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UNIVERSITY OF PÉCS
SZENTAGOTHAI RESEARCH CENTRE

Extreme-high-field THz Pulse Sources and their Application for Particle Acceleration

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HIGH-FIELD THz LAB


High-Brightness Budapest, Hungary, 23-25 March 2022

Deliverables Report

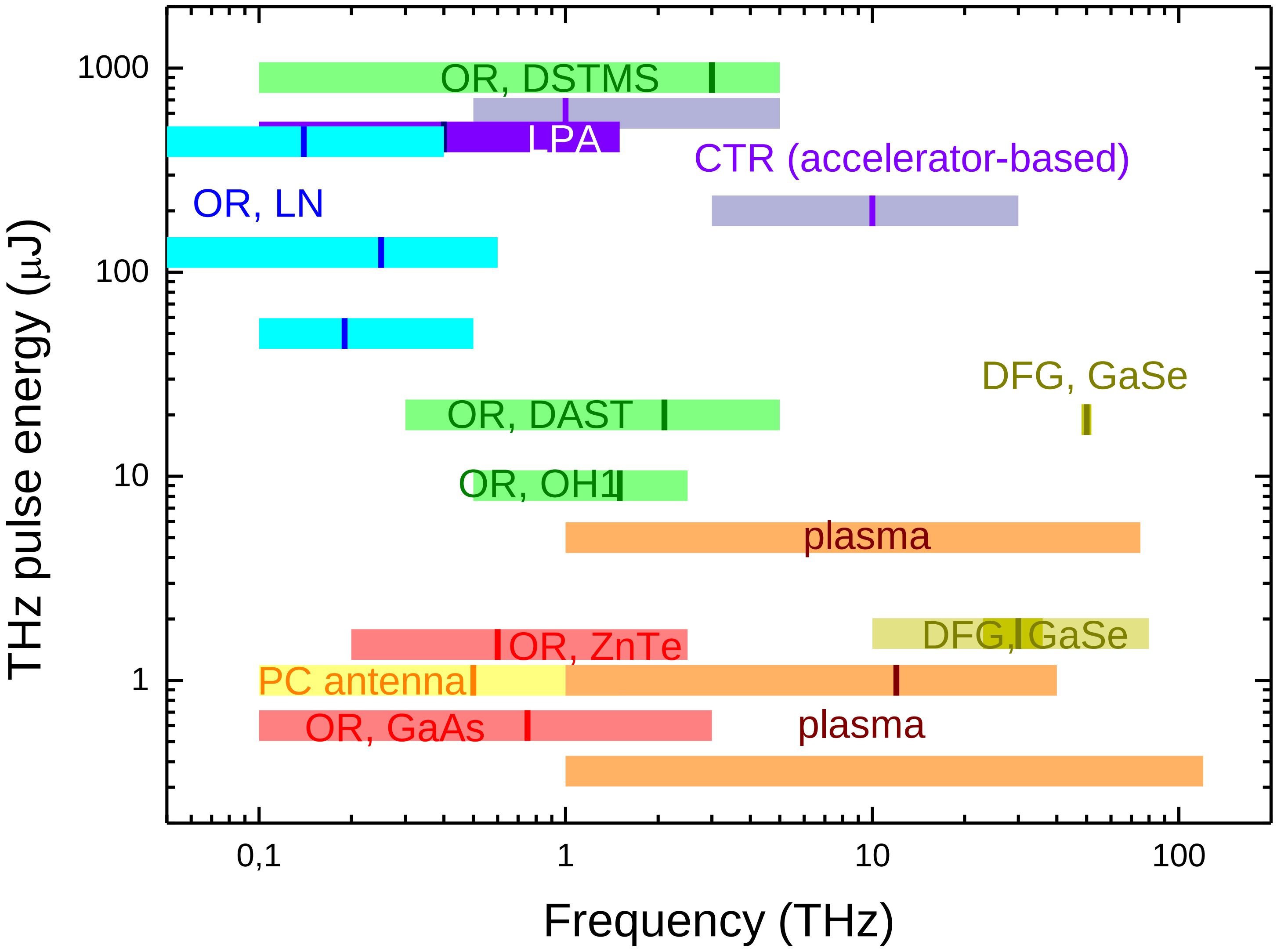
EuPRAXIA PP
Grant Number 101079773
DELIVERABLE: WP14-D14.1

1. **Plasma targetry and diagnostics for ultrabright, high rep-rate beams.** The diagnostics work of this Working Group would focus on ultrabright electron beam slices, while targetry has a much wider impact on many aspects of EuPRAXIA. Given the fact that many labs involved in EuPRAXIA already have related structures in place, the main goal here will be to coordinate the activities and seek tailor-made solutions to the various aspects of EuPRAXIA's target needs. Diagnostics and beam manipulation via THz fields is another strong drive for the full control over relativistic beams for applications.

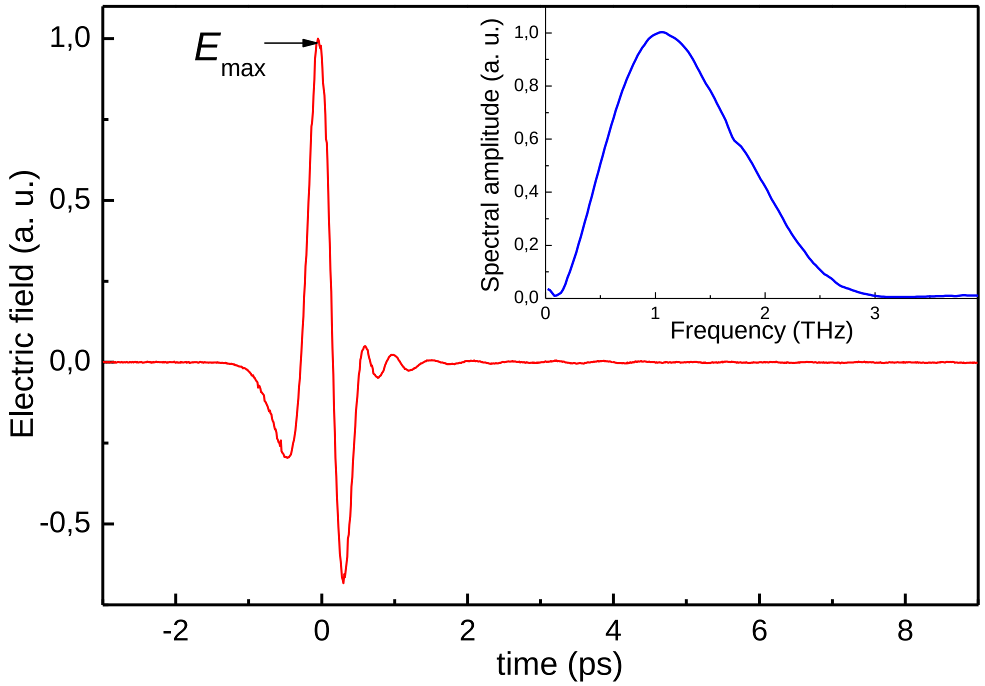
- Ultrabright electron beam slice measurements require mostly personnel on the level of one or two PDRAs, and, given the access to one of the consortium's high-brightness beam sources, a moderate amount of consumables
- Target development will focus on simulation and construction work, and requires two dedicated PDRAs for development of new approaches and coordination work between EuPRAXIA partners.
- THz source development will require a dedicated THz beamline, initially to be installed at CALA's kHz PFS-pro laser system for early concept studies performed in strong collaboration with UP, the inventor of the highly efficient THz sources. This will be complemented by personnel funding for one PDRA and one PHD student to verify highly efficient THz generation concepts and study their application on an electron beamline, as is being constructed for the P-MOPA effort.

The five international Working Groups to be installed for the five identified areas would organize themselves to carry out the work. There are already very good track records in some of the areas and across many partners, institutions and people. Within these Working groups, a flat hierarchy would be adopted. For example, a rotating Working Group chair mechanism would be used. In addition to these Working Groups, participation in Doctoral Training Centres are required.

High-energy THz pulse sources



Classification of THz pulses by peak electric field and energy

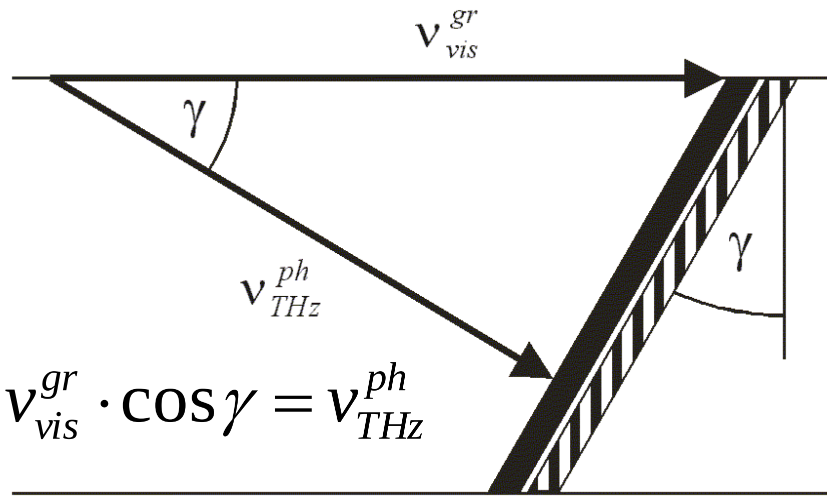


- Linear (TDTS) THz spectroscopy (E_{max} ≈ 100 V/cm → 10 fJ energy)
- High field THz pulses (E_{max} ≈ 100 kV/cm → 1 μJ energy)

  THz pump – probe measurement, nonlinear THz optics
- Extreme high field THz pulses (E_{max} ≈ 100 MV/cm → 10 mJ energy)

  enhancement of HHG, particle manipulation, etc.

Limitations for further upscaling the THz field strength



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

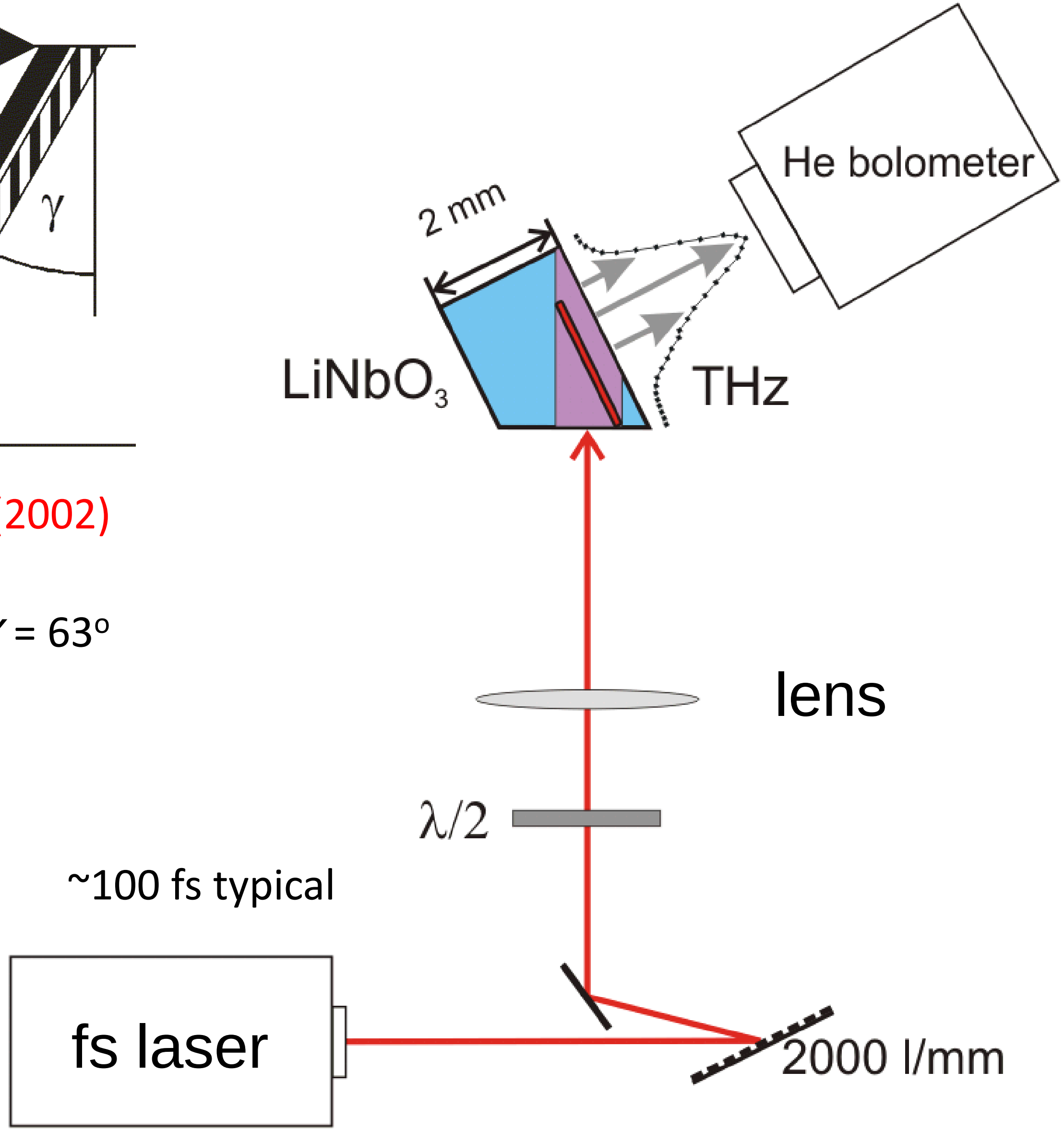
Hebling et al., Opt. Expr. **10**, 1161 (2002)

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

γ = 63°

pulse front tilt

angular dispersion



Success of TPFP LiNbO3 THz sources

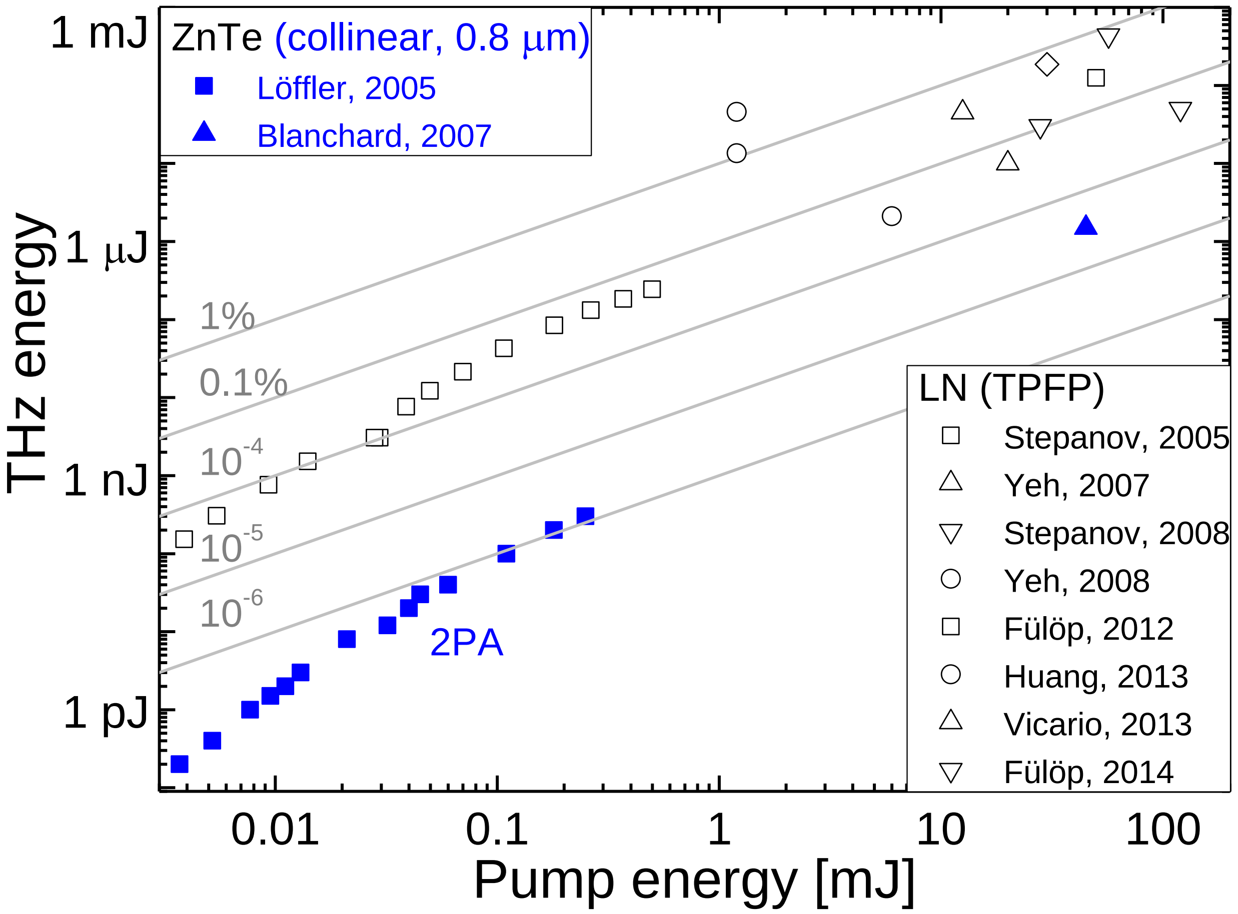
Material	ZnTe	GaP	GaAs	LN
d_{eff} [pm/V]	68.5	24.8	65.6	168

Large nonlinear coefficient

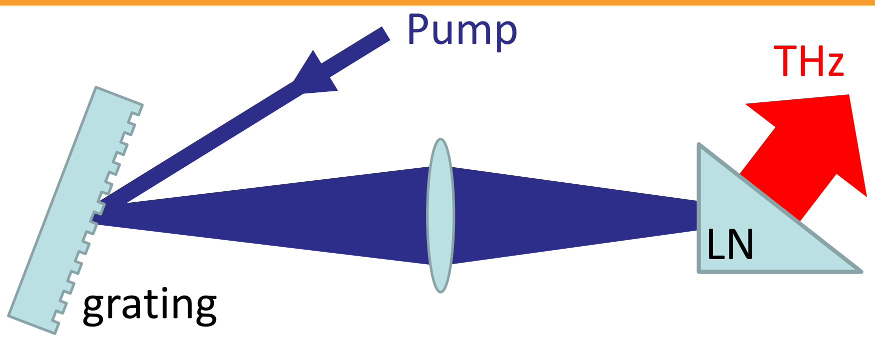
Large bandgap, absence of low-order multi-photon absorption (MPA)

Very efficient THz pulse source on the 0.2 – 2 THz range

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



Limitations of TPFP in LN



Conventional tilted-pulse-front pumping (TPFP)
Hebling et al., Opt. Express, 2002

- **Limited interaction** length due to large angular dispersion ($\gamma \approx 63°$)

  Fundamental limit

- **Imaging errors** at large spot sizes

  Special limit

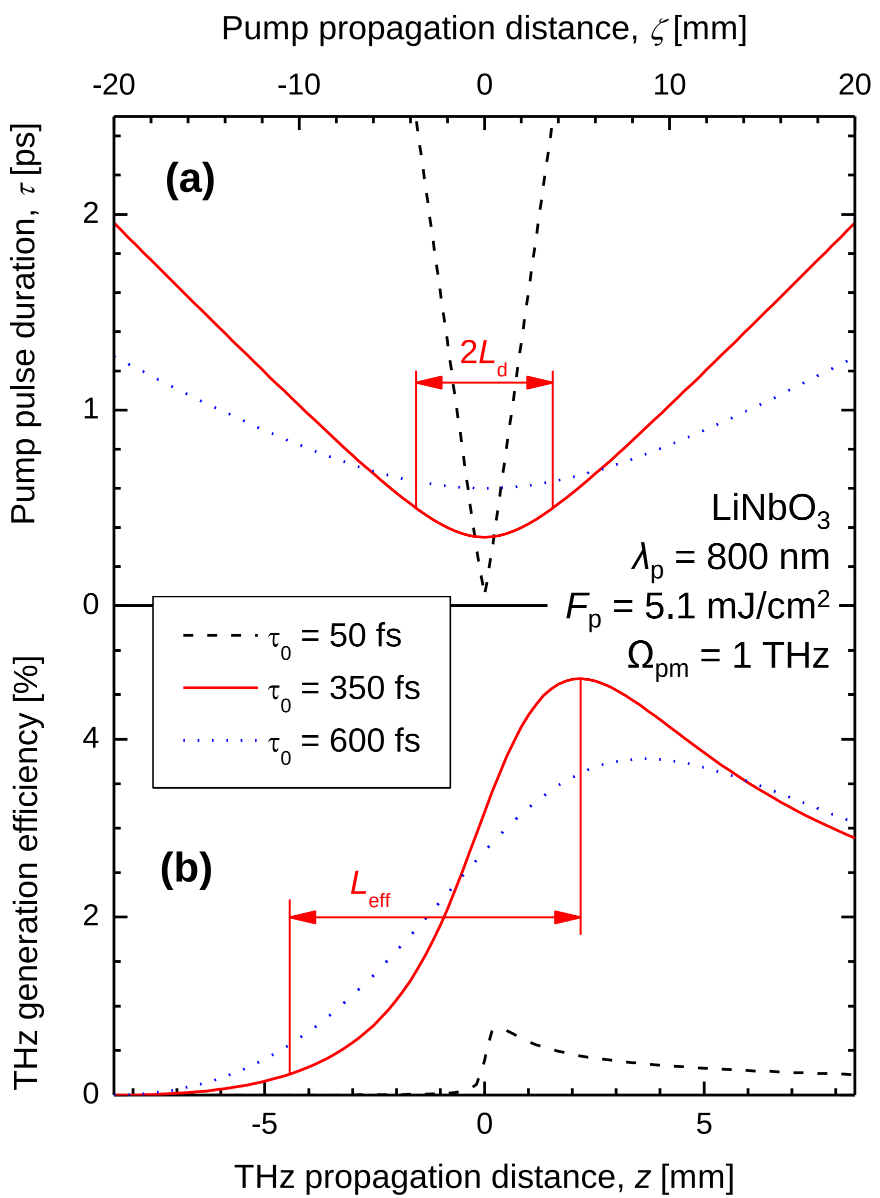
- **Prism shaped LN crystal with large wedge angle leads to THz pulse and beam distortions**

  Special limit

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

→ **Limited focusing (field increasing)**

Effective interaction length



Pulse front tilt:

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

angular dispersion

material dispersion

GVD parameter:

$$D = \frac{d\left(v_g^{-1}\right)}{d\lambda} = \frac{\lambda}{c}\left[n\left(\frac{d\varepsilon}{d\lambda}\right)^2 - \frac{d^2 n}{d\lambda^2}\right]$$

J. Hebling, Opt. Quant. Elect., **28**, 1759 (1996)

J. A. Fülöp et al., Opt. Express, **18**, 12311 (2010)

Mitigate/lesser fundamental limitation

Limited interaction length due to angular dispersion

Solution 1: Longer pump pulse duration

Suggested based on simulations:

J. A. Fülöp et al., Opt. Express, **18**, 12311 (2010)
J. A. Fülöp et al., Opt. Express, **19**, 15090 (2011)

Experimentally demonstrated
E_{THz} = 125 μJ, 460 μJ

J. A. Fülöp et al., Opt. Lett., **37**, 557 (2012)
J. A. Fülöp et al., Opt. Express, **22**, 20155 (2014)

Using cryogenically cooled LN
$\eta = 2$ % conversion efficiency

S.-W. Huang et al., Opt. Lett. **38**, 796 (2013)

Mitigate/lesser fundamental limitation

Limited interaction length due to angular dispersion

Solution 2: Nonlinear crystal with smaller needed tilt angle

Reconsidering semiconductors for THz generation!

Reconsidering semiconductors for THz generation

☺ **Collinear phase matching** at 800 nm pump laser wavelength

☹ **Smaller nonlinear coefficient**

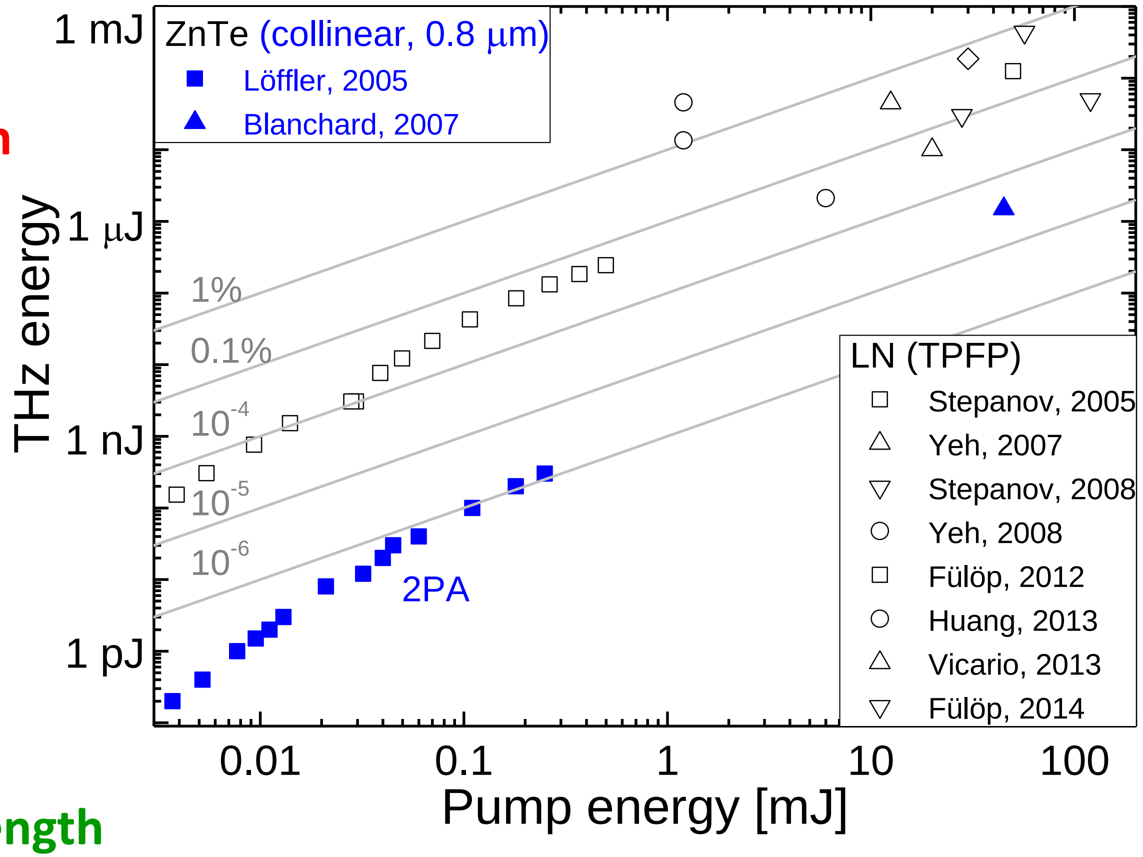
☹ Strong **two-photon absorption** (2PA) at such pump wavelengths

→ Free carrier absorption of THz

→ Limited useful pump intensity

→ Low efficiency for THz generation

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



☺ **Solution: longer pump wavelength**

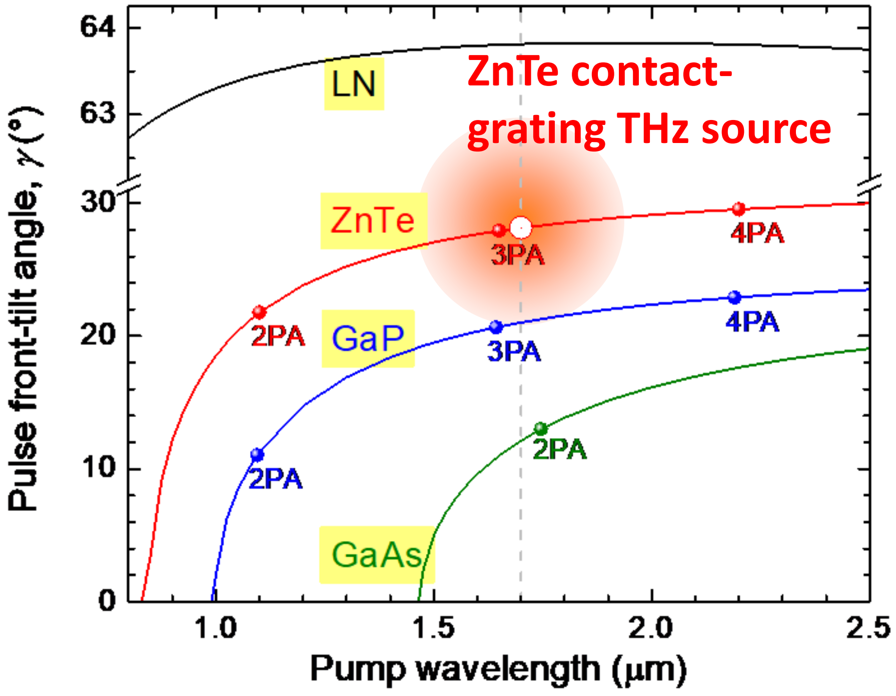
→ Allows for higher pump intensity and more efficient THz generation

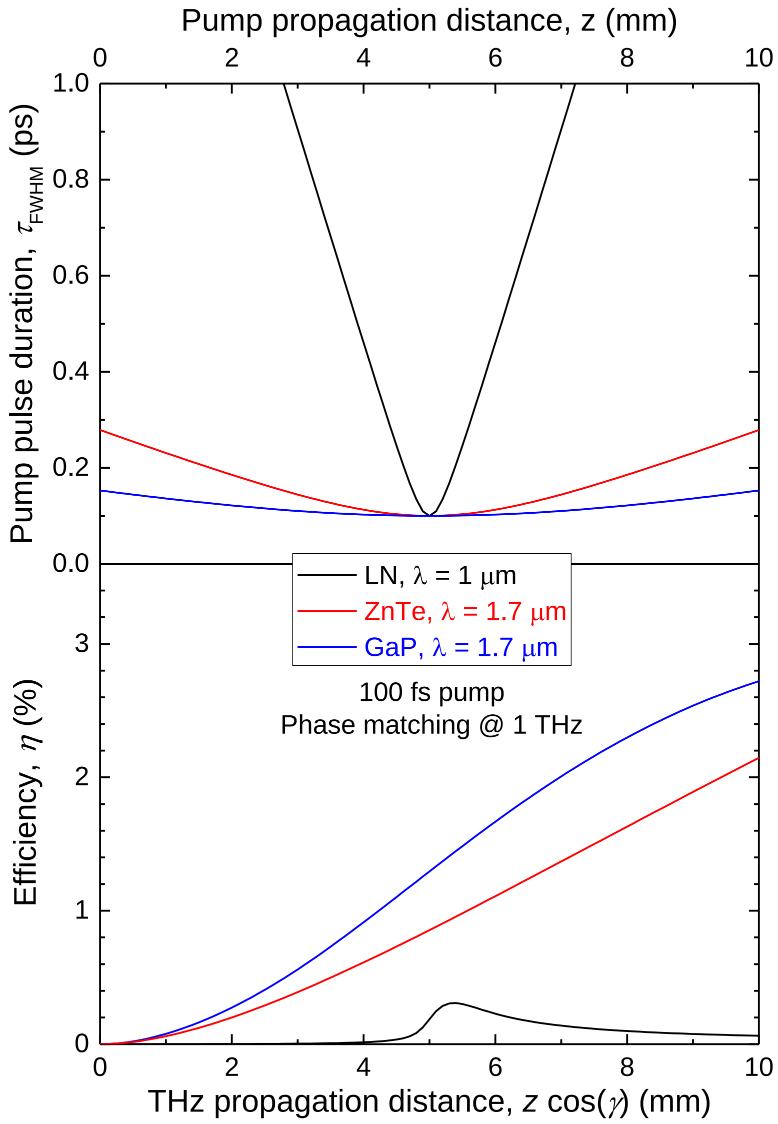
→ Requires tilted-pulse-front pumping

J. A. Fülöp et al., Opt. Express **18**, 12311 (2010) F. Blanchard et al., Appl. Phys. Lett. **105**, 241106 (2014)

Tilted-pulse-front pumping semiconductors

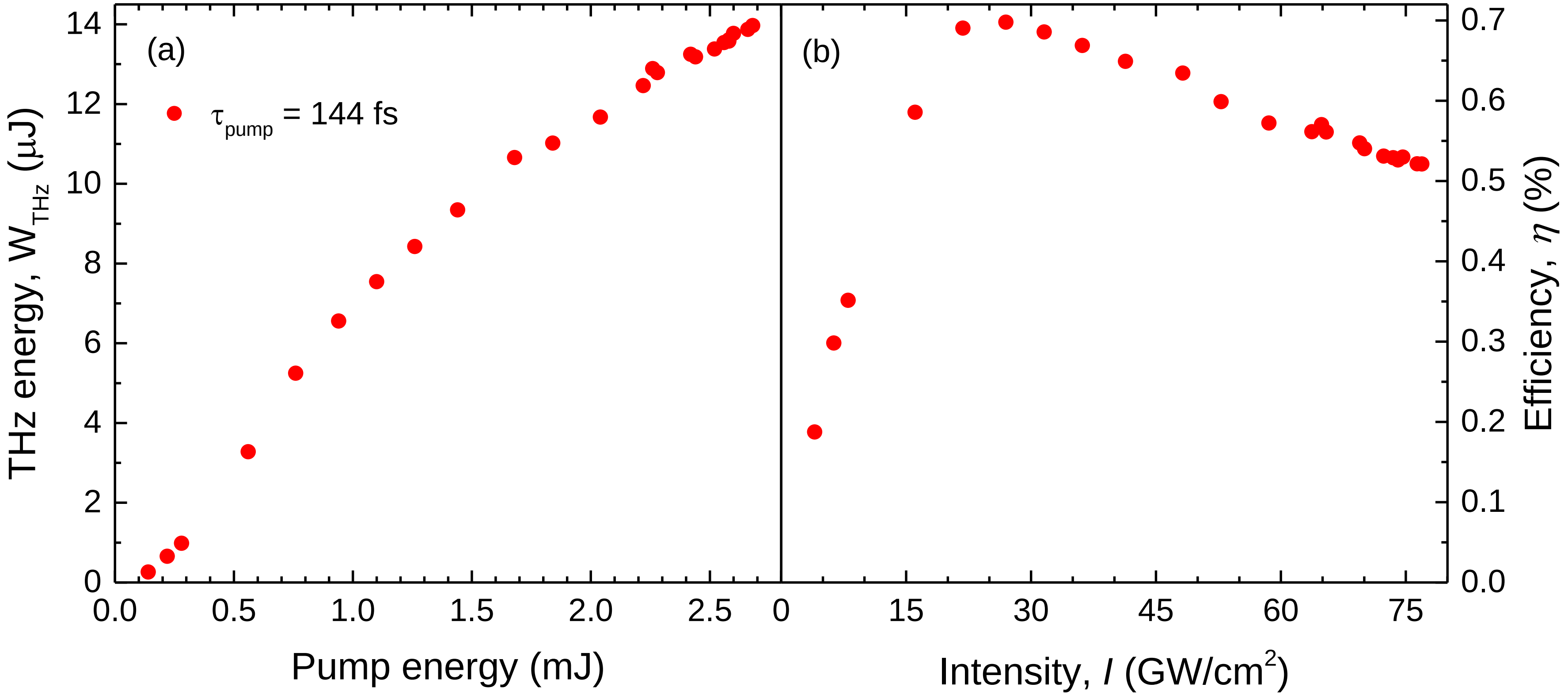
Small tilt angle for semiconductors ($\gamma \lesssim 30°$)





- **Large interaction length** for THz generation compensates for smaller nonlinear coefficient
- More **uniform crystal length** across the pumped area
- Advantageous for the implementation of a **contact grating**

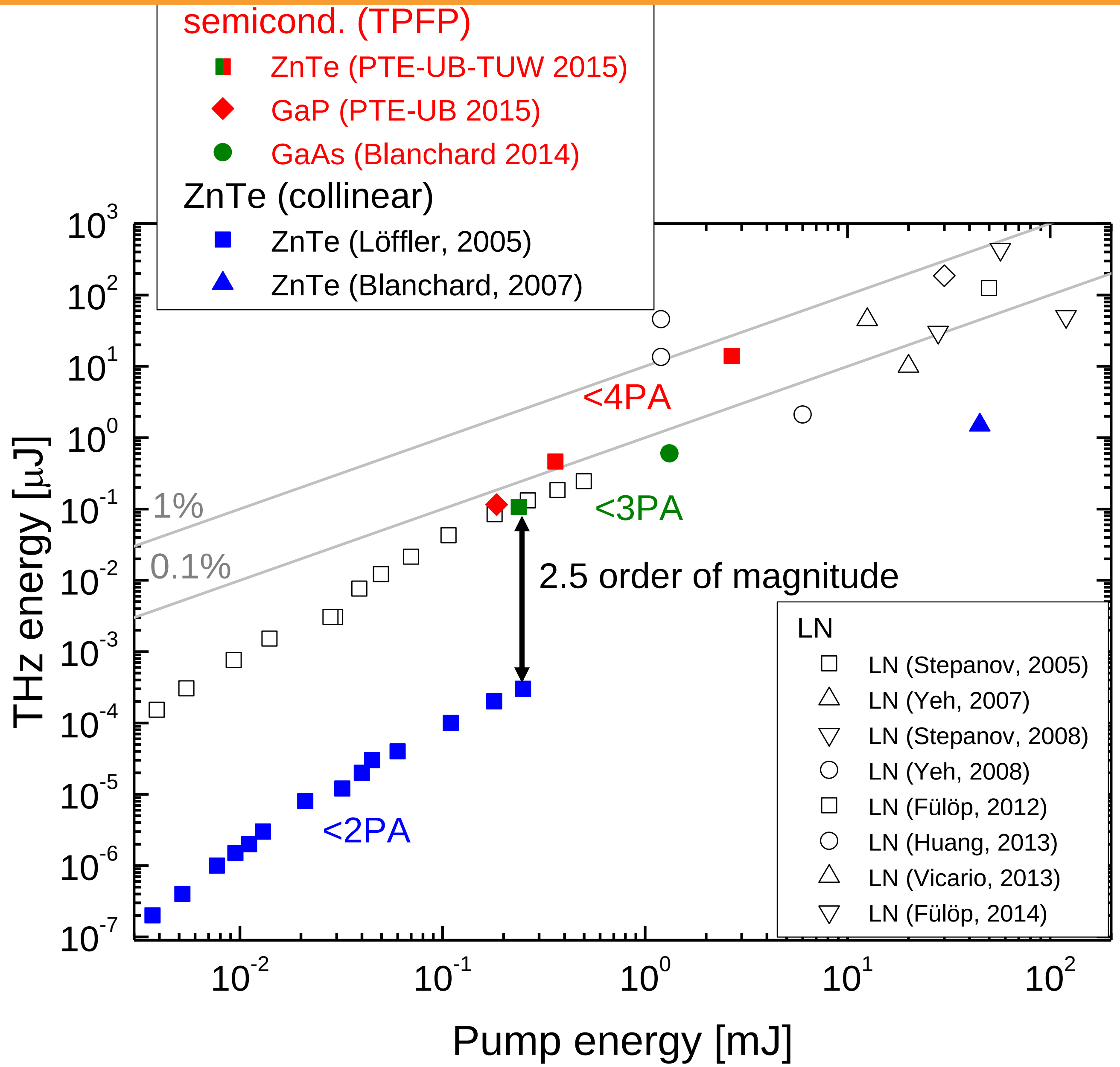
ZnTe pumped at 1.7 μm wavelength, excluding 2 or 3 PA



For comparison:

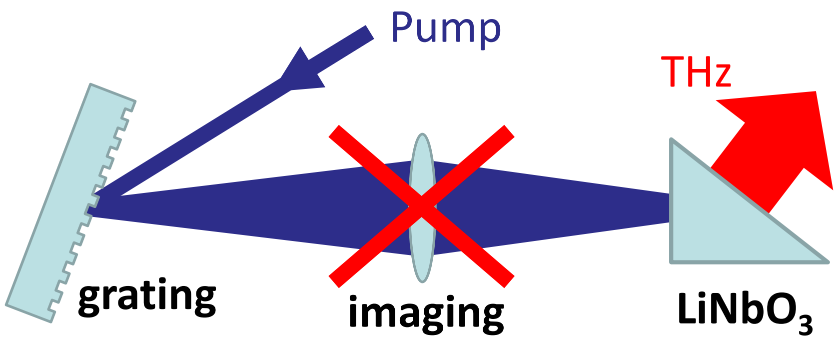
- ZnTe: η = $3.1\text{x}10^{-5}$ with 48 mJ pump @ 0.8 μm (Blanchard et al., Opt. Express, 2007)
- GaAs: $5\text{x}10^{-4}$ with 1.44 mJ pump @1.8 μm (Blanchard et al., Appl. Phys. Lett., 2014)

Semiconductor TPFP THz sources compared to LN ones

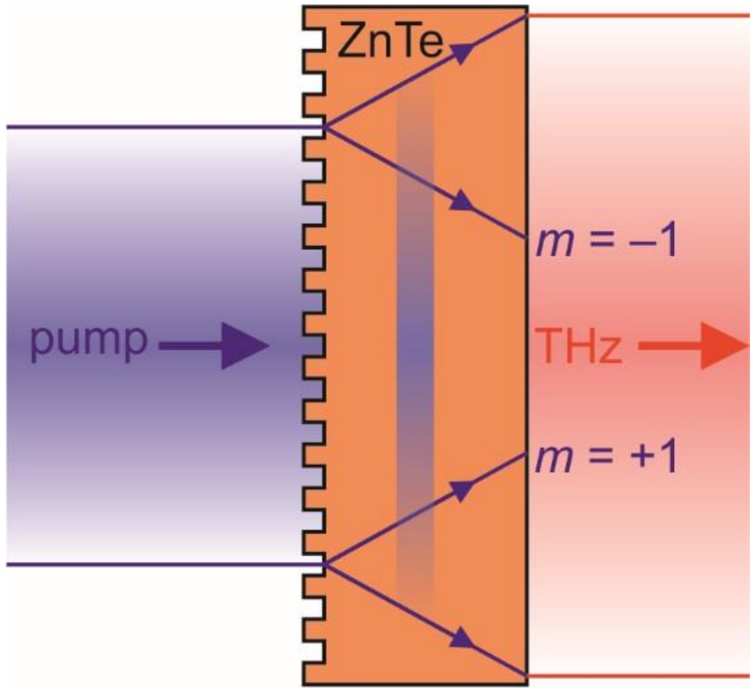


ZnTe contact-grating THz source

Elimination of the imaging error and prism shaped nonlinear crystal



Conventional TPFP (with imaging)



TPFP with contact grating (no imaging)

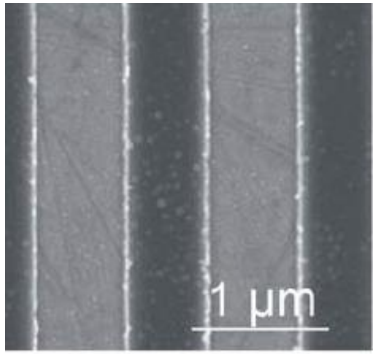
L. Pálfalvi et al., Appl. Phys. Lett. **92**, 171107 (2008)

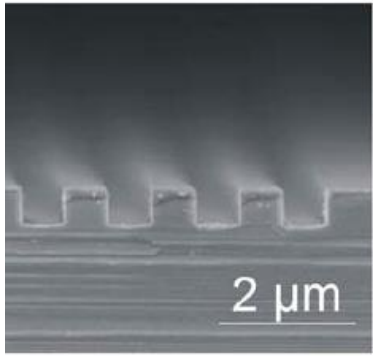
- Collinear geometry possible (with symmetrically propagating diffraction orders *m* = ±1)

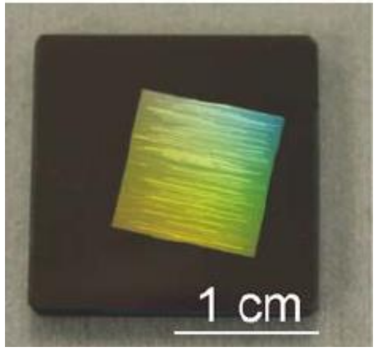
  Bakunov et al., J. Opt. Soc. Am. B, 2014

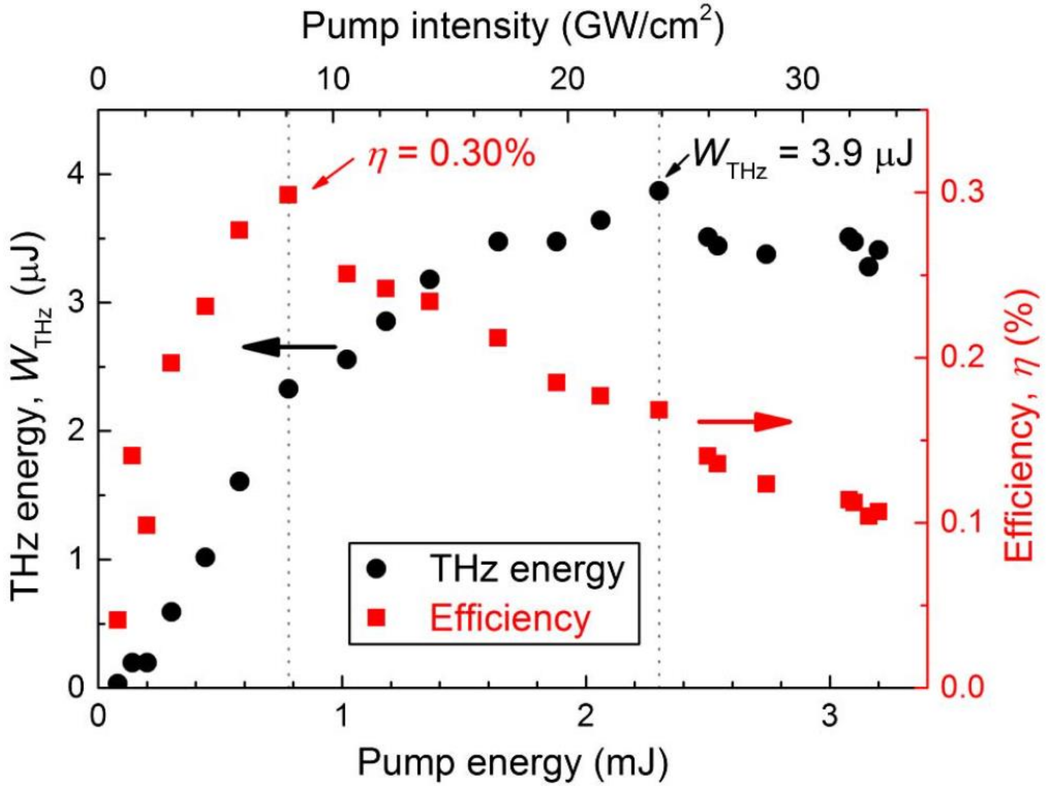
- THz energy easily increased by using larger pumped area
- Excellent THz beam quality

ZnTe contact-grating THz source









X100 efficiency compared to 800 nm pumped ZnTe

X10 efficiency compared to 2 μm pumped GaAs

J. A. Fülöp et al., Optica **3**, 1075 (2016)

Gy. Polónyi et al.: IEEE JSTQE **23**, 8501208 (2017) GaP η = 2%

Multi-cycle: P. S. Nugraha et al.: J. Phys. B **51**, 094007 (2018),

G. Krizsán et al.: EOS-TST 2018, poster, GaP up to η = 7%

Source of pump pulses (DCOPA) Gy. Tóth et al.: Opt. Express **25**, 28258 (2017)

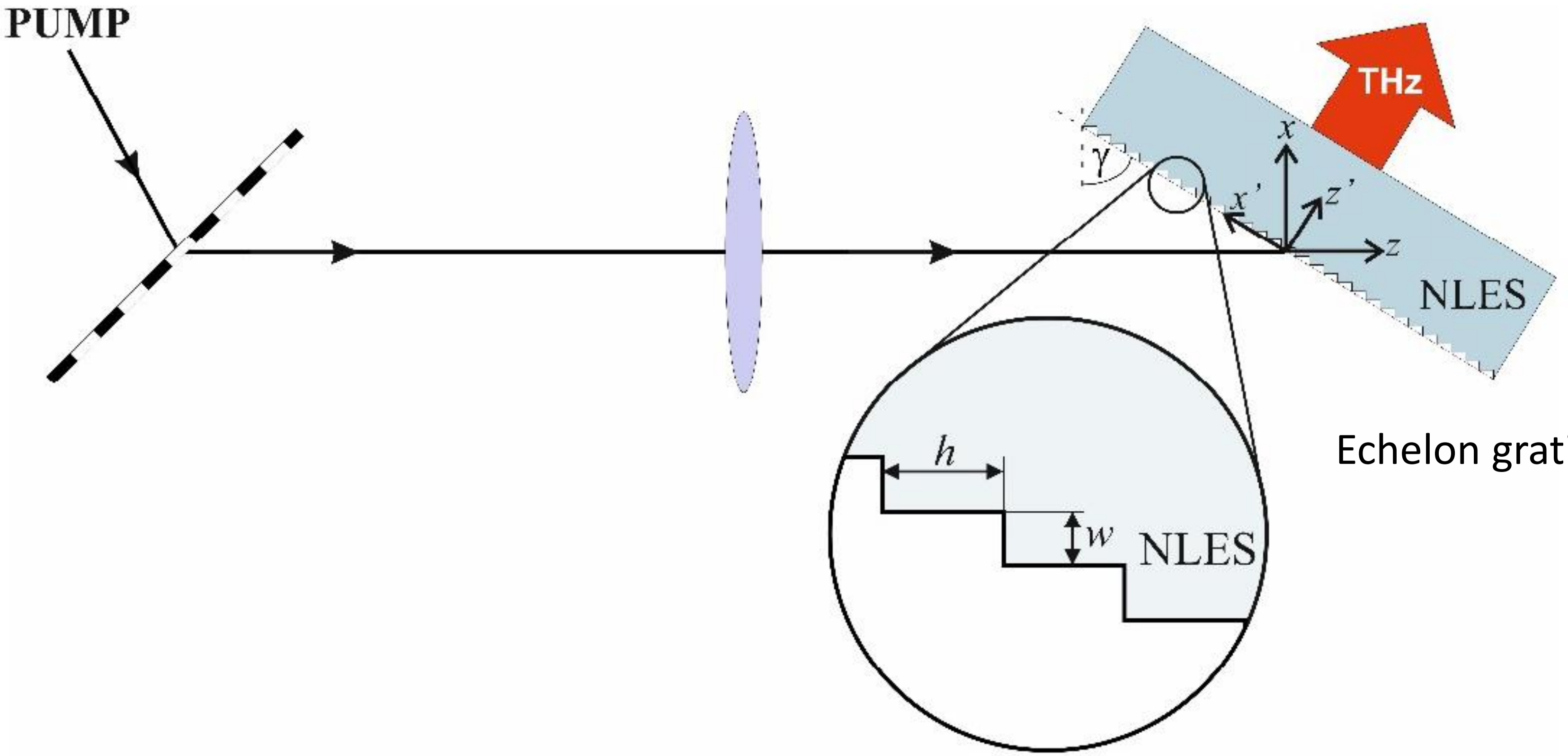
Gy Tóth et al.: EOS-TST 2018, poster η = 50%

Mitigate/lesser special limitation

Hybrid TPFP with Echelon-type CG

Limitations: 1. Imaging error caused pulse lengthening
2. Prism shaped LN

Strongly reduced limitation in case of hybrid TPFP schemes

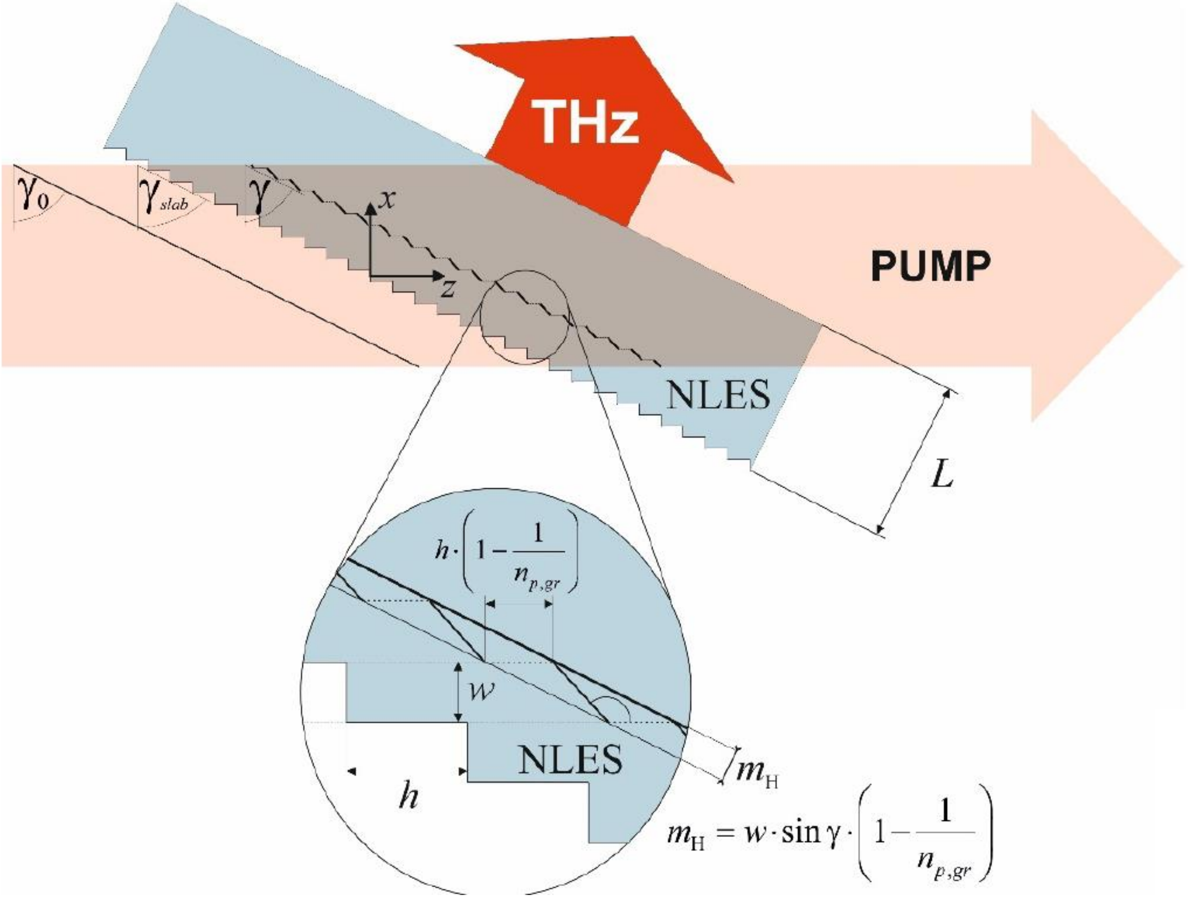


Echelon grating

B. K. Ofori-Okai et al.: Optics Express **24**, 5057 (2016) reflection Echelon, LN prism
L. Pálfalvi et al.: Optics Express **25,** 29560 (2017)
P.S. Nugraha et al.: EOS-TST 2018, today 15.05

Mitigate/lesser special limitation

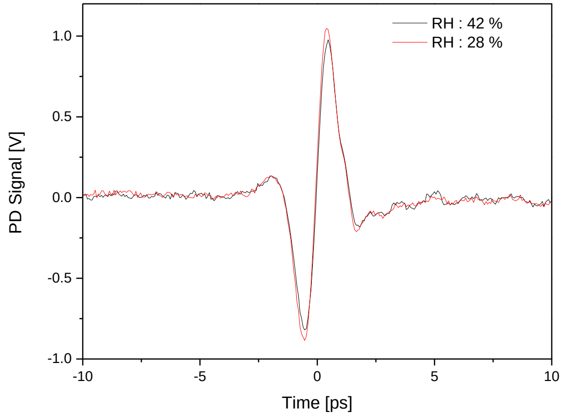
Hybrid TPFP with Echelon-type CG

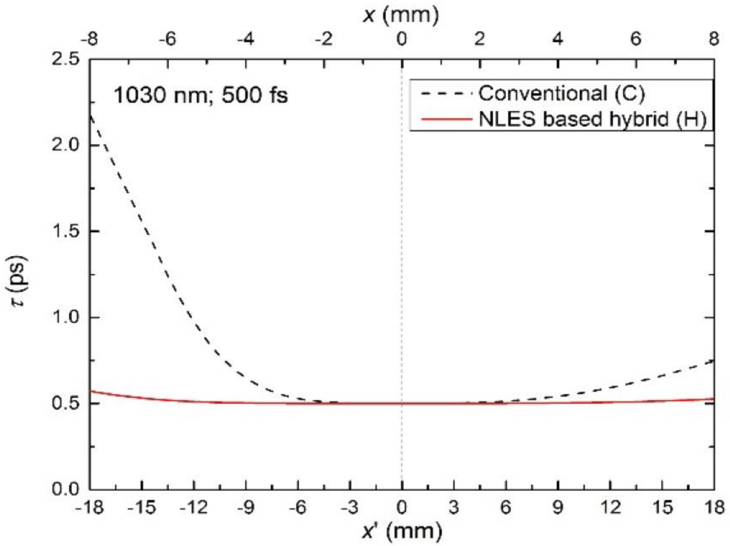


Preliminary results: 1.2 μJ THz

η = 0.06 % at 300 K

Perfect single-cycle THz pulse shape





1 mJ THz predicted at T = 100 K for 500 fs, 100 mJ pump from 1 inch diameter LN with η = 1 %

L. Pálfalvi et al.: Opt. Express **25,** 29560 (2017)

P.S. Nugraha et al.: EOS-TST 2018, today 15.05

Sources of THz pulses with extremely high field strength

A. Sell et al.: Optics Lett. **33,** 2767 (2008)   OR in GaSe 100 MV/cm at 30 THz

C. Vicario et al.: Optics Lett. **39**, 6632 (2014) OR in DSTMS 40 MV/cm at 5 THz

X. Wu et al.: Optics Express 26, 7107 (2018) TPFP OR in LN 4 MV/cm at 0.5 THz

Generation of THz pulses with > 10 MV/cm on the 0.3 – 3 THz range by CG TPFP or hybrid TPFP schemes is expected in the near future

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

E-acceleration in gas jet:

Z. Tibai et al., Journal Physics B: At. Mol. Opt. Phys. 51 134004 (8pp) (2018)

Sz. Turnár et al., Applied Physics B, 127, 38 (2021)

E-acceleration in waveguide:

D. Zhang et al., Phys. Rev. X 10, 011067 (2020)

D. Zhang, et al., Optica 6, 872-877 (2019)

THz linacs:

E. A. Nanny et al.: Nature Comm. **6**, 8486 (2015)

H. Xu et al., Nat. Photonics 15, 426–430 (2021)

Inverse free electron laser (IFEL):

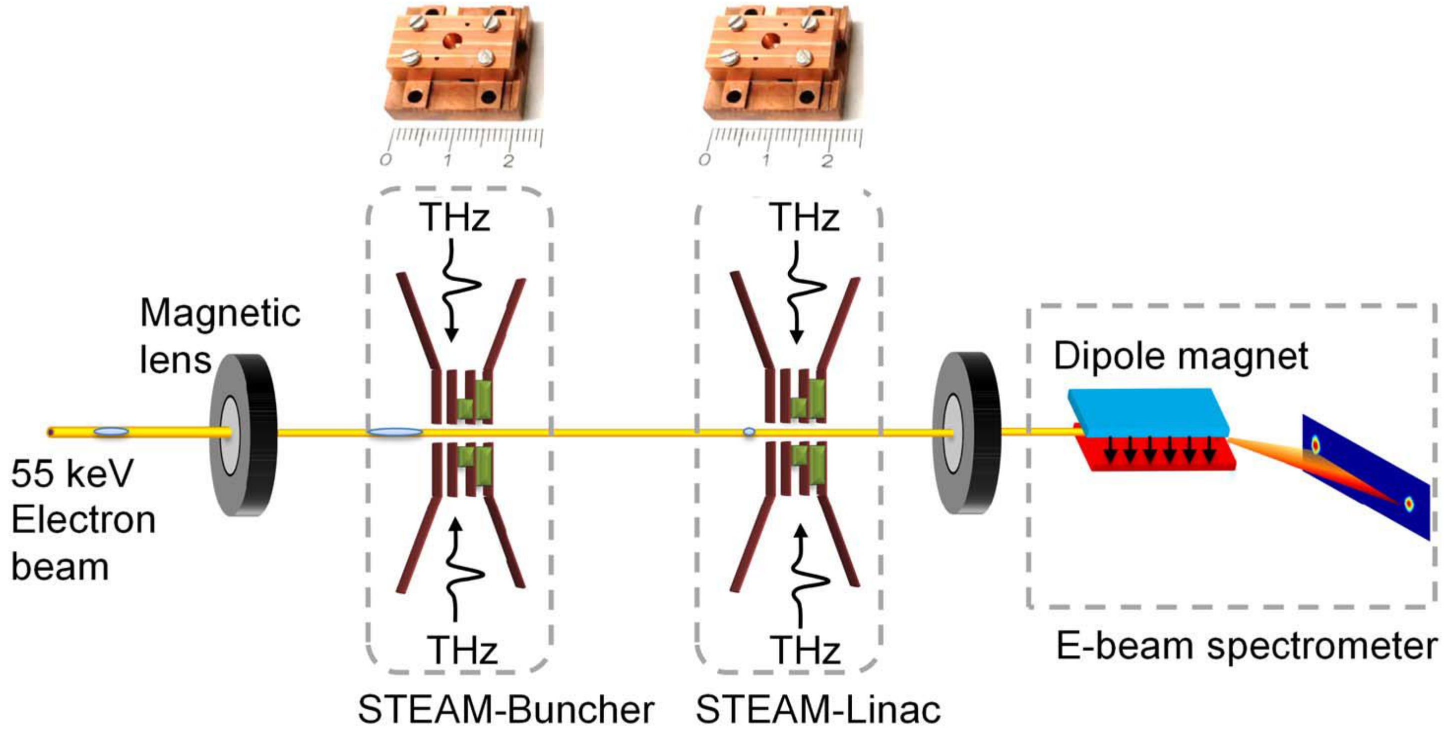
E. Curry et al.: Phys. Rev. Lett. **120**, 094801 (2018),

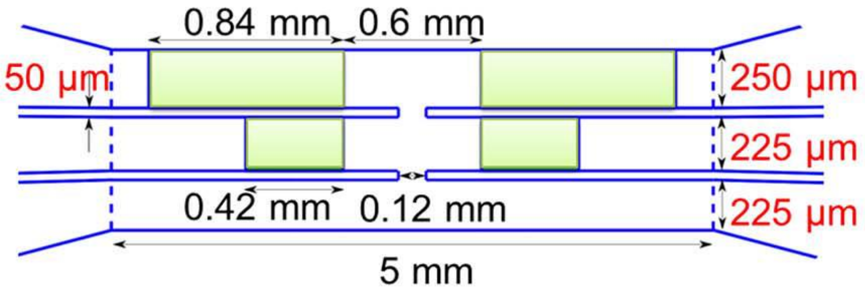
E. Curry et al., New Journal of Physics 18(11), 113045 (2016)

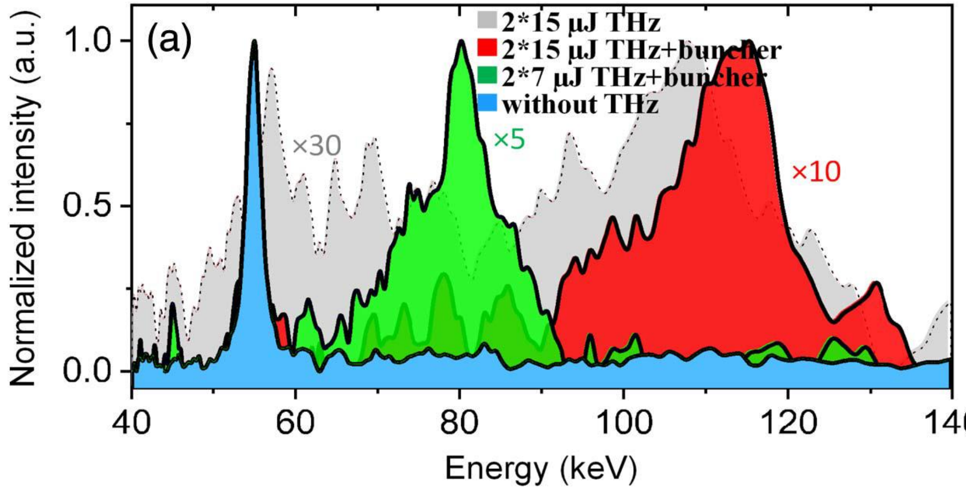
4., Carrier-envelop-phase (CEP) stable attosecond pulse generation

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

Segmented-terahertz-electron-accelerator-and-manipulator (STEAM)

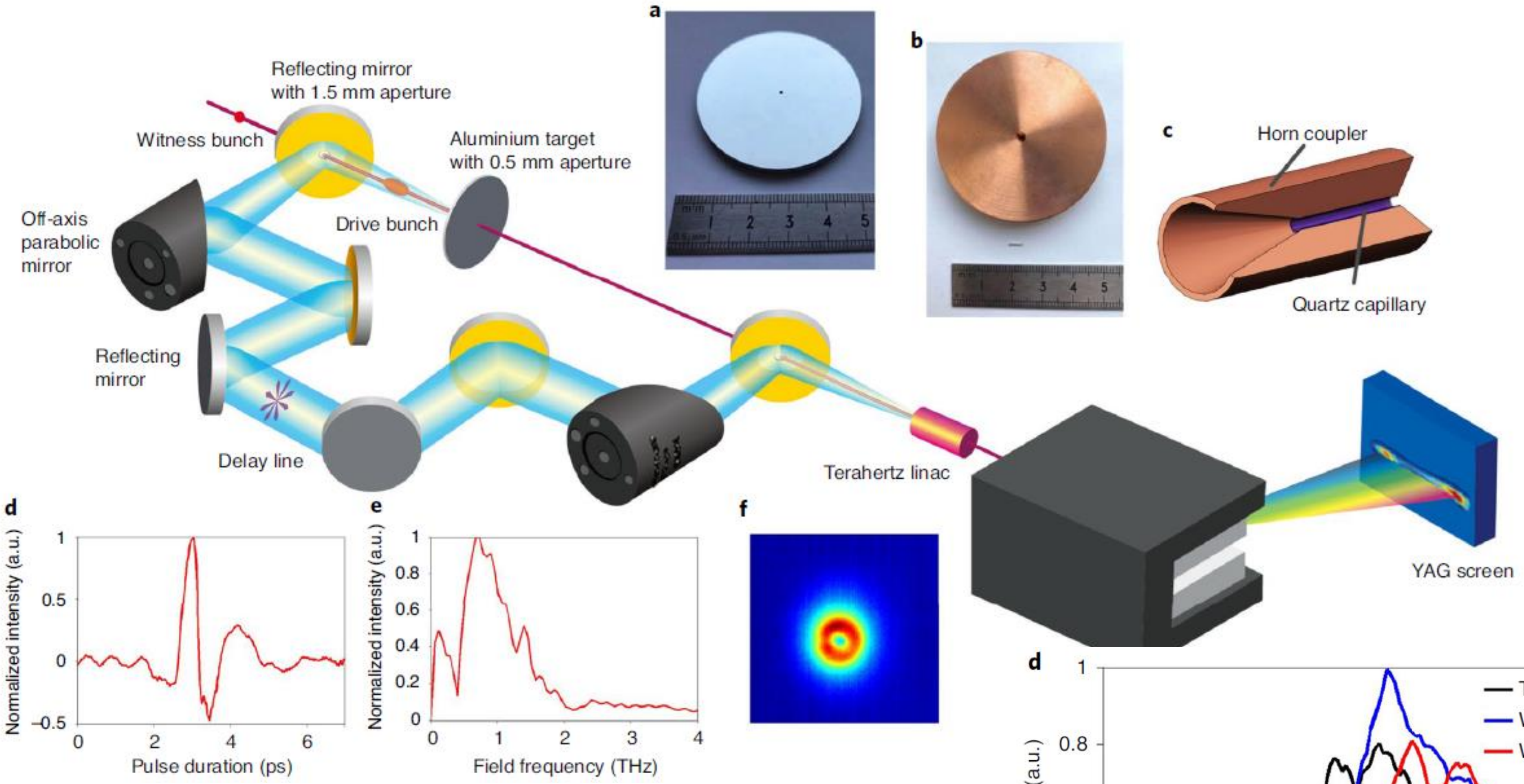


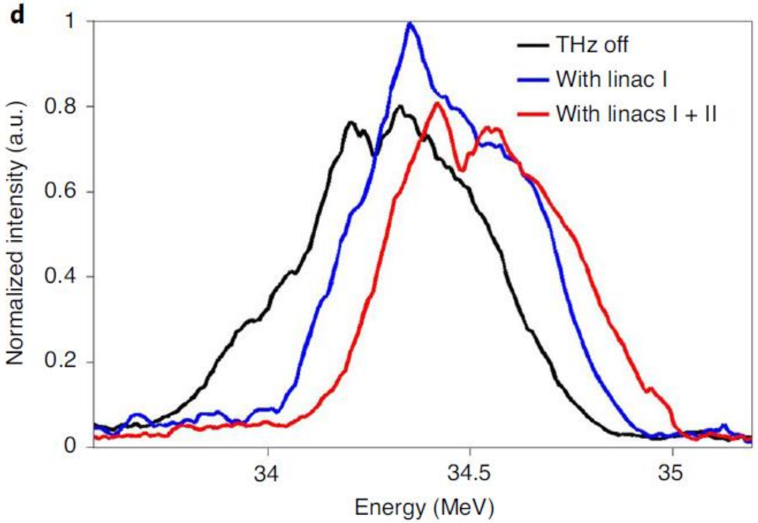




D. Zhang, et al., Optica 6, 872-877 (2019)

THz linac II





E_{THz} = 1.1 GV/m

85 MV/m average accelerating gradient

H. Xu et al., Nat. Photonics 15, 426–430 (2021)

TWAC – Multicycle THz source design

Slides from
Gy. Tóth (Gyuri)

Pump laser paramete

FL pulse duration: 40 fs
Central wavelength: 800 nm
Pump energy: max. 1.5 J

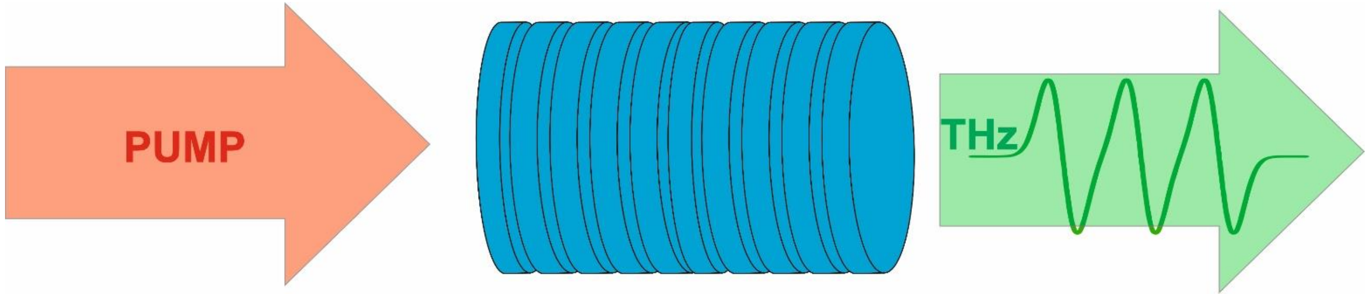
THz pulse paramete

Central frequency: 0.165 THz
Number of cycles: 10
Energy: as high as we can

Needed wafer structure

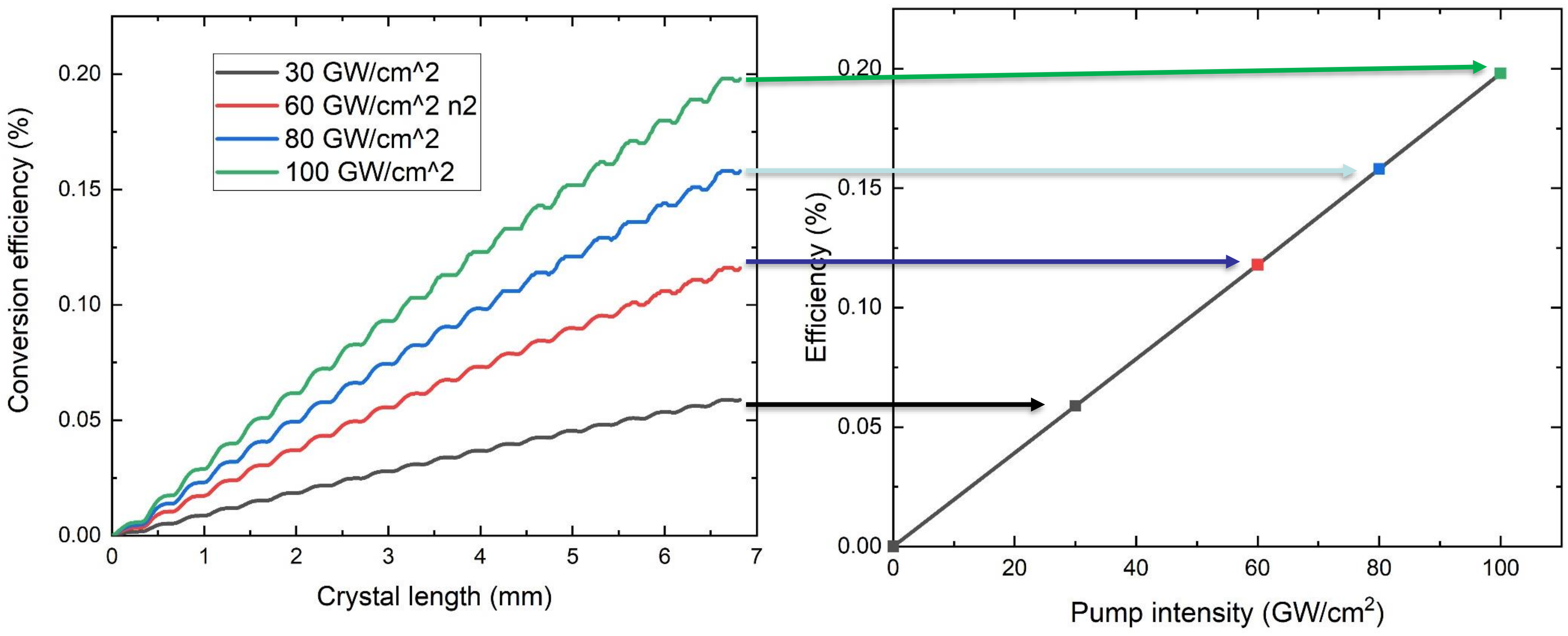
$$\nu = \frac{2c}{L\left(n_{THz} - n_{IR}^{gr}\right)}$$

Wafer thickness: 336 µm
Number of wafer: 20
Length of the structure: 6.7 mm

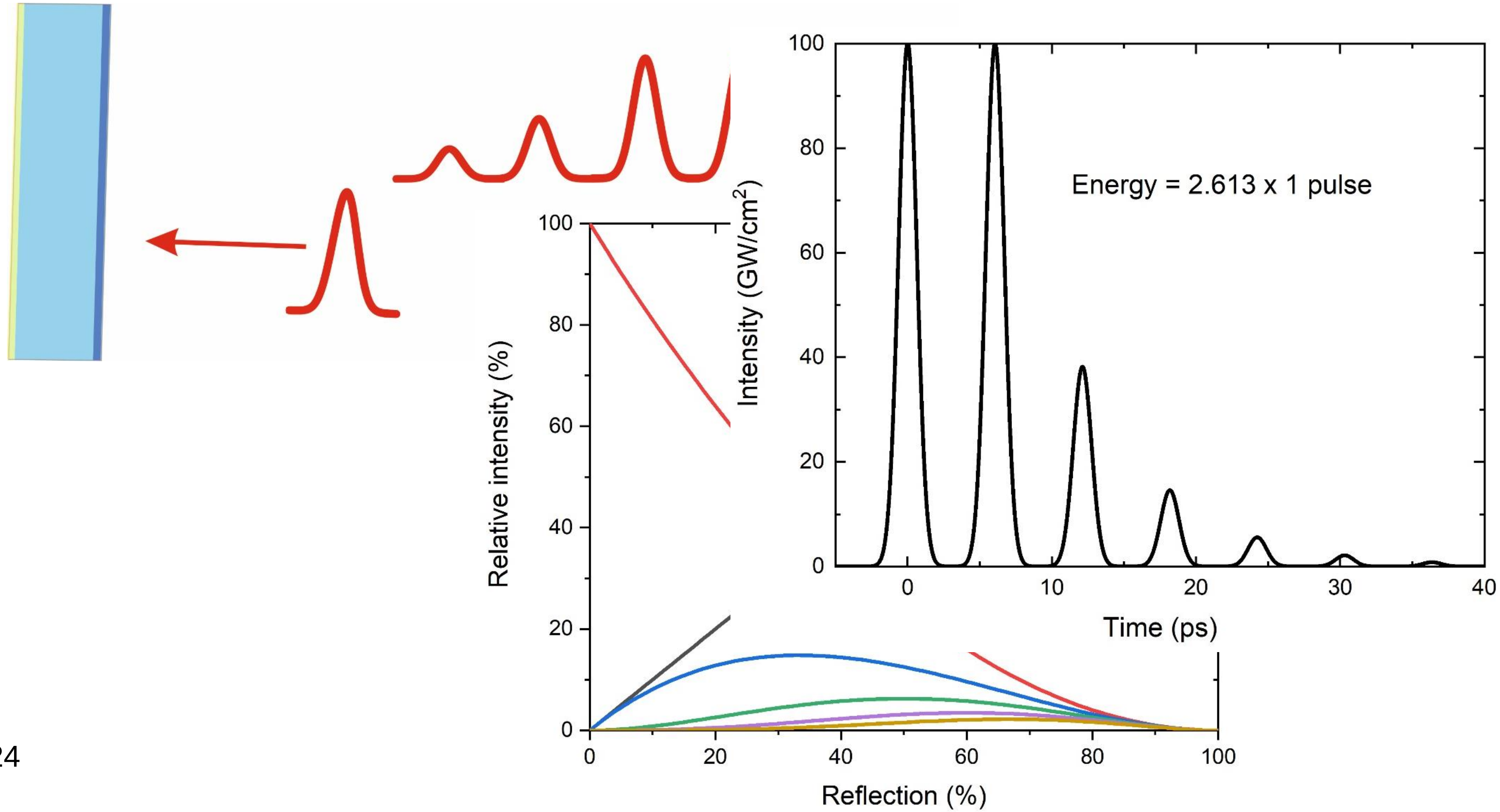


TWAC – THz source

Investigation of the intensity depende



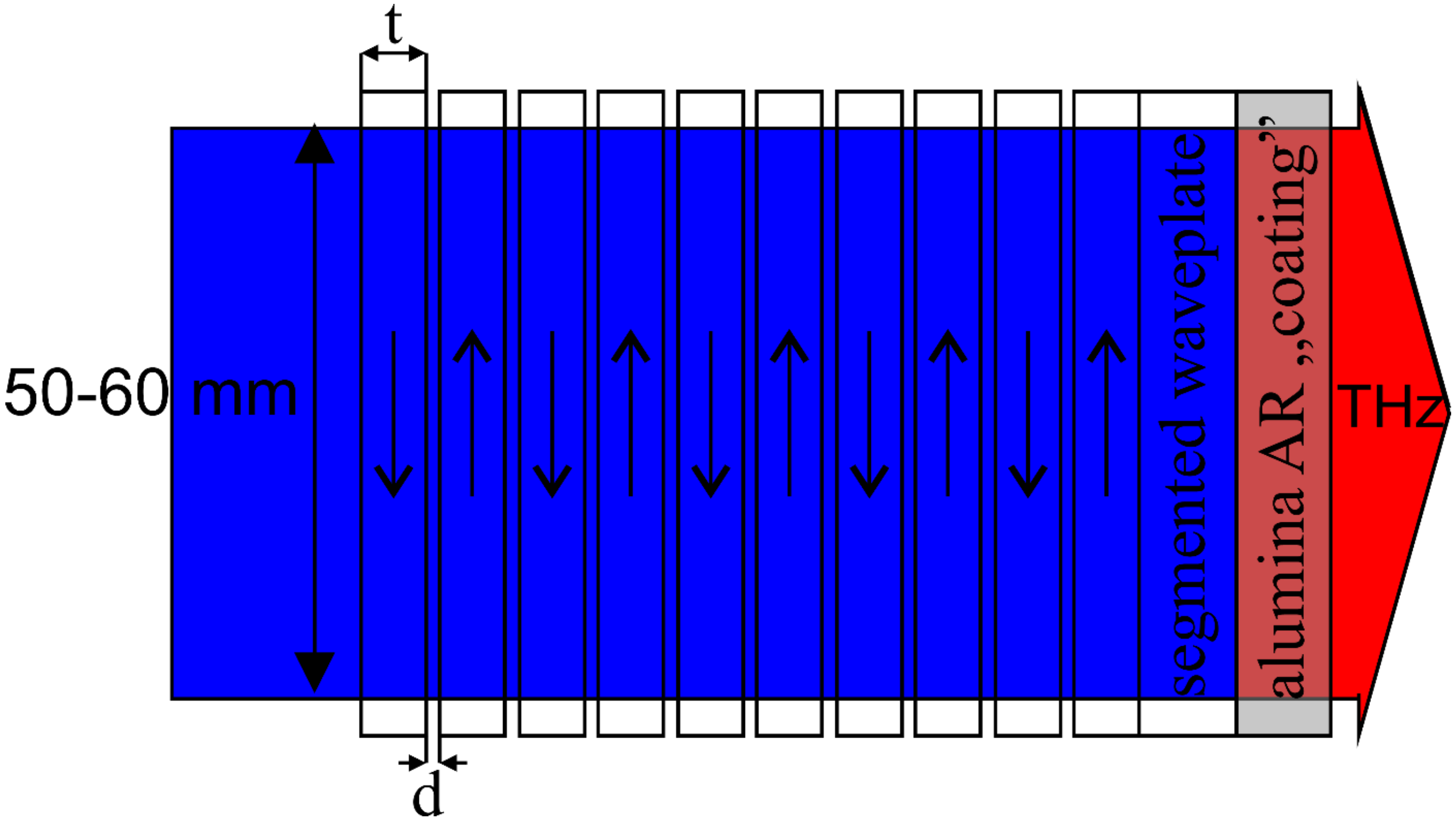
Gires-Tournois interferometer for multipulse gener



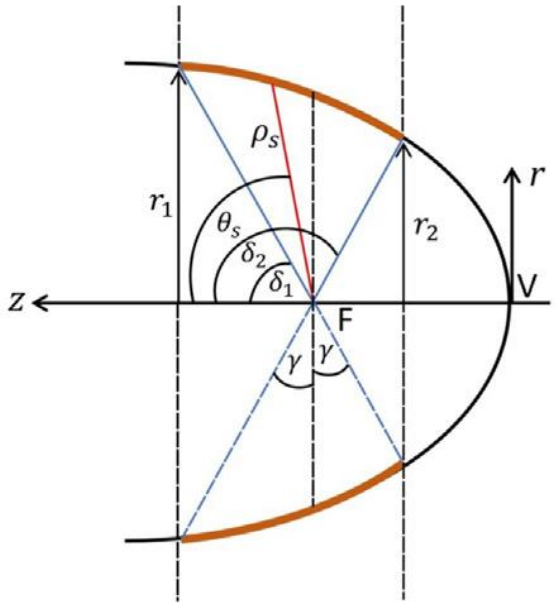
TWAC – THz source realization

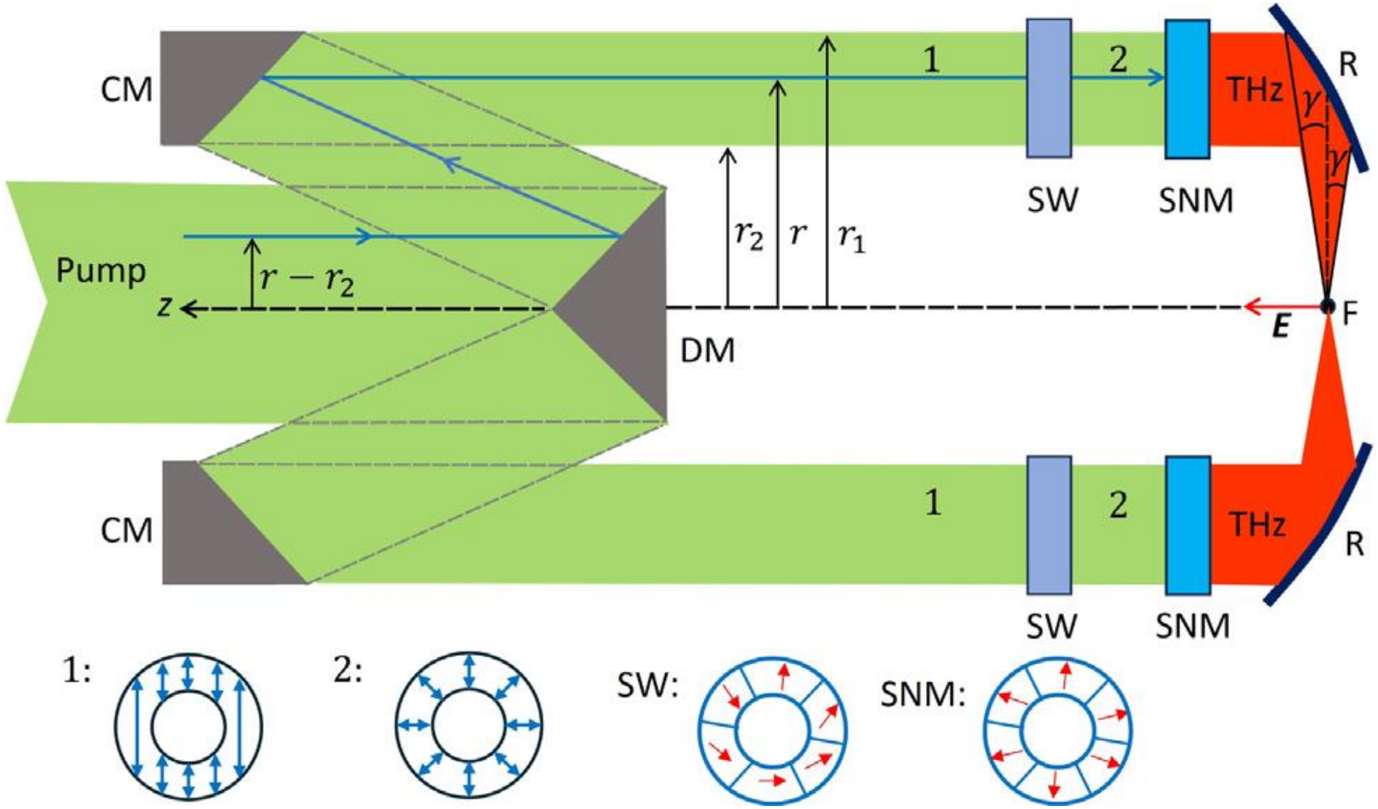
Radially polarized THz pulses are needed!

For linear-to-radial polarization conversion a segmented THz HWP will be used



Considered focusing set-ups





- L. Pálfalvi, Z.T. Godana, Gy. Tóth, J.Hebling: Generation of extremely strong accelerating electric field by focusing radially polarized THz pulses with a paraboloid ring
  Opt. & Las. Techn. **180**, 111554 (2025)

Summary

- TPFP LN is a highly efficient and widely used source of high energy , high field single-cycle THz pulses (appl. in THz pump – probe and control experiments)
- Both fundamental and special limitations hindering generation of THz pulses with mJ level energy and few MV/cm peak field strength by TPFP LN is identified.
- Solutions for mitigate or eliminate these limitations has been suggested or demonstrated. (Semiconductor CG pumped at wavelength not allowing 3-photon-absorption (η = 0.7% demonstrated, η > 2% predicted), hybrid-CG, hybrid-Echellon grating LN.) Generation of single-cycle THz pulses with extremely high field by TPFP sources are foreseen.
- Many important application possibilities of THz pulses with extremely high field strength generated by TPFP, GaSe or organic NC identified. A few examples are:
  - Enhancement of HHG
  - Field-free orientation of molecules
  - Acceleration and other manipulation of charged particles
  - Carrier-envelop-phase (CEP) stable attosecond pulse generation