Compact terahertz time-domain spectroscopy systems enabled by mode-locked laser diodes

Jan C. Balzer

University of Duisburg-Essen, Germany

Academic career

FH Dortmund
Dipl.-Ing. (FH) 2008
(telecommunication engineering)

University of Duisburg-Essen
W1 Professor “THz Systems”
Since Nov. 2017

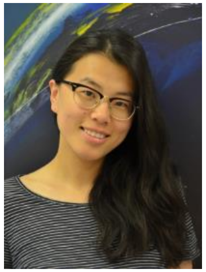
Ruhr-University Bochum
M.Sc. 2010 (nanotechnology)
Dr.-Ing. 2014 (Laser diodes)

Philipps University Marburg
Group leader from 2015-2017
(Terahertz technology)

Chair for communication systems


Prof. Czylwik


Prof. Balzer


Xuan Liu


Vladyslav Cherniak


Tobias Kubiczek


Dr. Schultze


Kevin Kolpatzeck


Riu Liu

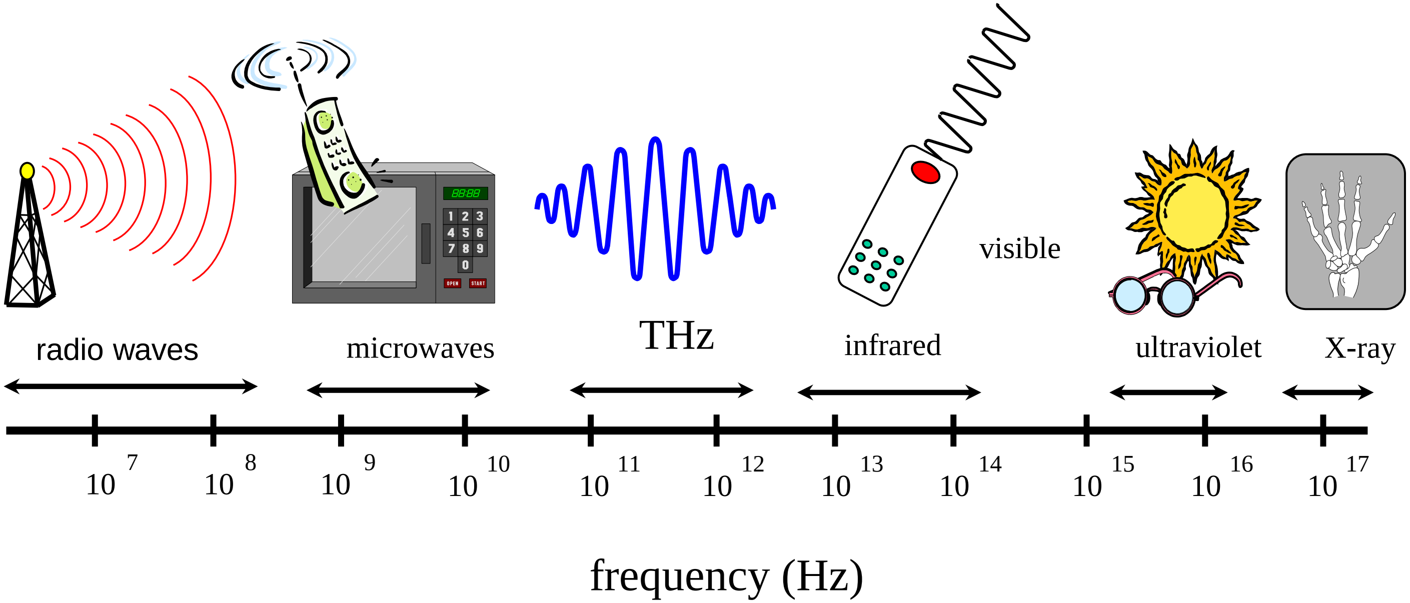

Kai Tybussek


Dilyan Damyanov

Outline

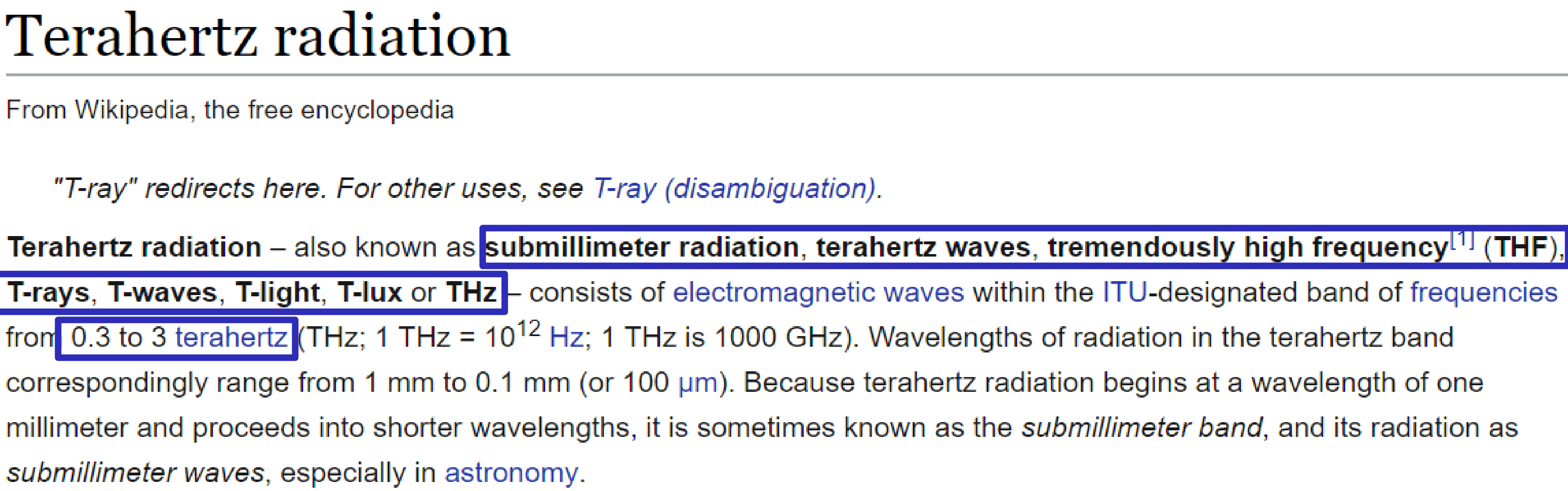
- Brief introduction to terahertz technology
- Enhancing the performance of THz CCS/QTDS
- Theoretical model for MLLD-driven THz-TDS
- Model-driven optimization of THz-TDS
- Conclusion and outlook

What is Terahertz radiation?



- THz radiation is located between microwaves and infrared
- It took some time to address this spectral range

What is Terahertz radiation?



Terahertz radiation

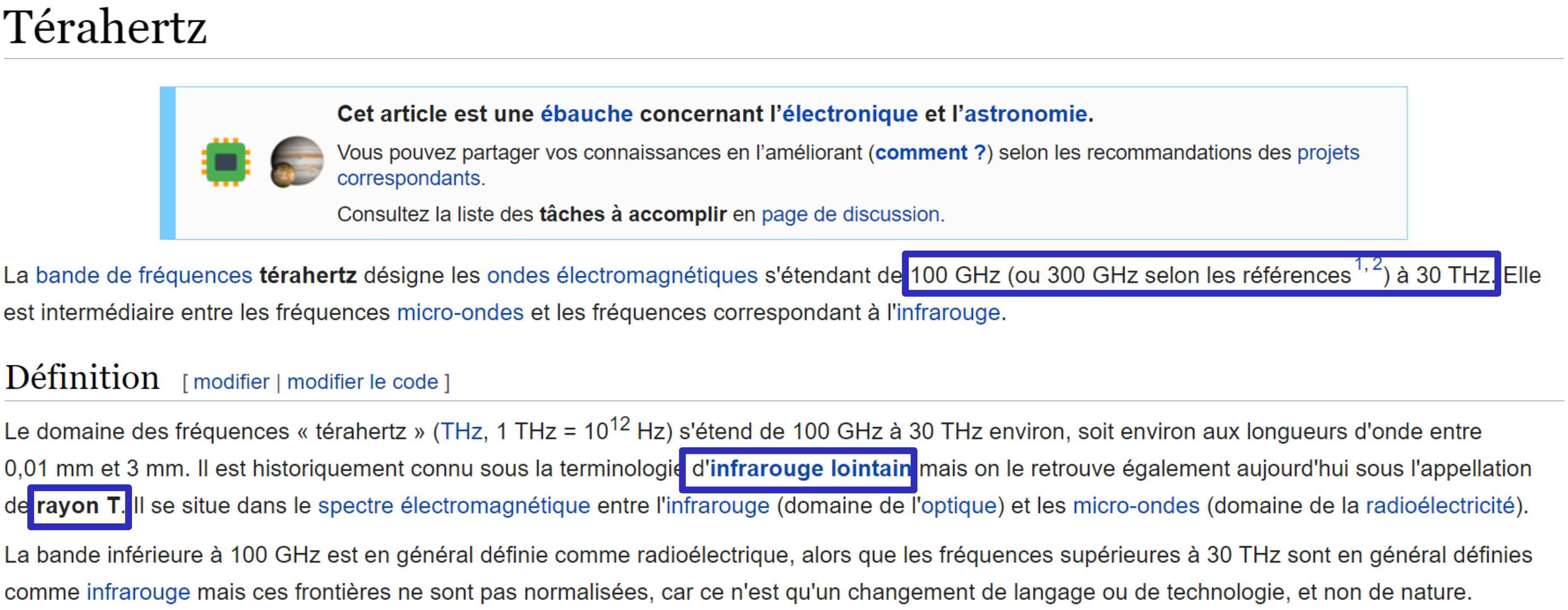
From Wikipedia, the free encyclopedia

"T-ray" redirects here. For other uses, see T-ray (disambiguation).

Terahertz radiation – also known as **submillimeter radiation**, **terahertz waves**, **tremendously high frequency**[1] (**THF**), **T-rays**, **T-waves**, **T-light**, **T-lux** or **THz** – consists of electromagnetic waves within the ITU-designated band of frequencies from 0.3 to 3 terahertz (THz; 1 THz = 10^{12} Hz; 1 THz is 1000 GHz). Wavelengths of radiation in the terahertz band correspondingly range from 1 mm to 0.1 mm (or 100 µm). Because terahertz radiation begins at a wavelength of one millimeter and proceeds into shorter wavelengths, it is sometimes known as the *submillimeter band*, and its radiation as *submillimeter waves*, especially in astronomy.

- Definition on English Wikipedia: 300 GHz to 3 THz
- Further: many different names like submillimeter radiation, terahertz waves, tremendously high frequency (THF), T-rays, T-waves, T-light, T-lux or THz

What is Terahertz radiation?



Térahertz

Cet article est une ébauche concernant l'électronique et l'astronomie.

Vous pouvez partager vos connaissances en l'améliorant (**comment ?**) selon les recommandations des projets correspondants.

Consultez la liste des **tâches à accomplir** en page de discussion.

La bande de fréquences **térahertz** désigne les ondes électromagnétiques s'étendant de 100 GHz (ou 300 GHz selon les références [1, 2]) à 30 THz. Elle est intermédiaire entre les fréquences micro-ondes et les fréquences correspondant à l'infrarouge.

Définition [modifier | modifier le code]

Le domaine des fréquences « térahertz » (THz, 1 THz = 10^{12} Hz) s'étend de 100 GHz à 30 THz environ, soit environ aux longueurs d'onde entre 0,01 mm et 3 mm. Il est historiquement connu sous la terminologie d'**infrarouge lointain** mais on le retrouve également aujourd'hui sous l'appellation de **rayon T**. Il se situe dans le spectre électromagnétique entre l'infrarouge (domaine de l'optique) et les micro-ondes (domaine de la radioélectricité).

La bande inférieure à 100 GHz est en général définie comme radioélectrique, alors que les fréquences supérieures à 30 THz sont en général définies comme infrarouge mais ces frontières ne sont pas normalisées, car ce n'est qu'un changement de langage ou de technologie, et non de nature.

- Definition on French Wikipedia: 100 (300) GHz to 30 THz
- Further names: d'infrarogue lointain or rayon T

Why is THz radiation interesting?

First of all: The frequency range exists!

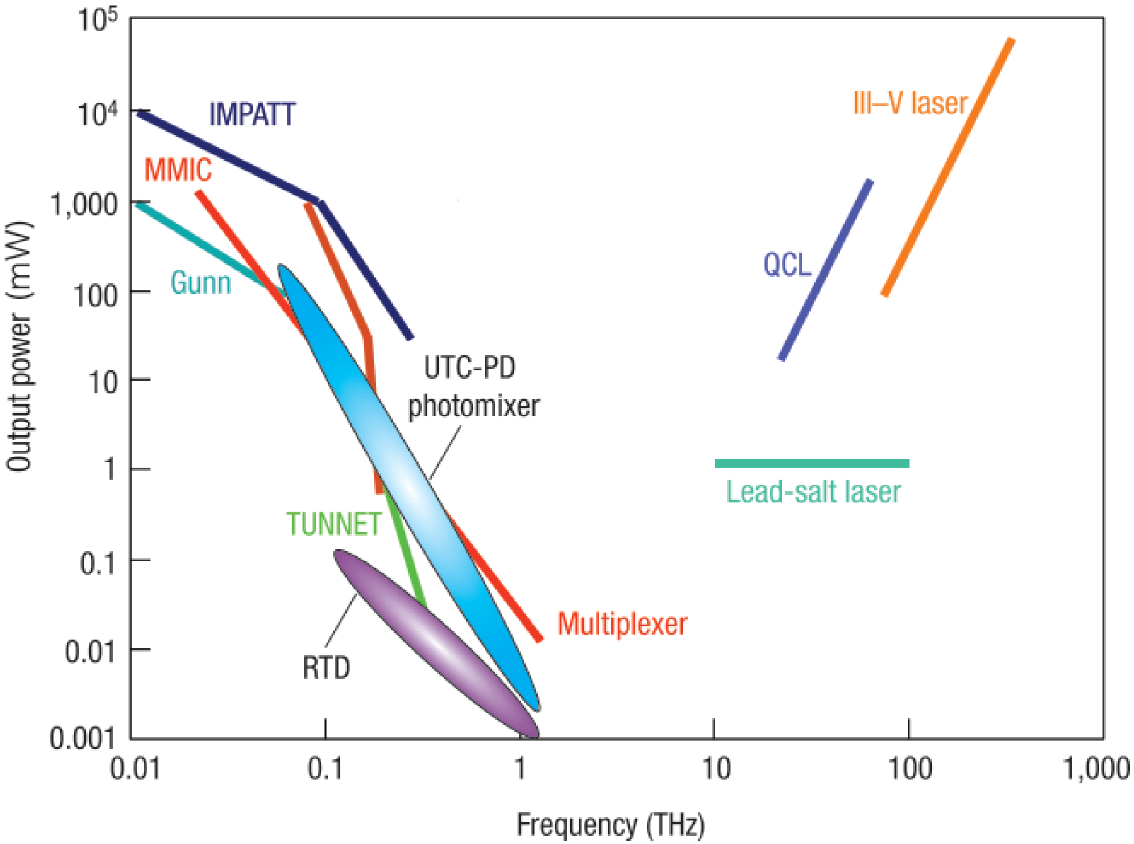
1. THz waves have low photon energy
    - No photoionization in biological tissues like X-rays
    - Safe for sample and operator
    - Even if not: Cannot penetrate human body like microwaves

2. THz waves have longer wavelength than VIS or IR
    - Less affected by Mie scattering
    - Dry dielectric materials like cloth, paper, wood and plastic are transparent
    - Higher spatial resolution than microwaves

3. Spectral fingerprint of materials
    - Many molecules exhibit strong absorption and dispersion
    - Allows to identify specific materials like explosives or drugs

4. Detection of amplitude and phase
    - Coherent techniques give access to absorption and dispersion spectroscopy
    - Complex refractive index (or permittivity) can be directly measured

The “THz gap”



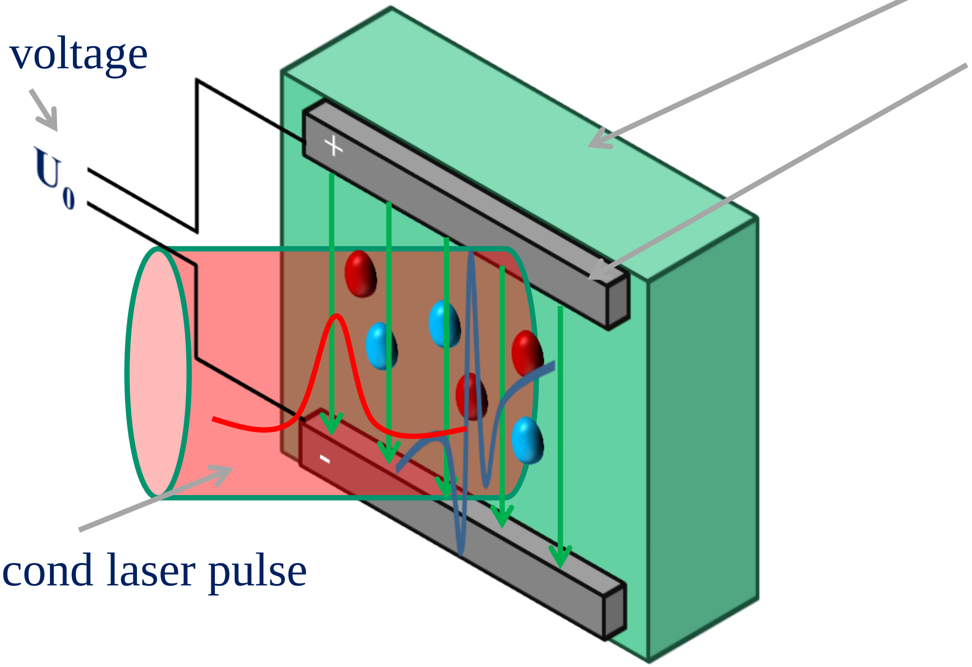
Tonouchi, M. (2007). Cutting-edge terahertz technology. *Nature photonics*, *1*(2), 97.

- The output power of purely electronic system is decreasing for higher frequencies
- Also for optoelectronic photomixers
- Output power of lasers is decreasing with lower frequencies

➡ THz gap

Optoelectronic THz generation

Bias voltage



Femtosecond laser pulse

Semiconductor

Antenna structure

Electron

Hole

Accelerated carriers generate electromagnetic field

→ Here in the THz frequency range

Optoelectronic THz generation

- The electric field from an electric dipole can be described as follows:

$$\mathrm{E}(r,t)\frac{1}{4\pi\varepsilon_0}\left[\frac{\mathbf{1}}{\boldsymbol{r^3}}\vec{\boldsymbol{p}}(\boldsymbol{t_r})+\frac{\mathbf{1}}{\boldsymbol{cr^2}}\dot{\vec{\boldsymbol{p}}}(\boldsymbol{t_r})+\frac{\mathbf{1}}{\boldsymbol{c^2r}}\ddot{\vec{\boldsymbol{p}}}(\boldsymbol{t_r})\right]$$

- We can see 3 different contributions to the electric field:

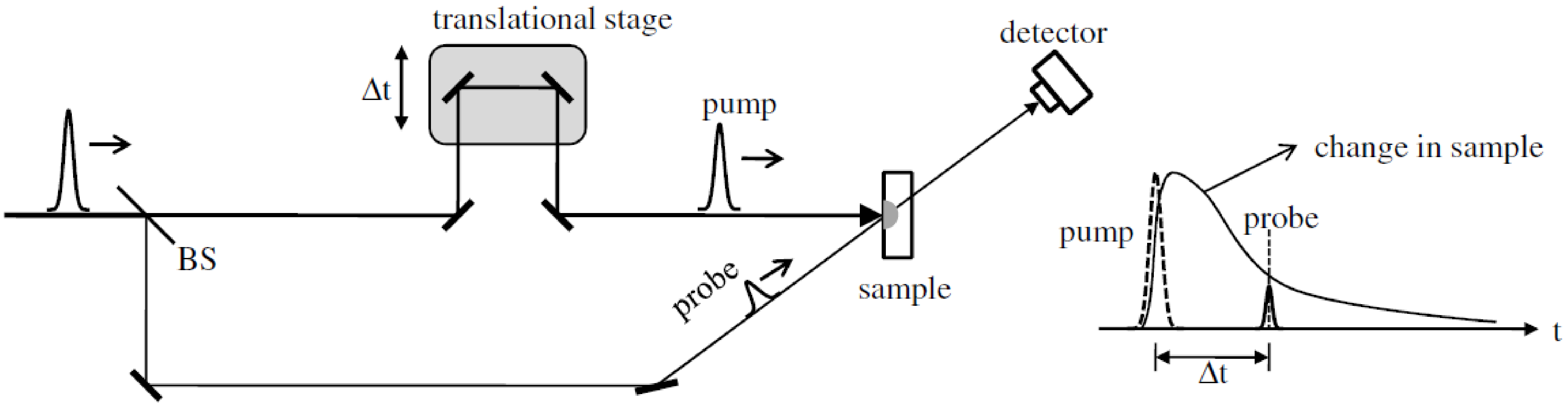
$\frac{1}{r^3}\vec{p}(t_r)$ = static electric field

$\frac{1}{cr^2}\dot{\vec{p}}(t_r) = j(t)$ current density

$\frac{1}{c^2r}\ddot{\vec{p}}(t_r) = \frac{dj(t)}{dt}$ first derivative of the current density

- Since we are interested in the far field ($r \gg \lambda \gg d$, where d is the size of the electric dipole) only the last term survives due to the ${}^{1}/_{r}$ propornationaliy
- The radiated (far) field is hence proportional to a change in current density

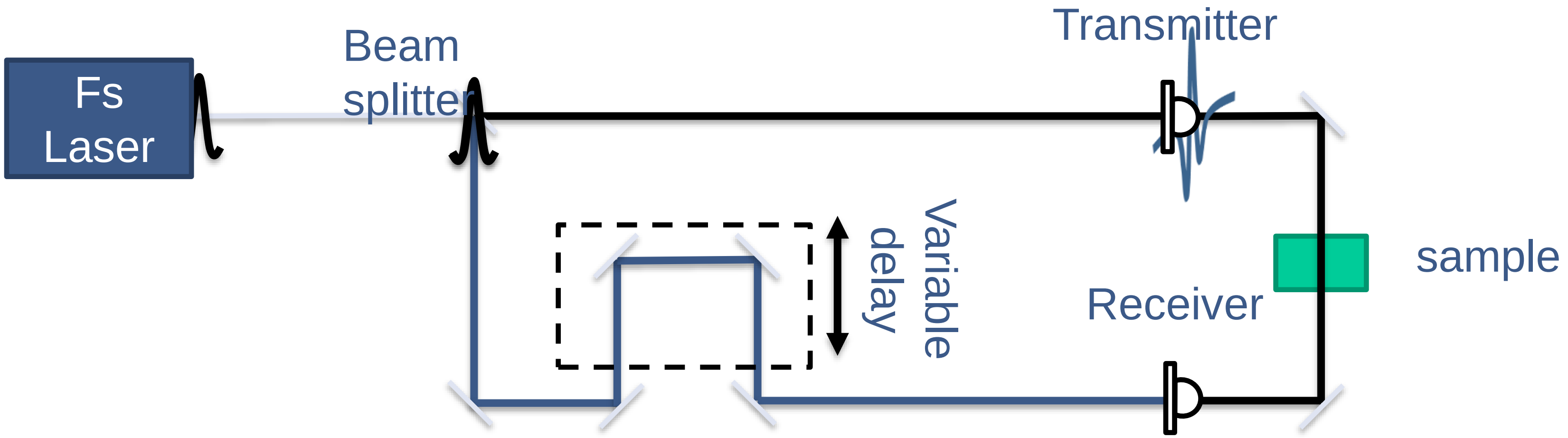
Time-resolved pump-probe technique



- Due to the short duration and high peak power, femtosecond lasers can be used to excite effects like ionization, polarization changes and measure the duration
- With time-resolved pump probe experiments, **reversible** processes can be measured with a high temporal resolution only limited by pulse duration (~6 fs) and accuracy of translation stage (~0.1 fs)
- Samples are measured by probe in transmission, reflection and/or scattering
- Important: Mode locked lasers emits periodic pulse train

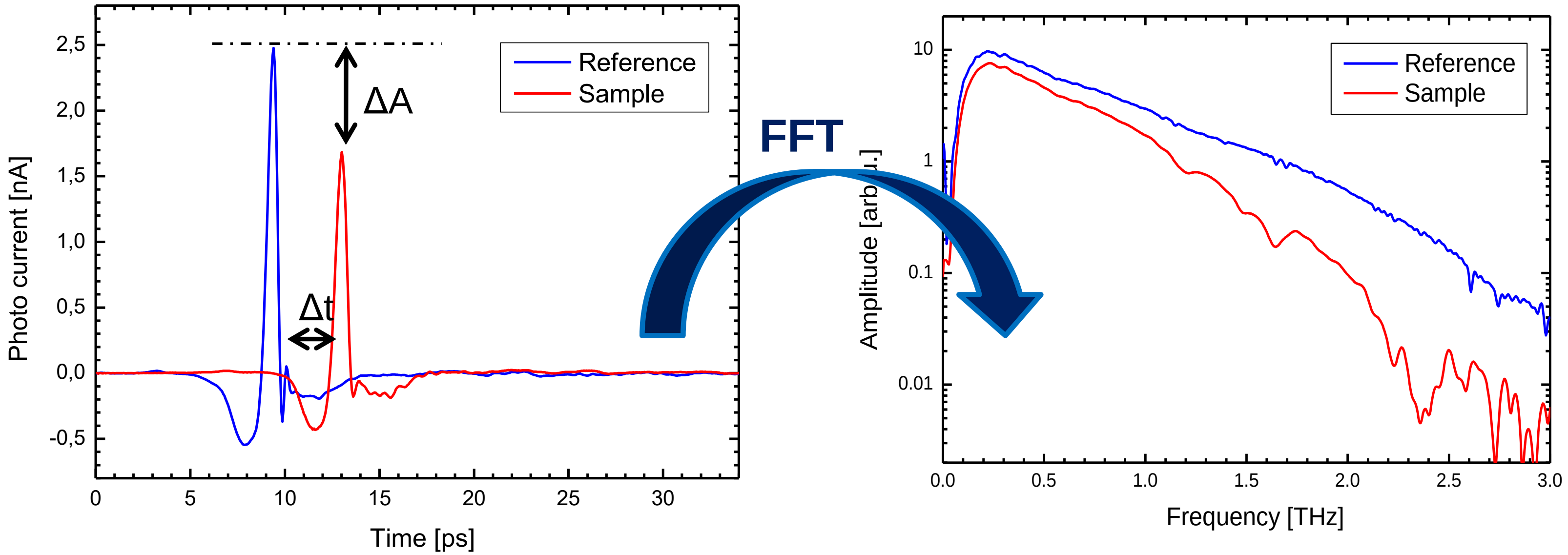
Lee, Yun-Shik. *Principles of terahertz science and technology*. Vol. 170. Springer Science & Business Media, 2009.

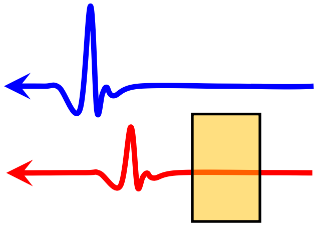
Terahertz time-domain spectroscopy (THz-TDS)



- Setup like pump-probe technique: Transmitter and Receiver are used for frequency conversion
- The THz pulse is sampled at the receiver with the shorter optical probe pulse
- The variable delay line (translation stage) enables optical sampling of the THz field amplitude
- For spectroscopy: THz pulses are measured with and without sample
- Fourier transforms of the waveforms give information about dispersion and absorption

Terahertz time-domain spectroscopy (THz-TDS)



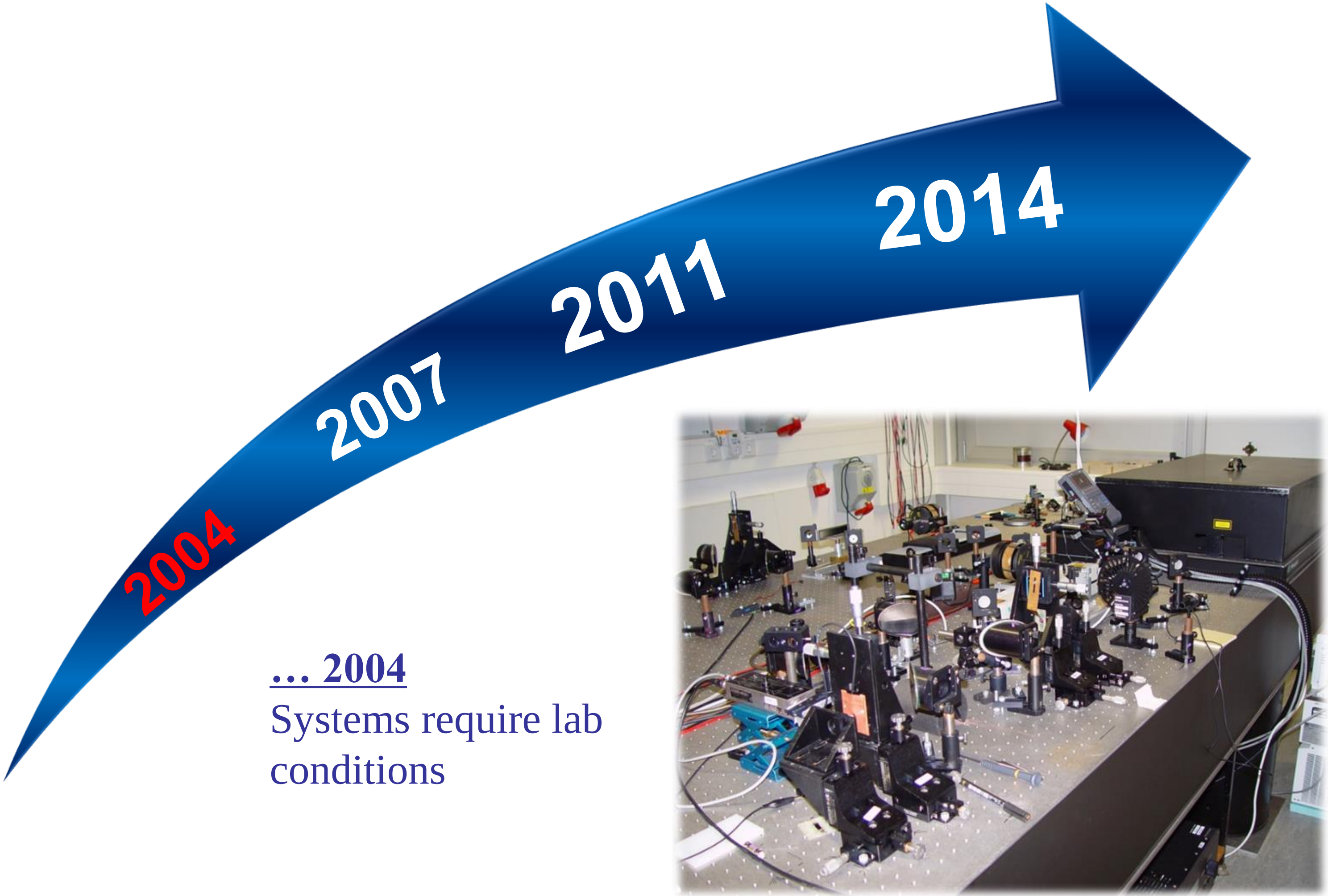


Delay (Δt) ↔ thickness (d) & refractive index (n)

Attenuation (ΔA) ↔ absorption coefficient (α)

<u>Material parameters:</u> Calculation of transfer function: $H(\omega) = \frac{Sample(\omega)}{Reference(\omega)}$

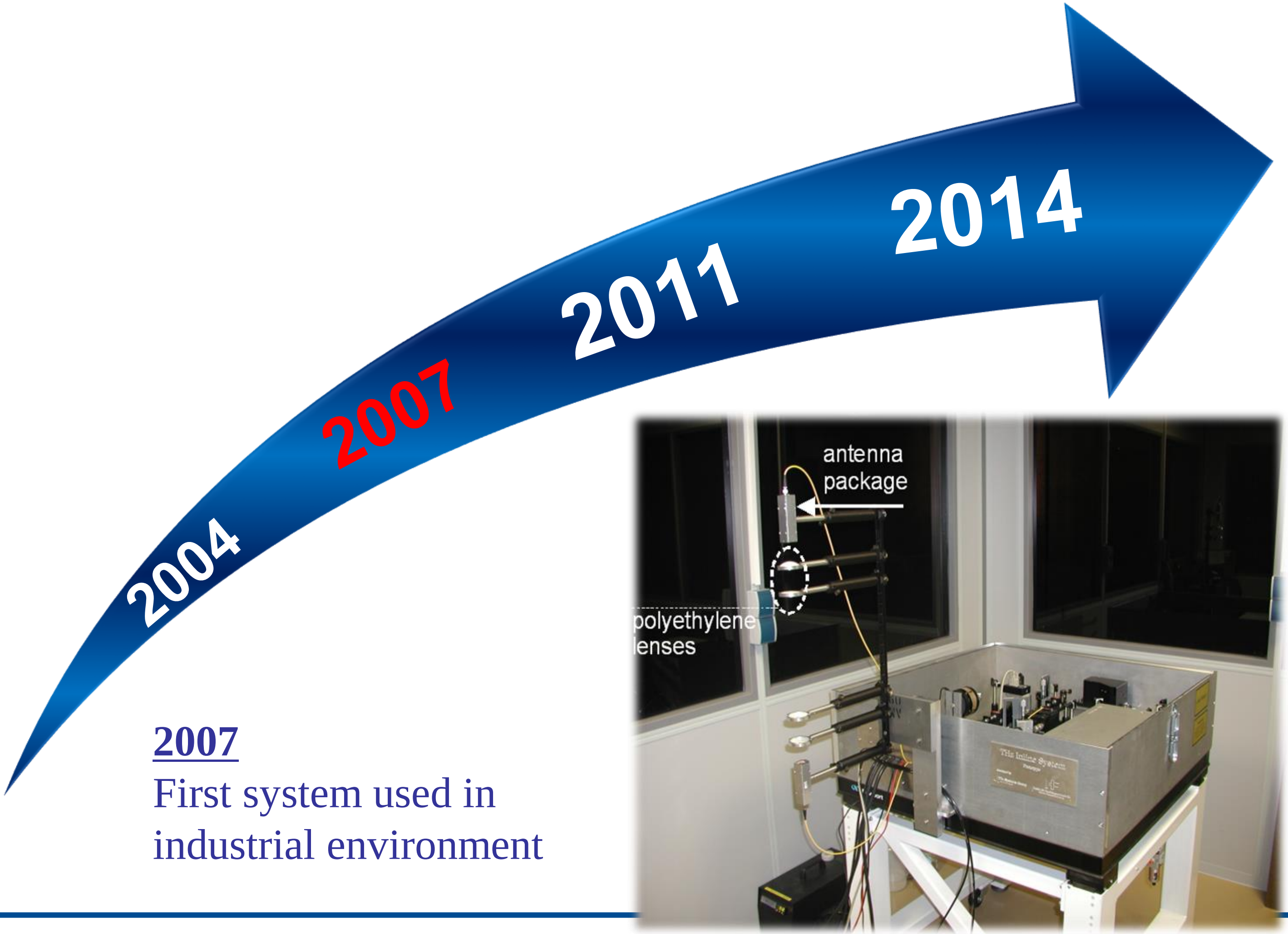
Development of THz-TDS systems



… 2004

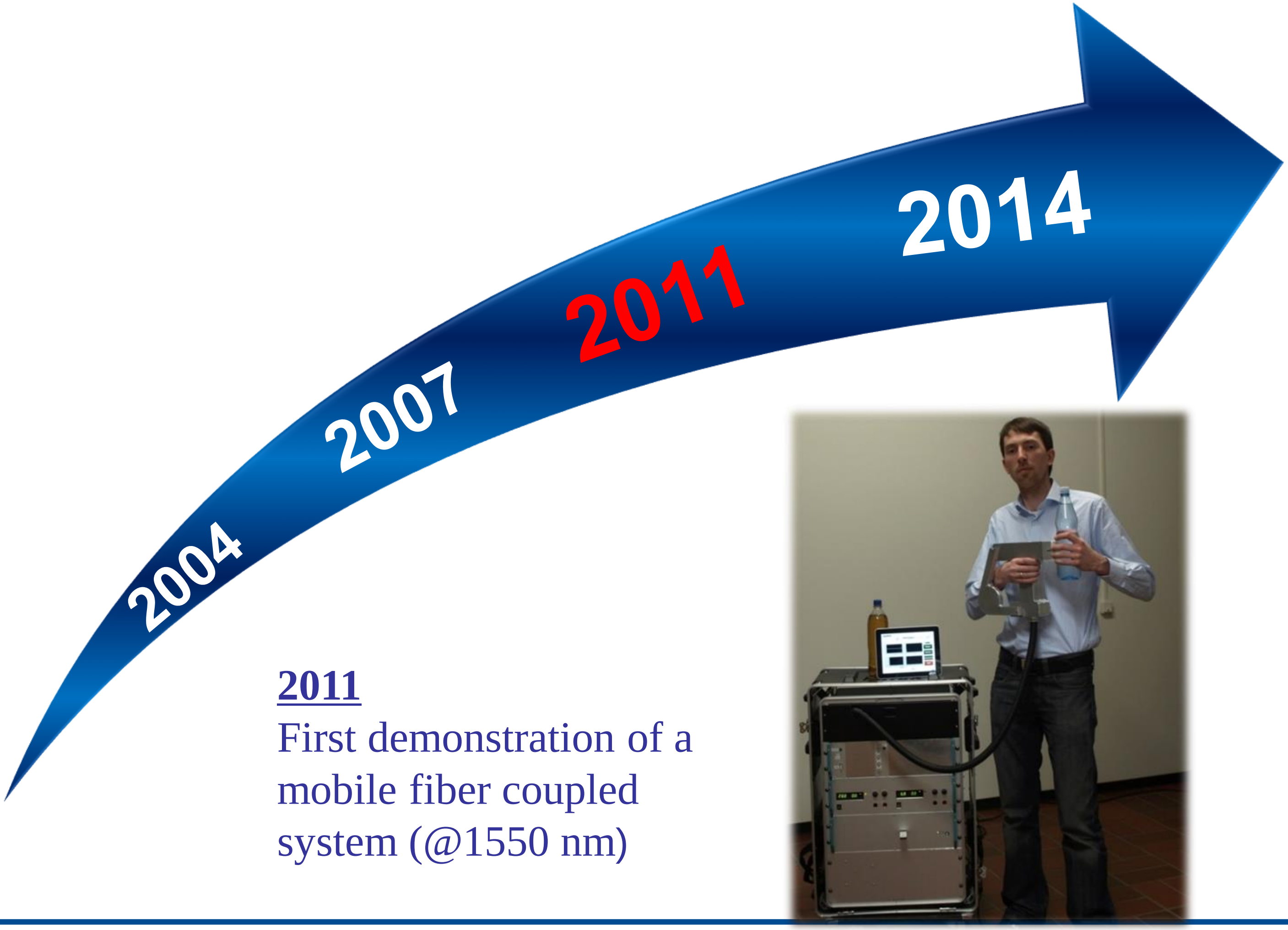
Systems require lab conditions

Development of THz-TDS systems



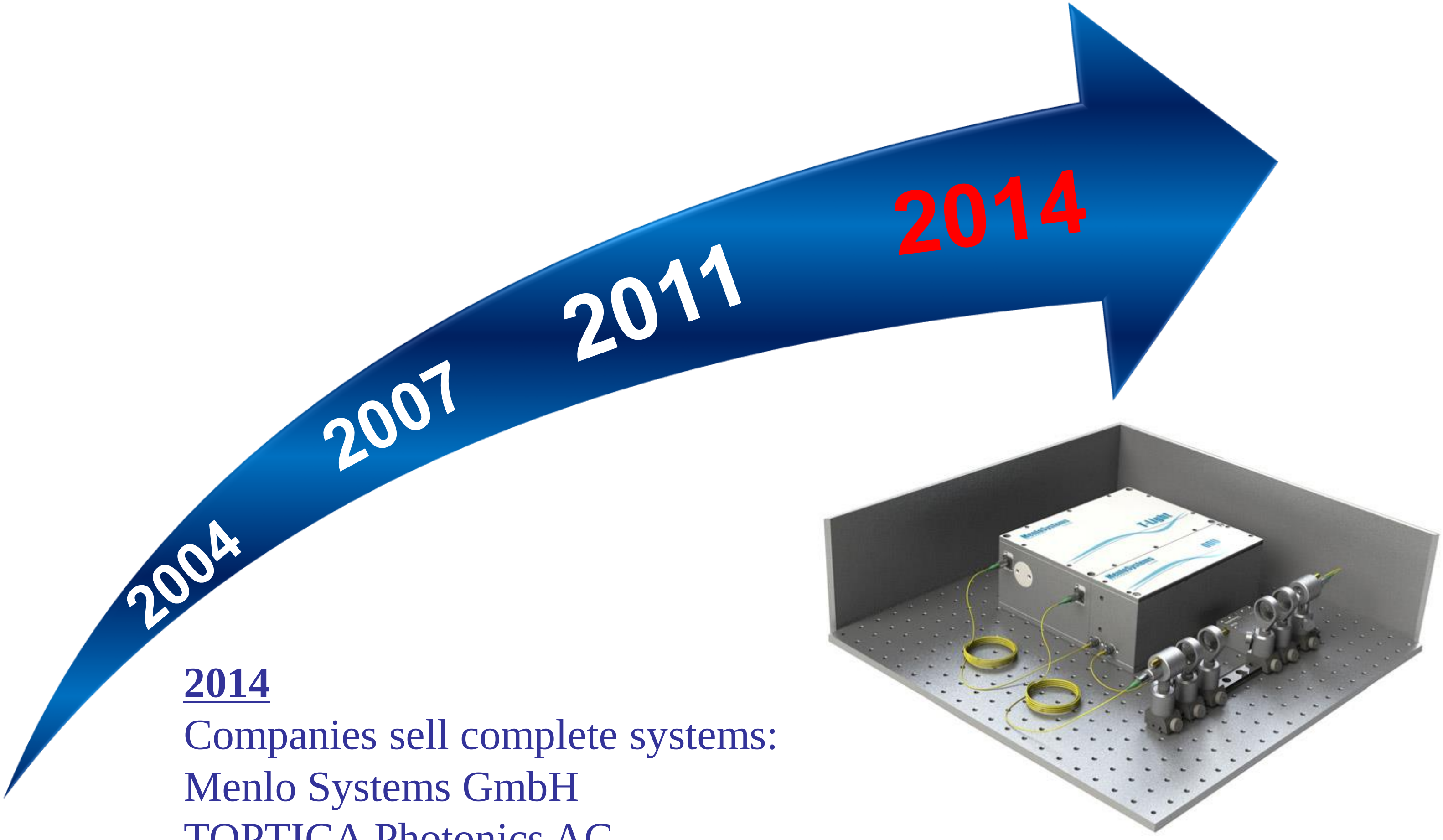
2007
First system used in
industrial environment

Development of THz-TDS systems



2011
First demonstration of a mobile fiber coupled system (@1550 nm)

Development of THz-TDS systems



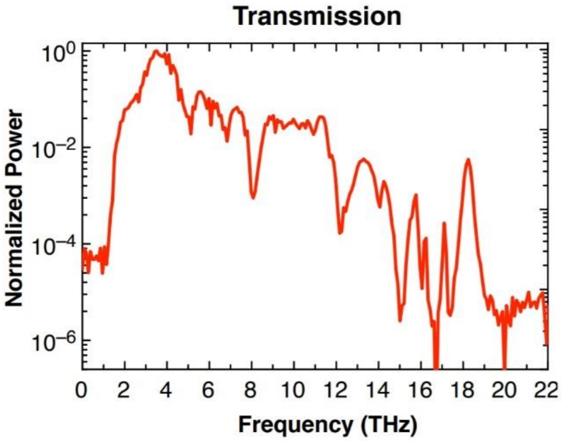
2014

Companies sell complete systems:
Menlo Systems GmbH
TOPTICA Photonics AG
Hübner GmbH & Co. KG

TOPTICA Photonics AG

Hübner-Photonics

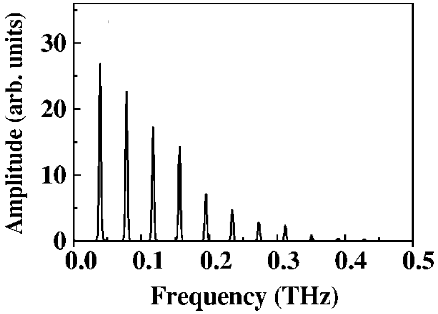
Rainbow Photonics



Development of LD-driven THz-TDS systems

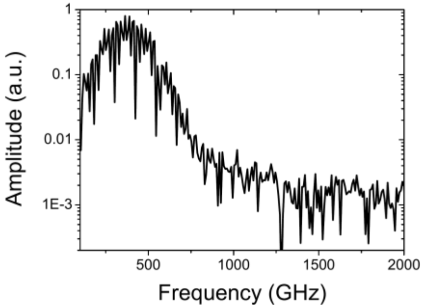
Morikawa et al. (2000) [1]:

- Cross-correlation spectroscopy (CCS)
- THz-TDS setup using a commercial 810 nm multi-mode laser diode



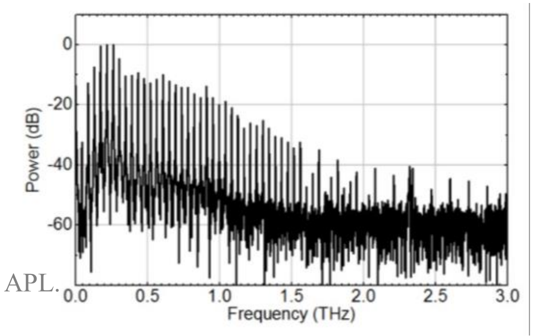
Scheller et al. (2009) [2]:

- Quasi time-domain spectroscopy (QTDS)
- Adapted the CCS approach using a commercial 660 nm multi-mode laser diode



Kohlhaas et al. (2017) [3]:

- QTDS using a 1550 nm multi-mode laser diode
- Using photodiode-based emitter and photoconductive receiver

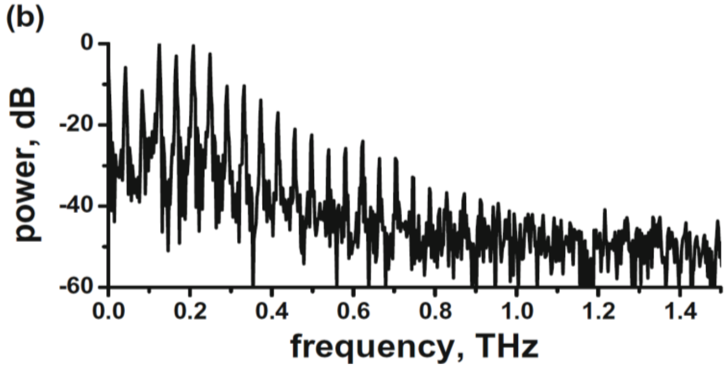


[1] O. Morikawa et al., “A cross-correlation spectroscopy in subterahertz region using an incoherent light source”, APL.
[2] M. Scheller and M. Koch, “Terahertz quasi time domain spectroscopy,” Opt. Express.
[3] R.B. Kohlhaas, A. Rehn et al., “Terahertz quasi time-domain spectroscopy based on telecom technology for 1550 nm” Opt. Express.

Development of LD-driven THz-TDS systems

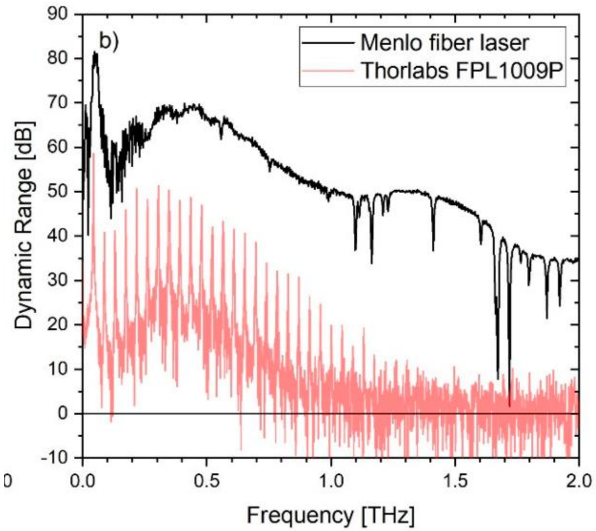
Merghem et al. (2017) [1]:

- THz-TDS using a research-grade monolithic 1550 nm QDash mode-locked laser diode (MLLD)
- Ultra-high repetition rate THz-TDS (UHRR THz-TDS)



Tybussek et al. (2019) [2]:

- TDS using a low-cost “accidentally mode-locked” Fabry-Perot laser diode
- Comparison between conventional THz-TDS and UHRR THz-TDS



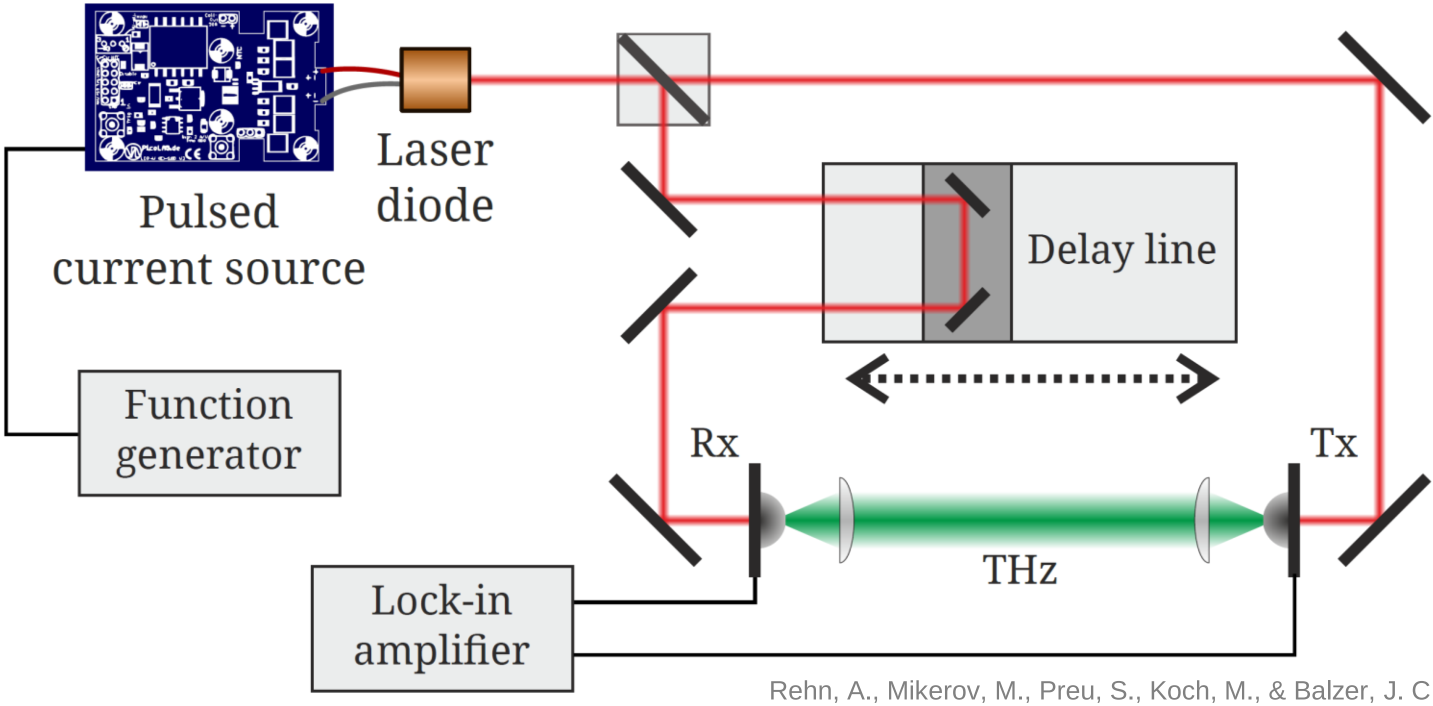
[1] K. Merghem et al., “Terahertz time-domain spectroscopy system driven by a monolithic semiconductor laser”, J. Infrared, Millimeter, Terahertz Waves (2017).

[2] K.-H. Tybussek et al., “Terahertz time-domain spectroscopy based on commercially available 1550 nm fabry–perot laser diode and ErAs:In(Al)GaAs photoconductors”, Appl. Sci. (2019).

ENHANCING THE PERFORMANCE OF QTDS/CCS

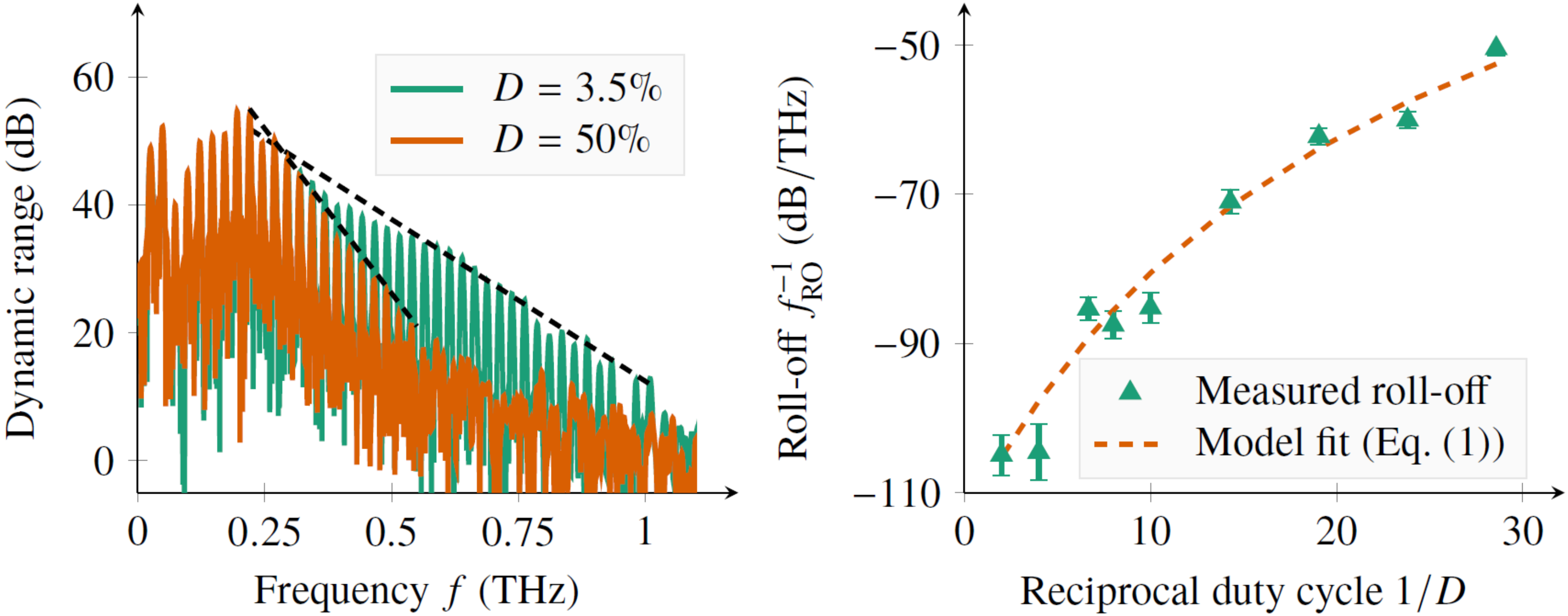
How to increase the photocurrent?

- Idea: $E_{THz} \propto \frac{d}{dt} I_{opt} \Rightarrow$ Increase the photocurrent!
- However, thermal damage threshold for PCA's around $P_{avg} \approx 30mW$
- Reduce the duty cycle to gain a higher peak intensity at constant P_{avg} :



Rehn, A., Mikerov, M., Preu, S., Koch, M., & Balzer, J. C. (2018). Enhancing the performance of THz quasi time-domain spectroscopy systems by low duty cycle laser operation. *Optics Express*, *26*(25)

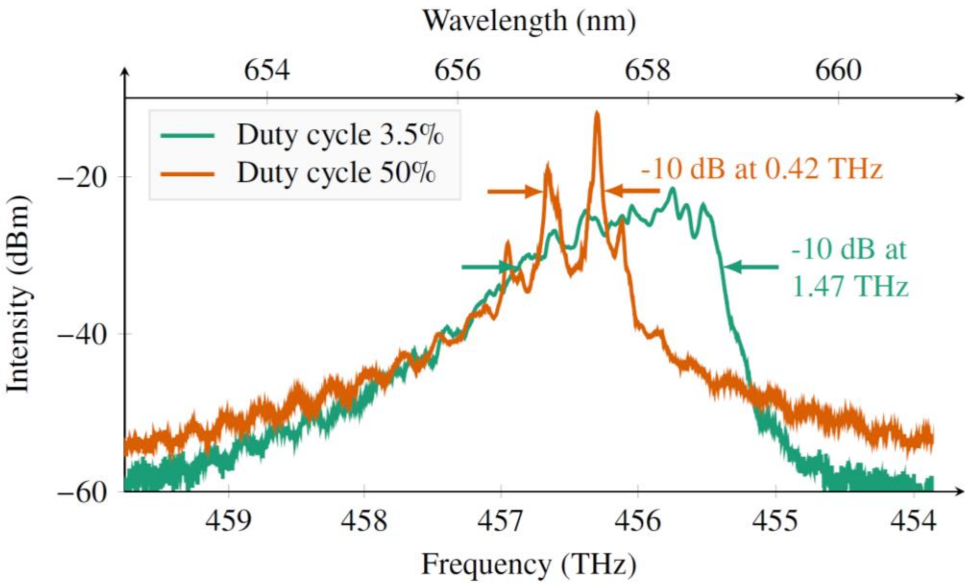
Variation of the duty cycle

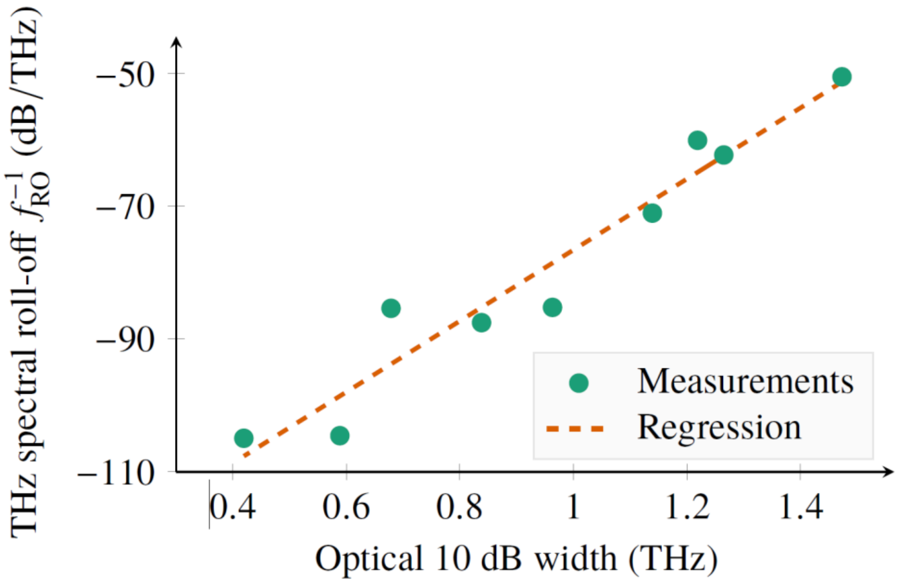


- Measurements have shown that not only the emitted power is increased
- Lower duty cycle decreases the roll-off and increases the spectral bandwidth!

Rehn, A., Mikerov, M., Preu, S., Koch, M., & Balzer, J. C. (2018). Enhancing the performance of THz quasi time-domain spectroscopy systems by low duty cycle laser operation. *Optics Express*, *26*(25)

Variation of the duty cycle



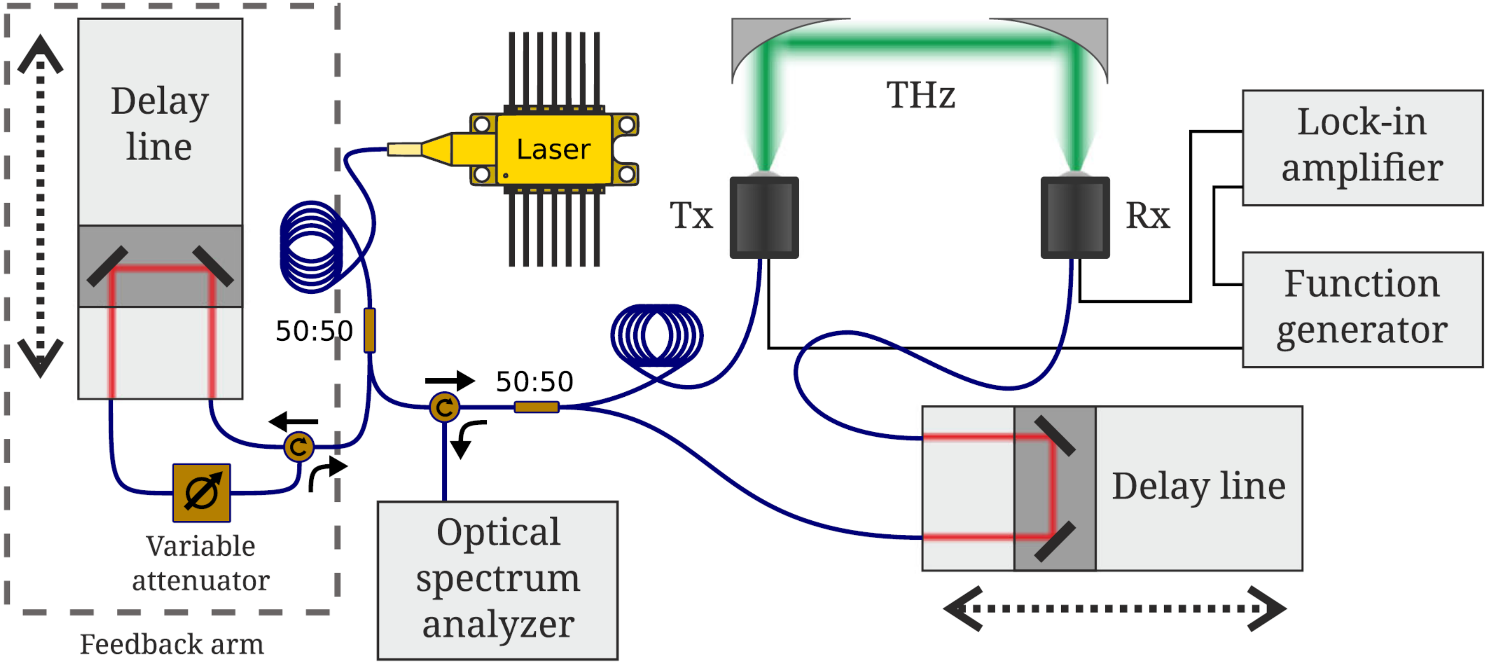


- The increased bandwidth stems from an increase optical bandwidth
- Linear correlation between optical bandwidth and THz roll-off
- However, very low duty cycle destroys laser diode

Rehn, A., Mikerov, M., Preu, S., Koch, M., & Balzer, J. C. (2018). Enhancing the performance of THz quasi time-domain spectroscopy systems by low duty cycle laser operation. *Optics Express*, *26*(25)

Optical feedback

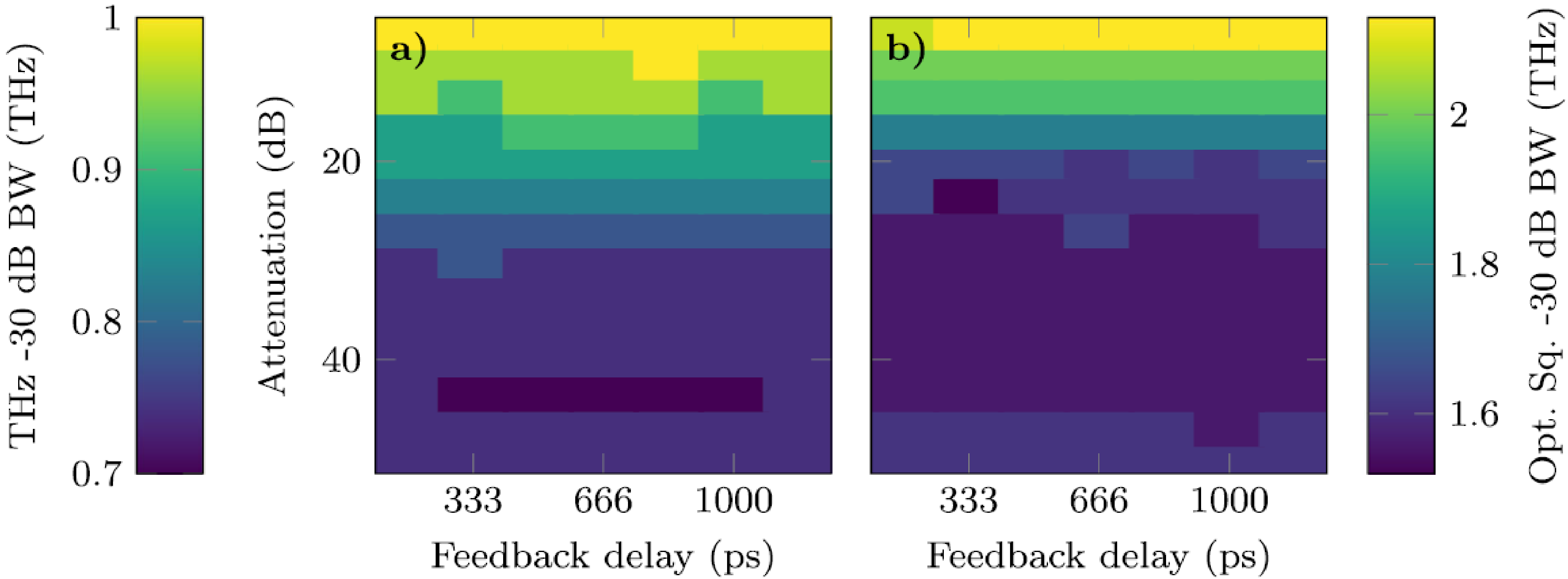
- Idea: Feedback has shown some potential to stabilize laser diodes
- Here, we used a variable attenuator with a delay line to manipulate feedback strength and delay:



Rehn, A., Kohlhaas, R., Globisch, B., & Balzer, J. C. (2019). Increasing the THz-QTDS Bandwidth from 1.7 to 2.5 THz Through Optical Feedback. Journal of Infrared, Millimeter, and Terahertz Waves, 40(11), 1103-1113.

Optical feedback

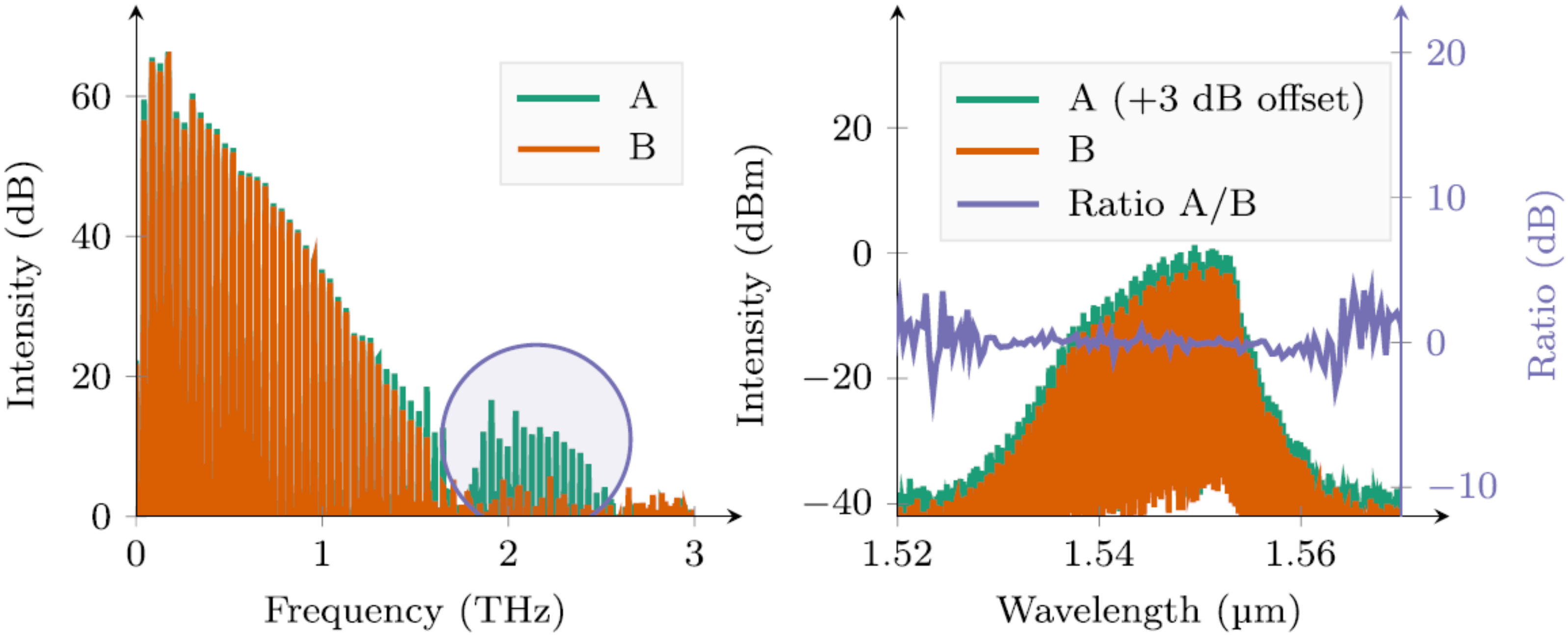
- Our observation: stronger feedback (i.e. lower attenuation) broadens the optical spectrum
- Broad optical spectrum leads to an increased THz bandwidth



Rehn, A., Kohlhaas, R., Globisch, B., & Balzer, J. C. (2019). Increasing the THz-QTDS Bandwidth from 1.7 to 2.5 THz Through Optical Feedback. Journal of Infrared, Millimeter, and Terahertz Waves, 40(11), 1103-1113.

Optical feedback

- But what about the phase?
- A closer look reveals quite different THz spectrum for identical optical spectrum
- This must be related to the phase of the optical spectrum!

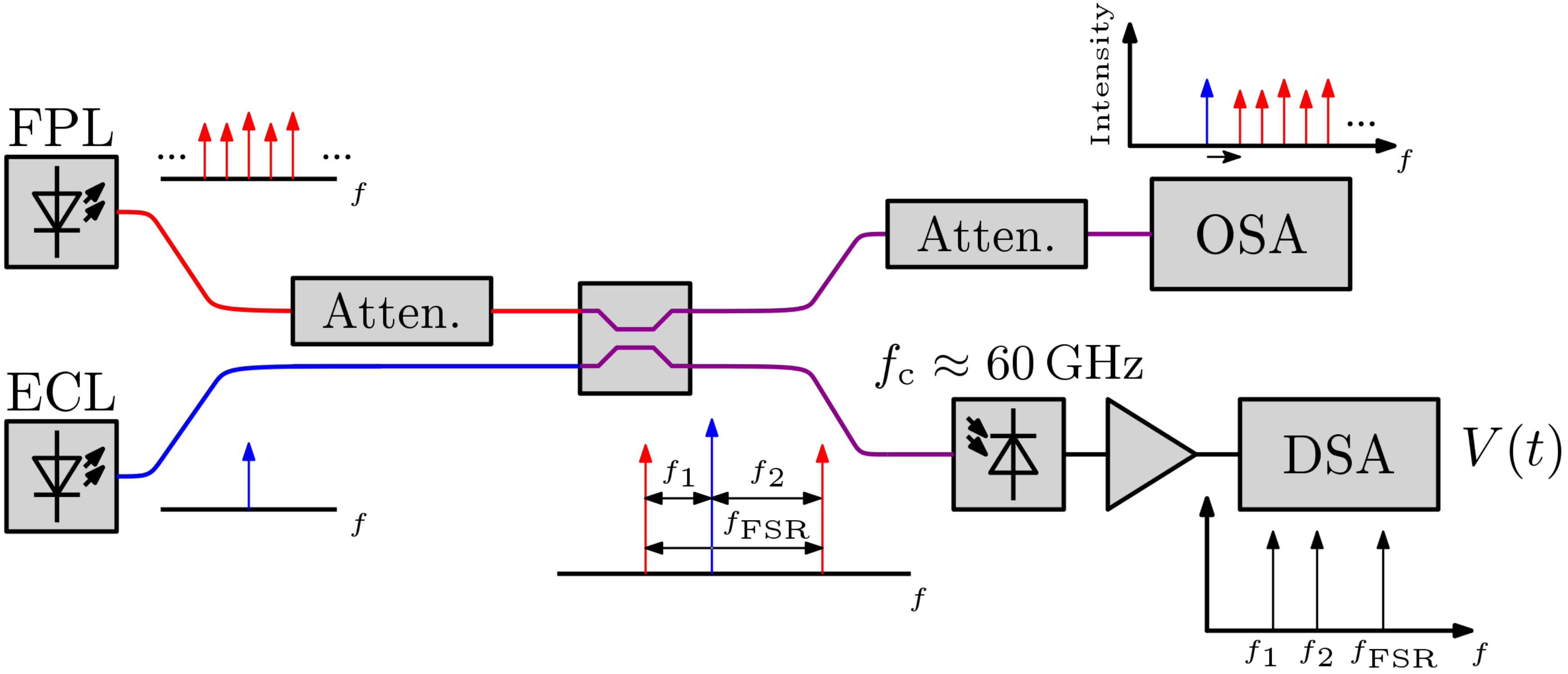


Rehn, A., Kohlhaas, R., Globisch, B., & Balzer, J. C. (2019). Increasing the THz-QTDS Bandwidth from 1.7 to 2.5 THz Through Optical Feedback. Journal of Infrared, Millimeter, and Terahertz Waves, 40(11), 1103-1113.

THEORETICAL MODEL FOR MLLD-DRIVEN THZ TDS

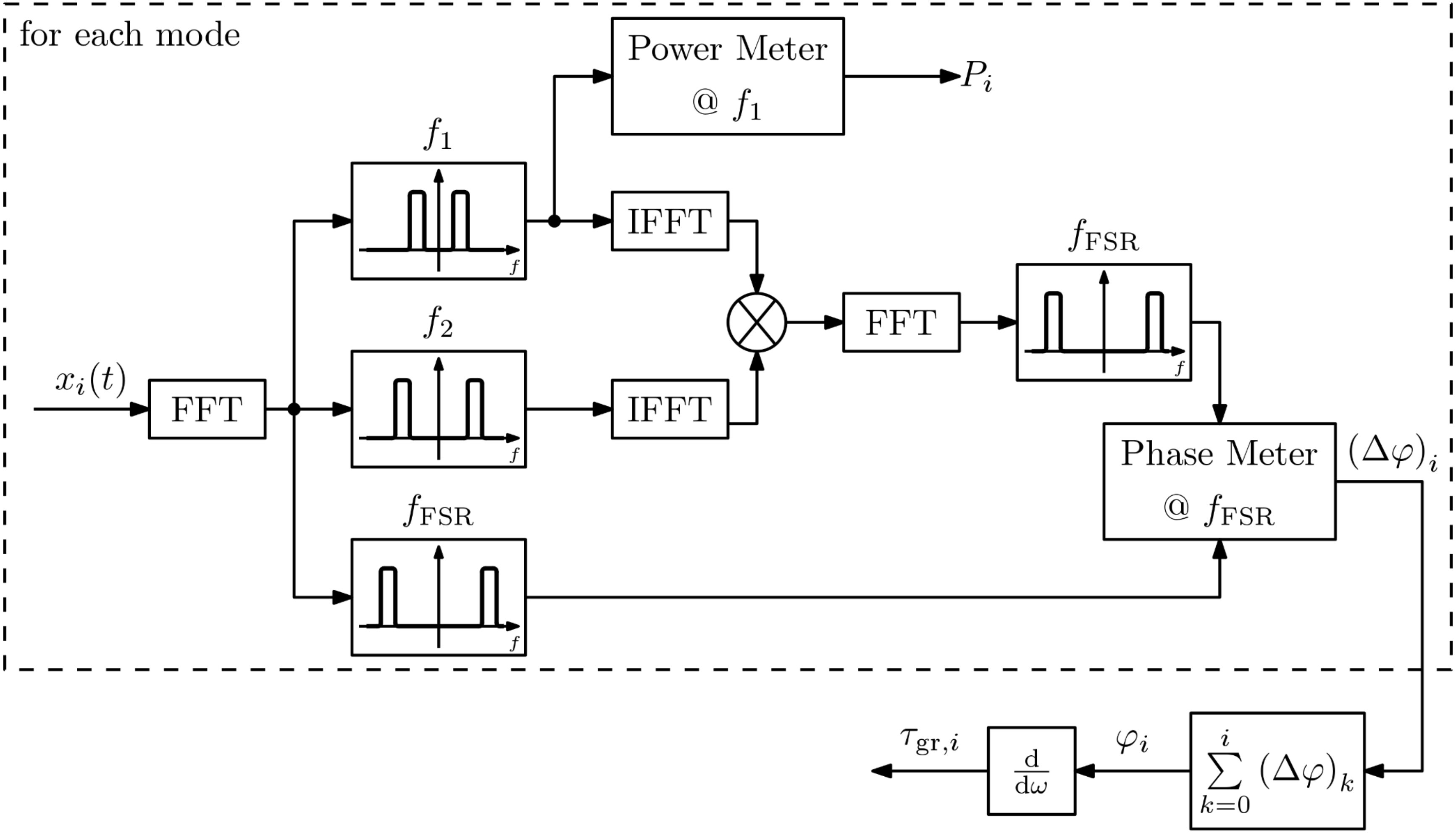
How to measure the optical phase

- Indirect methods: frequency resolved optical gating (FROG)
- Direct measurement: Stepped-heterodyne optical complex spectrum analyzer [*]



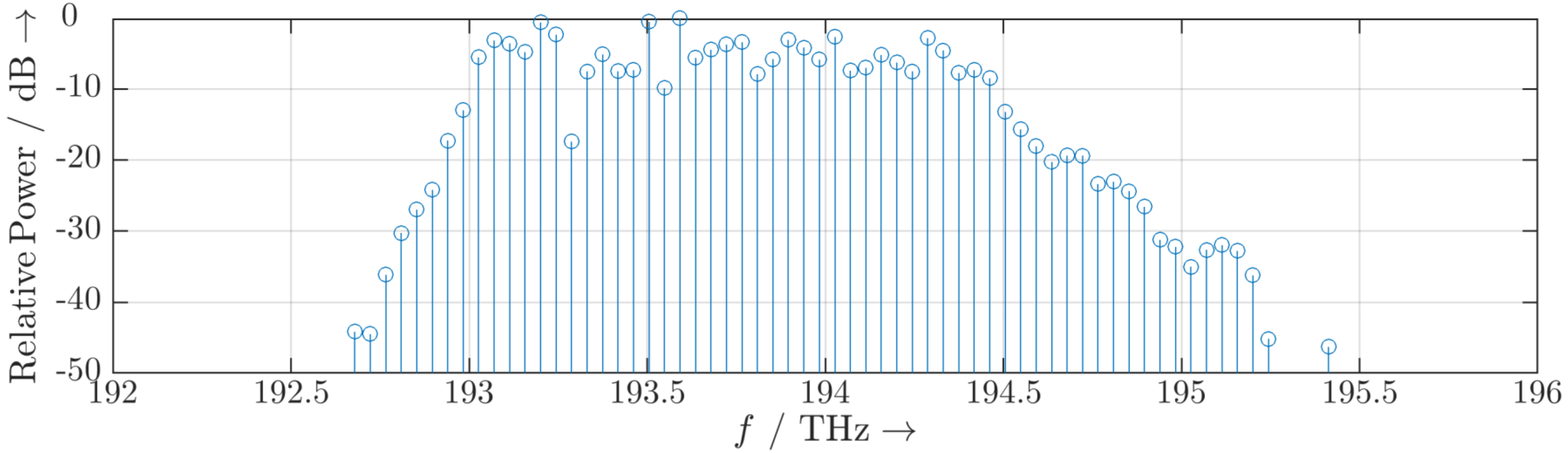
[*] Reid, D. A., Murdoch, S. G., & Barry, L. P. (2010). Stepped-heterodyne optical complex spectrum analyzer. Optics express, 18(19), 19724-19731.

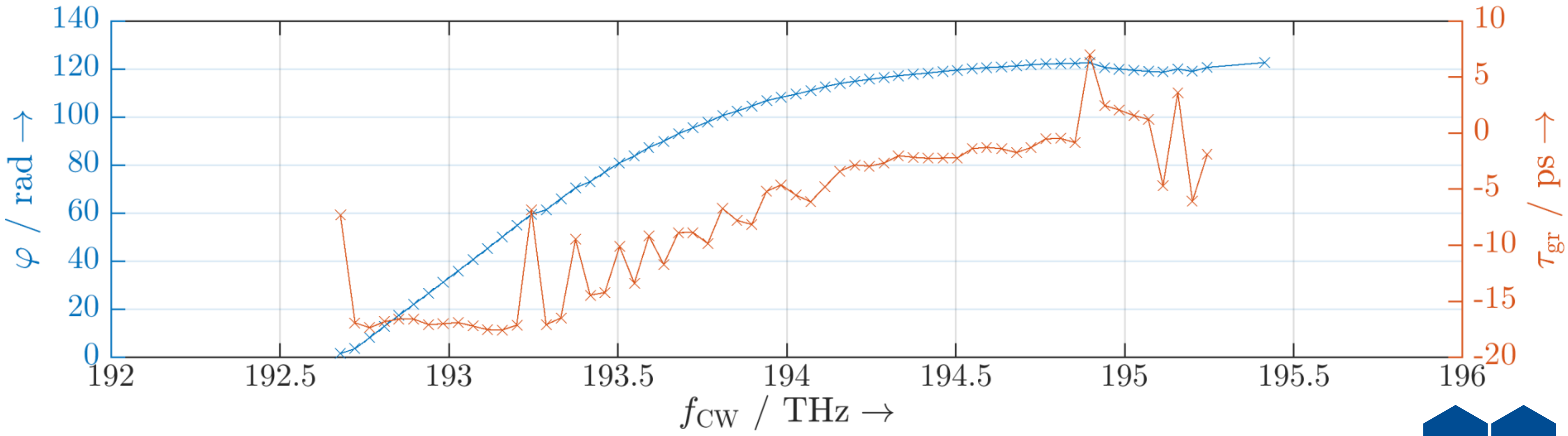
How to measure the optical phase



jan.balzer@uni-due.de

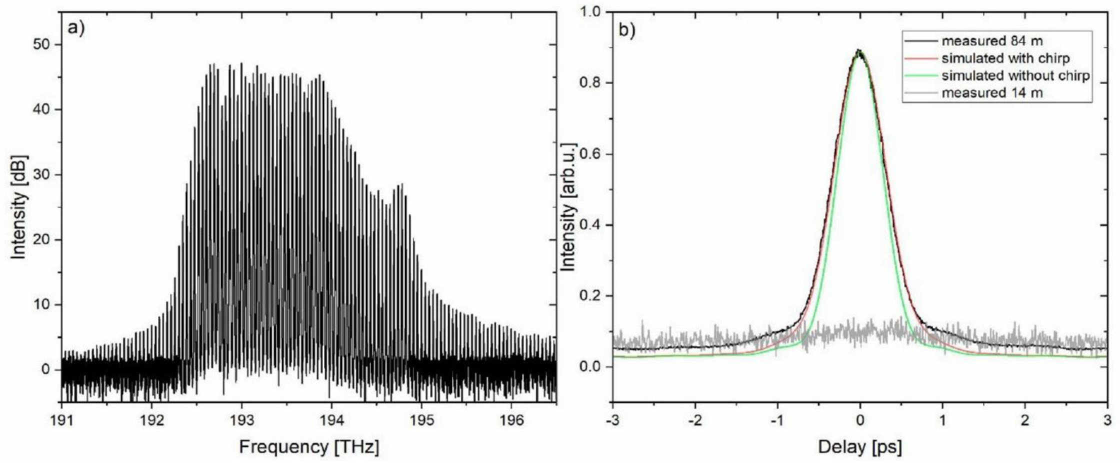
Results for the commercial laser diode





Results for the commercial laser diode

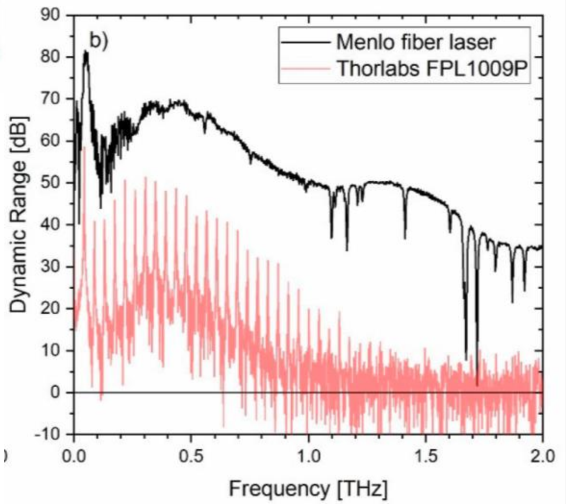
- Laser is mode-locked but strongly chirped!
- With dispersion compensation: conventional THz-TDS is feasible



- -10 dB-bandwidth: 1.61 THz
- 544 fs deconvoluted pulse duration
- Fourier-limited pulse duration: ~450 fs

Comparison to fiber laser:

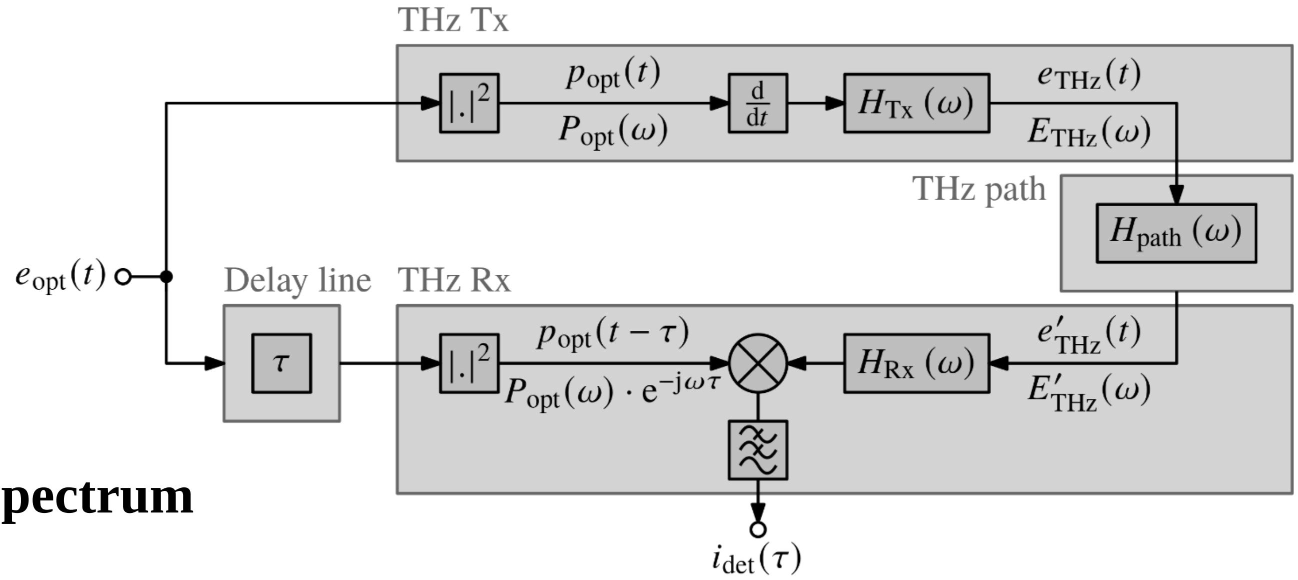
- Fiber laser: 70 dB peak SNR and > 4 THz bandwidth
- Laser diode: 51 dB peak SNR and ~ 1.4 THz bandwidth



Tybussek, K. H., Kolpatzeck, K., Faridi, F., Preu, S., & Balzer, J. C. (2019). Terahertz Time-Domain Spectroscopy Based on Commercially Available 1550 nm Fabry–Perot Laser Diode and ErAs: In (Al) GaAs Photoconductors. Applied Sciences, 9(13), 2704.

System theoretical model

- **System model**



- **Complex optical spectrum**

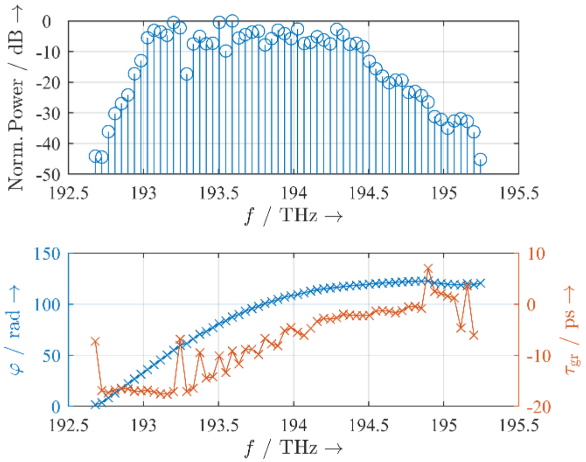
$$e_{\mathrm{opt}}(t) = \sum_{k=0}^{N-1} E_k \cdot \mathrm{e}^{\mathrm{j}[(\omega_0+k\Omega)\cdot t+\varphi_k]}\ ,\ \Omega = 2\pi\cdot F$$

- **Transfer function of the terahertz system**

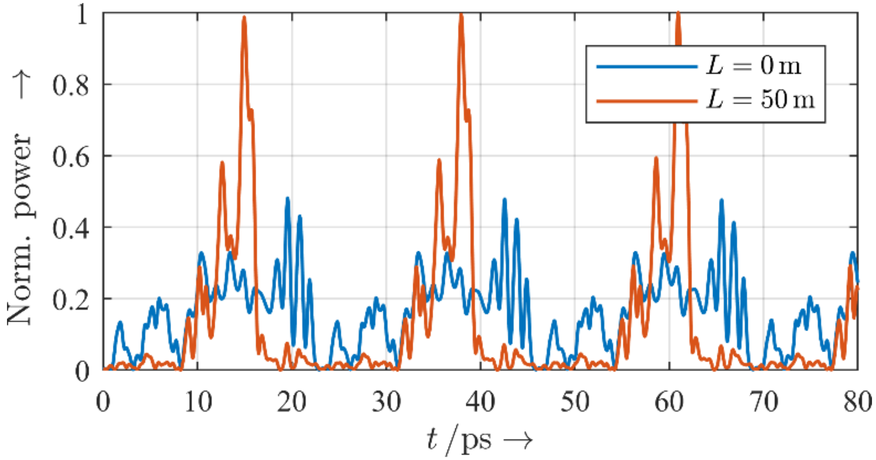
$$H_{\mathrm{THz}}(m\Omega) = m\Omega\cdot H_{\mathrm{Tx}}(m\Omega)\cdot H_{\mathrm{path}}(m\Omega)\cdot H_{\mathrm{Rx}}(m\Omega)\ ,\ m = 1\ldots N-1$$

Kolpatzeck, K., Liu, X., Tybussek, K. H., Häring, L., Zander, M., Rehbein, W., ... & Balzer, J. C. (2020). System-theoretical modeling of terahertz time-domain spectroscopy with ultra-high repetition rate mode-locked lasers. Optics Express, 28(11), 16935-16950.

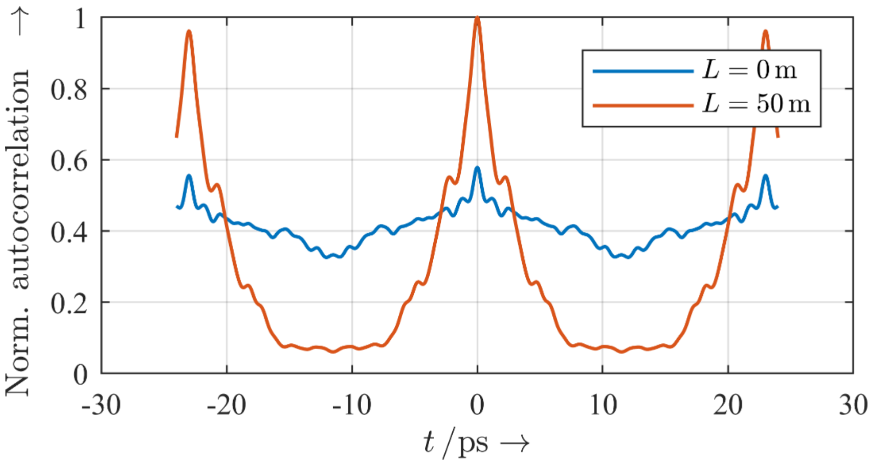
Verification of the complex optical spectrum



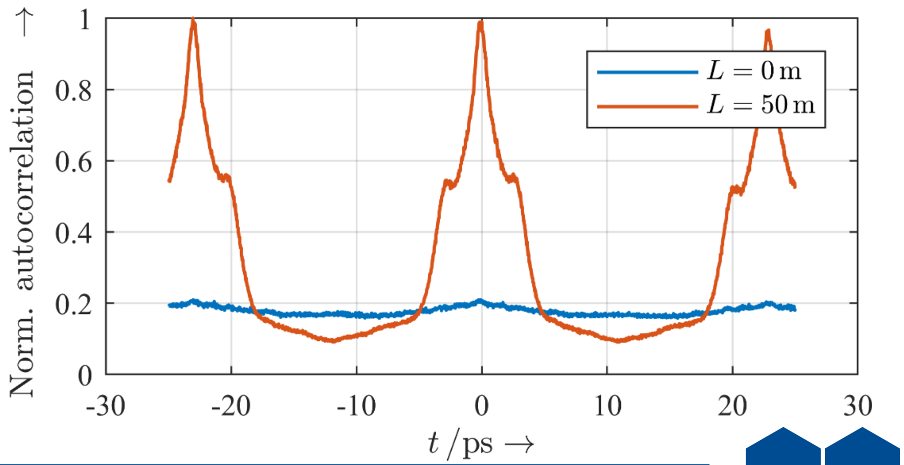
Calculate E-Field



Calculated autocorrelation



Measured autocorrelation



Verification of the model

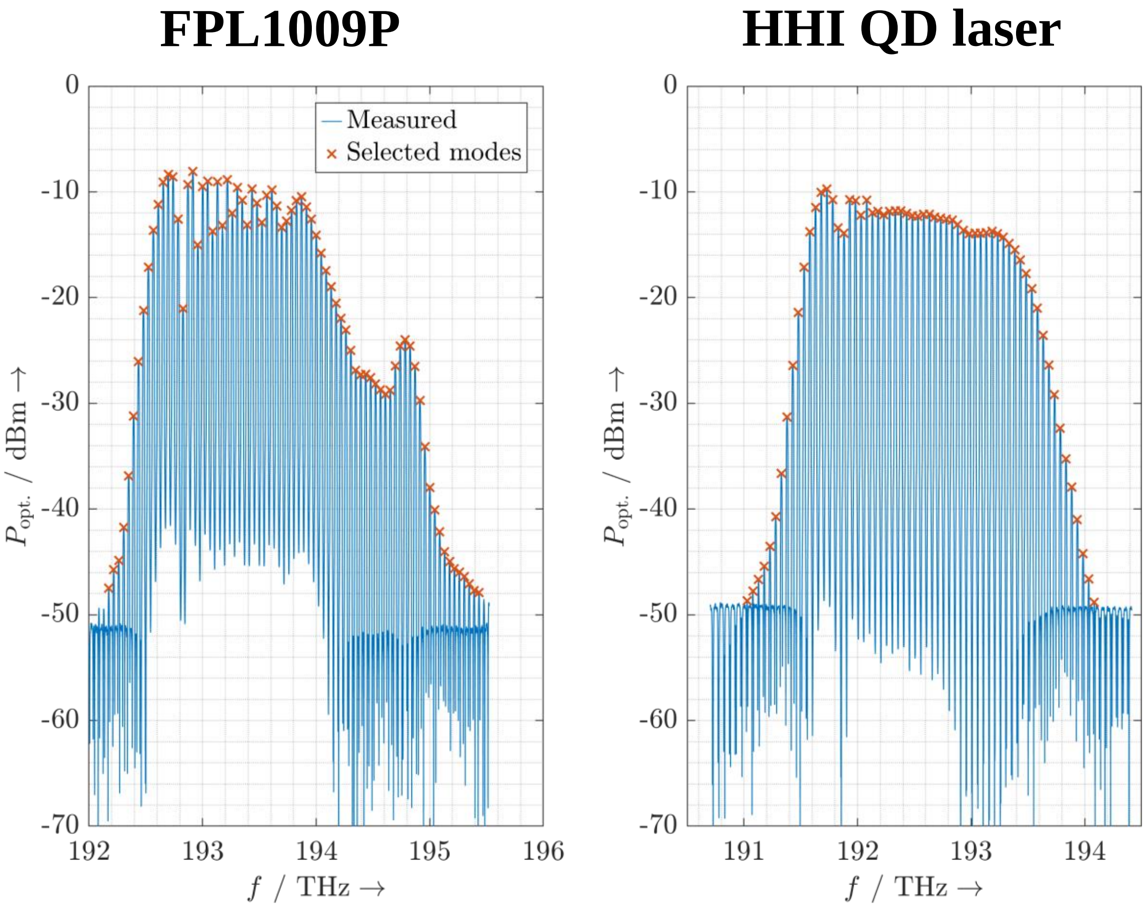
Two different MLLDs

- Thorlabs FPL1009P: Single-section Fabry-Perot laser
- QD laser from Fraunhofer HHI: Two-section quantum dot laser
- Different amounts of chirp generated by adding different lengths of single-mode fiber between laser and spectrometer

Measured quantities

- Complex optical spectrum using optical spectrum analyzer and stepped-heterodyne technique
- Terahertz transfer function by frequency-domain spectroscopy (FDS)
- Intensity autocorrelation with SHG autocorrelator
- Terahertz spectrum using TDS setup with MLLD

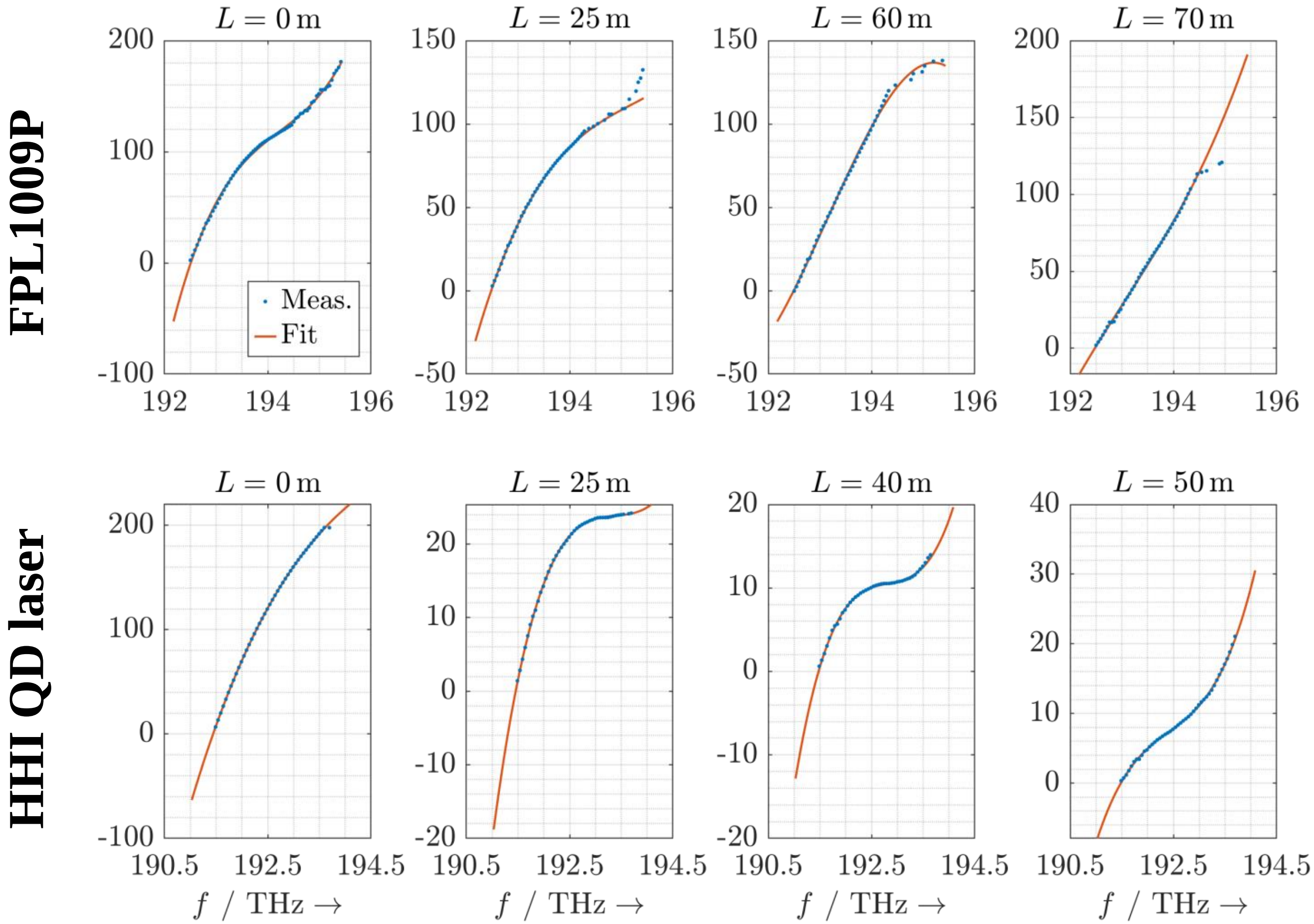
Optical spectrum


FPL1009P

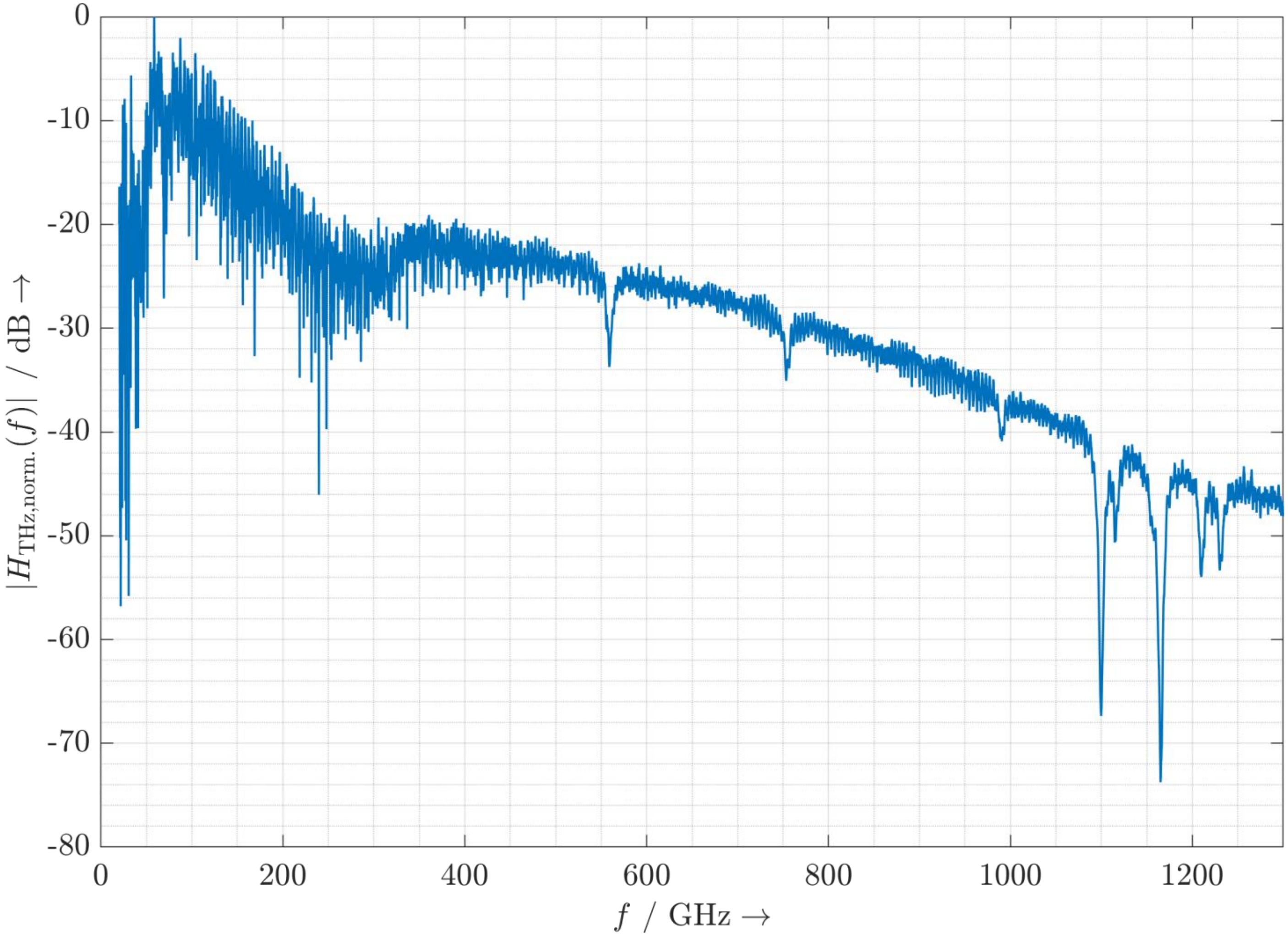
HHI QD laser

Kolpatzeck, K., Liu, X., Tybussek, K. H., Häring, L., Zander, M., Rehbein, W., ... & Balzer, J. C. (2020). System-theoretical modeling of terahertz time-domain spectroscopy with ultra-high repetition rate mode-locked lasers. Optics Express, 28(11), 16935-16950.

Optical phase spectra



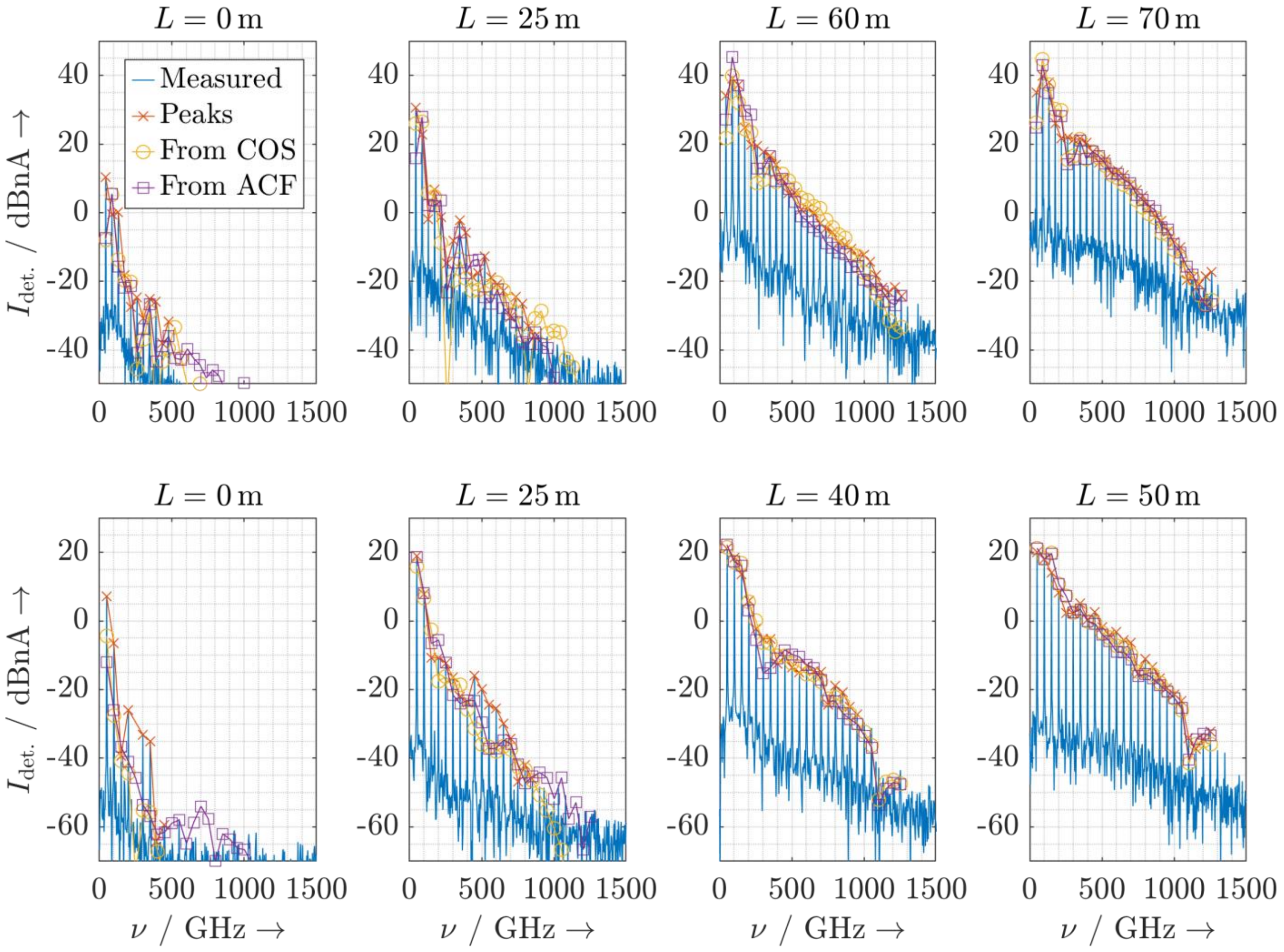
Terahertz path transfer function from THz FDS



Measured and calculated TDS spectra

FPL1009P

HHI QD laser

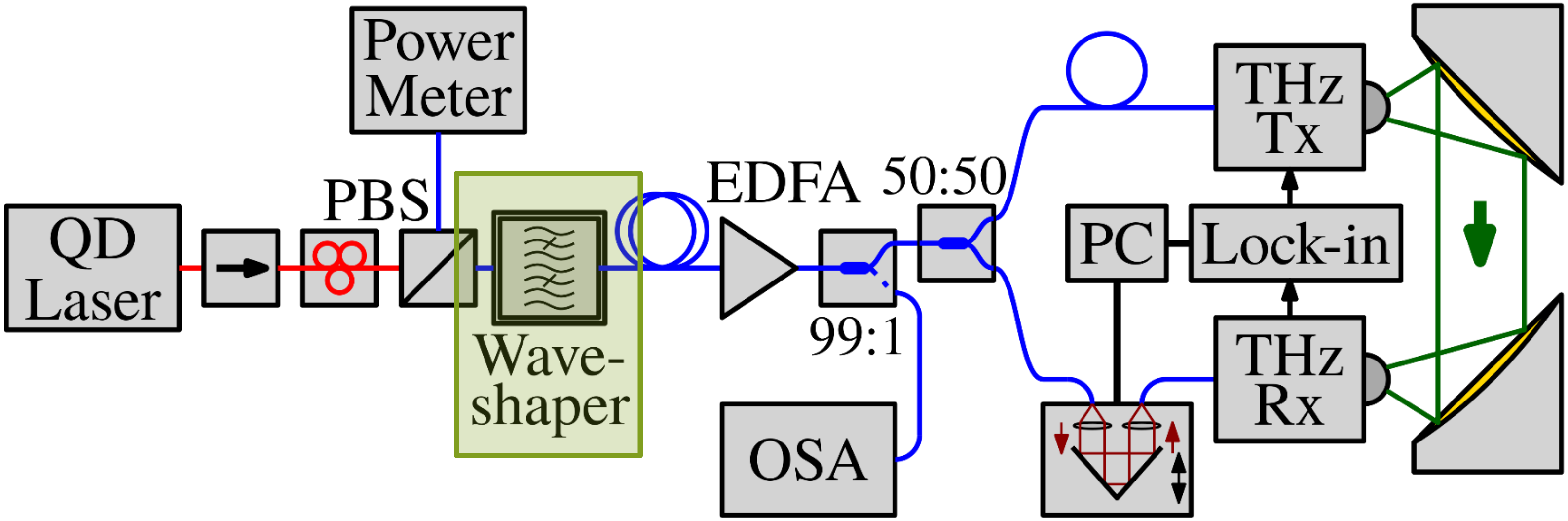


Compact THz TDS system enabled by MLLD's
jan.balzer@uni-due.de

MODEL-DRIVEN OPTIMIZATION OF THZ TDS

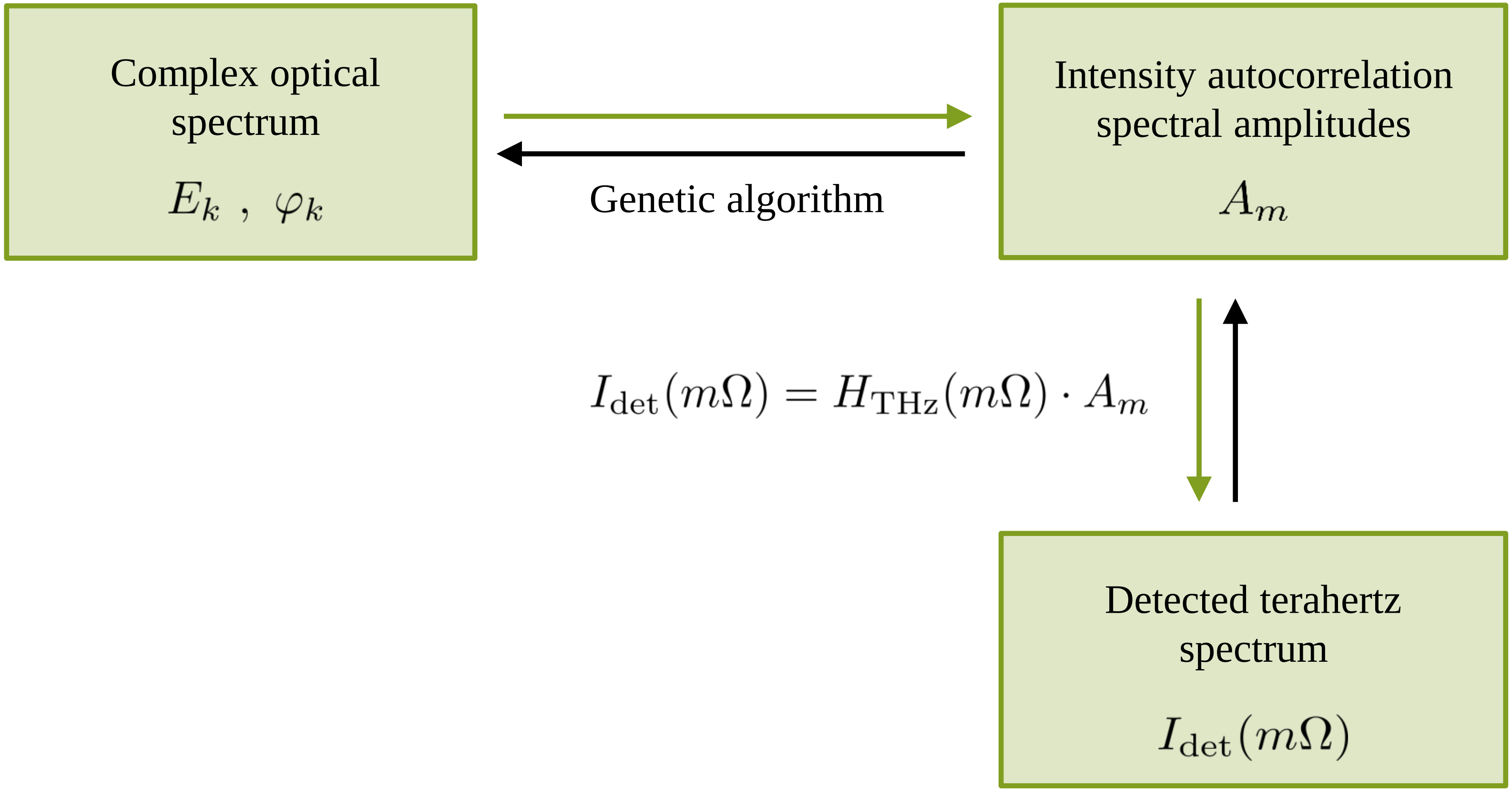
Model-driven spectral shaping

- Systematic enhancement of terahertz spectra in UHRR THz-TDS.
- Optical spectrum is shaped with a programmable optical filter (=waveshaper) to synthesize a desired terahertz spectrum.
- Fast offline optimization of the optical spectrum using a genetic algorithm based on an analytical model of UHRR THz-TDS system.



Spectral synthesis

$$A_m = \sum_{k=m}^{N-1} \sum_{l=m}^{N-1} E_k E_{k-m} E_l E_{l-m} \cdot \cos\left[(\varphi_k - \varphi_{k-m}) - (\varphi_l - \varphi_{l-m})\right]$$



Complex optical spectrum

$E_k \;,\; \varphi_k$

Genetic algorithm

Intensity autocorrelation spectral amplitudes

A_m

$$I_{\mathrm{det}}(m\Omega) = H_{\mathrm{THz}}(m\Omega) \cdot A_m$$

Detected terahertz spectrum

$I_{\mathrm{det}}(m\Omega)$

Synthesis procedure

1. Determination of the unshaped optical spectrum:
   - Measurement of the unshaped optical amplitude spectrum.
   - Linearization of the optical phase spectrum.
2. Definition of an optimization goal.
3. Optimization with a genetic algorithm:
   - Genetic algorithm in MATLAB determines the optimized optical amplitudes E_k.
   - Calculation of the amplitude coefficients of the programmable optical filter from the desired amplitudes E_k and the unshaped optical spectrum.
4. Implementation of the amplitude coefficients in the programmable optical filter.
5. Measurement of the terahertz spectrum.

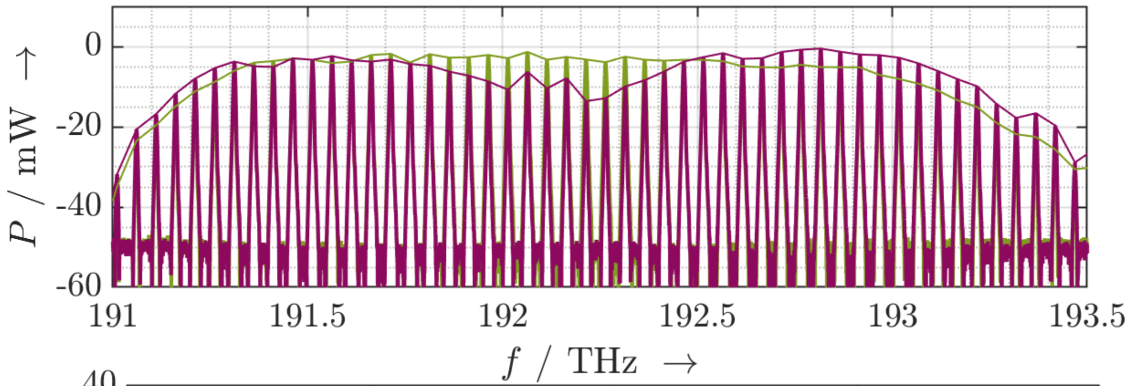
Spectral shaping for maximum bandwidth

- Spectral amplitude of the weakest spectral component in the terahertz spectrum is maximized. Step by step, the spectral components are “pulled up” by the GA

- Optical spectra

  unshaped

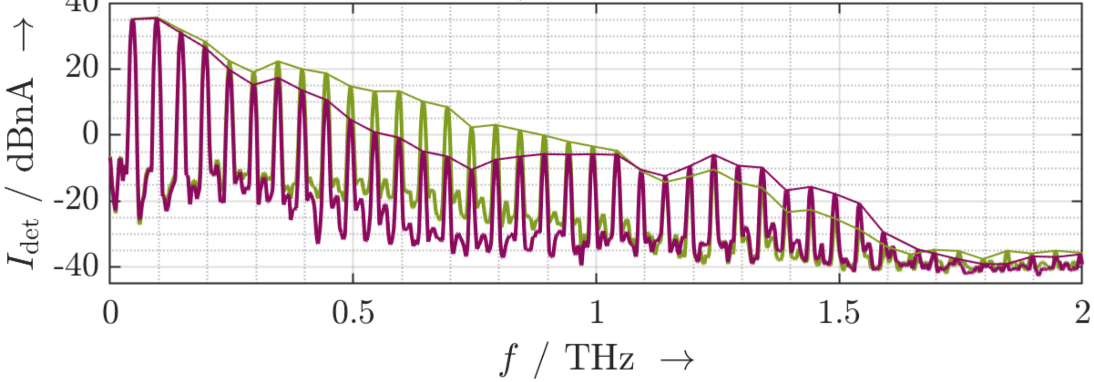
  shaped



- Terahertz spectra

  unshaped

  shaped

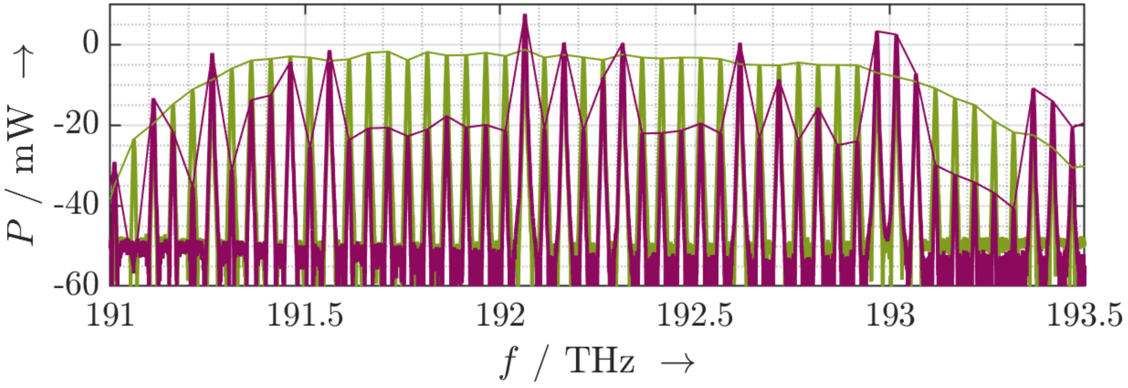


- Increase of up to 8 dB for spectral components between 1.1 and 1.6 THz

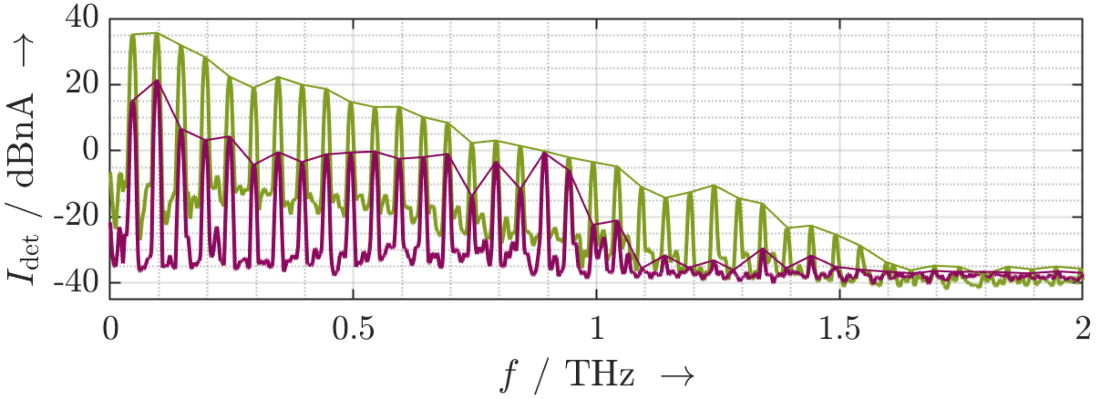
Spectral shaping for rectangular THz spectrum

- Amplitude variation of the spectral components in the detected terahertz spectrum is minimized. Tradeoff between flatness and amplitude.

- Optical spectra
  - — unshaped
  - — shaped



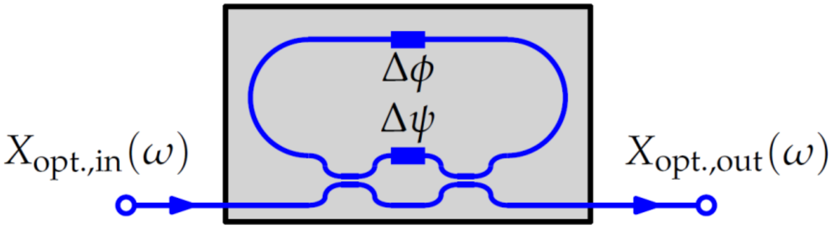
- Terahertz spectra
  - — unshaped
  - — shaped



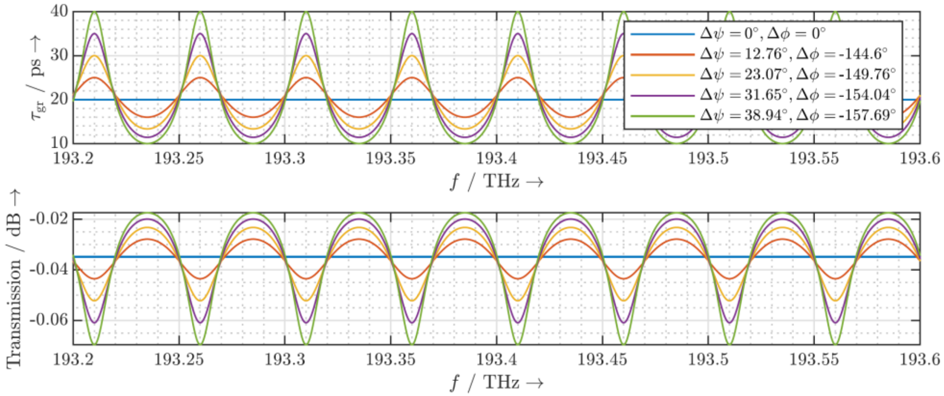
- Good flatness in the frequency range from 150 GHz to 950 GHz.

Conclusion & outlook

- MLLD’s can be used to built compact and cost-effective THz-TDS systems
- Performance of MLLD crucial for performance of THz-TDS (especially the pulse duration/spectral bandwidth)
- Optimization methods must be combined to achieve a bandwidth of 3 THz
- Complete integration of MLLD-driven THz-TDS systems is a big challenge, but we have a concept:



Liu, X., Kolpatzeck, K., Häring, L., Balzer, J. C., & Czylwik, A. (2020). Wideband Beam Steering Concept for Terahertz Time-Domain Spectroscopy: Theoretical Considerations. *Sensors*, *20*(19), 5568.



Thank you for your attention!

Marie SFB/TRR 196

DFG Deutsche Forschungsgemeinschaft
German Research Foundation