Ultrabroadband spectroscopy for explosives detection

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A joint initiative of:

Liberté • Égalité • Fraternité
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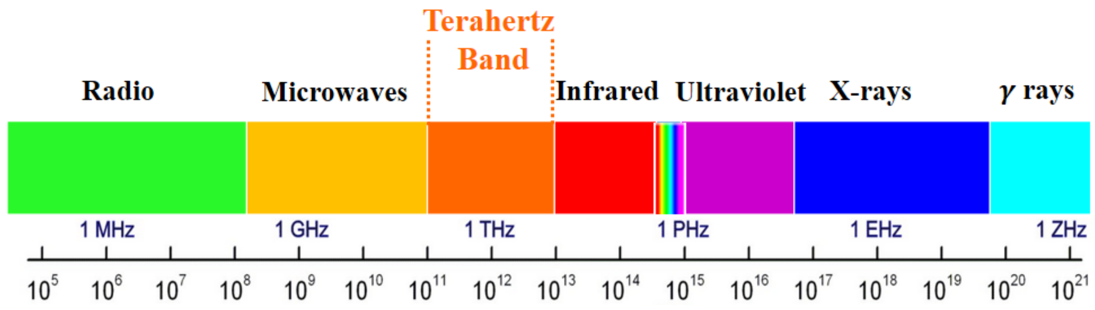
Bundesministerium der Verteidigung

06/06/2017

Terahertz Radiation and Chemical Recognition



SHORT FACTS

- Coherent (phase sensitive), broadband detection method
- Current time domain spectroscopy (TDS) systems working
  in the frequency band from 0.1-4.0 THz
- Characterization of spectral fingerprints allow for chemical
  recognition of illicit substances (e.g. explosives, drugs etc.)
- Contact-free method, identification possible through packaging
- Non-ionizing

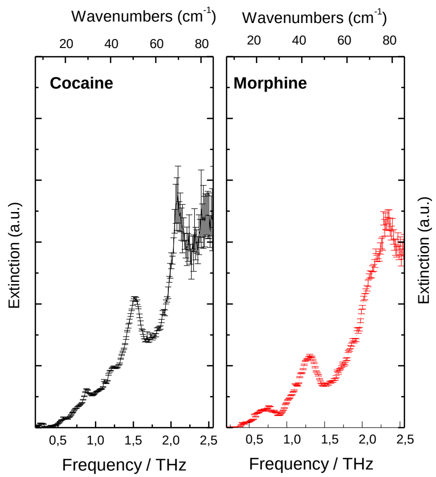
DRAWBACKS

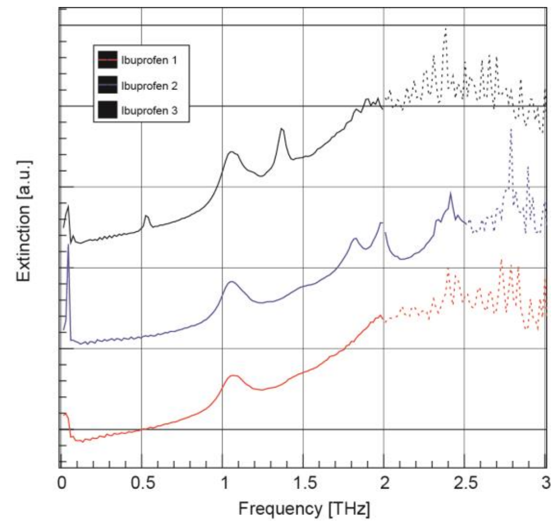
- Limited bandwidth
- Strong absorption due to water vapor
  in atmosphere
- High quality spectroscopy only possible
  in close proximity of the sample

THz spectroscopy for detection purposes

"Civilian" applications: Detection of hazardous substances through packaging

Spectra of cocaine and morphine through plastic bag





Ibuflam 800 Zentiva ®

Ibu 800 Hexal ®

Ibuprofen 600 Stada ®

Spectra of different generica of Ibuprofene

In collaboration with the **institut national de la police scientifique de Lyon**

CSOSG project "AlyPOTEC" 2013-2017

Air plasma based THz stand-off detection

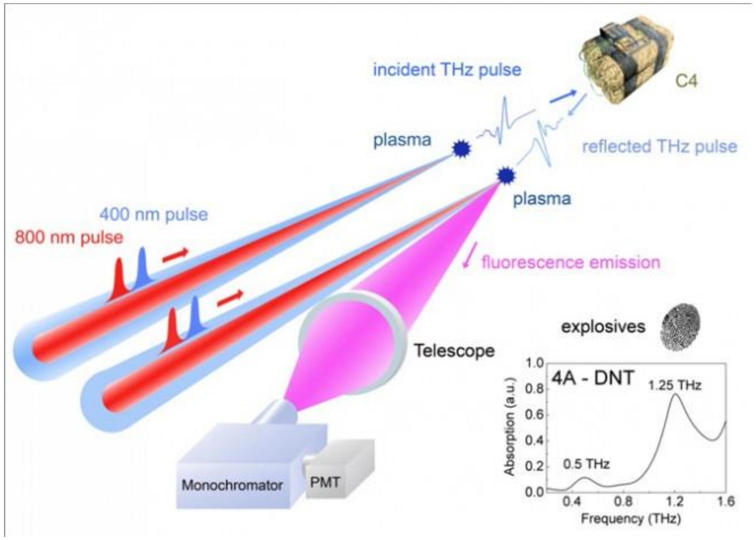


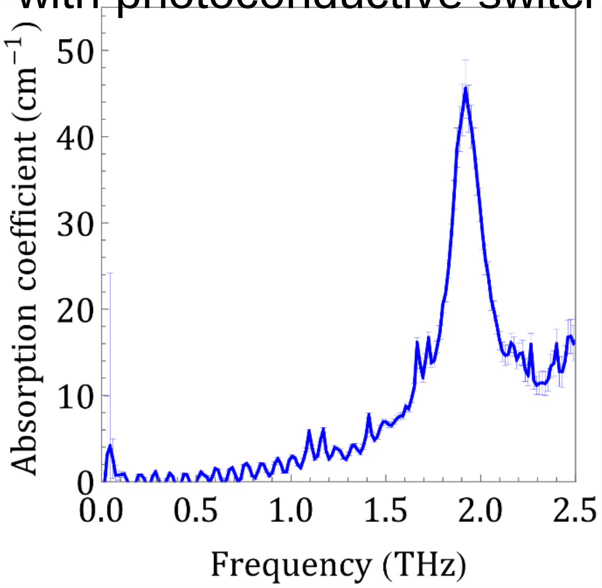
Figure adapted from www.wired.com

Air plasma based THz stand-off detection has been proposed in the US and has been considered as „breakthrough“ in THz remote detection.

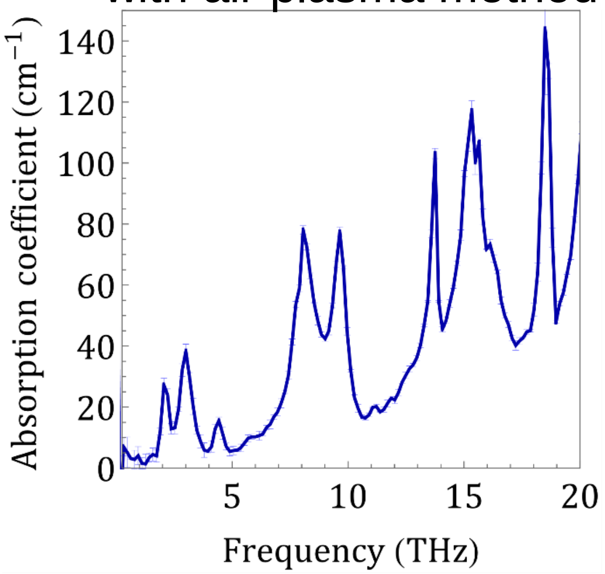
This approach overcomes the hurdles of transmission of Thz radiation over long distances through air and reduced identification performance due to limited bandwidth.

Traditional THz TDS vs. ultrabroadband THz TDS

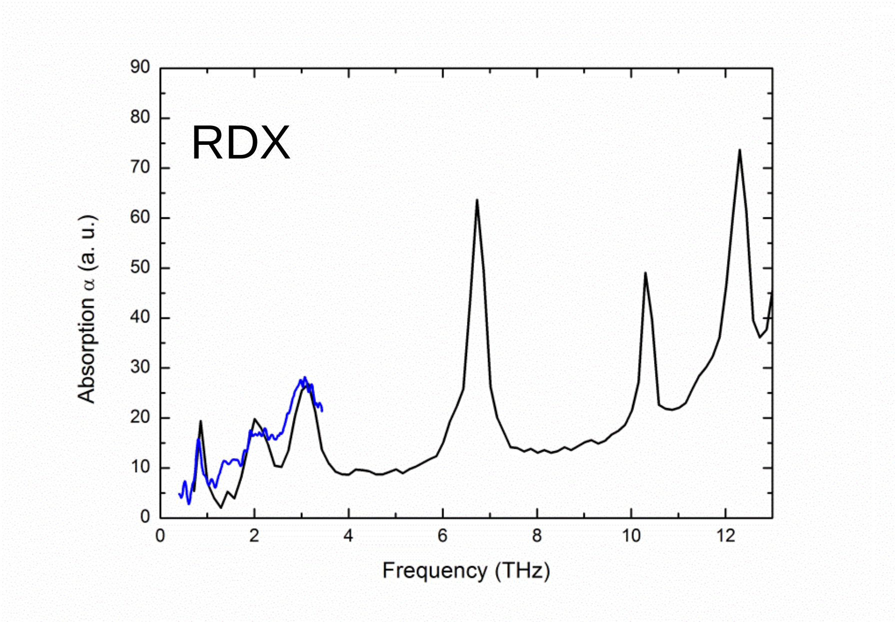
THz-TDS spectrum measured with photoconductive switches

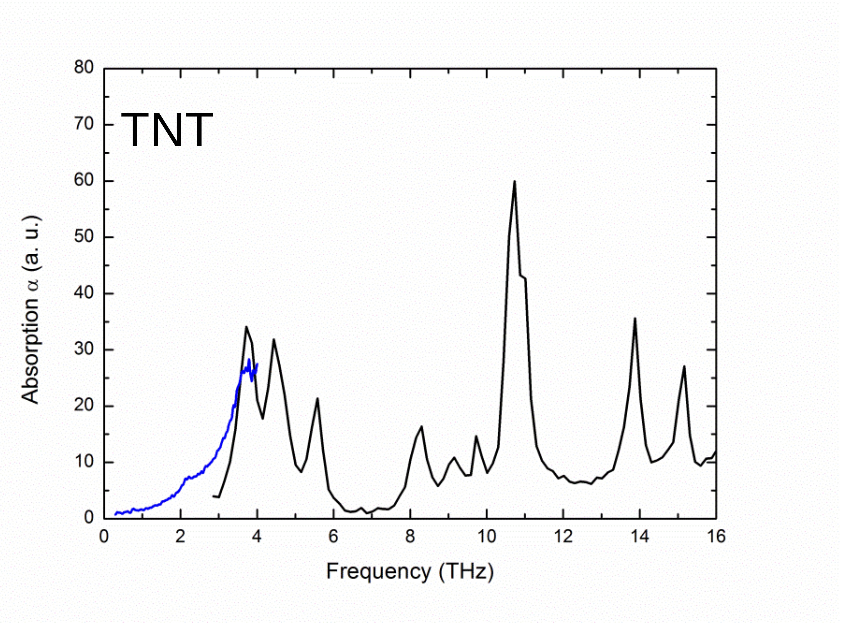


THz-TDS spectrum measured with air plasma method



Traditional THz TDS vs. ultrabroadband THz TDS





THz-TDS spectrum measured with photoconductive switches

THz-TDS spectrum measured with air plasma method

Stand-off detection of explosives

Goal: Study of the stand-off generation of THz radiation using air plasmas

Funded by: DGA, ASTRID

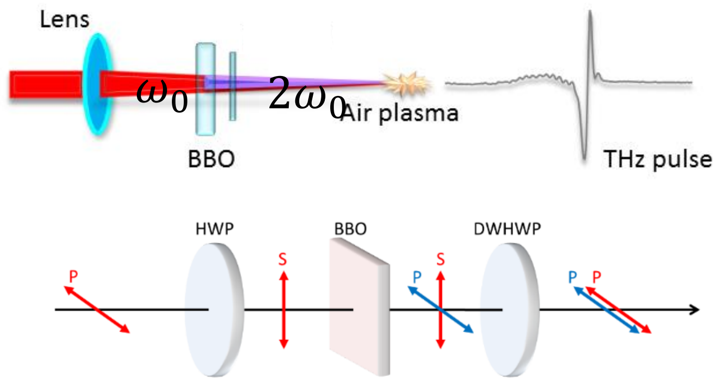
In cooperation with: CEA (de la recherche à l'industrie), CELIA, DTU Technical University of Denmark, Université de Bordeaux, CNRS

ALTESSE
Stand-off detection of explosives

- **Joint theoretical & experimental project**
- **Currently: Plasma generation by 2-color excitation using 400 nm/800 nm**
- **Laser parameters Spectra-Physics Spitfire Pro XP:**
  - Wavelength: 800nm
  - Repetition rate: 1 kHz
  - Pulse duration: 35 fs
  - Pulse energy: 3.5 mJ
  - Average power: 3.2 W
  - → safety issues

- **Objective: Plasma generation using eye-safe lasers**
- **Advantage: THz generation efficiency scales with $\lambda^{4.6}$**
- **Evaluation of methods to increase THz yield/bandwidth**
  - Change from focused geometry to filamentation
  - Multi-color excitation
- **Setting-up of a spectral database for the extended spectral range (ca. 40 different materials, explosives and simulants)**
- **Ab-initio density functional theory (DFT) and molecular dynamics (MD) simulations to get a deeper insight to the physical nature of the observed spectral features**

Two-color plasma THz generation



Tianwu Wang et al., J. Infrared Millim. Terahertz Waves 37, 592–604 (2016)

Asymmetric photocurrent model

Asymmetric field + plasma

$$E_L(t) = E_1\cos(\omega t + \phi) + E_2\cos[2(\omega t + \phi) + \theta]$$

Drift velocity

$$v_d = eE_1 \sin\frac{\phi}{m_e\omega} + eE_2\frac{\sin(2\phi+\theta)}{2m_e\omega}$$

Current

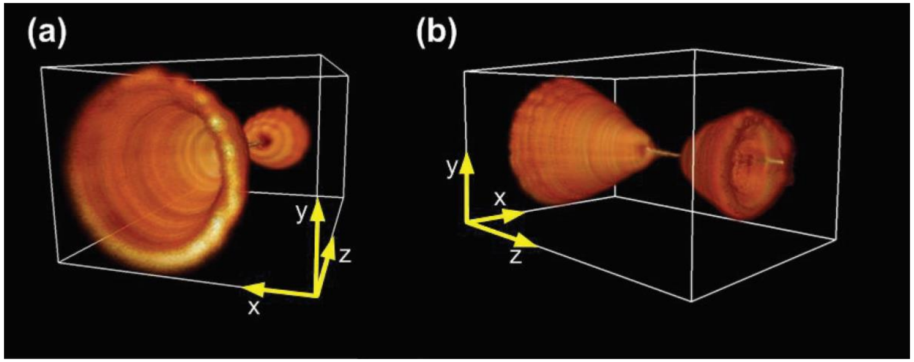
$$J_\perp(t) = \int_{t_0}^{t} e\, v_e(t,t')N_e(t')dt'$$

THz emission

$$\boldsymbol{E_{THz} \propto \frac{\partial J_\perp(t)}{\partial t}}$$

K.-Y. Kim, Optics Express, vol. 15, pp. 4577–4584 (2007)

Beam Shape

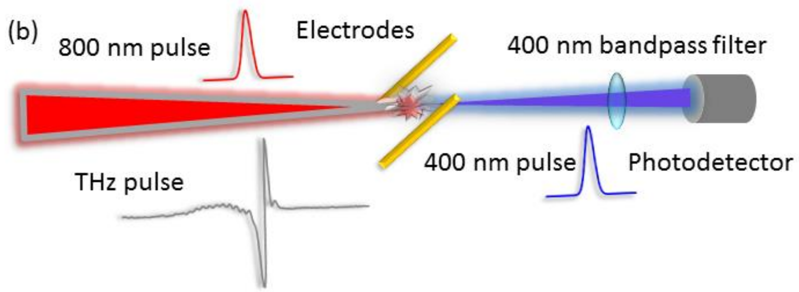


Pernille Karlskov et al., New Journal of Physics 15 (7), 075012 (2013)

- Due to the elongated generation region, the THz emission profile from the plasma is conical (Bessel-Gauss beam)

- When focused, the beam profile collapses to a tight central spot

Air Biased Coherent Detection



Generation of second harmonic by four-wave mixing

$$E_{2\omega} \propto \chi^{(3)} E_\omega E_\omega E_{THz}$$

By adding varying strong bias field

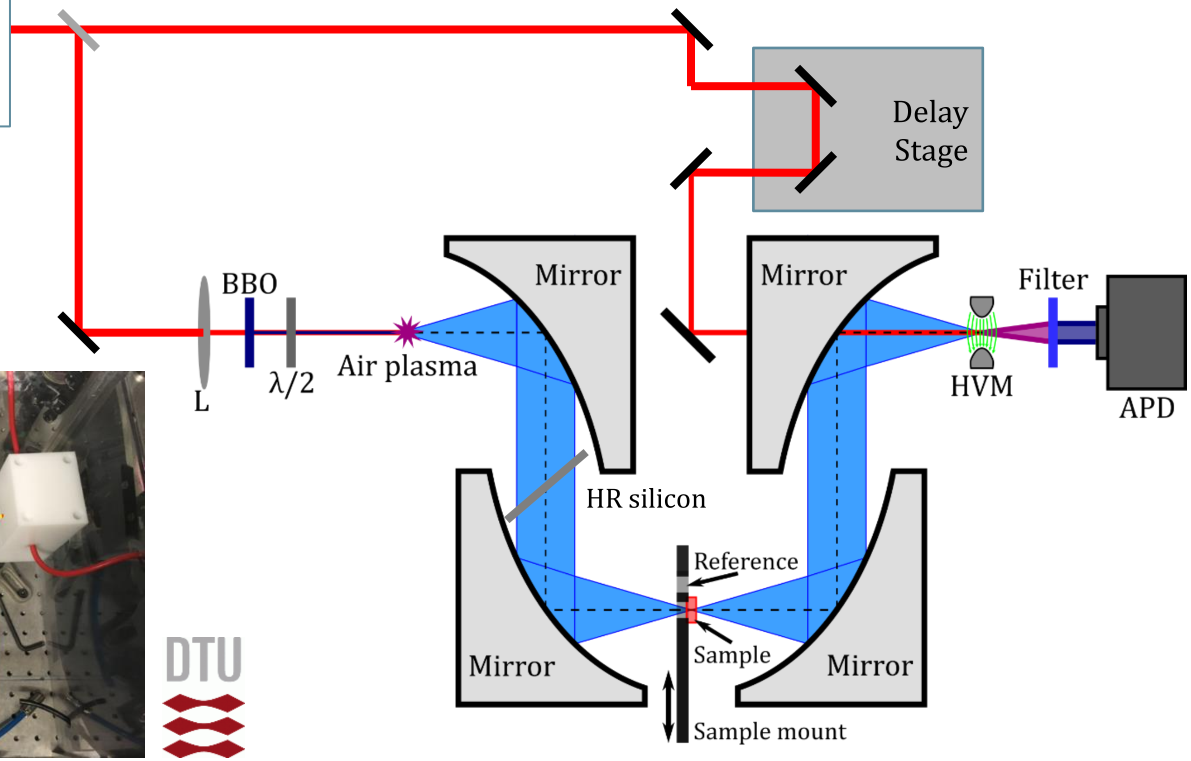
$$E_{2\omega} \propto \chi^{(3)} E_\omega E_\omega (E_{THz} \pm E_{DC})$$

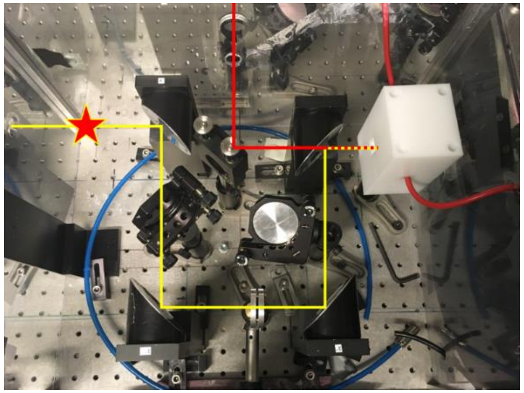
THz signal is picked up by lock-in amplifier

$$I_{2\omega} \propto |E_{2\omega}|^2 \propto \left(\chi^{(3)} I_\omega\right)^2 2E_{THz}E_{DC}$$

Ultra-broadband Terahertz Spectroscopy Setup

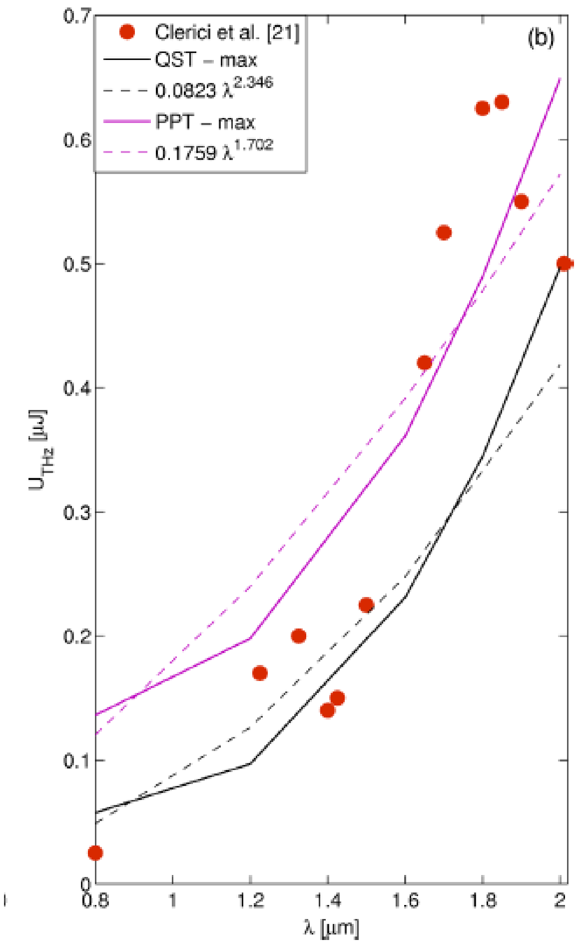
Spitfire XP (3.5 mJ)
Wavelength: 800 nm
Pulse Duration: 35 fs
Repetition Rate: 1 kHz





DTU

Investigation of deendance of driving wavelength and generated THz field



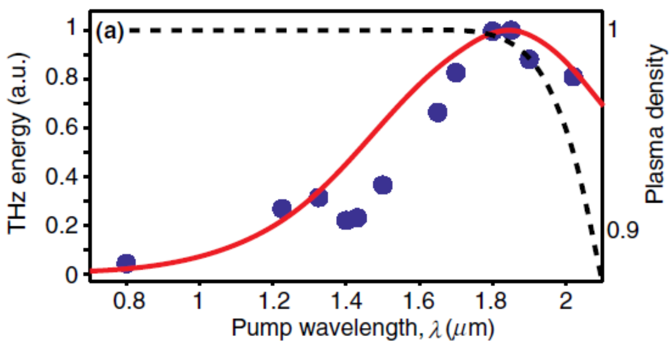
The THz yield is strongly dependent on the driving **field wavelength**

Example with:

- 2-color field
- Gaussian beams
- O2 and N2 molecules
- Maximum THz energy for ν < 80 THz

Calculations performed:

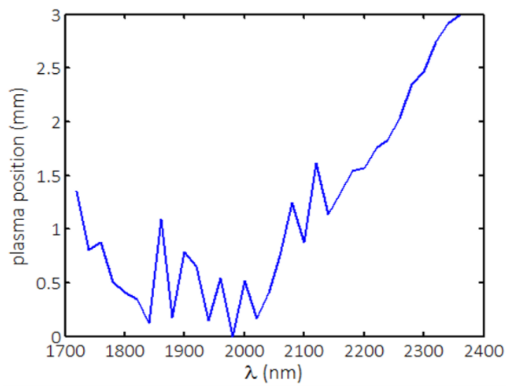
- QST (quasi-static tunneling rate)
- PPT (Perelomov, Popov and Terent'ev ionization rate) [6]

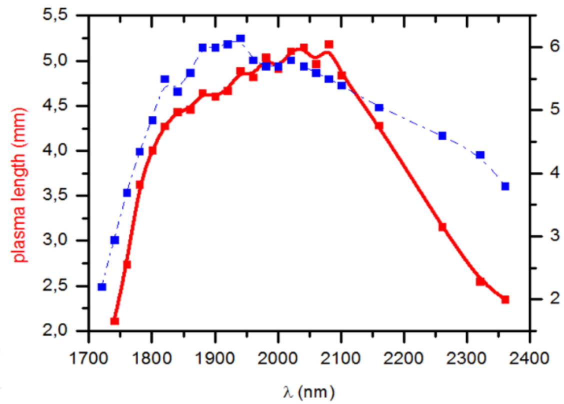


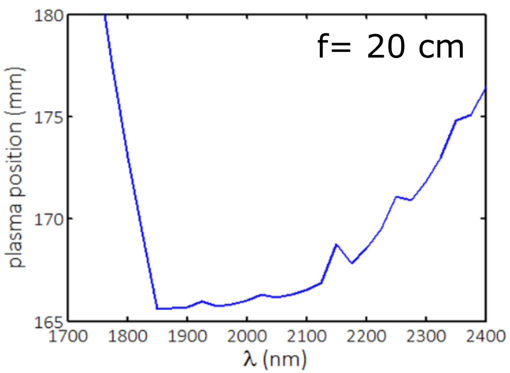
Drop in energy corresponds to plasma density decreasing after 1.8 µm (black dotted curve)

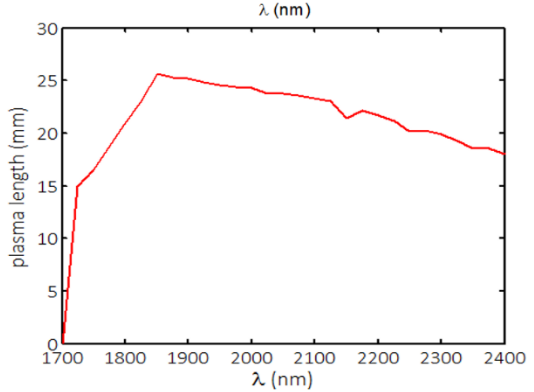
Plasma characterization

$P_{laser} \approx 6GW << P_{crit} \left(\approx 60\,GW\right)$: **multiphoton ionization (MPI) regime**







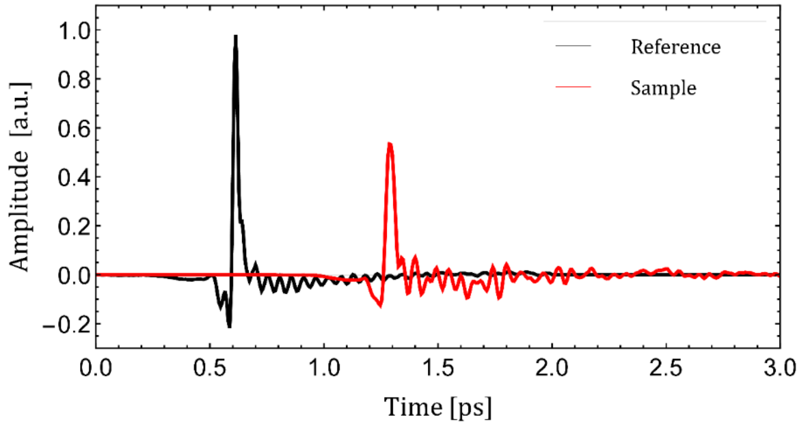


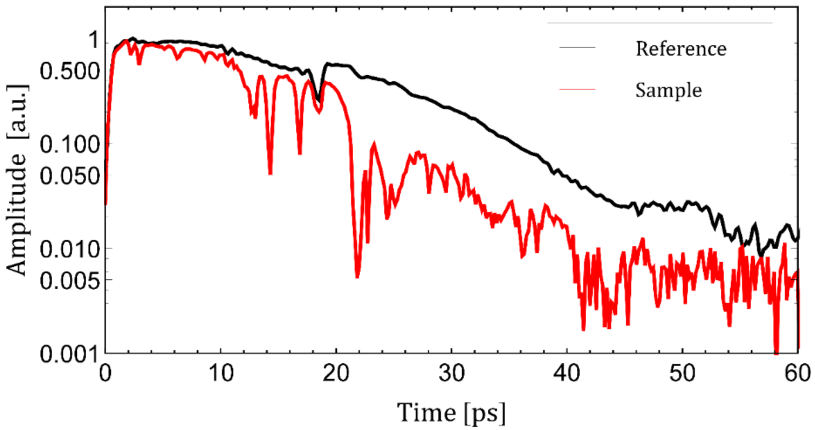
Model :
two-scale variational method (pulse duration and transverse radius), suitable for MPI regime

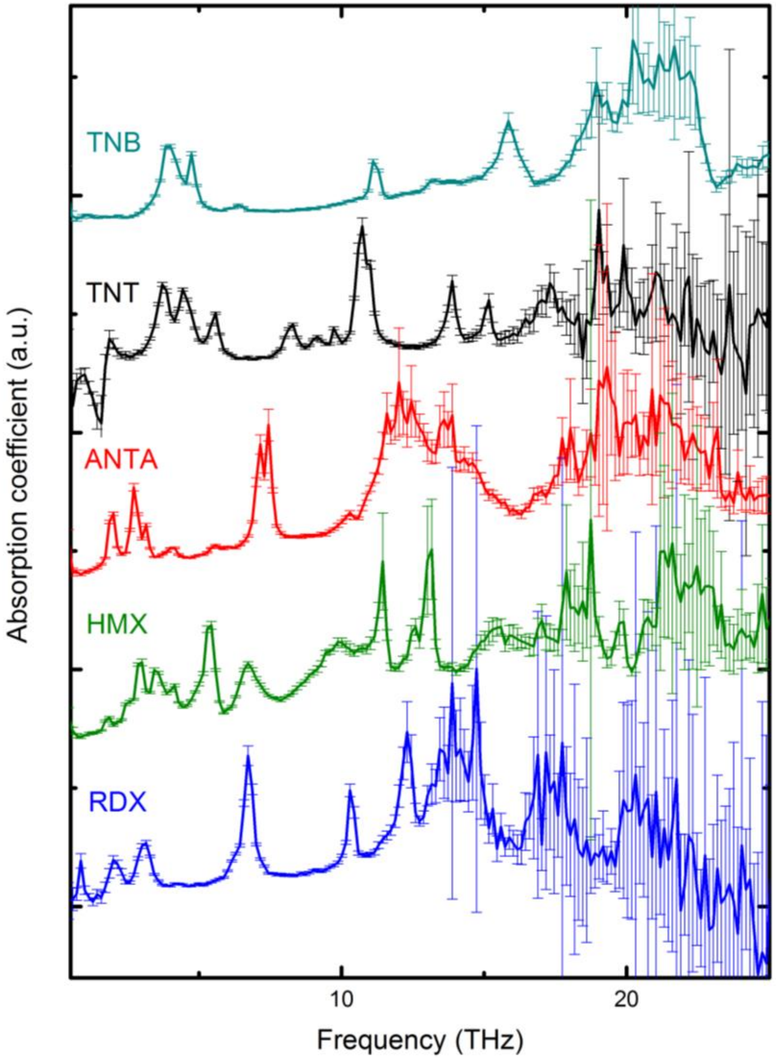
Experiment :
the plasma length approximatelly follows the energy variation

Simulations :
qualitative accordance with the plasma experimental behavior. The calculations are performed for gaussian pulses and differing from actual laser beam mode.

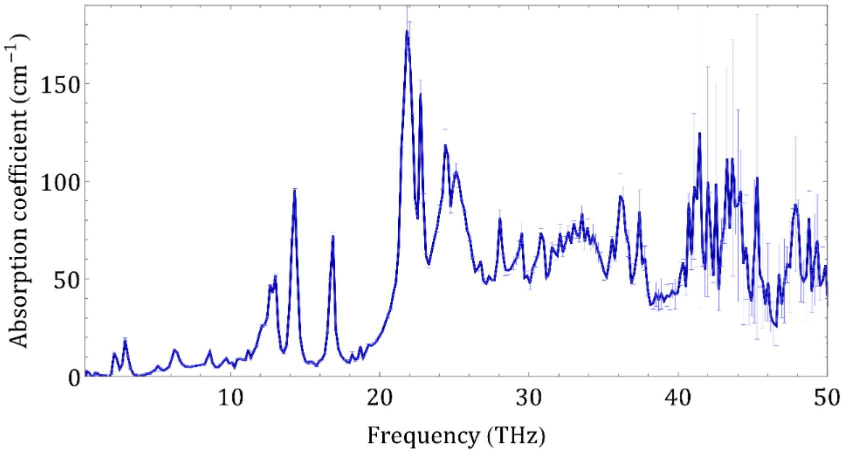
Ultra-broadband THz spectroscopy results

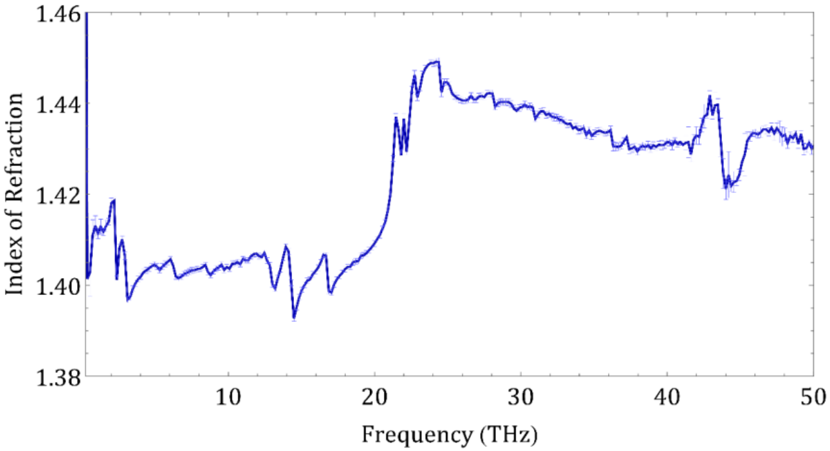






Data Evaluation





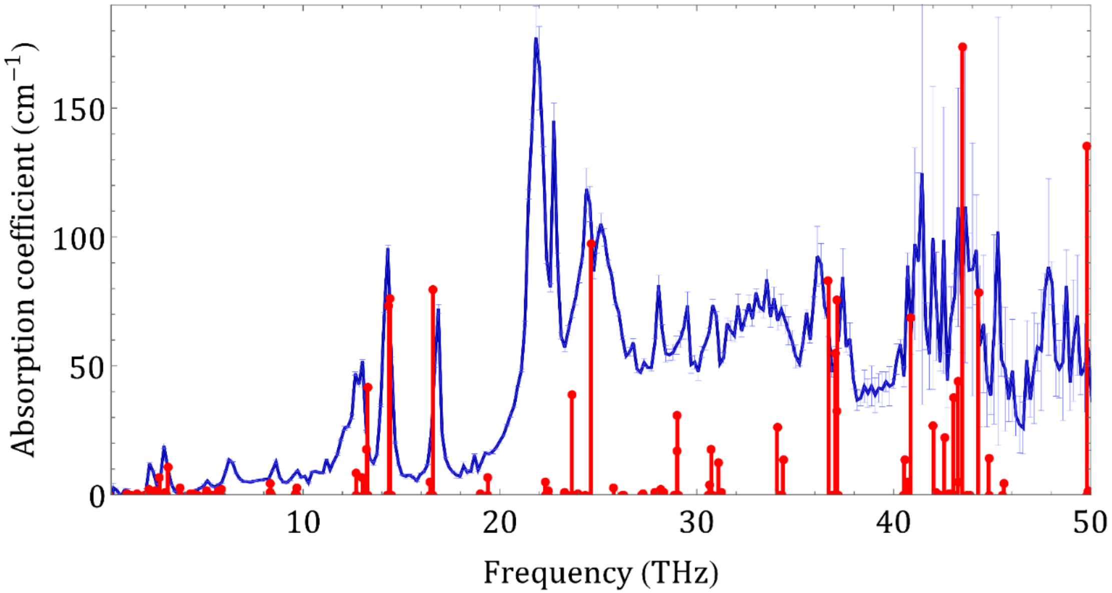
$$\frac{E_{sam}(\omega)}{E_{ref}(\omega)} = Ae^{i\phi(\omega)} = Te^{-\frac{\alpha d}{2}}e^{\frac{i(n-1)\omega d}{c}}$$

$$n(\omega) = 1 + \frac{\phi c}{\omega d}$$

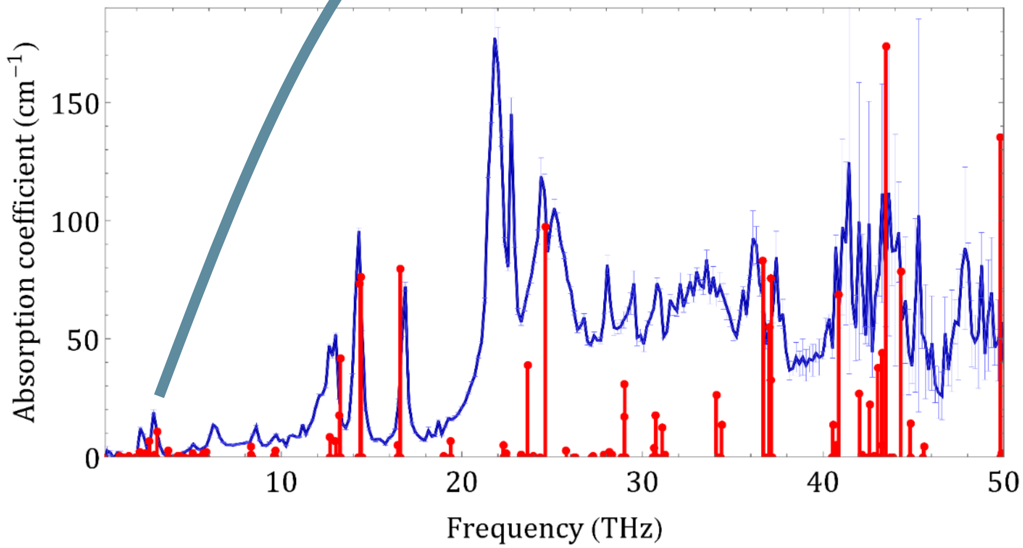
$$\alpha(\omega) = -\frac{2}{d}\ln\left(\frac{(n+1)^2}{4n}\cdot\frac{|E_{sam}(\omega)|}{|E_{ref}(\omega)|}\right)$$

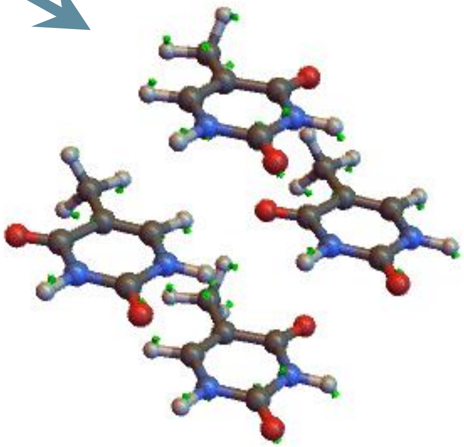
Simulations

- **Castep: ab-initio DFT and MC simulation**
  - Geometry Optimization
  - Phonon and Energy
  - Molecular Dynamics (time dependet)



Simulations





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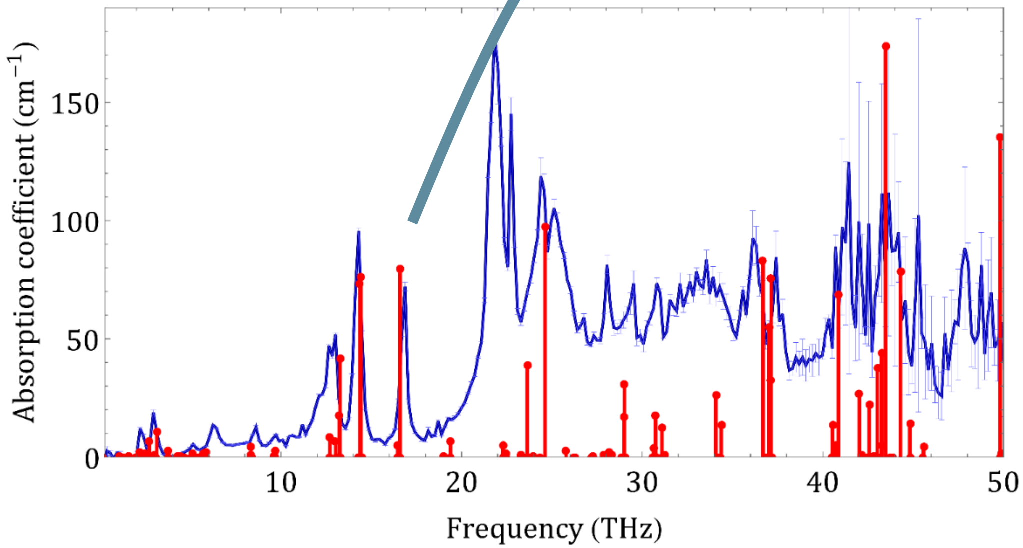

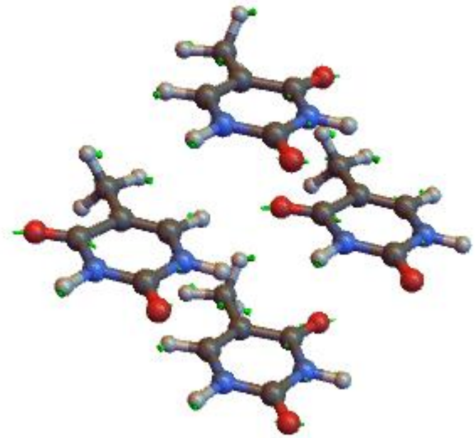

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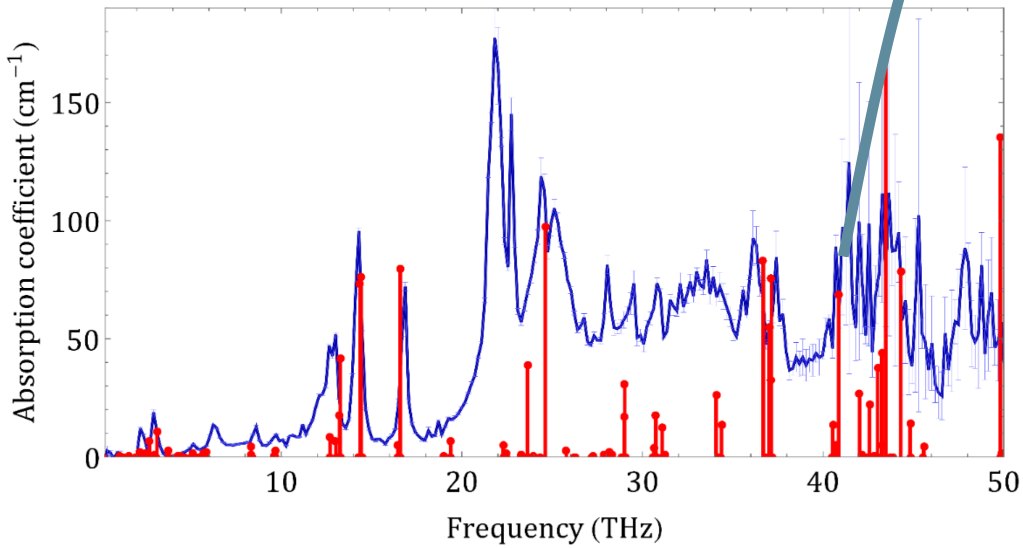
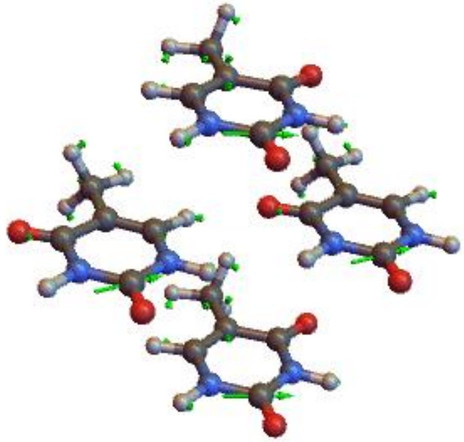
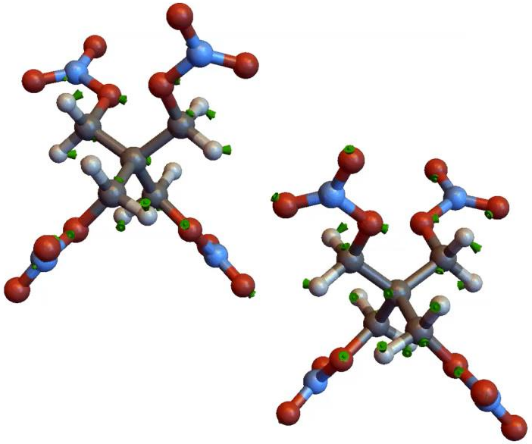
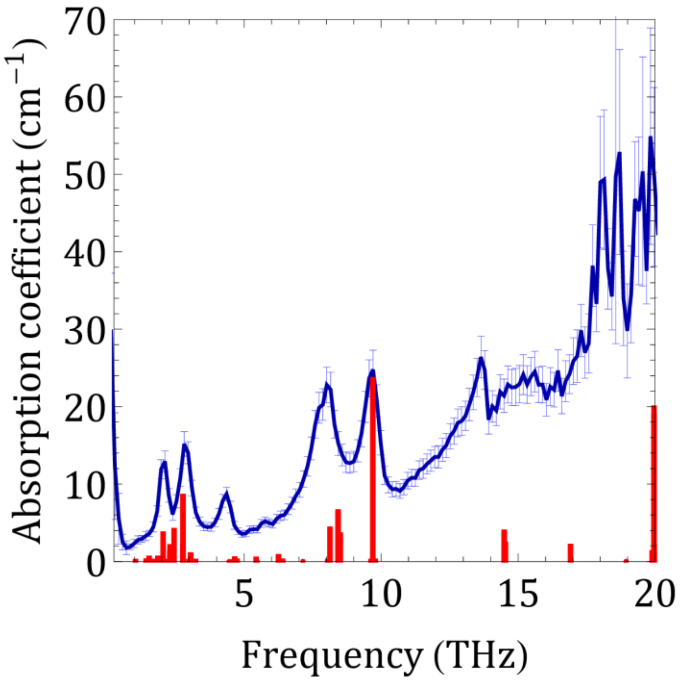




PETN





Absorption coefficient (cm^{-1})
70
60
50
40
30
20
10
0
5
10
15
20
Frequency (THz)

Conclusion & Outlook

- **Air plasma based THz is a promising method for realization of stand-off detection of explosives**
- **Allowing for unambiguous identification of materials**
- **Comprehensive knowledge about the ABCD system, its components construction and operation has been aquired**
- **We have set up a spectral database of over 40 materials in the ultrabroadband frequency band (0.5-50.0 THz)**
- **We are able to adequately simulate molecular crystalline materials of medium size (ca. 60 molecules per unit cell) to further study the nature of the observed spectral features**

- **Currently we are investigating the wavelength dependence of the THz yield at DTU in cooperation with CELIA using pump wavelength of up to 2.6 µm**
- **Amplified laser system has just arrived at ISL**
- **Air plasma based THz system for remote generation of THz pulses will be installed within the next weeks at ISL**
- **We will now focus on the experimental investigation of this method for remote detection of explosives**

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