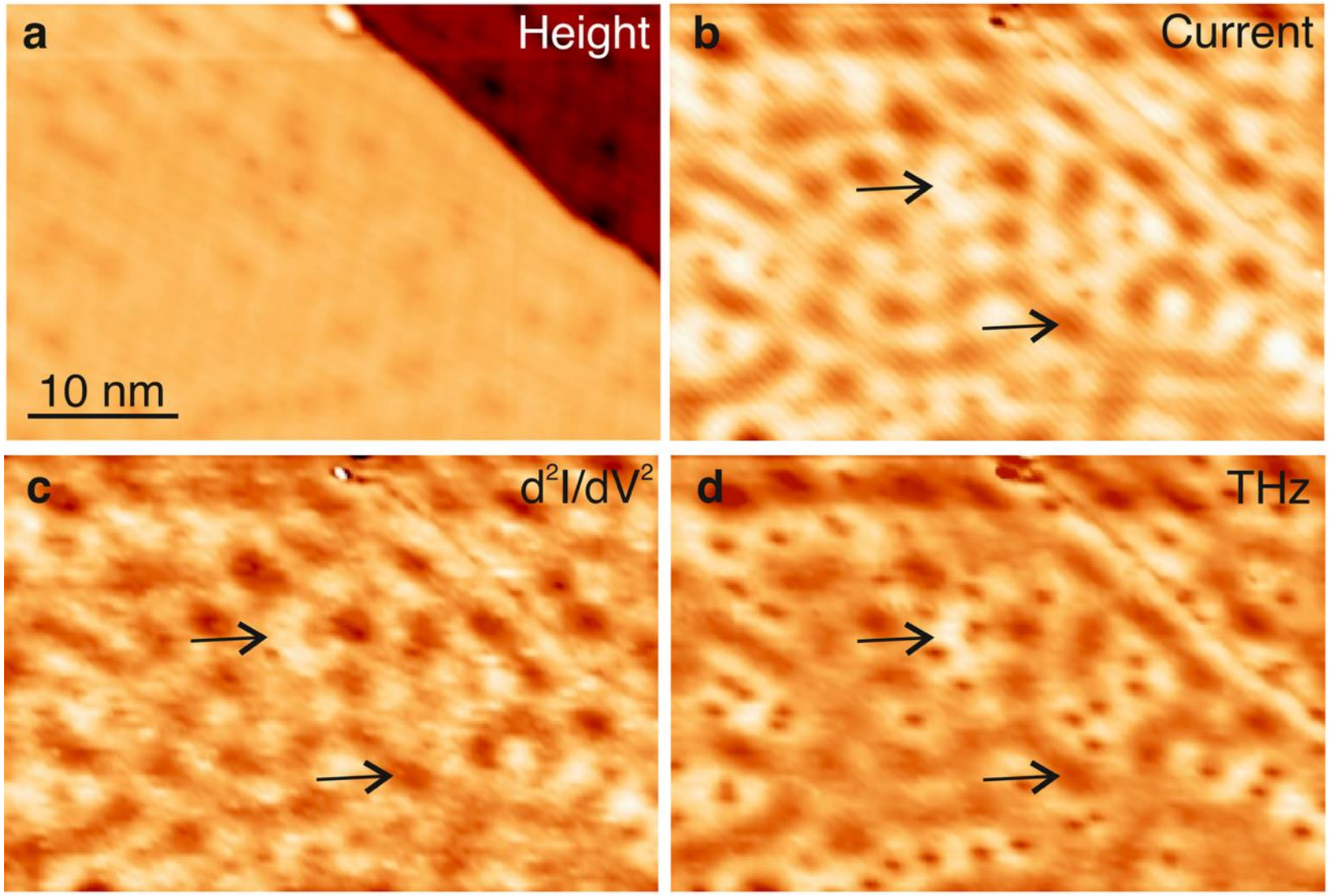
THz current versus far-field amplitude



- Non-linear evolution
- Complex: depends on the details of the *I*-*V* characteristic
- Mind the voltage (μV) because of metallic substrate

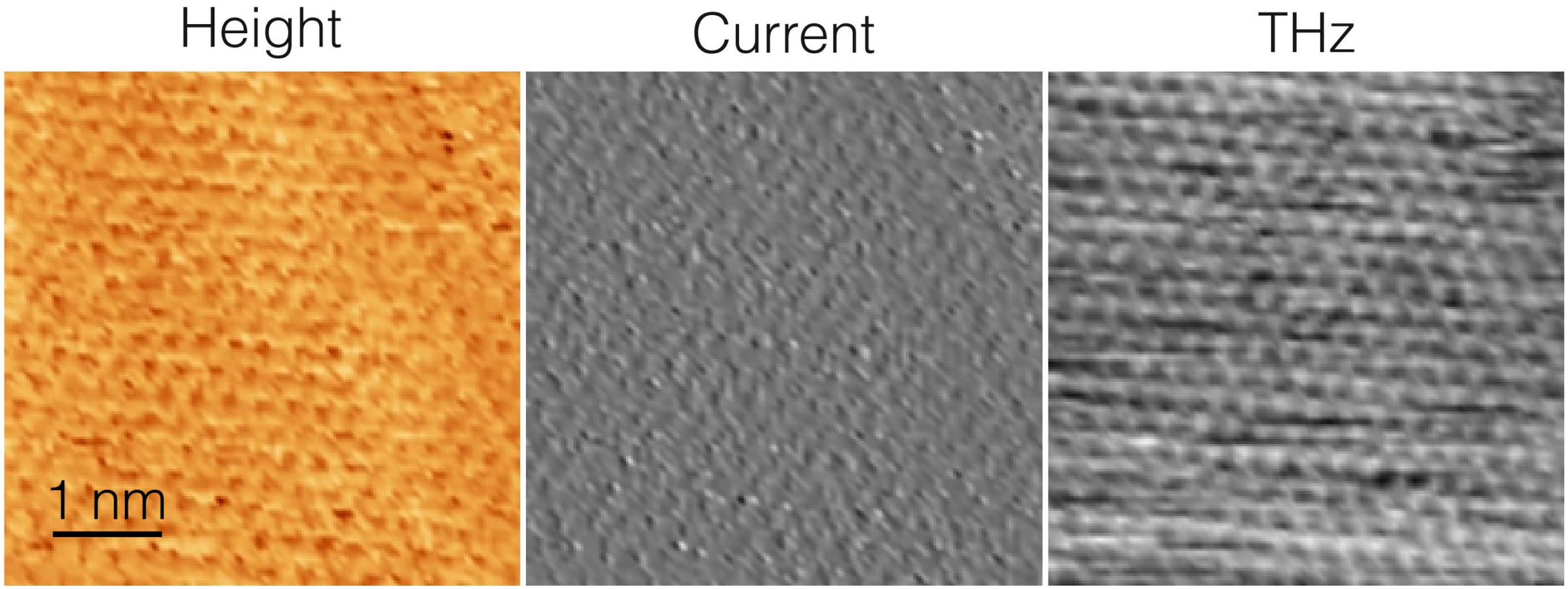
Azazoglu, …, MG, Rev. Sci. Instrum. **95**, 023703 (2024)

Sensitivity to non-linearity



- Measurement at $V_{DC} = 0$
- Modulation $V_{AC} = 280$ µV @ 977 Hz
- Rectification provides DC current used for STM operation
- In these conditions THz image similar to current image

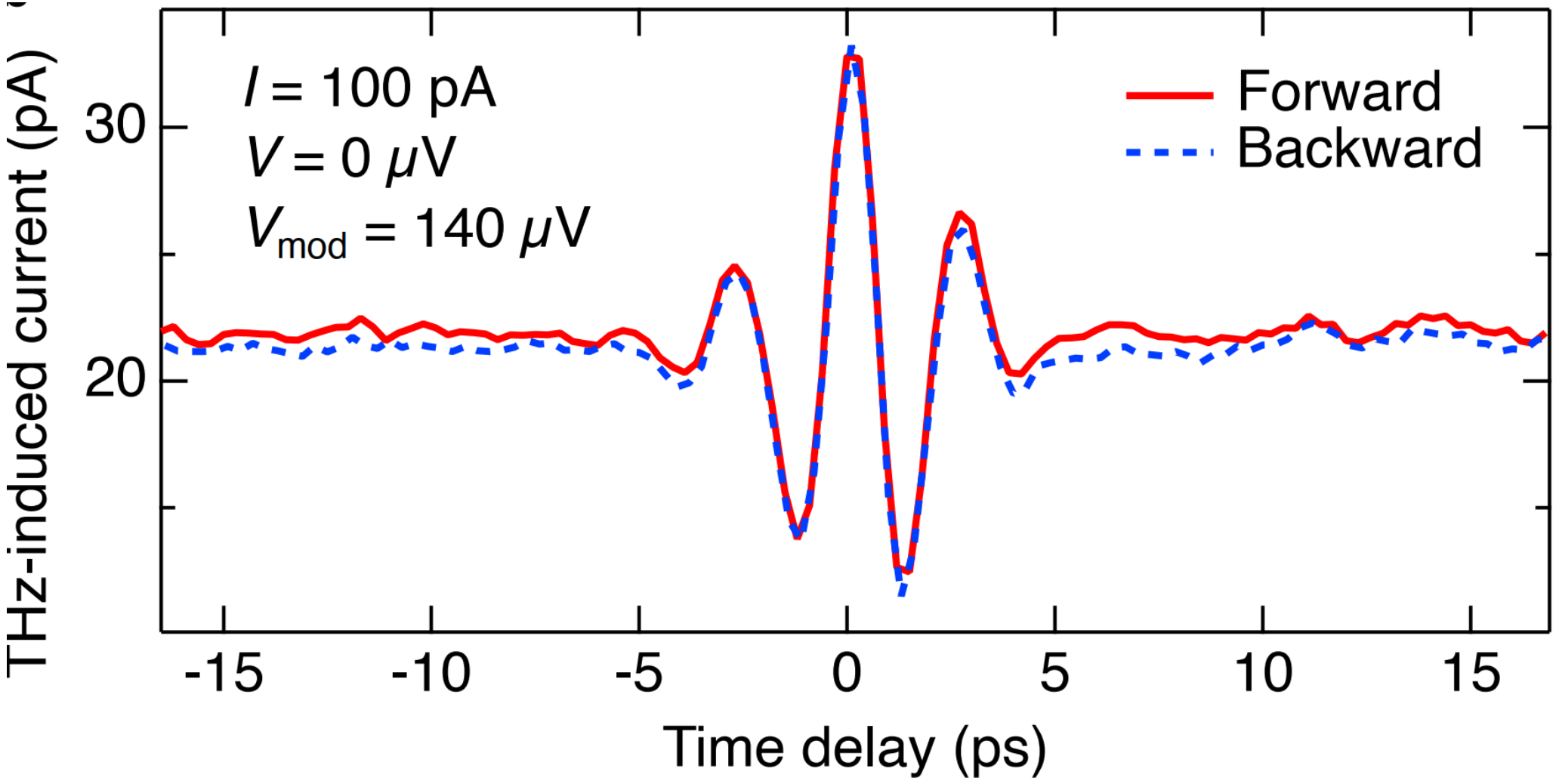
Atomic resolution



- Contrast sometimes better in THz channel

Azazoglu, …, MG, Rev. Sci. Instrum. **95**, 023703 (2024)

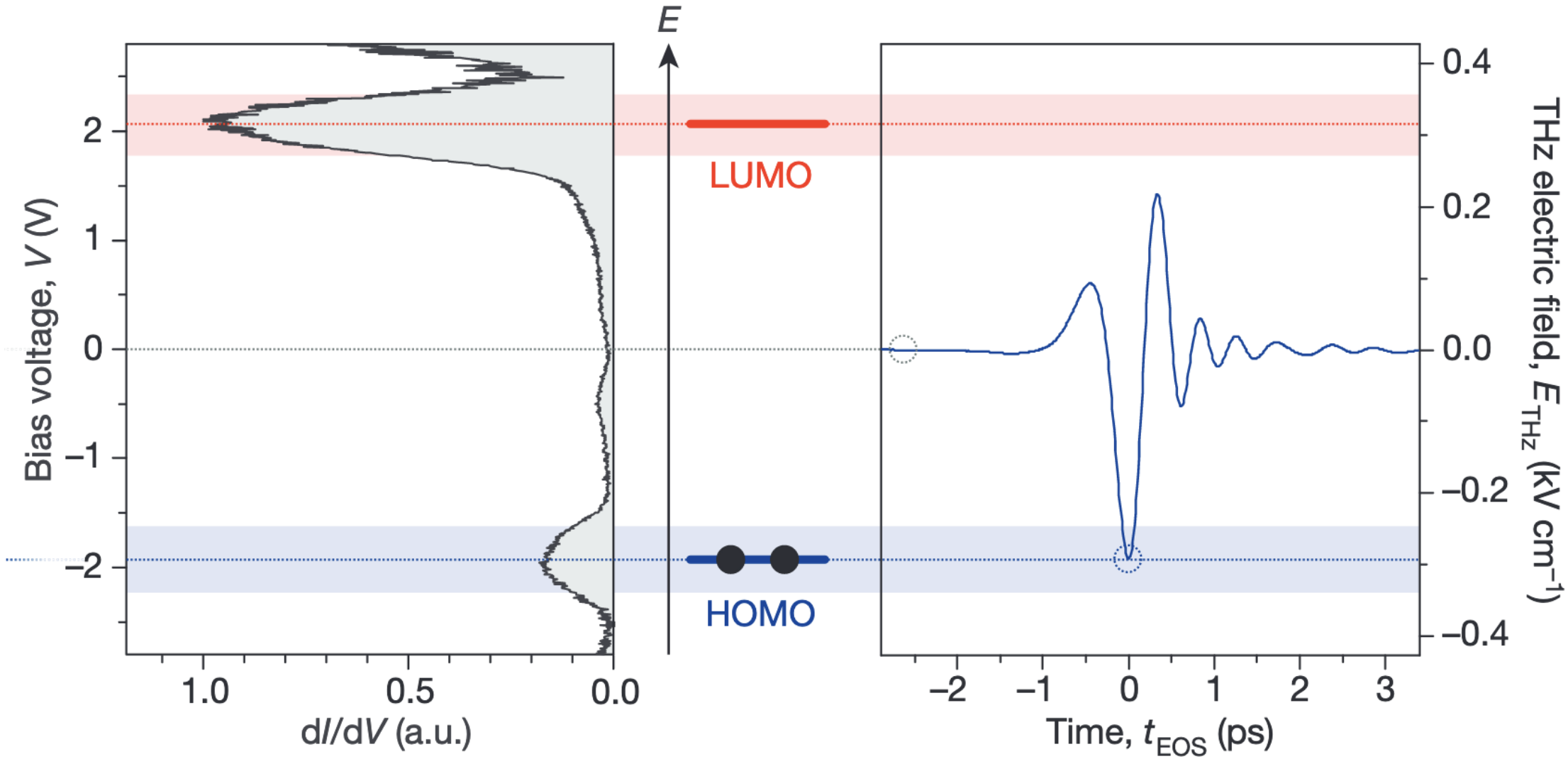
Pump and probe pulse – Cross-correlation measurement



- Time resolution is on the order of 1 ps

Azazoglu, …, MG, Rev. Sci. Instrum. **95**, 023703 (2024)

But what's the time resolution?

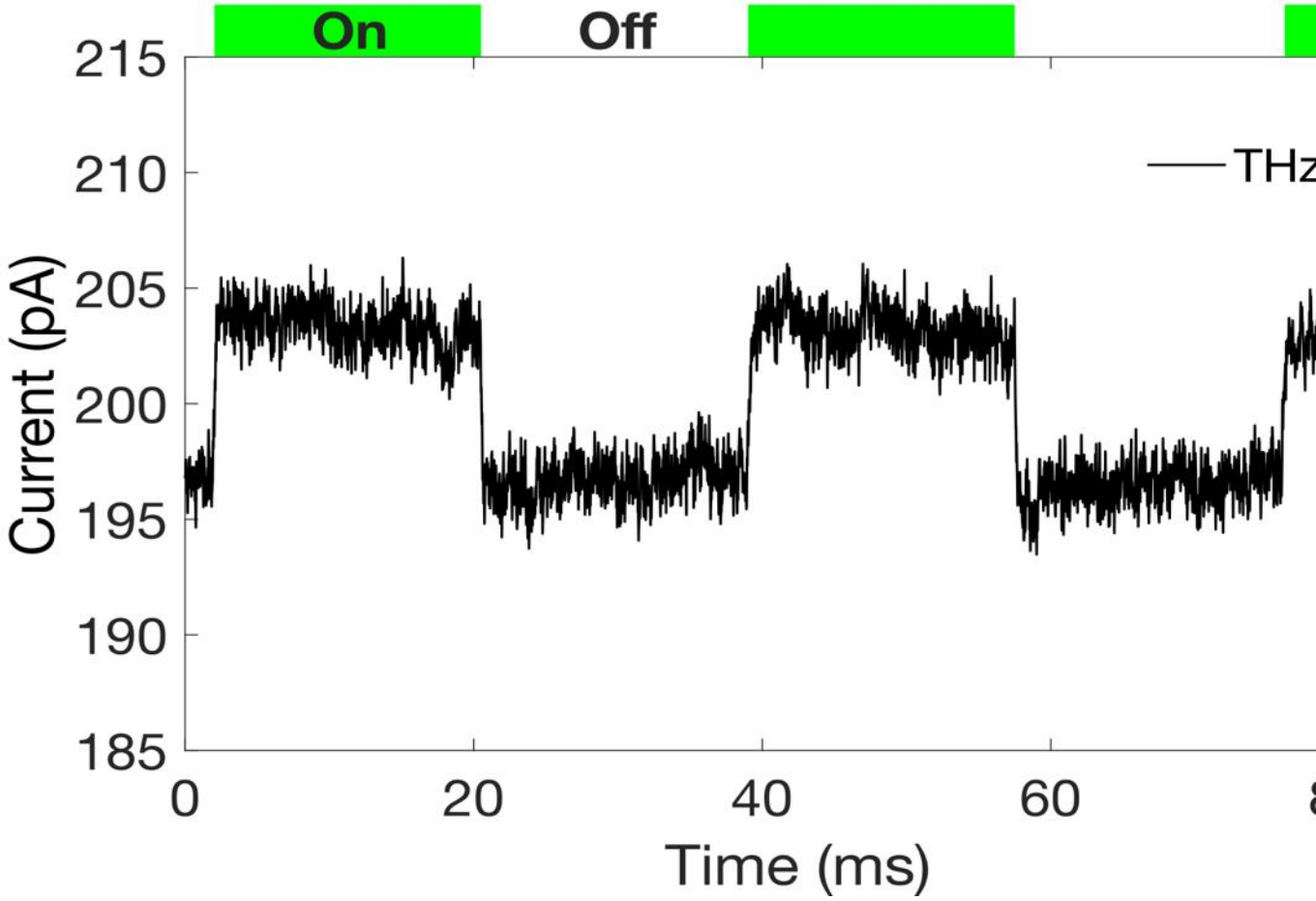


- Only the time during which the pulse can extract an e– from the HOMO is of interest
- Sub-picosecond time resolution in general
- The true time resolution depends on the sharpness of the spectroscopic features

Cocker et al., Nature 539, 263 (2016)

Importance of the repetition rate

$$I_{\mathrm{THz}} = f_{\mathrm{Rep}} \int_0^{1/f_{\mathrm{Rep}}} i_{\mathrm{THz}}(t) \cdot dt$$



- The amplitude of the THz field is important to select the excitation process, but not for the signal intensity
- I_{THz} scales with (i) the rectification (waveform) and (ii) repetition rate

Acknowledgments

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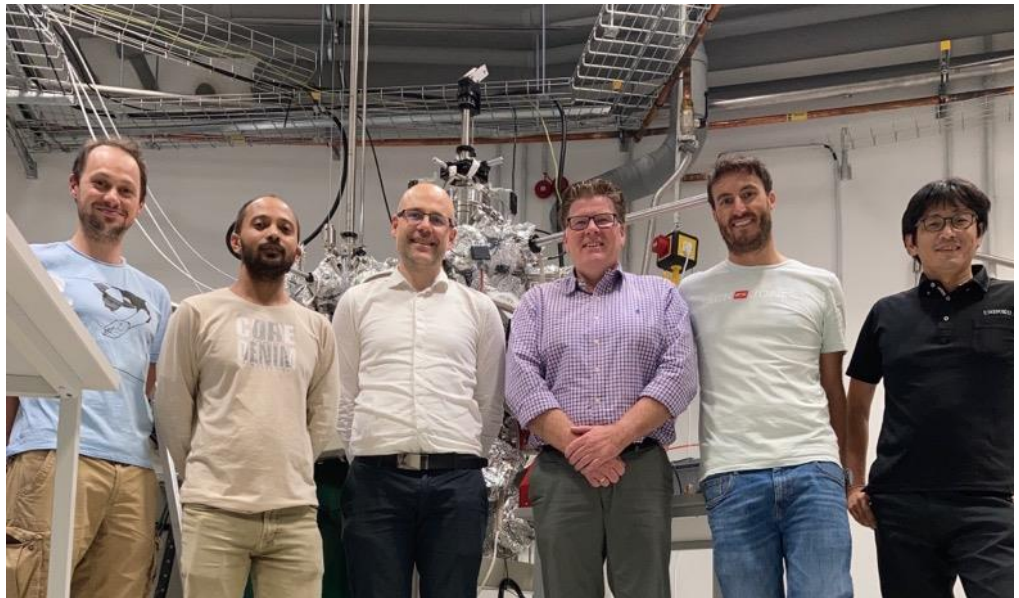
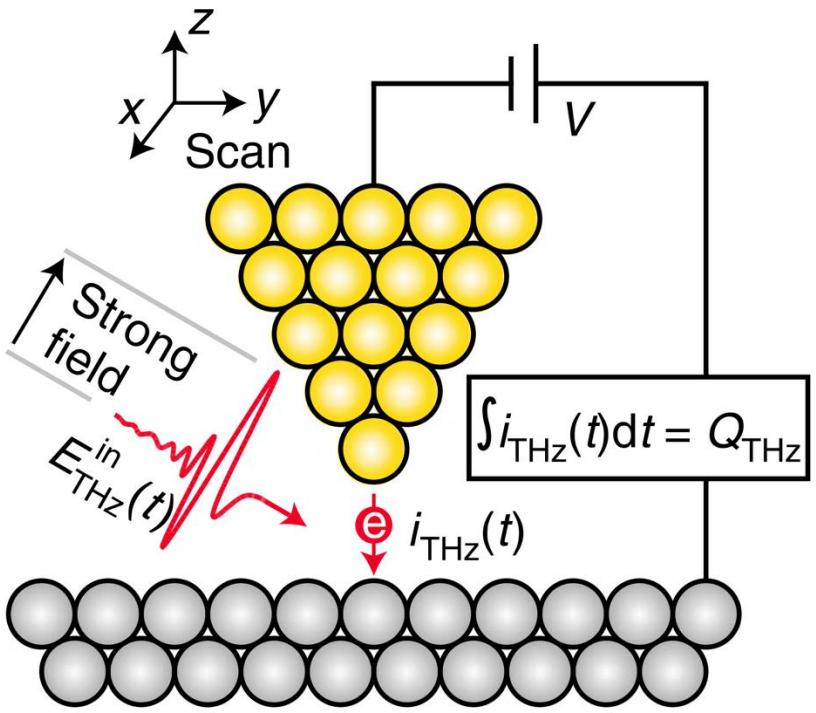
CRC 1242

The group (AG Gruber)



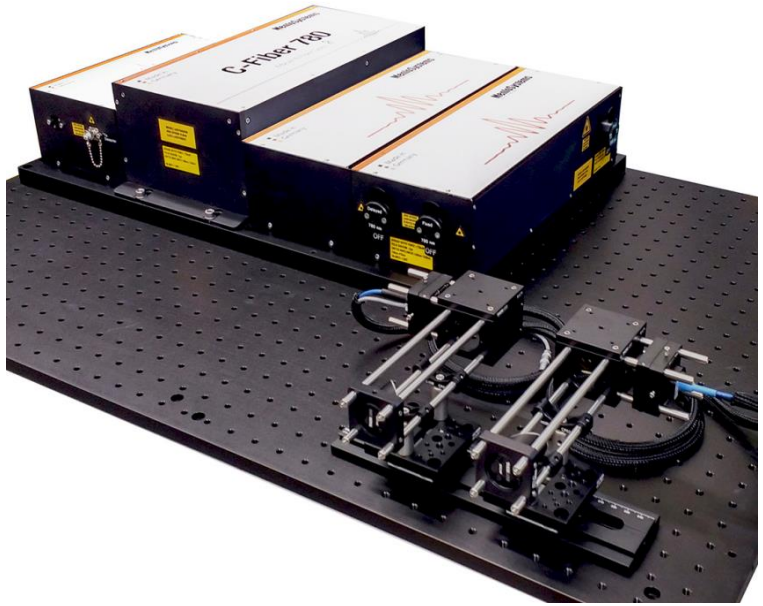
Conclusion

Lightwave-driven STM to combine spatial & time resolutions

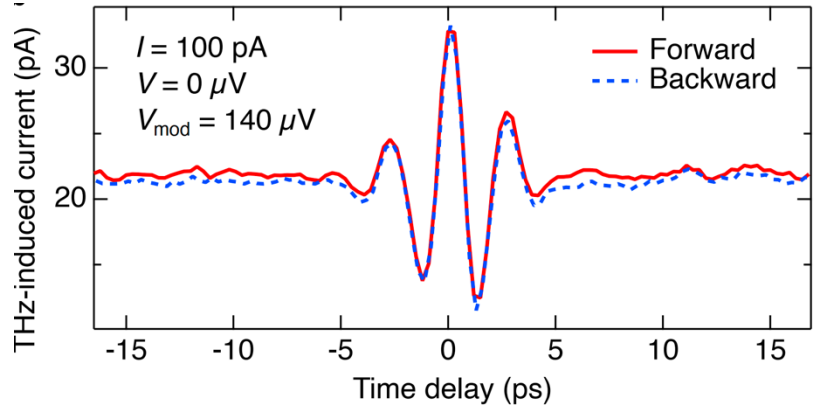


Cocker *et al.*, Nat. Photonics **15**, 558 (2021)

Implementation based on commercial THz-generation system



Evidence for sub-nm and sub-ps resolutions



Azazoglu, …, MG, Rev. Sci. Instrum. **95**, 023703 (2024)