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SZENTAGOTHAI RESEARCH CENTRE

Tilted-pulse-front-pumped THz pulse sources for femtosecond material science

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HIGH-FIELD
THz LAB

FemtoMat2025, February 24-27, Mauterndorf, Austria

Outline

- Properties of THz pulses
- Classification of single-cycle THz pulses
- Most important generation schemes:
  - 1., Photoconductive switches (photoconductive antennae)
  - 2., Optical rectification (detection: electro-optic sampling (EOS)
  - 3., Two- color laser plasma
  - 4., Spintronic THz emitters
- Tilted-pulse-front-pumping set-up (past and future)
- Nonlinear THz spectroscopy
- THz driven electron gun
- Coulomb explosion as high energy ion source

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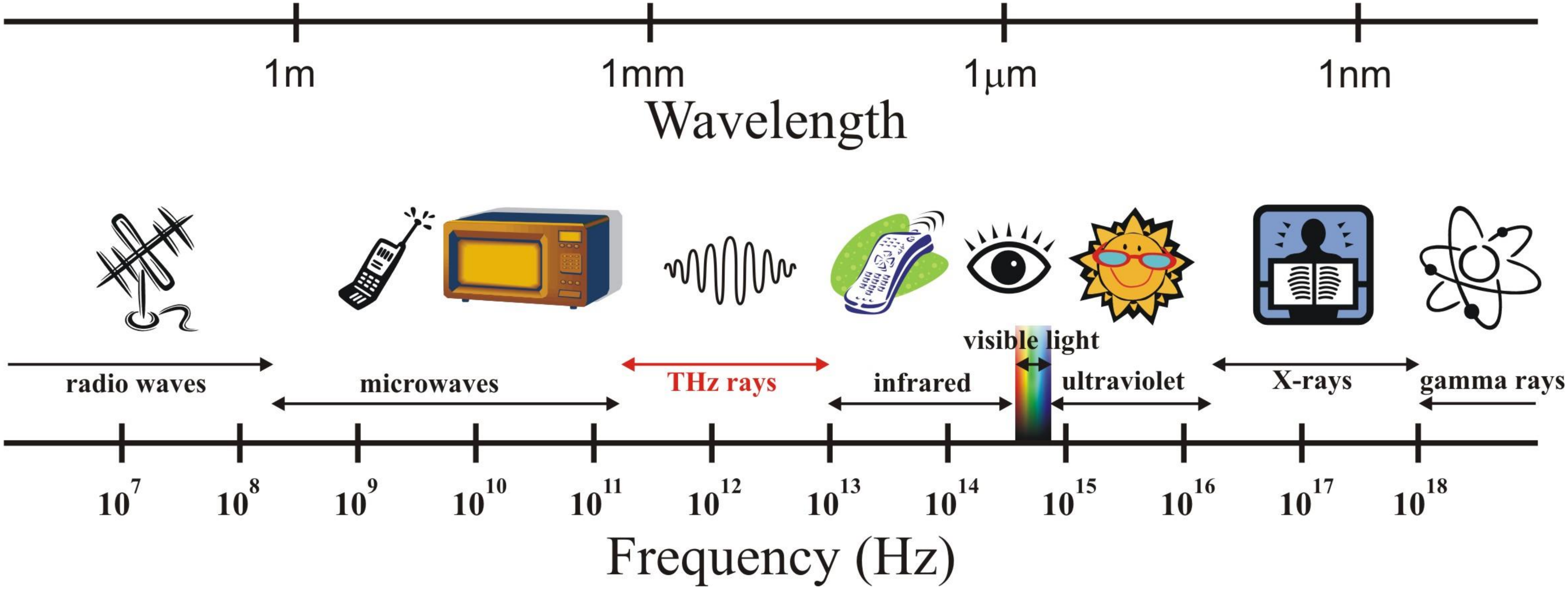
Terahertz radiation (T-ray) in the EM spectrum



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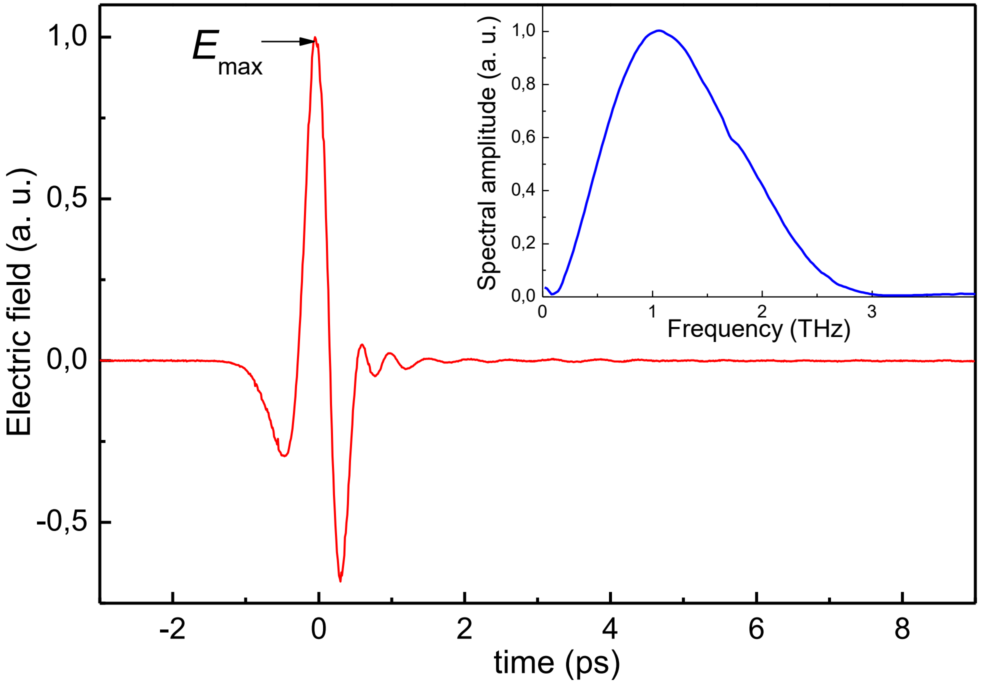
Correspondence of 1 THz frequency

- 1 THz ÷ 4.15 meV photon energy ($h\nu$)
- 1 THz ÷ 33.3 cm^{-1} wave-number
- 1 THz ÷ 300 µm wavelength (λ)
- 1 THz ÷ 1 ps temporal period (T)
- 1 THz ÷ 48.1 K temperature ($T=h\nu/k_B$)

Classification of THz pulses by peak electric field and energy

Single-cycle THz sources are based on fs lasers



OR in LN

- Low field THz pulses (E_{max} ≈ 100 V/cm → 10 fJ energy)

  Linear THz spectroscopy (TDTS)

- High field THz pulses (E_{max} ≈ 100 kV/cm → 1 μJ energy)

  THz pump – probe measurement, nonlinear THz optics, mater.- excitation, control

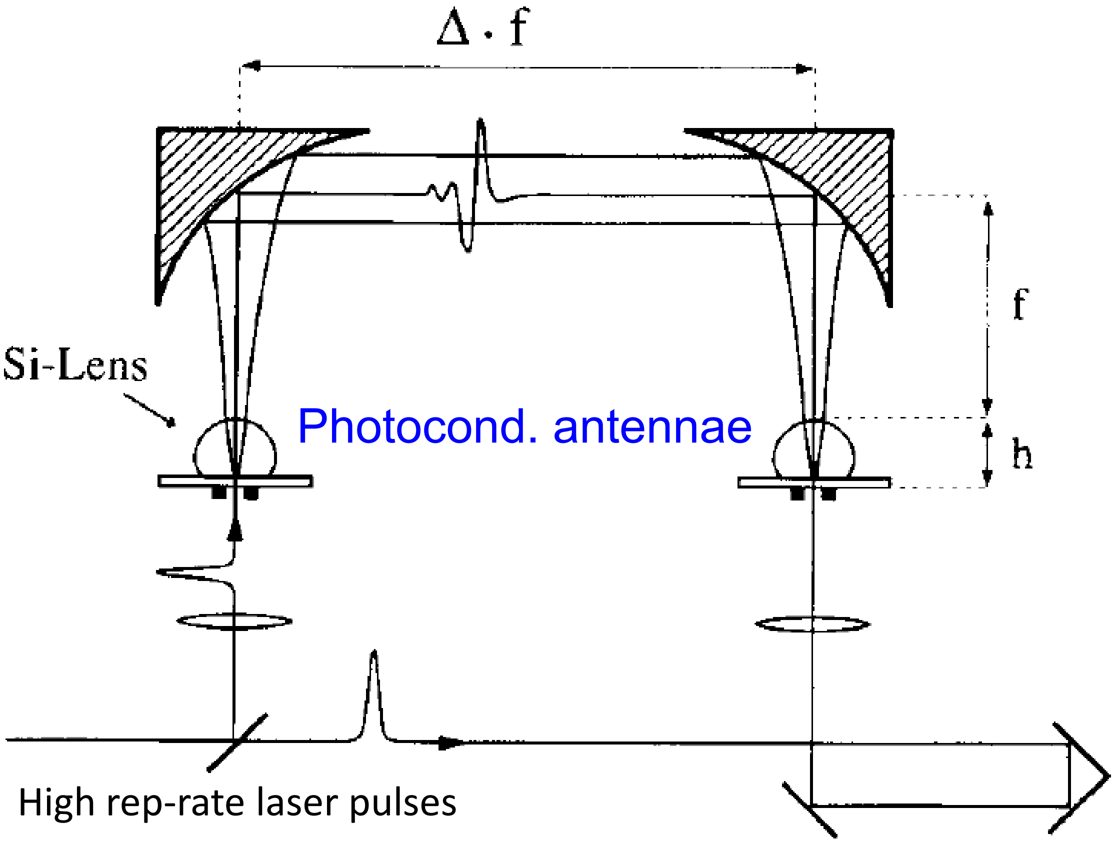
- Extreme high field THz pulses (E_{max} ≈ 100 MV/cm → 10 mJ energy)

  enhancement of HHG, particle acceleration (manipulation), material processing

Photoconductive switches

Time domain THz spectrometer (TDTS) is usually based on photoconductive switches (photoconductive antennae)

Set-up for transmission measurement



Photocond. antennae

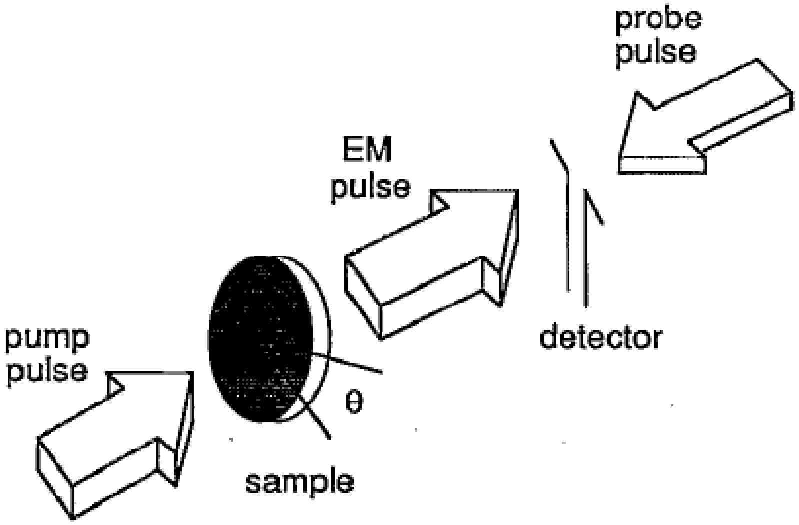
High rep-rate laser pulses

The sample is placed between the two off-axis parabolic mirrors

$W_{THz} \approx 10$ fJ

P. U. Jepsen: JOSA B **13**, 2424 (1996)

THz pulse generation by optical rectification (OR) of ultrashort laser pulses in nonlinear optical crystals (NOC)



NOC: GaAs, DAST, $LiNbO_3$, ZnTe

X.-C. Zhang et al., Appl. Phys. Lett. **61**, 3080 (1992)
A. Rice et al., Appl. Phys. Lett. **64**, 1324 (1994)

Velocity matched: $v^{gr}_{pump} = v^{phase}_{THz}$
$n^{gr}_{pump} = n^{phase}_{THz}$

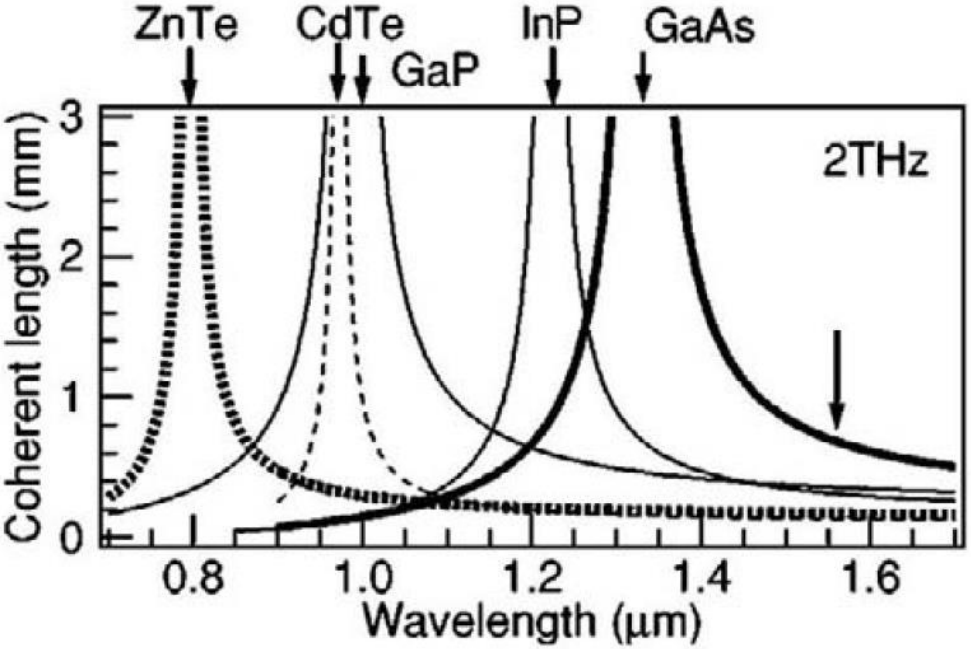
Extensively used: Ti:sapphire laser (820 nm) pulse OR in ZnTe
phase-matched (velocity matched)

Velocity matching by tilted-pulse-front pumping in $LiNbO_3$ (LN)
Much higher THz generation efficiency

Optical rectification

Velocity matching

Coherent length: $l_c = \frac{\pi}{\Delta k} = \frac{c}{2\nu_{THz}\left|n_p^{gr} - n_{THz}\right|}$



	ZnTe	CdTe	GaP	InP	GaAs
λ_p (µm)	0.8	0.97	1.0	1.22	1.35

Velocity matched pump wavelengths for 2 THz generation

M. Nagaia et al., Appl. Phys. Lett. **85**, 3974 (2004)

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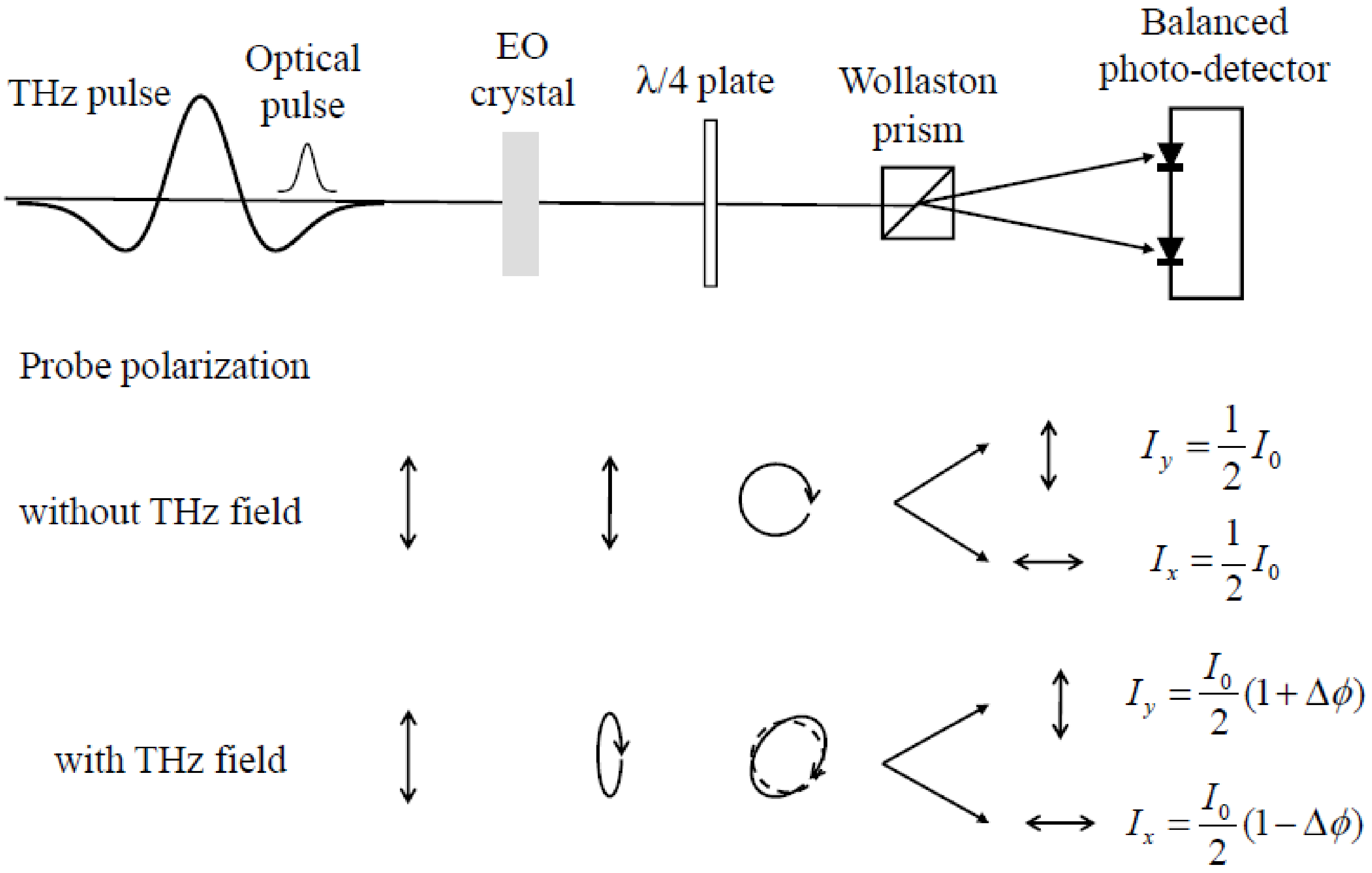
Electro-optic sampling

Using EO effect for measuring THz pulse shape

The quasi-static field of THz pulse can induce birefrigence through the Pockels effect



Lee Y.-S.: Principles of THz Sci. & Tech., Springer 2009

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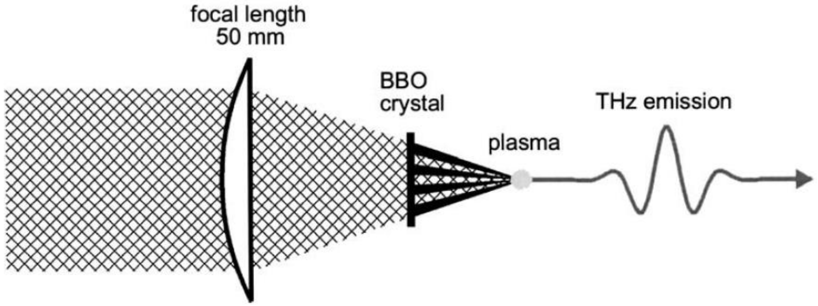
THz pulse generation by two-color laser plasma

Bartel T. et al., Opt. Lett. **30**, 2805 (2005)



Cook D. J., Hochstrasser R. M.: Opt. Lett. **25**, 1210 (2000)

Origin of THz radiation: four-wave rectification, special form of four-wave mixing:

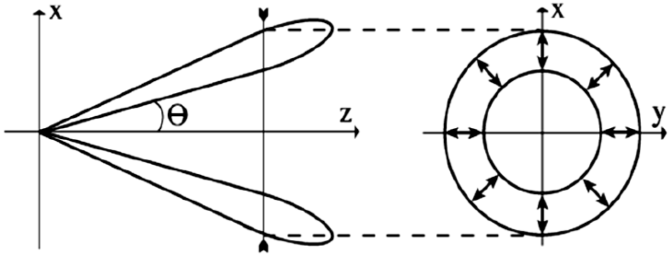
$$P(\omega_{THz}) = \varepsilon_0 \chi^{(3)}(\omega_{THz}, 2\omega - \omega_{THz}, -\omega, -\omega) E(2\omega - \omega_{THz}) E^*(\omega) E^*(\omega)$$

Kim K. Y. et al., Opt. Express **15**, 4577 (2007)

Origin of THz radiation: asymmetric current generated by the two-color electric field

Kouloukl idis A.D. et al., Nat. Commun. **11**, 292 (2020)

Advantage of long wavelength pumping: 185 μJ THz energy, 100 MV/cm at 3.9 μm



The conical emission is usually inconveninet

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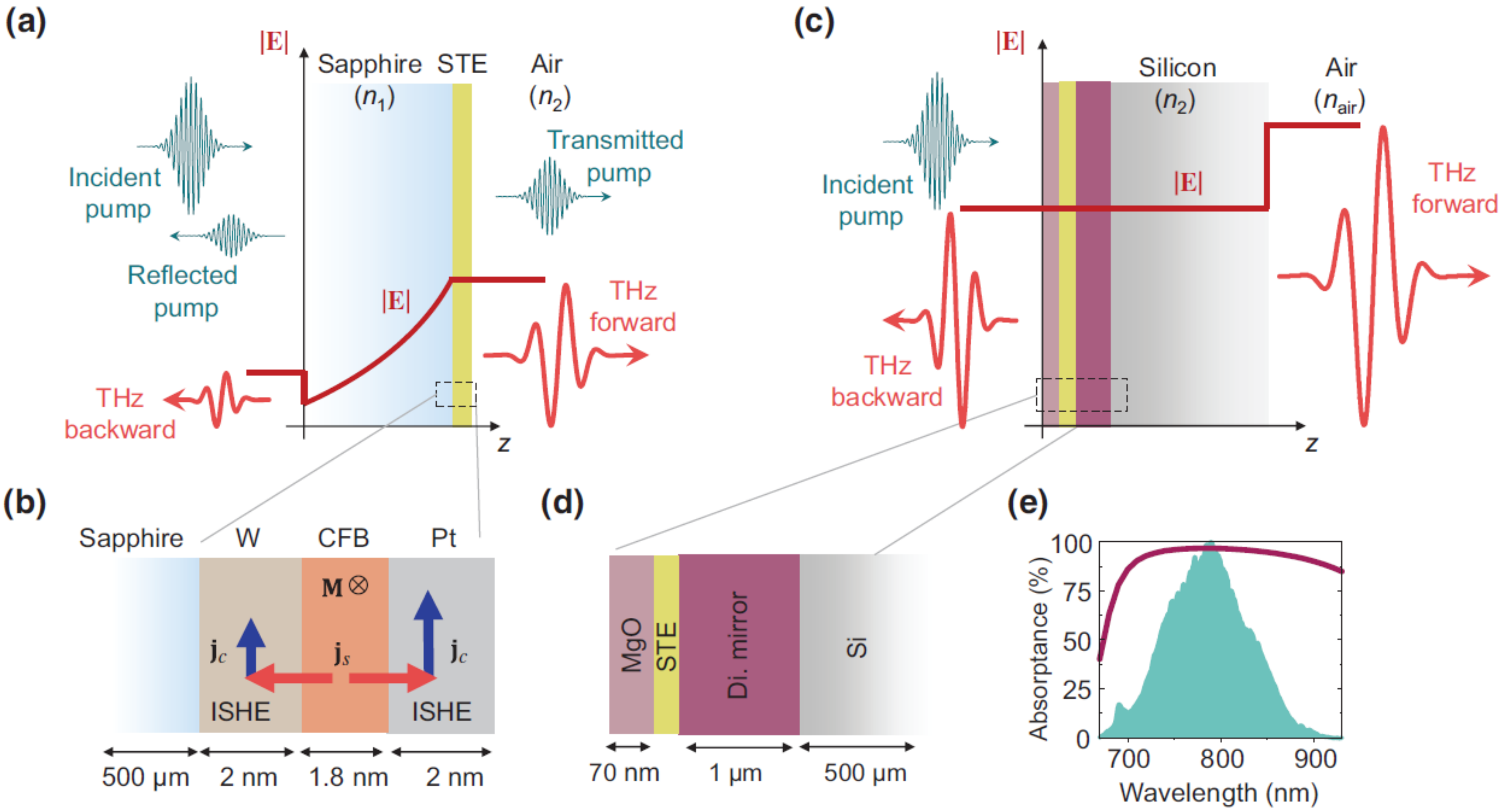
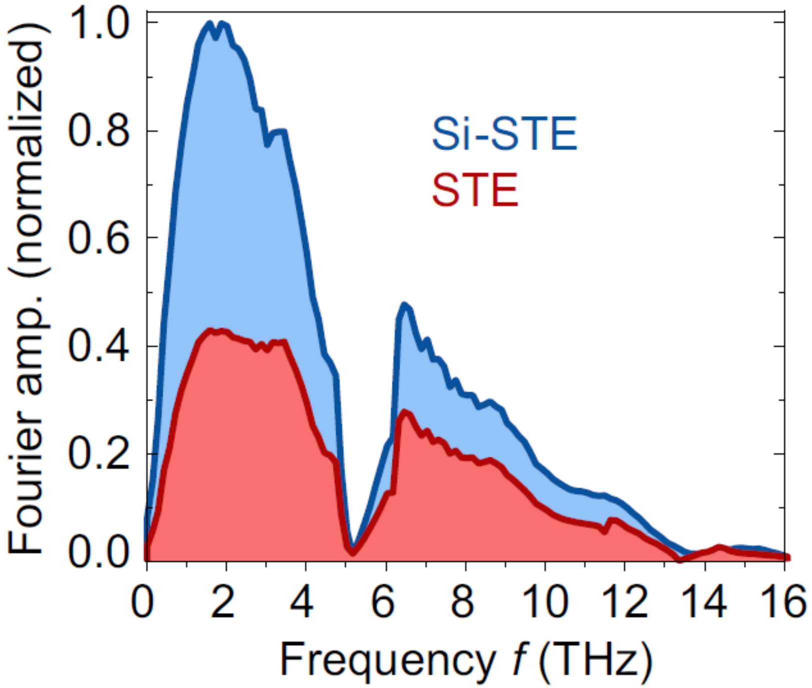
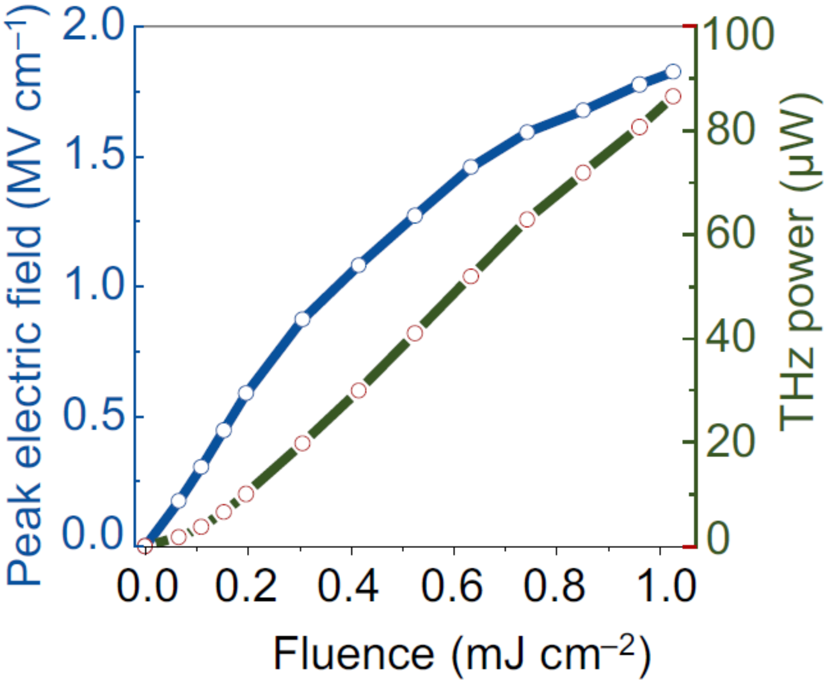
Spintronic THz emitter (STE)



Phys. Rev. Appl. **19**, 034018 (2023) Tobias Kampfrath, FUB

Spintronic THz emitters





Phys. Rev. Appl. **20**, 034037 (2023) Tobias Kampfrath, FUB

Zeeman-torque sampling

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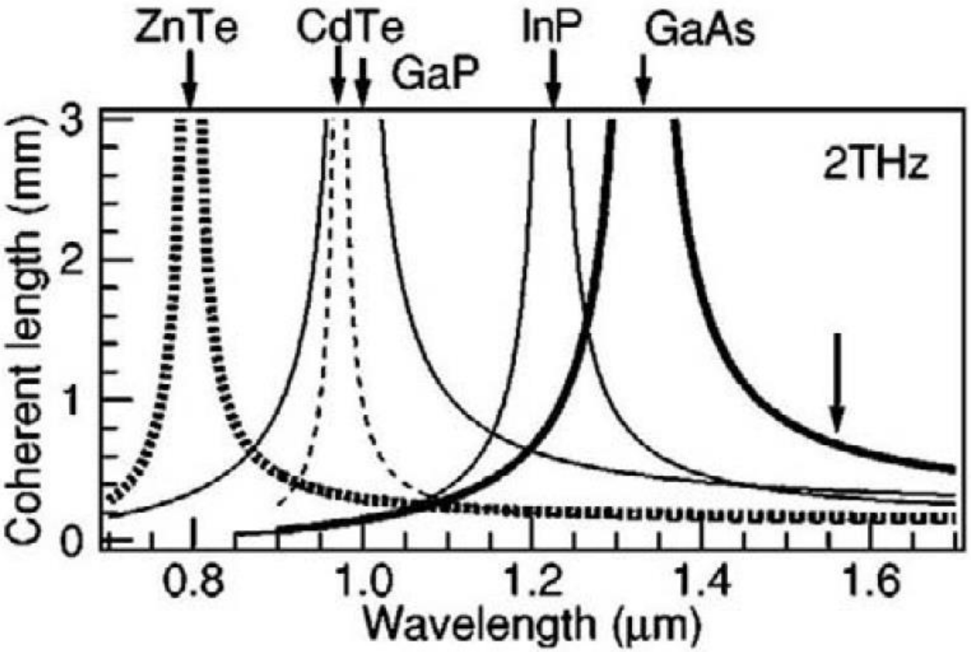
Optical rectification

Collinear velocity matching

Coherent length: $$l_c = \frac{\pi}{\Delta k} = \frac{c}{2\nu_{THz}\left|n_p^{gr} - n_{THz}\right|}$$



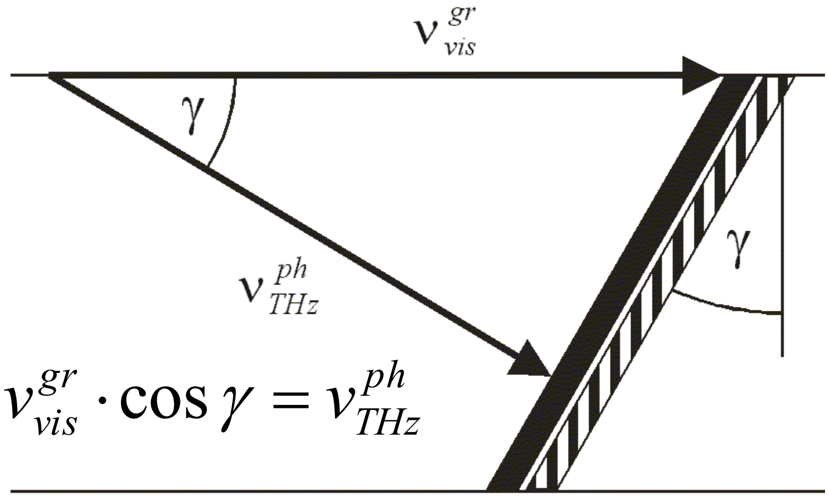
	ZnTe	CdTe	GaP	InP	GaAs
λ_p (µm)	0.8	0.97	1.0	1.22	1.35

High energy THz source:
ZnTe pumped by Ti:saphire laser

$W_{THz} \approx 1$ nJ ÷ 10 nJ

M. Nagaia et al., Appl. Phys. Lett. **85**, 3974 (2004)

Velocity matching by tilted-pulse-front pumping (TPFP) setup



$$v_{vis}^{gr} \cdot \cos\gamma = v_{THz}^{ph}$$

Material	ZnTe	GaP	GaAs	LN
d_{eff} [pm/V]	68.5	24.8	65.6	168

$n^{gr}_{pump} \approx n^{phase}_{THz}/2$ $\quad$ $v^{gr}_{pump} = 2 * v^{phase}_{THz}$

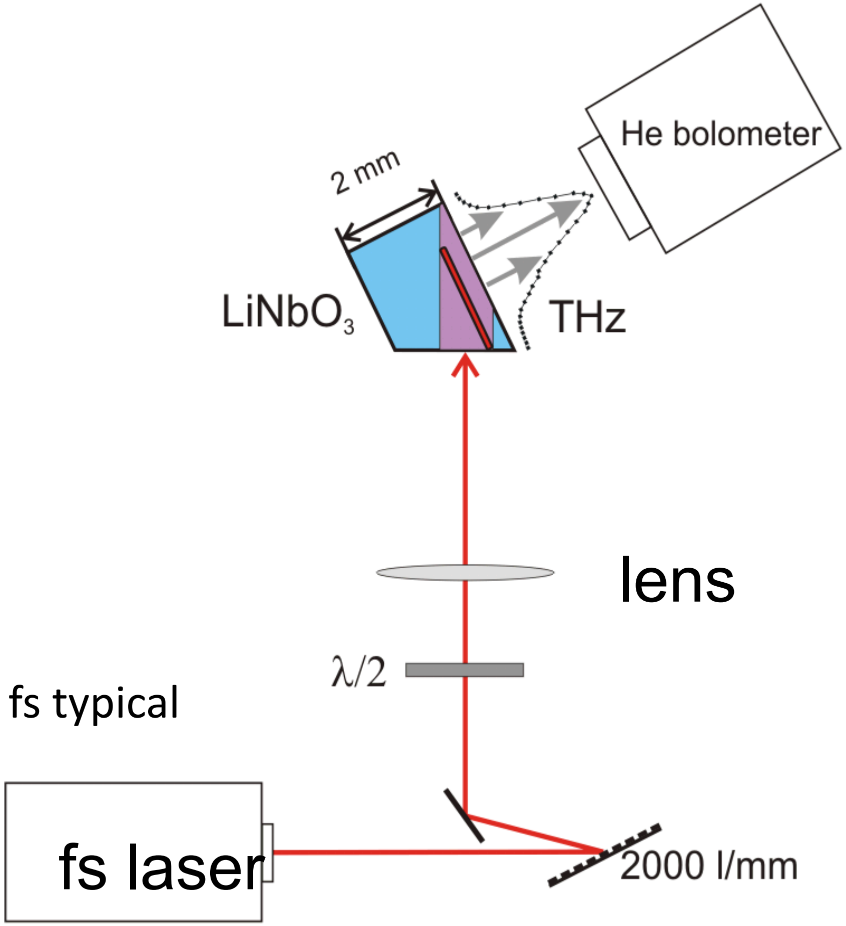
Hebling et al., Opt. Expr. **10**, 1161 (2002)

$$\tan\gamma = -\frac{n}{n_g}\lambda\frac{d\varepsilon}{d\lambda}$$

pulse front tilt

angular dispersion

~100 fs typical



Success of TPFP LiNbO3 THz sources

Material	ZnTe	GaP	GaAs	LN
d_{eff} [pm/V]	68.5	24.8	65.6	168

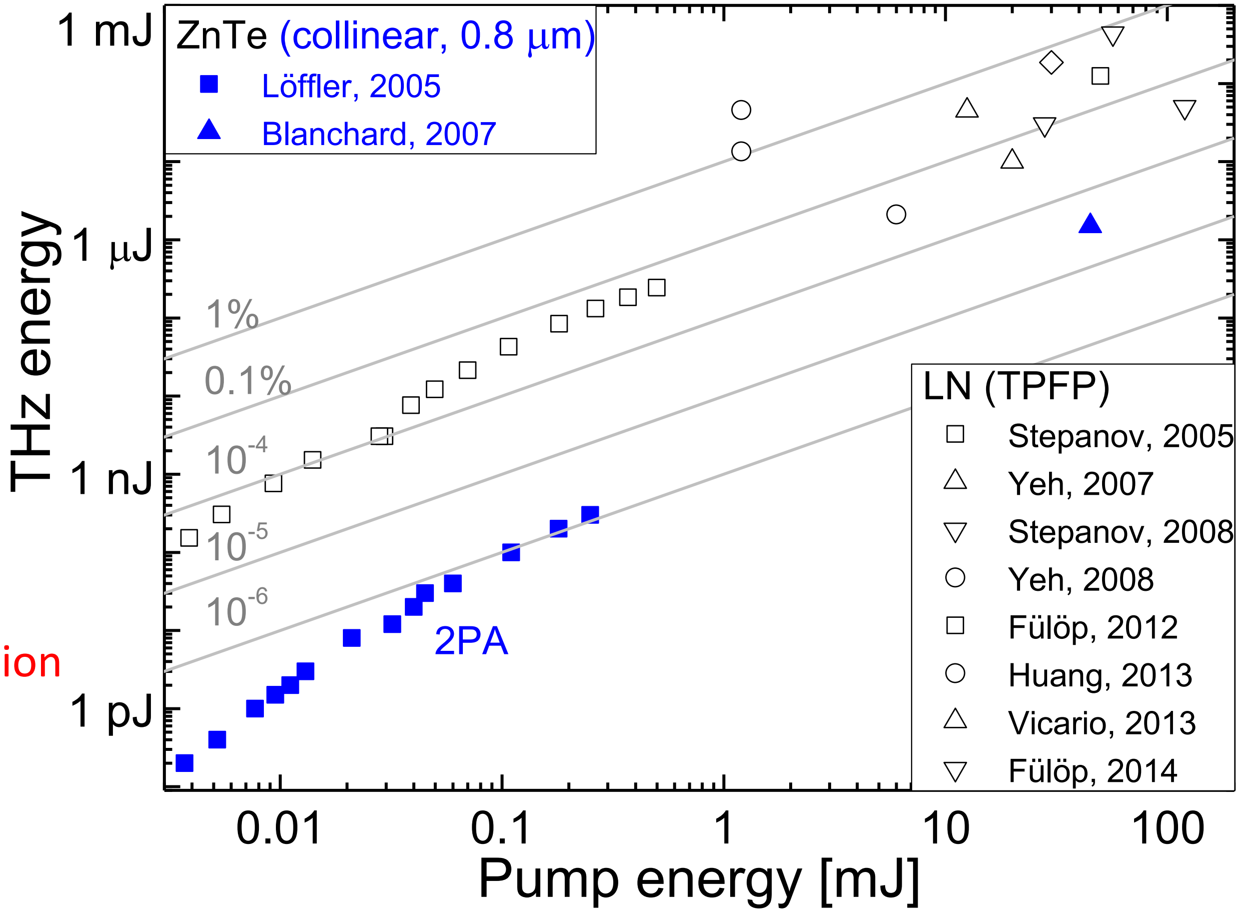
Advantages of LN:

LN has large nonlinear coefficient, large bandgap, → absent of low-order multi-photon absorption

LN is a very efficient THz pulse source on the 0.2 – 2 THz range

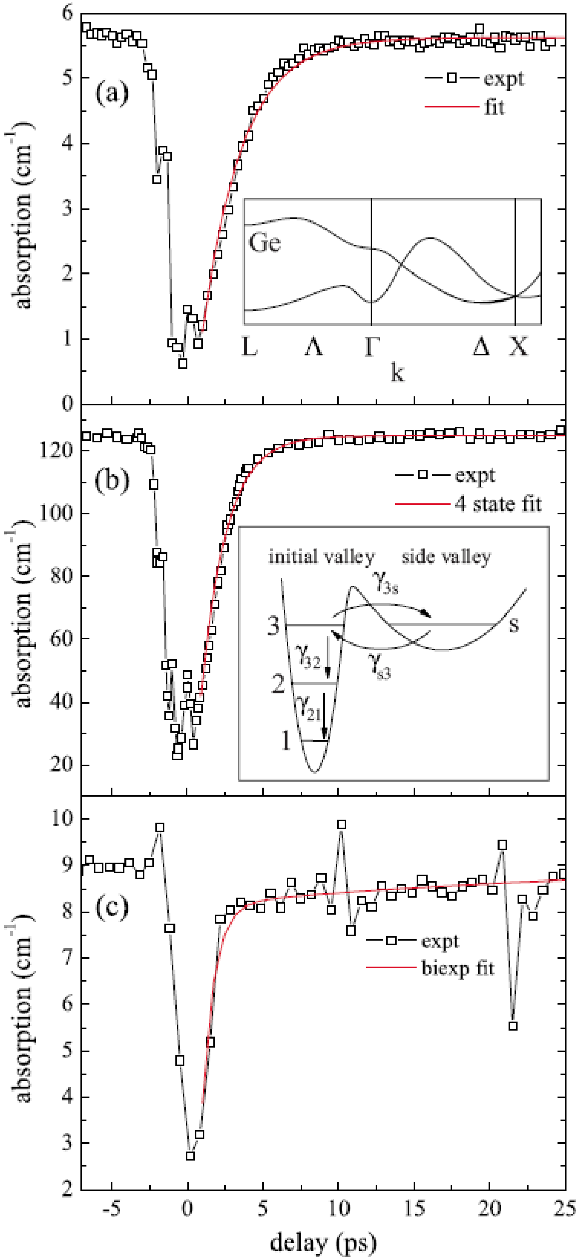
μJ level enough for pump in pump – probe and control experiments

Salen, P. et al. Matter manipulation with extreme terahertz light…. Phys. Rep. **836-837**, 1 (2019).



Following the carrier dynamics by THz pump – probe meas.

Transient bleaching of free carrier absorption



n-type Ge, $n_c = 5\times10^{14}$ cm^{-3}

$$\alpha_c(\omega)=\frac{\varepsilon_b\omega_p^2\gamma}{nc\left(\omega^2+\gamma^2\right)}=\frac{e^2N_c\gamma}{\varepsilon_0 ncm^*\left(\omega^2+\gamma^2\right)}=\frac{eN_c\gamma^2\mu}{\varepsilon_0 nc\left(\omega^2+\gamma^2\right)}$$

$$\alpha(t)\propto e^{-t/\tau_r}$$ τ_r = 2.7 ps

n-type GaAs, $n_c = 8 \times 10^{15}$ cm^{-3}

γ_{S3} = 2 ps^{-1}, γ_{3S} = 20 ps^{-1}, γ_{32} = 9 ps^{-1}

$$\alpha(t)\propto\sum_i \mu_i n_i(t)$$ τ_r = 1.9 ps

n-type Si, $n_c = 5 \times 10^{14}$ cm^{-3}

$$\alpha(t)\propto e^{-t/\tau_{r1}}+e^{-t/\tau_{r2}}$$ τ_{r1} = 0.8 ps, τ_{r2} = 24 ps

Hebling et al.: Phys. Rev. B **81**, 035201 (2010)

Free carrier absorption increase by impact ionization in InSb

InSb, 450 μm, Eg=0.24 eV at 77 K

Doped sample, N=2x10^{15} cm^{-3} at T=80 K

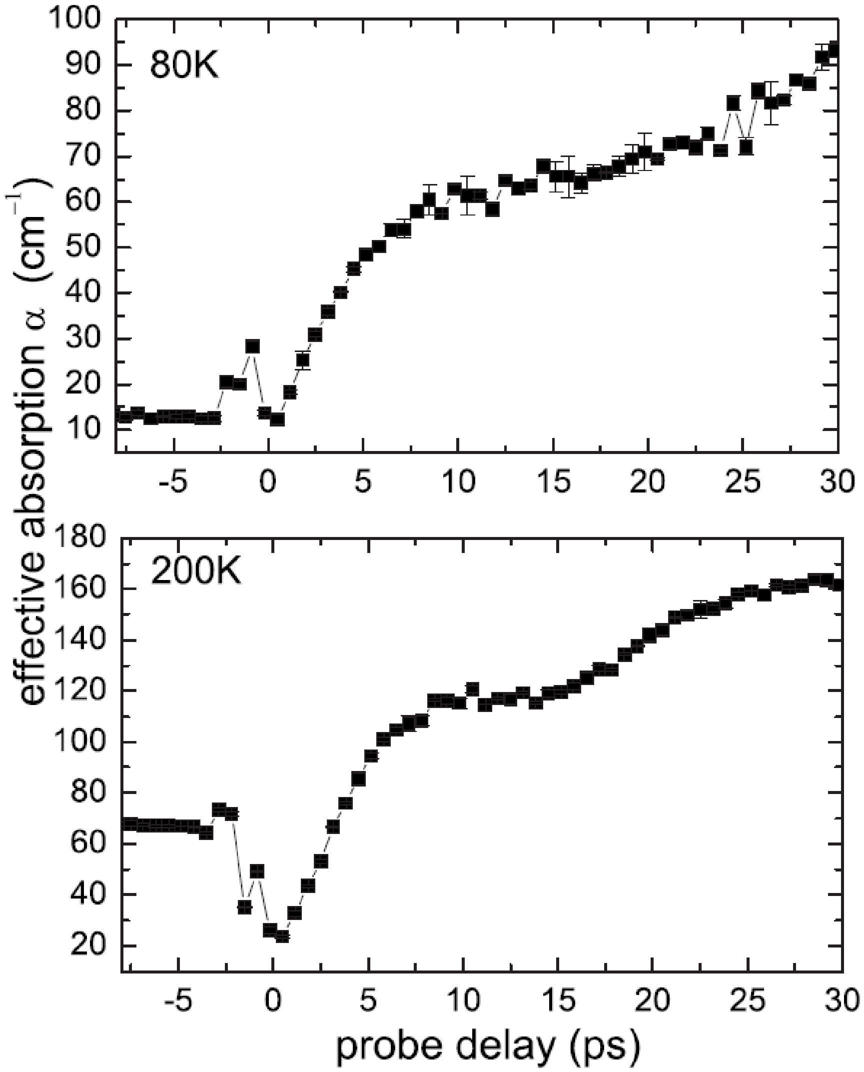
τ_m=2.5 ps > $T_{THz}/2$ →
accelerated electrons →
carrier multiplication by impact ionization

Absorption was calculated from the
0.2 – 1.6 THz part of the spectra

Absorption increase caused by impact ionization
For 200 K initial drop caused by nonparabolicity and scattering into side-valleys

$$\alpha_c(\omega) = \frac{\varepsilon_b \omega_p^2 \gamma}{nc(\omega^2 + \gamma^2)} = \frac{eN_c \gamma^2 \mu}{\varepsilon_0 nc(\omega^2 + \gamma^2)}$$

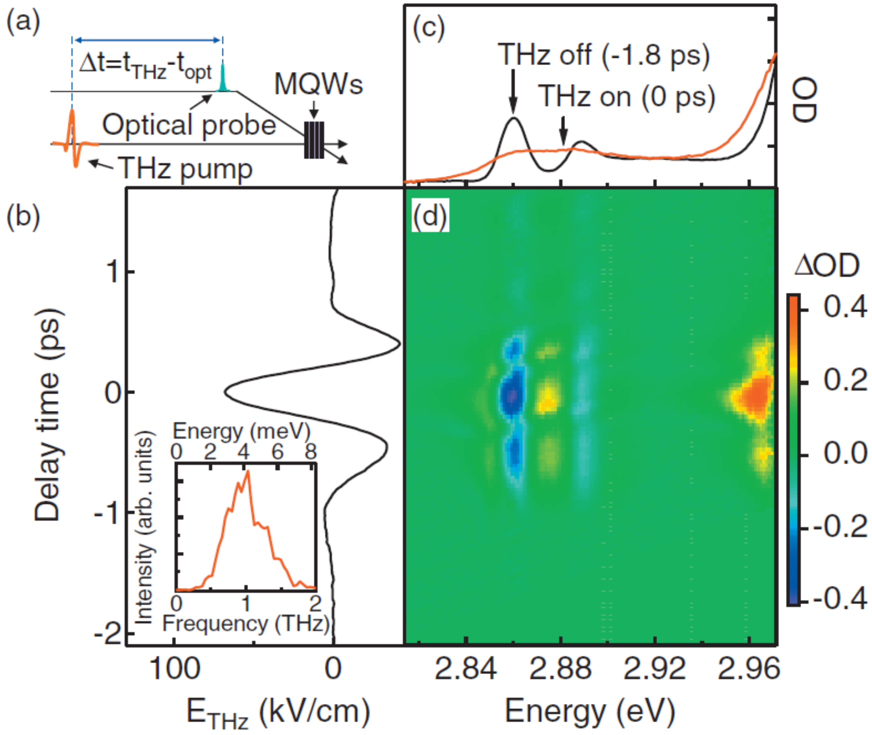
At 80 K eightfold increase in the absorption →
→ in the carrier concentration

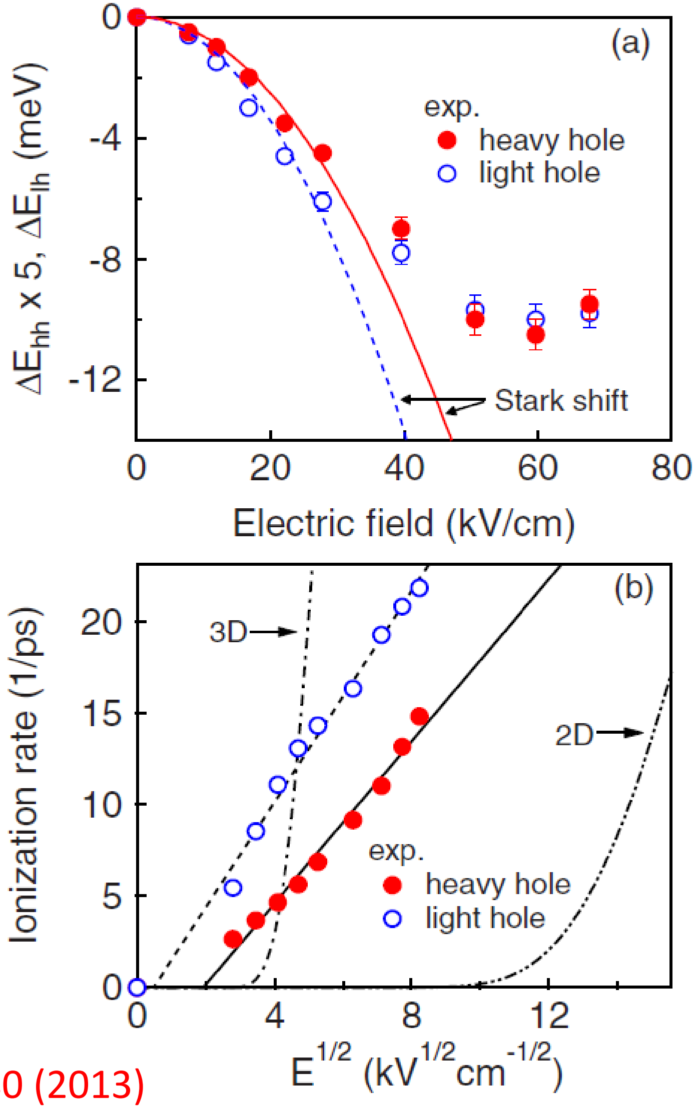


Hoffmann et al.: Phys. Rev. B **79**, 161201R (2009)

Other application examples of high-field TPFP LN THz sources

Hirori et al.: Phys. Rev. B **81**, 081305R (2010)





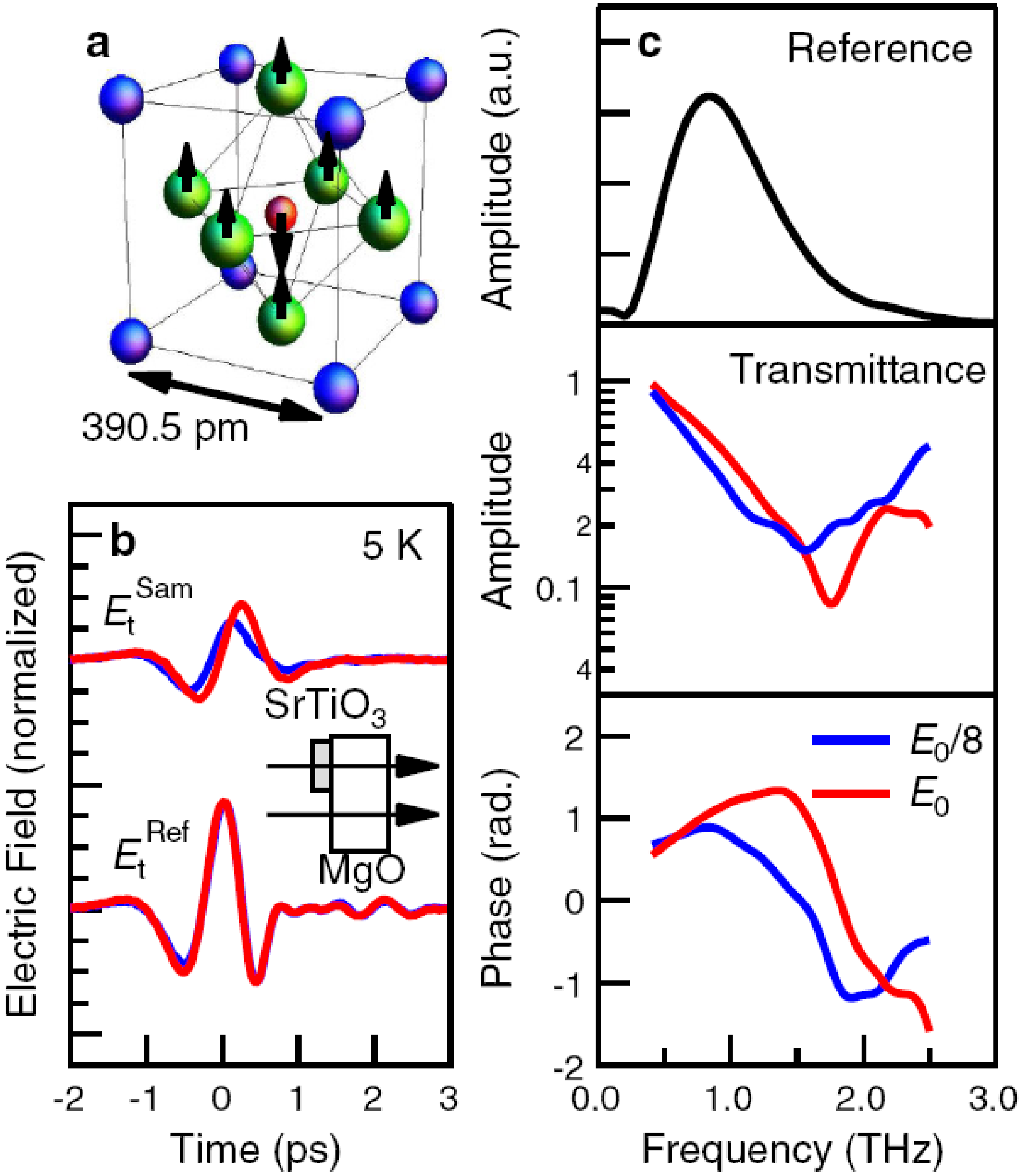
Review article:

T. Kampfrath, K. Tanaka, K. A. Nelson: Nat. Photonics **7**, 680 (2013)

D. Nocoletti, and A. Cavalleri: Adv. in Opt. and Photon. **8**, 401 (2016)

Coherent control of soft mode

Lattice anharmonicity



SrTiO$_3$ is a ferroelectric crystal soft mode at 1.5 THz

300-nm-thick SrTiO$_3$ on 0.5 mm MgO

Tilted-pulse-front pumped THz source
$E_{THz,max}$=80 kV/cm

Increasing E_{THz} increasing amplitude, decreasing period

$$t(\omega) = E_t^{Sam}(\omega) / E_t^{Ref}(\omega)$$

I. Katayama et al., Phys. Rev. Lett. **108**, 097401 (2012)

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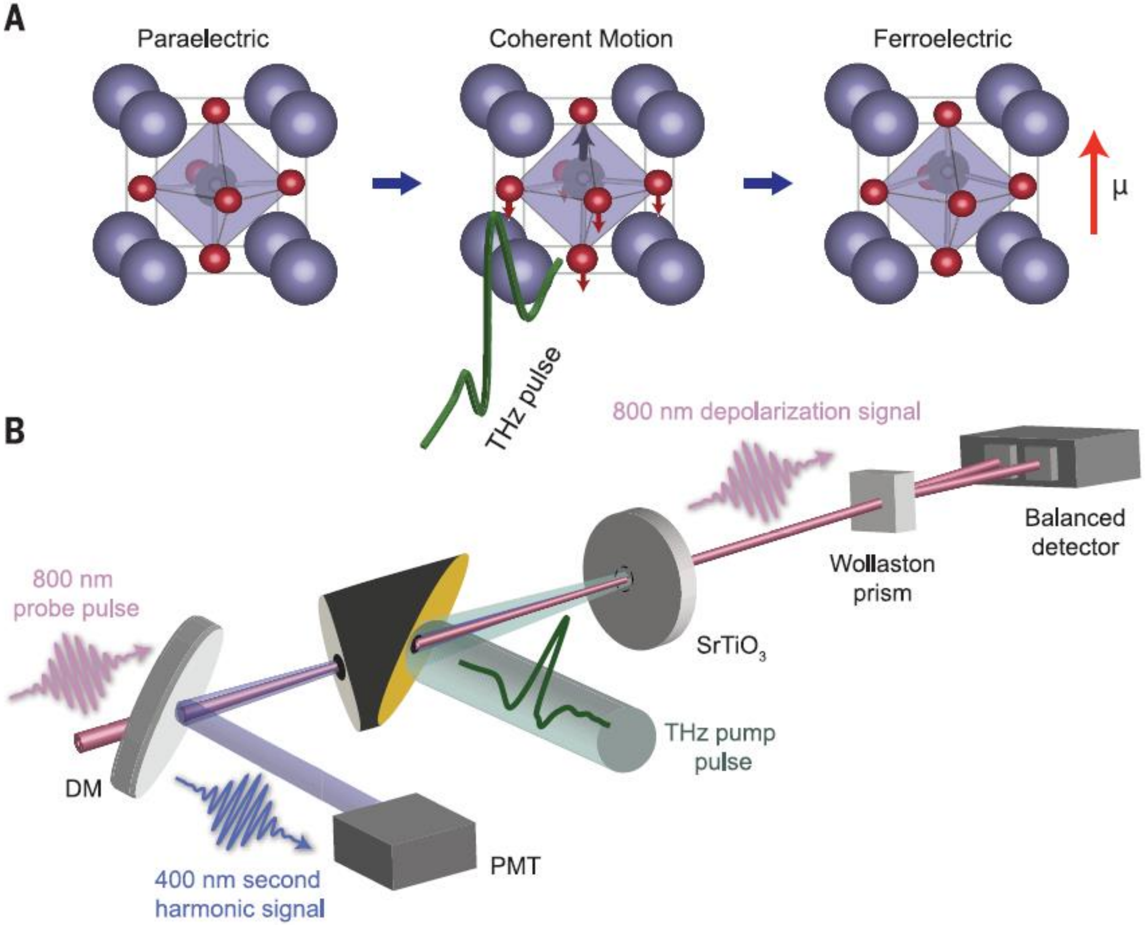
Control of material structure

Induction of Paraelectric → Ferroelectric phase transition in $SrTiO_3$

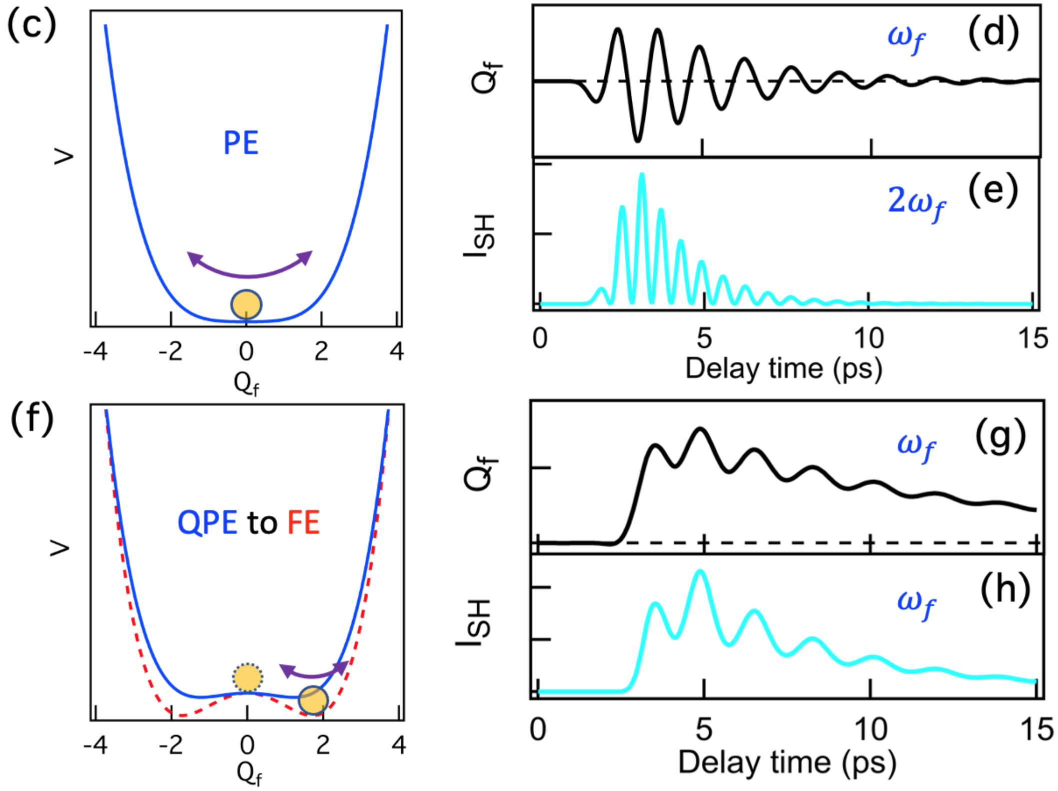


X. Li, et al., Science **364**, 1079 (2019)

Control of material structure?

B. Cheng, et al., Phys. Rev. Lett. **130**, 126902 (2024)

Induction of Paraelectric → Ferroelectric phase transition in $KTaO_3$ was NOT observed, INSTEAD a local dipolar correlation was observed



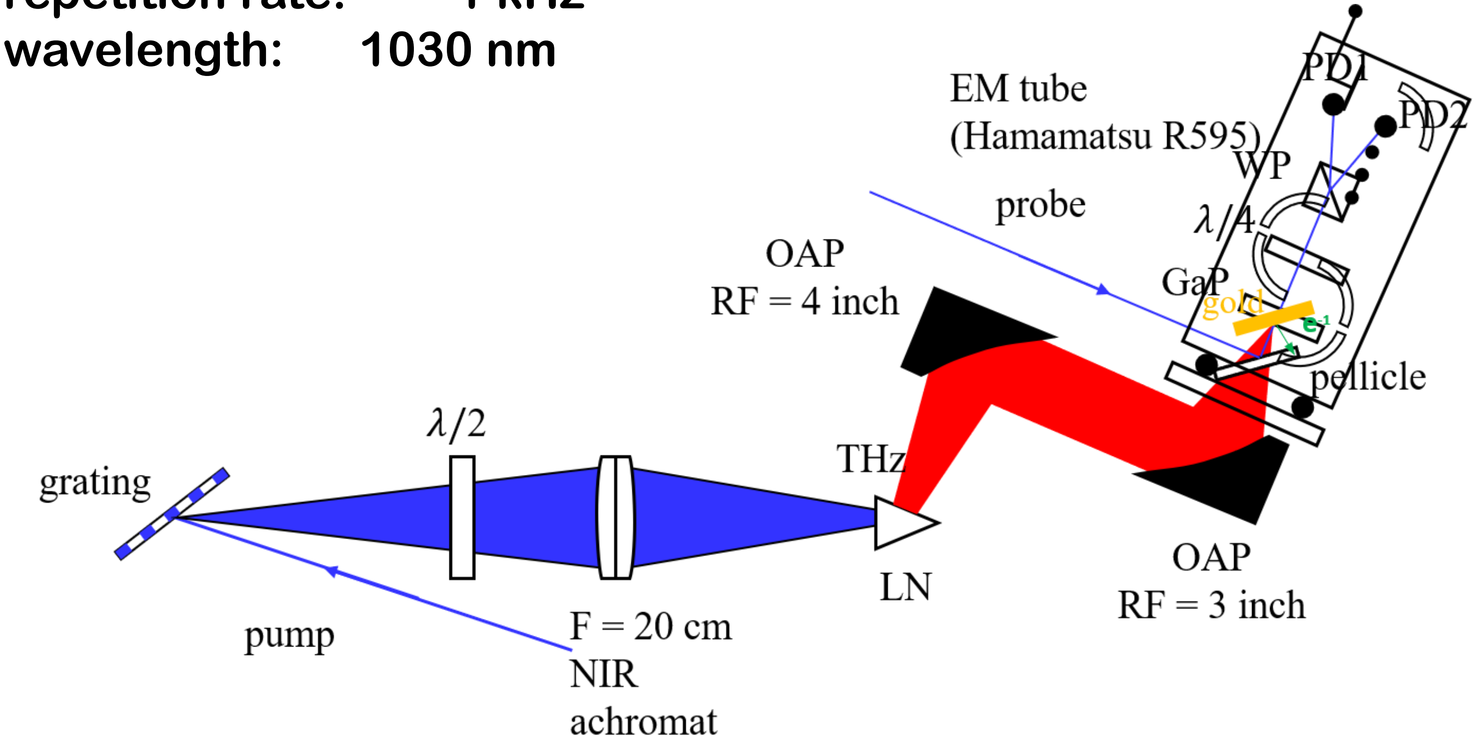
M. Basini et al., Nature **628**, 534 (2024)

THz electron emission

S. Li et al., N. Commun. **14**, 6596 (2023)

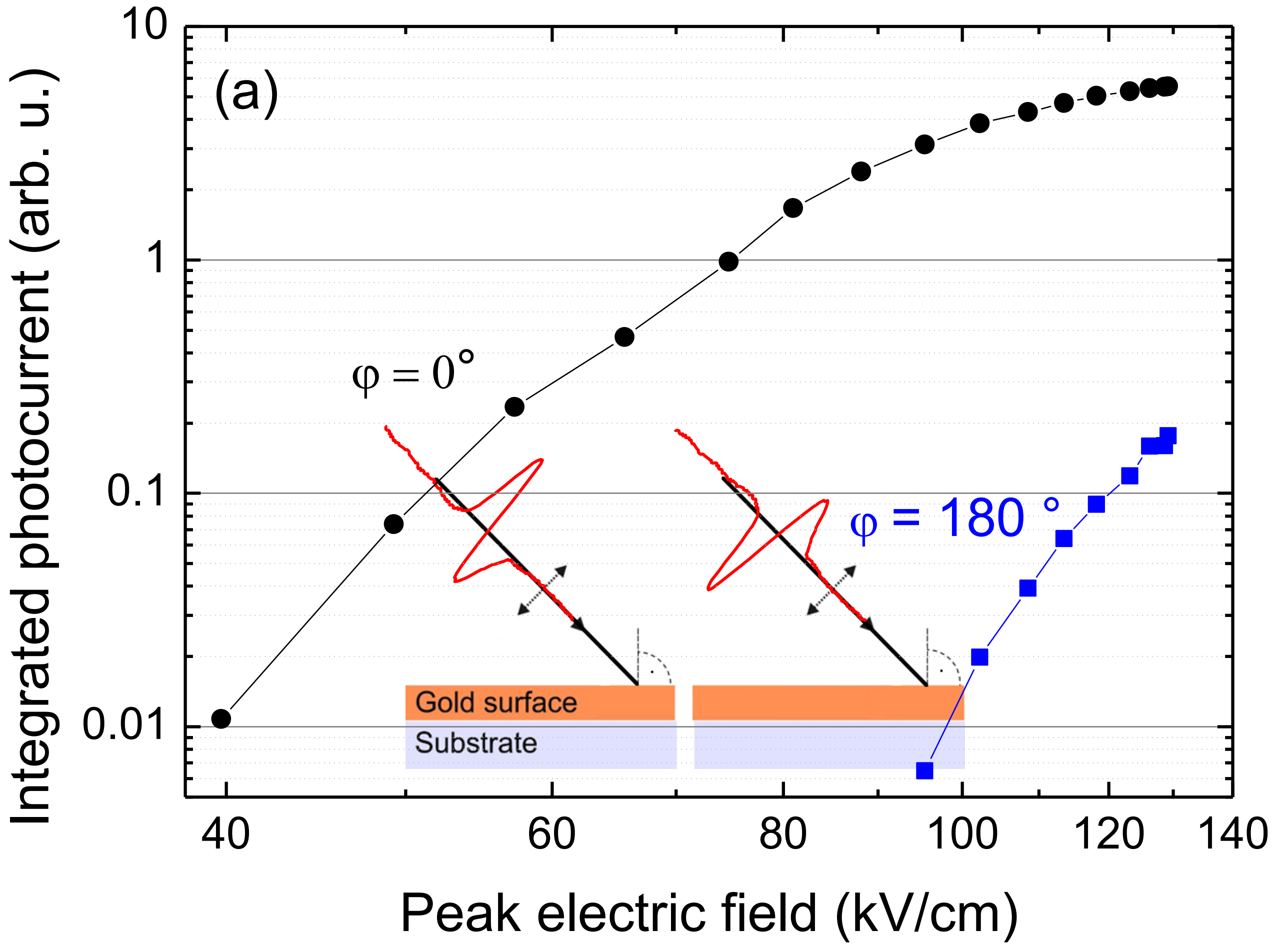
- **Pump pulse parameters:**
  - **- pulse duration: 220 fs**
  - **- pulse energy: up to 4 mJ**
  - **- repetition rate: 1 kHz**
  - **- wavelength: 1030 nm**

- ➢ EM signal vs E-field strength
- ➢ EM signal vs THz polarity



THz electron emission result

- **Photocurrent vs Electric filed:**



Application possibilities of THz pulses with extremely high field strength

1., Enhancement of HH generation (for attosecond pulse generation)

E. Balogh et al.: Phys. Rev. B **84**, 023806 (2011)
K. Kovács et al.: Phys. Rev. Lett. **108**, 193905 (2012)

2., Field-free orientation of molecules

S. Fleischer et al.: Phys. Rev. Lett. **107**, 163603 (2011)
K. N. Egodapitiya et al.: Phys. Rev. Lett. **112**, 103002 (2014)

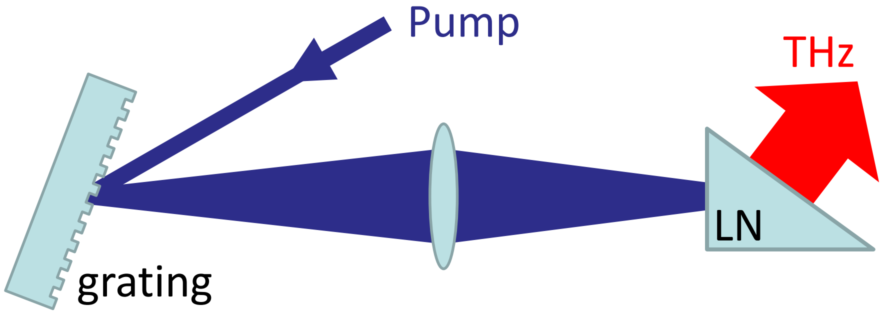
3., Acceleration (and other manipulations) of charged particles

J. Hebling et al.: arXiv 1109.6852 (2011)
L. Pálfalvi et al.: Phys. Rev. ST-AB **17**, 031301 (2014)
W. Ronny et al.: Sci. Reports **5**, 14899 (2015)
W. Ronny et al.: Optica **3,** 1209 (2016)
E. A. Nanny et al.: Nature Comm. **6**, 8486 (2015)
J. Ying et al.: Nature Photonics (2024)
E. Curry et al.: Phys. Rev. Lett. **120**, 094801 (2018), IFEL based
Sz. Turnár et al.: Appl. Phys. B 127, 38 (2021), Sz. Turnár et al., Fs-Mat 2025 poster

4., Carrier-envelop-phase (CEP) stable attosecond pulse generation

Gy. Tóth et al.: JOSA B **35**, A103 (2018)

Limitations of conventional TPFP in LN



$$\tan\gamma = -\frac{n}{n_g}\lambda\frac{d\varepsilon}{d\lambda}$$

- **Limited interaction** length due to large angular dispersion

  Fundamental limit

- **Imaging errors** at large spot sizes

  Special limit

- **Prism shaped LN crystal with large wedge angle leads to THz pulse and beam distortions**

  Special limit

→ **It is challenging to increase the THz energy & field strength further**

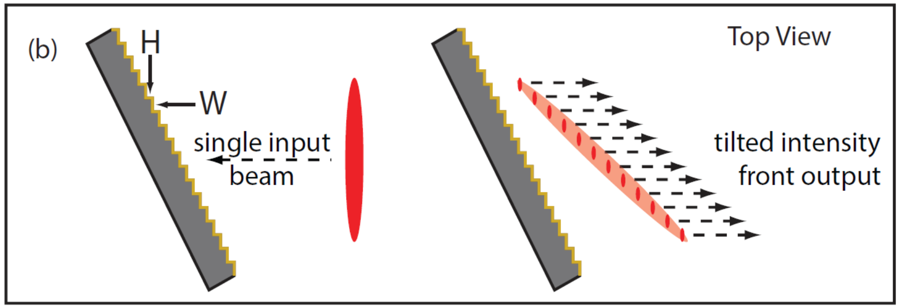
→ **Limited focusing (field increasing)**

Improving TPFP LN THz source setups

Ofori-Okai B.K. et al., Opt. Express **24**, 5057 (2016)

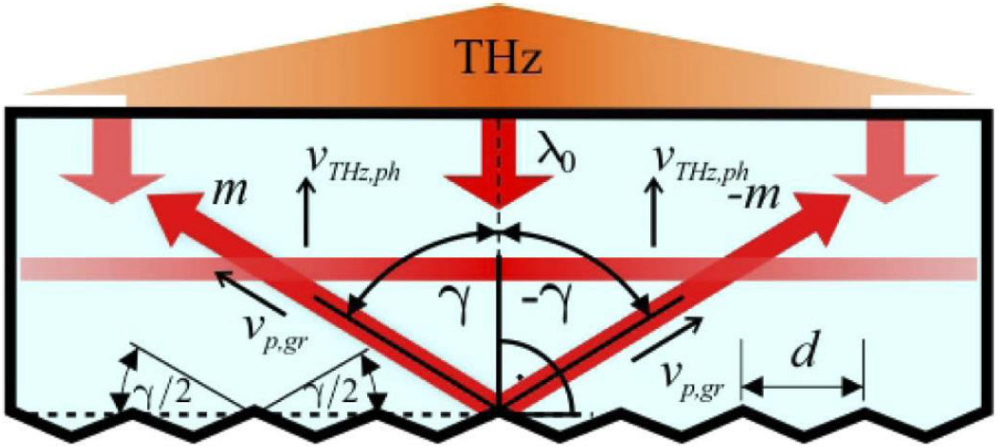
Guiramand L. et al., Phot. Res. **10**, 340 (2022)

Using echelon garting instead of optical grating



400 μJ pump pulse energy resulted 400 kV/cm focused THz field, η=1.3%

Reflective nonlinear slab (RNLS) Tóth Gy. et al., Opt. Express **27**, 30681 (2019)

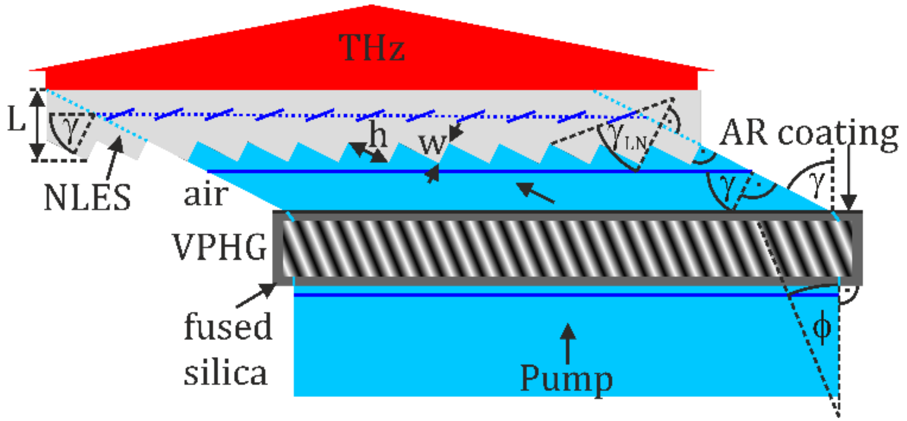


d = 3 ÷ 50 μm

Pump: 870 mJ, 1 ps, 1.03 mm with D = 7 cm, L = 4 mm LN RNLS → THz: 10 mJ, 50 MV/cm

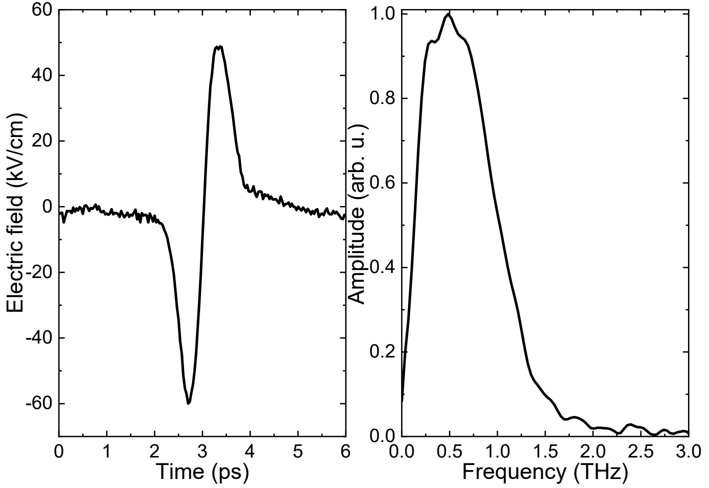
Improving TPFP LN THz source setups

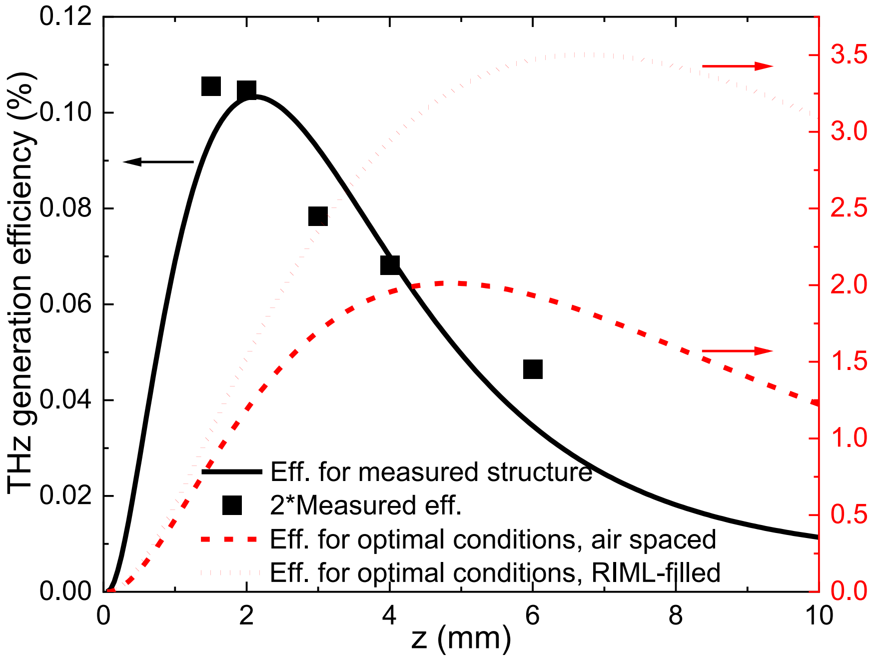
G. Krizsán et al., in preparation

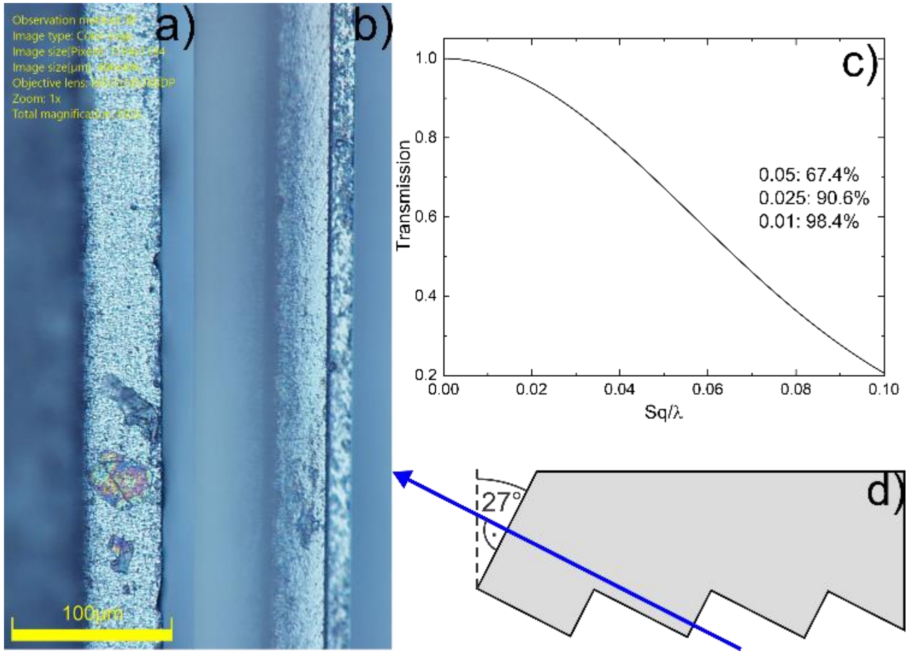


w = 50 μm

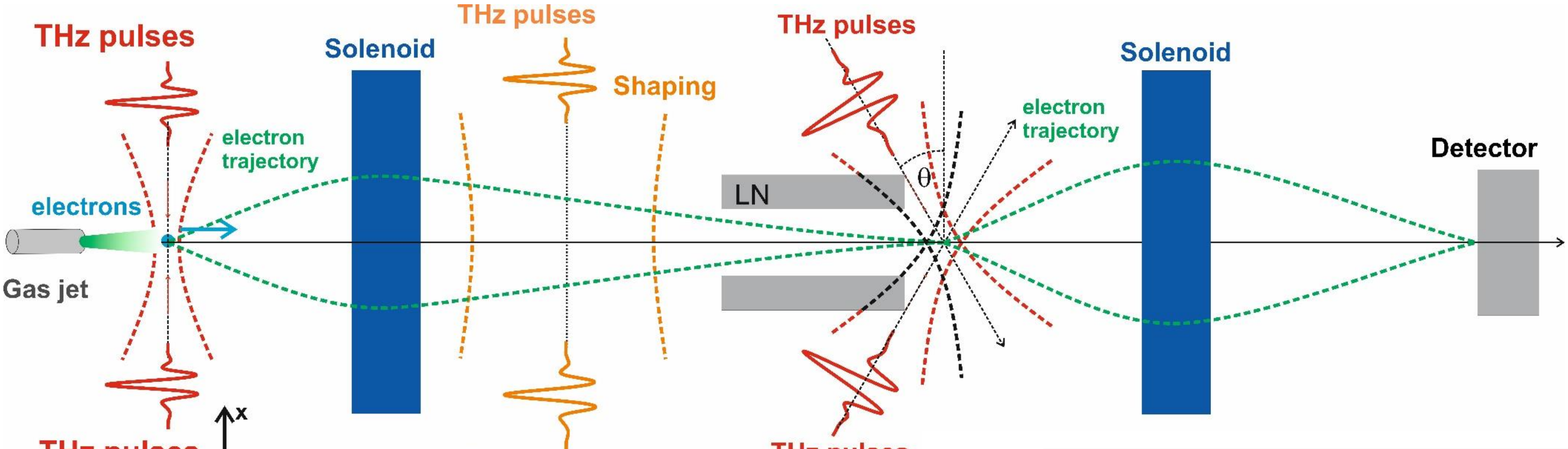
h = 70 μm







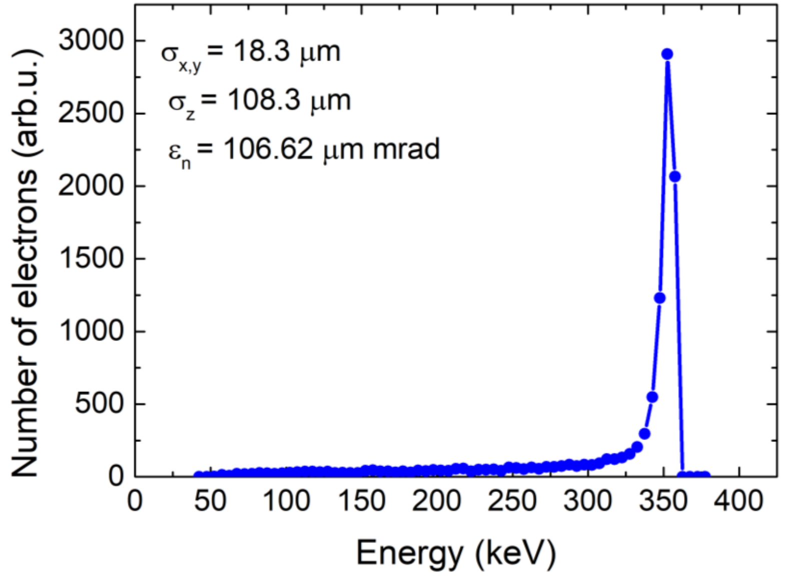
Plan of a THz-driven electron gun-accelerator system



2 x 4 x 0.5 mJ THz pulses

Application possibility: time resolved electron diffraction

Sz. Turnár et al.: Appl. Phys. B 127, 38 (2021)
Sz. Turnár et al.: Fs-Mat2025 P01.

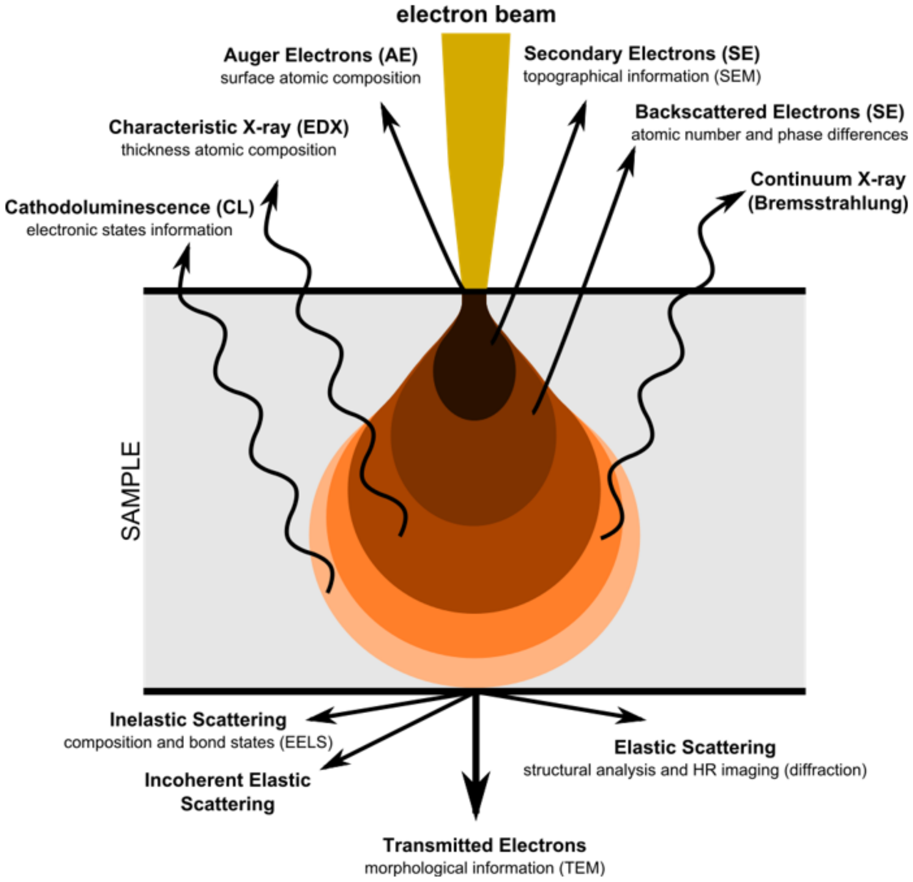


Accelerated electron bunches in material research

Areas of application (electron)

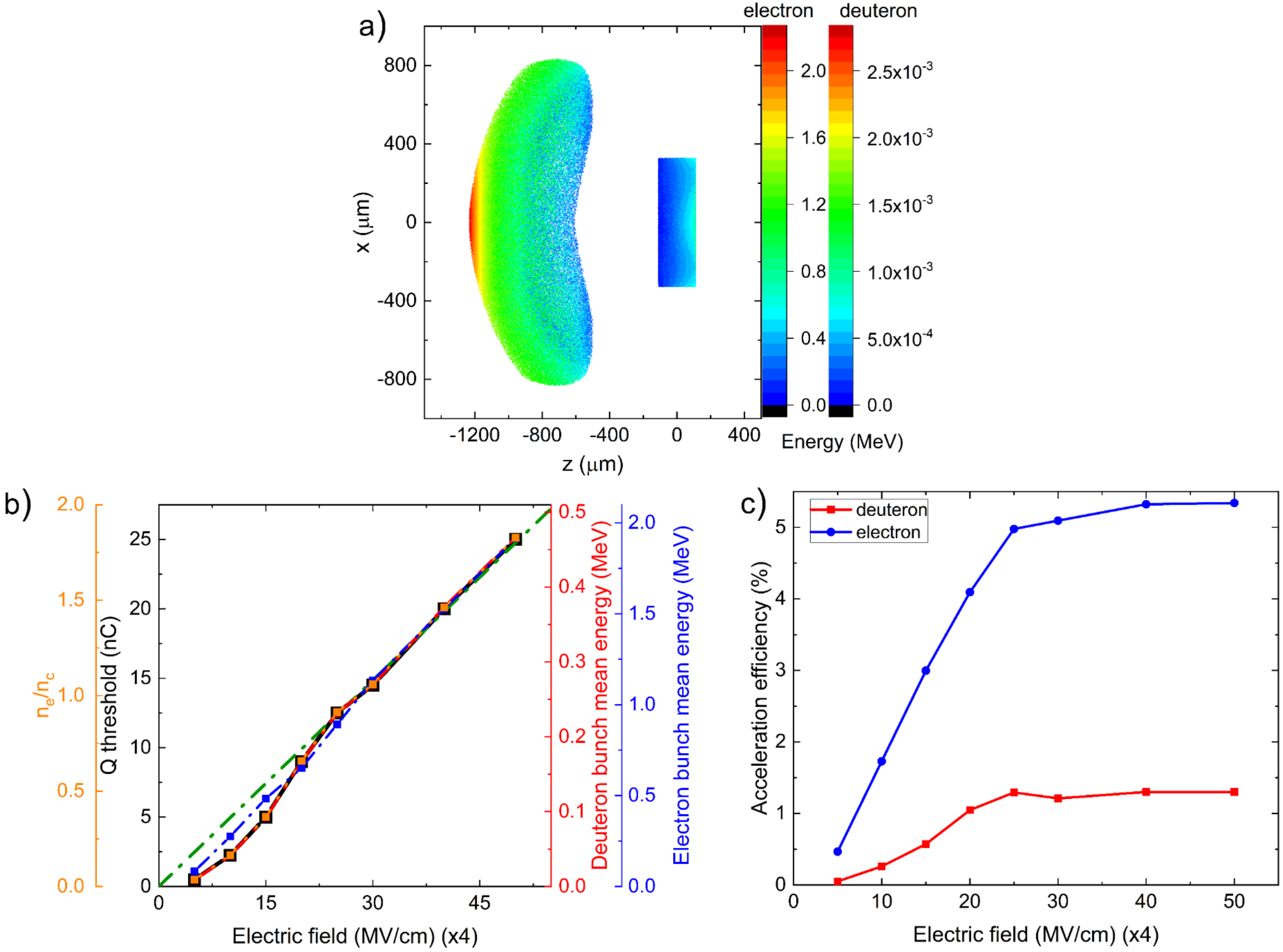
- Electron Microscopy
  - TEM (a few keV – a few 100 keV)
  - SEM (a few eV – a few keV)

- Auger electron spectroscopy (a few eV – 50 keV)
- Electron energy loss spectroscopy (10-30 keV)
- Electron stimulated desorption experiments (a few eV- a few keV)
- Electron diffraction (a few 100 keV)



https://en.m.wikipedia.org/wiki/File:Electron_Interaction_with_Matter.svg

Accelerated electron bunches in material research



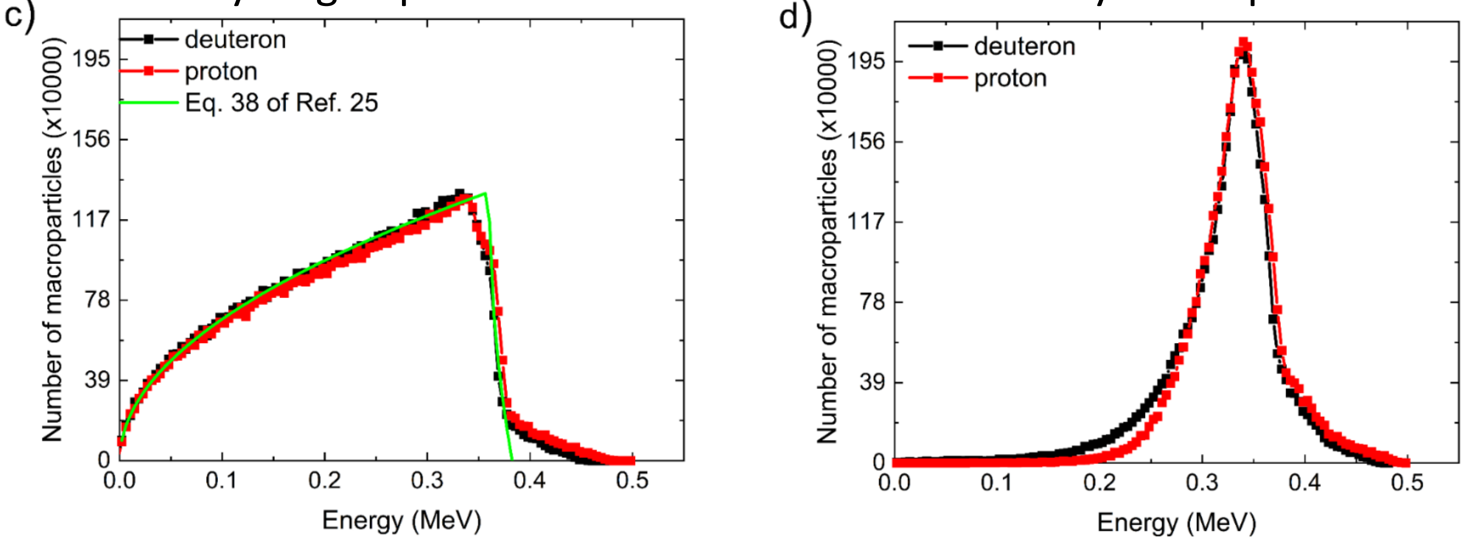
Sz. Turnár et al., Sci. Rep. submitted

Coulomb explosion following the ultrafast removal of electrons

Ion energy spectra using 4 x 25 MV/cm THz pulses and

hydrogen plasma

heavy water plasma



$6\text{x}10^4$ neutron/shot

Sz. Turnár et al., Sci. Rep. submitted

Summary

- OR, two-color laser plasma and STE can be used to generate THz pulses with mJ energy and few 100 keV/cm field strength
- TPFP LN is a highly efficient and widely used source of high energy , high field single-cycle THz pulses
- These pulses can be used to follow dynamics in different materials
- These pulses could be used for structure change
- A few reasons of limiting the generation efficiency of THz pulses with mJ level energy and few MV/cm peak field strength in conventional TPFP LN source is identified.
- A few solutions for mitigate or eliminate these limitations has been suggested or demonstrated. Generation of single-cycle THz pulses with extremely high field by new type TPFP sources are foreseen.
- Many important application possibilities of THz pulses with extremely high field strength generated by TPFP, GaSe or organic NC identified.