On Extracting the Maximum Terahertz Conversion Efficiency from Optical Rectification in Lithium Niobate

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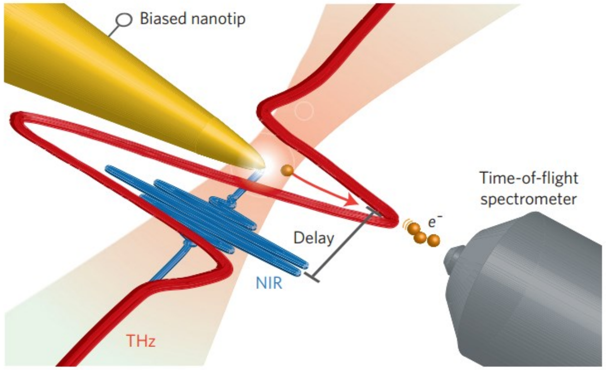
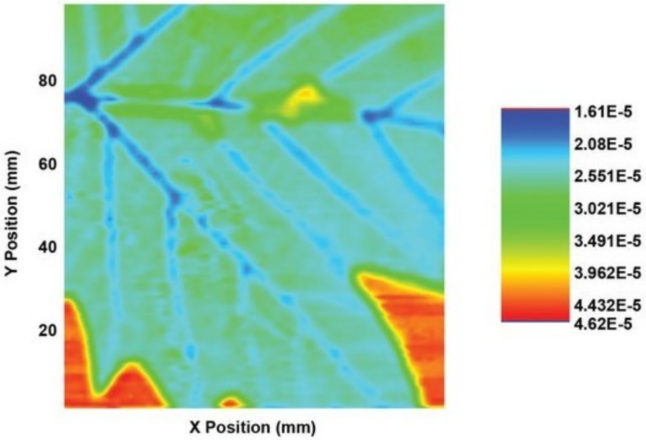
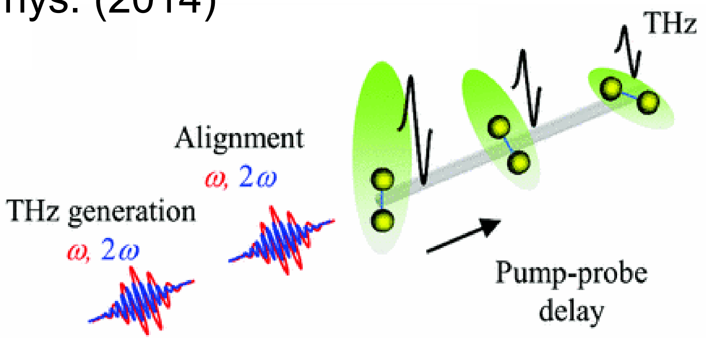
Motivation for efficient high-power terahertz generation



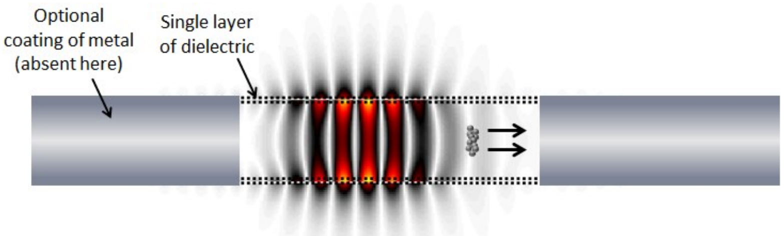
Wimmer, Nat. Phys. (2014)



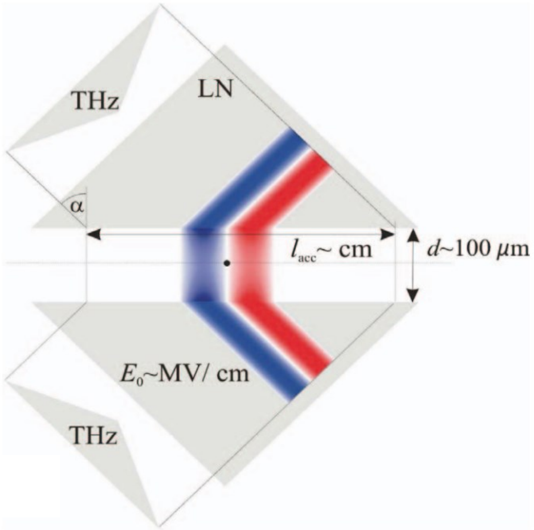
Jepsen, L&P Rev **5** (2011)



Fleischer, PRL **107** (2011)

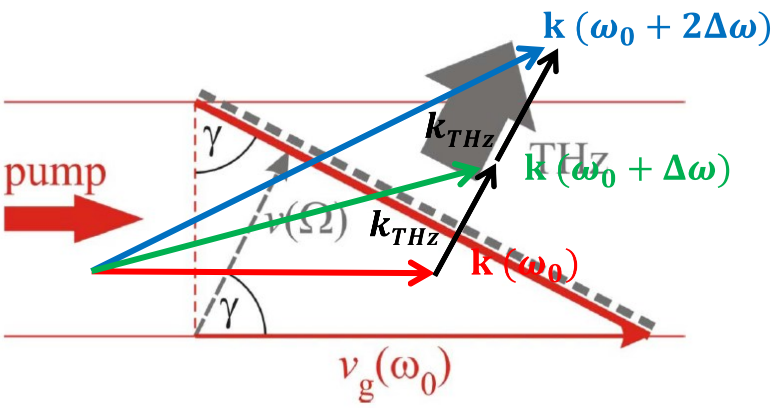


Wong, Opt. Exp. **21** (2013)



Palfalvi, Phys. Rev. STAB **17** (2014)

Tilted-pulse-front (TPF) in $LiNbO_3$ for terahertz generation



Phase-matching: tilted-pulse-front

$$k(\omega_0) - k(\omega_0 + \Delta\omega) = k_{THz}$$

$$v_{g,opt}\cos(\gamma) = v_{THz}$$

Attractive approach due to

- Relatively efficient: **%-level**
- Strong pumping: **>100 mJ**
- High repetition rate: **>1 kHz**

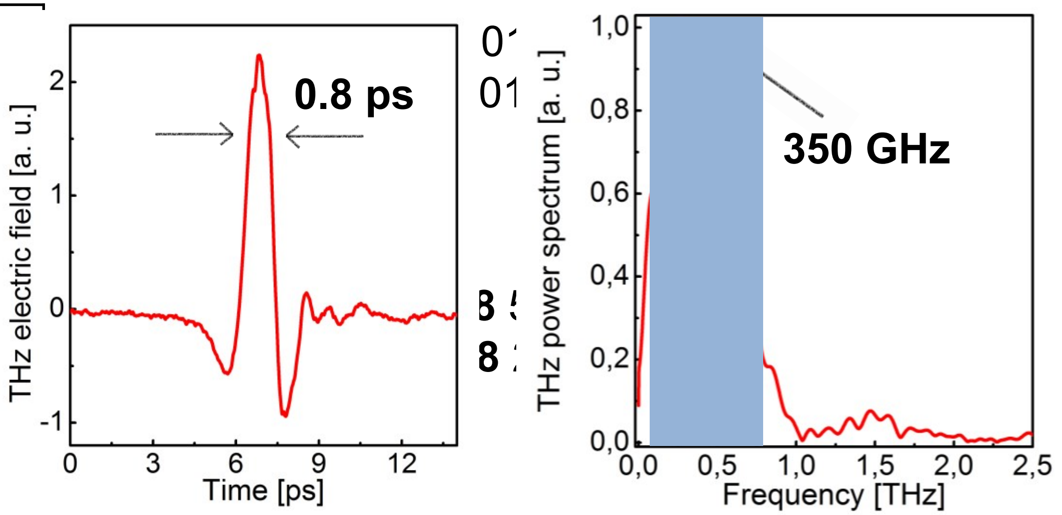
Theoretical efficiencies

10%

Experimental

~1%

Sub-cycle transients in 0.1-1 THz range



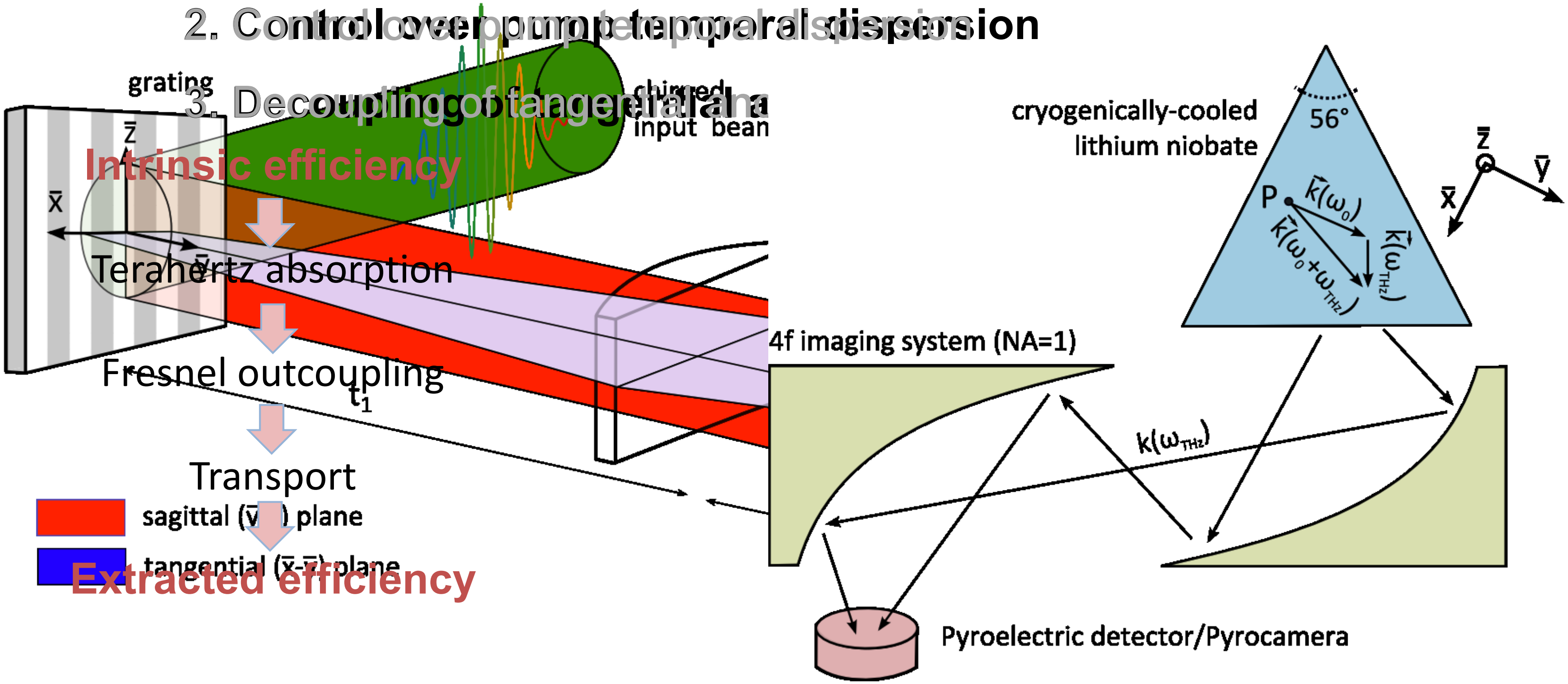
0
01
3 5
8 2

Hebling, Opt. Express **10** 1161 (2002)
Hoffmann, J. Phys. D: Appl. Phys. **44** (2011)

General Strategy to Increasing Extraction

Key aspects to a record extracted 2% conversion efficiency in lithium niobate from a Yb:KYW laser system at 1030 nm wavelength

1. **Cryogenic-cooling of the crystal for lower linear absorption**
2. **Control over pump temporal dispersion**
3. **Decoupling of tangential a**



Intrinsic efficiency

Terahertz absorption

Fresnel outcoupling

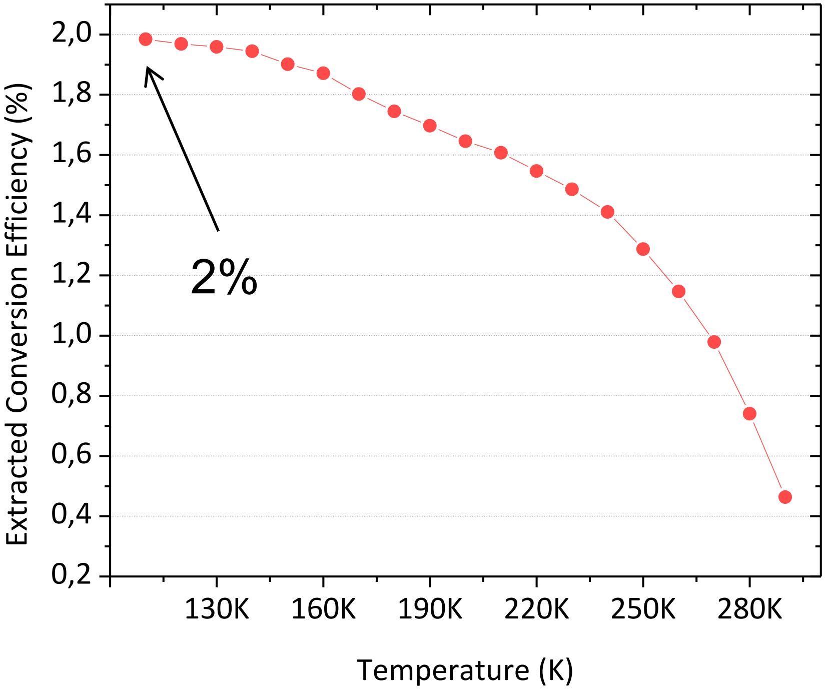
Transport

Extracted efficiency

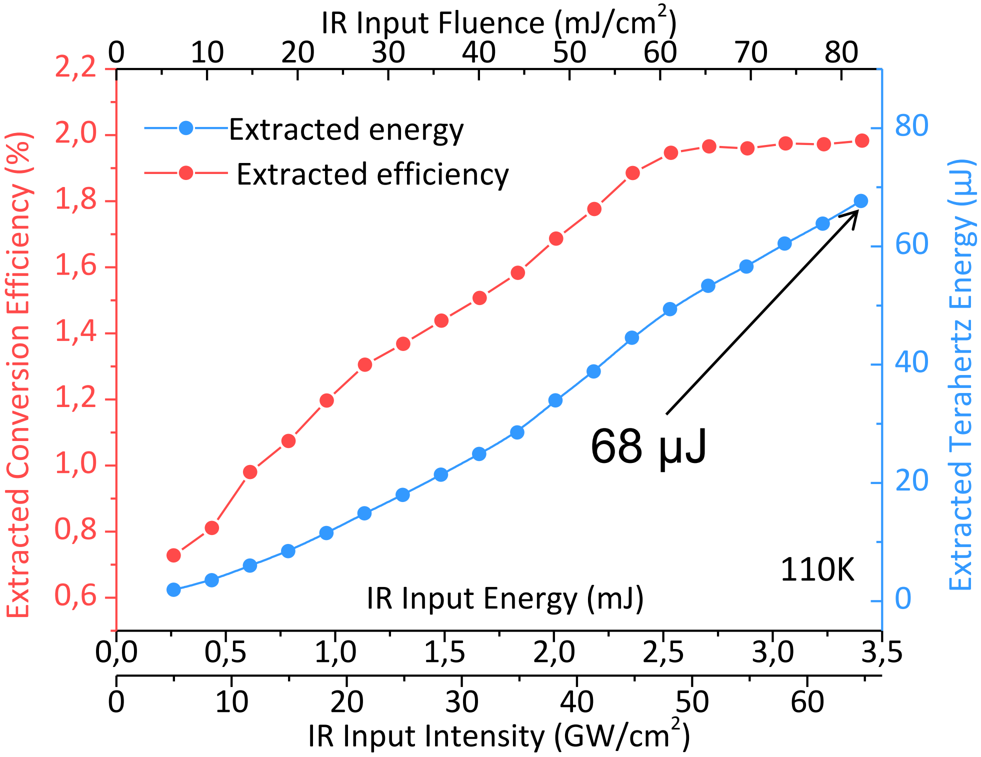
Reduced Linear Absorption at Cryogenic Temperatures

Results under moderate pump conditions: **ps-duration mJ-level** optical pulses

Cryogenically-cooling lithium niobate reduces linear terahertz absorption

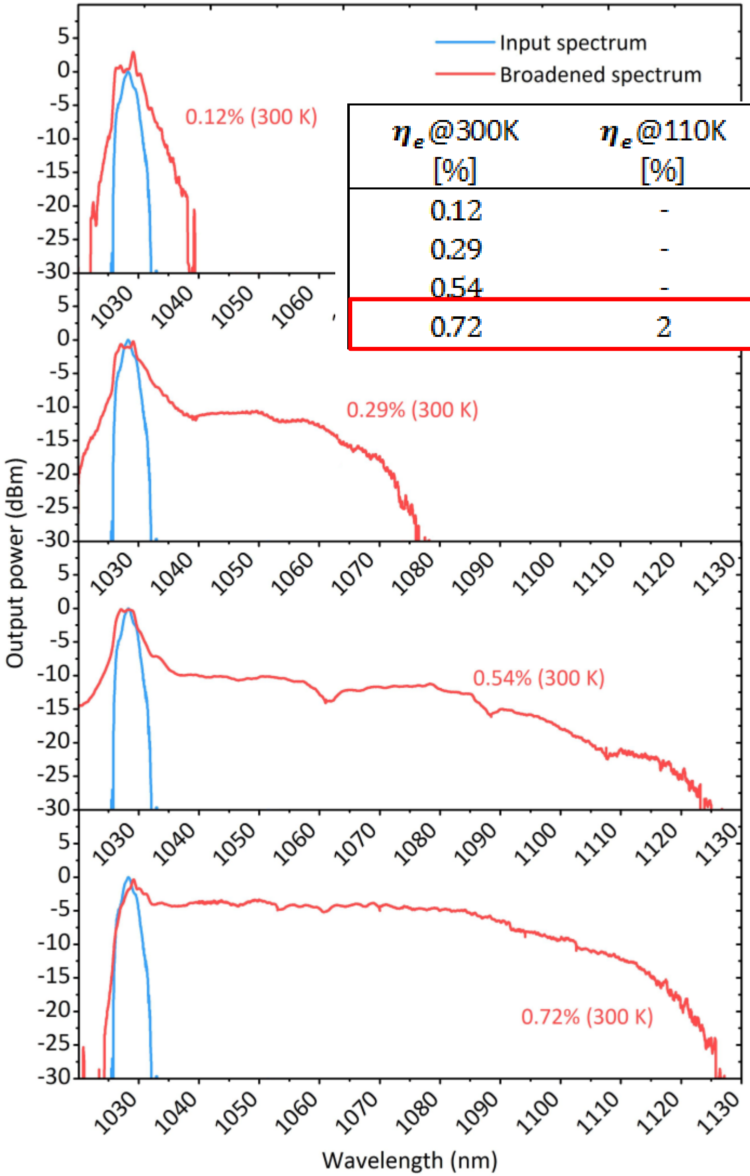


The extracted efficiency saturates at around 50 GW/cm^2 at 1 µm wavelength pump



A.L. Calendron et al., Opt. Lett. 22 (2014)

Efficiency verification from spectral broadening



η_e@300K [%]	η_e@110K [%]	N	λ_2 [nm]	η_i [%]	$\alpha_{0.3\text{THz}}$@300K/110K [cm^{-1}]	C [%]	T [%]
0.12	-	2*	1028.9	0.21*	-	18	5*
0.29	-	3*	1030.2	0.31*	-		
0.54	-	6*	1033	0.62*	-		
0.72	2	27*	1057.7	2.77*	10.97†/0.37‡		

**Estimated*
† Assumes 1 mm eff. length
‡ Assumes 2 mm eff. length

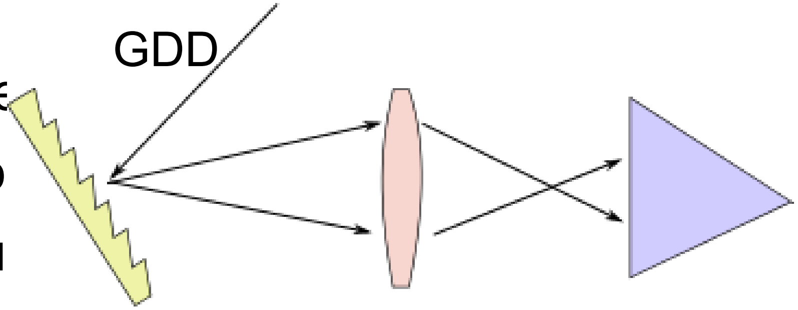
Number of Cascaded Cycles

$$N = \left({}^{c}/_{\nu_p}\right)\left(\lambda_1^{-1} - \lambda_2^{-1}\right)$$

Corresponding Intrinsic Efficiency

$$\eta_i \cong {}^{N\,h\nu_{THz}}\Big/_{h\nu_{opt}} \approx N\,10^{-3}$$

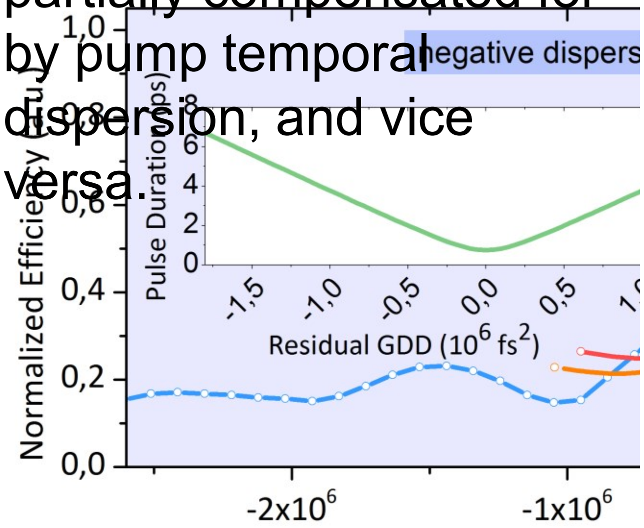
Influence of Temporal Chirp

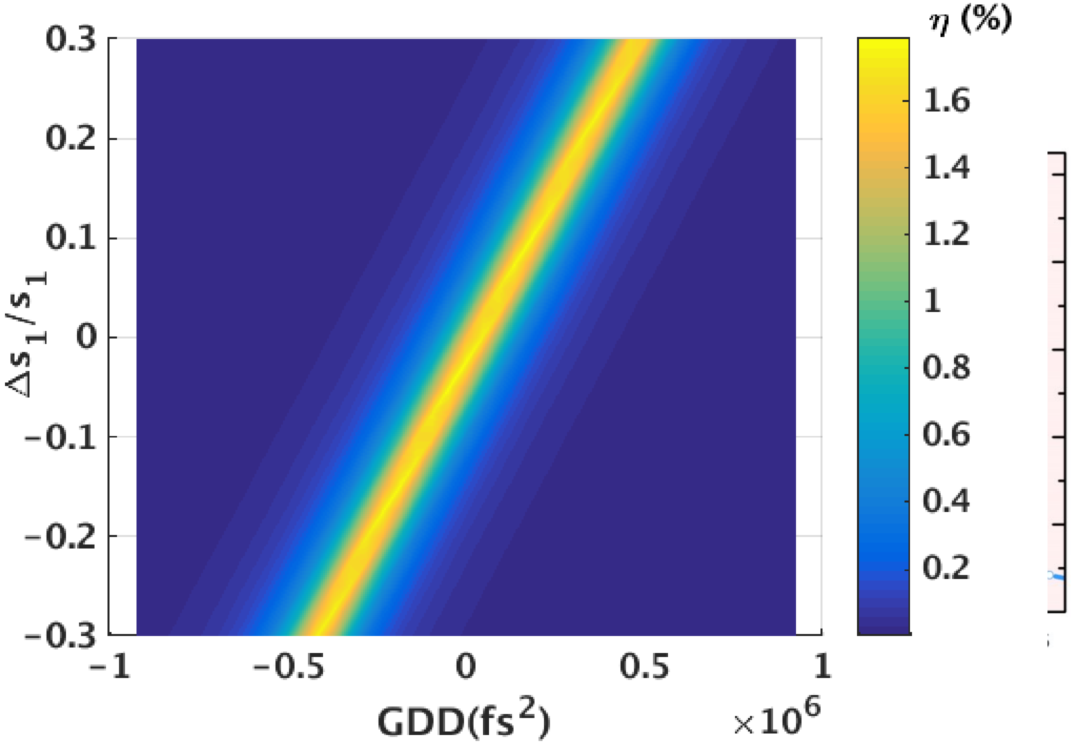


1. **Consensus:** compre
2. **Changing the temp**

- Causes the tilted pu
- Adds additional dispersion to s_1 to the system
- Will temporally

Example: Imperfect lens positioning can be partially compensated for by pump temporal dispersion, and vice versa.





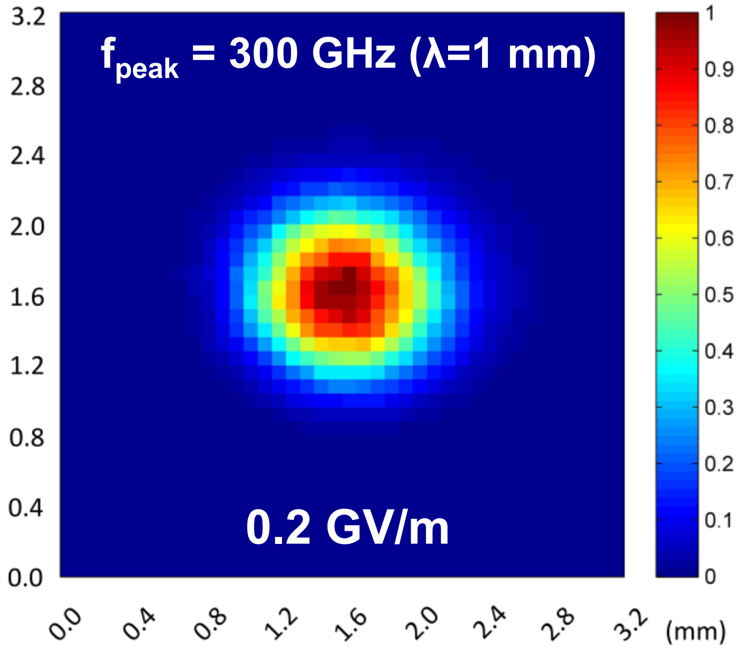
S. Akturk et al., Opt. Expr.19 (2004)

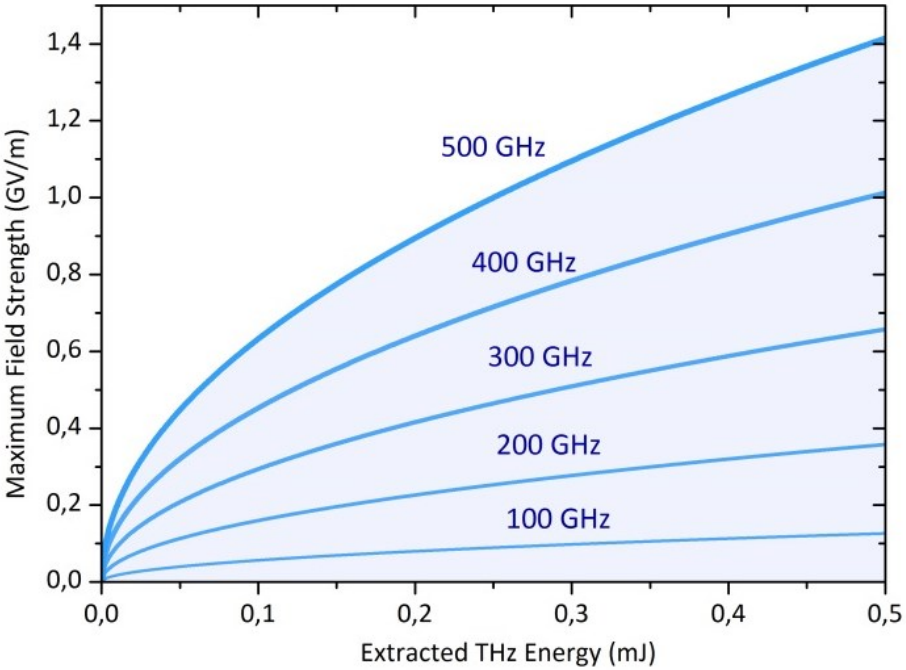
Towards GV/m Pulsed Field Strengths

Tangential and sagittal focusing decoupling of pump grants:

1. Decoupled phase-matching (tangential) and intensity (sagittal) tuning
2. High quality diffraction-limited Gaussian pulses

Scaling of diffraction-limited THz transients $\mathrm{E} \approx \frac{53.66}{c} \nu_p^{3/2} \sqrt{\epsilon}$





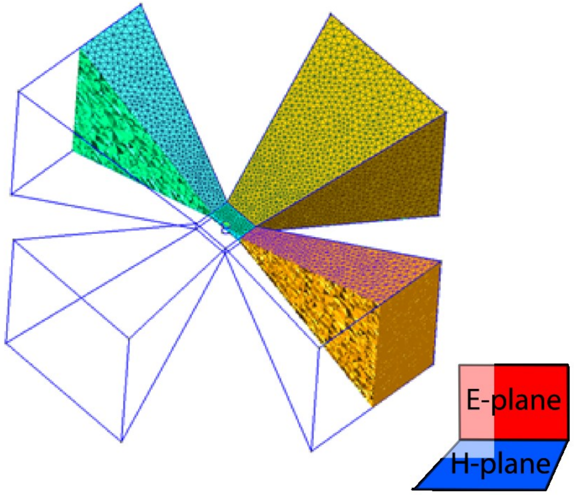
Summary

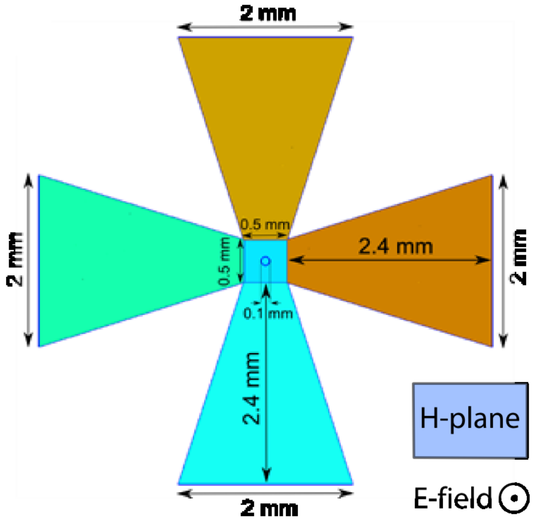
- **Refined** the method for **high-quality highly-efficient terahertz transients** in the **sub-THz range**

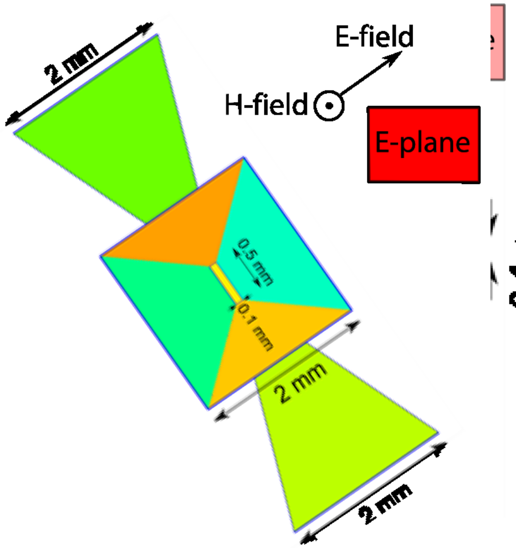
- **2% extracted efficiencies** are demonstrated from a generalized optimization process in **cryogenically cooled** lithium niobate exploiting **temporal and spatial beam shaping**

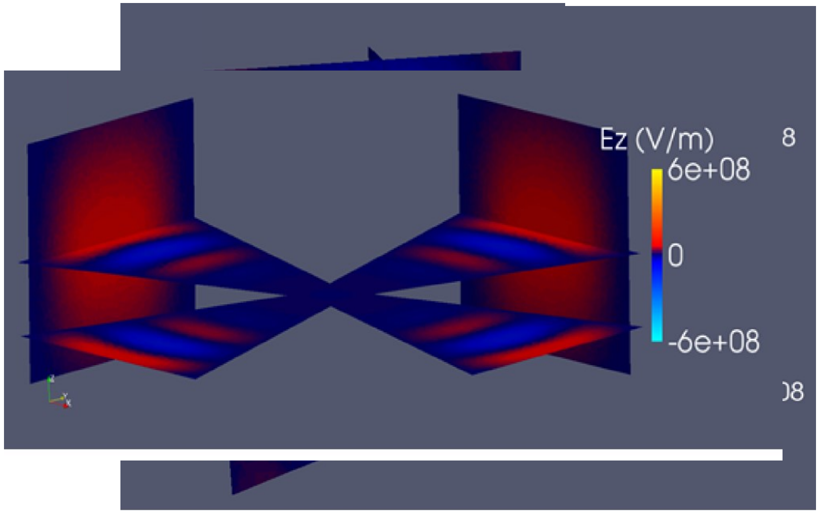
- Shown how to produce **GV/m-range** fields (approaching material breakdown limit) for nonlinear ultrafast THz science and technology using **mJ-level ps-duration optical pulses.**

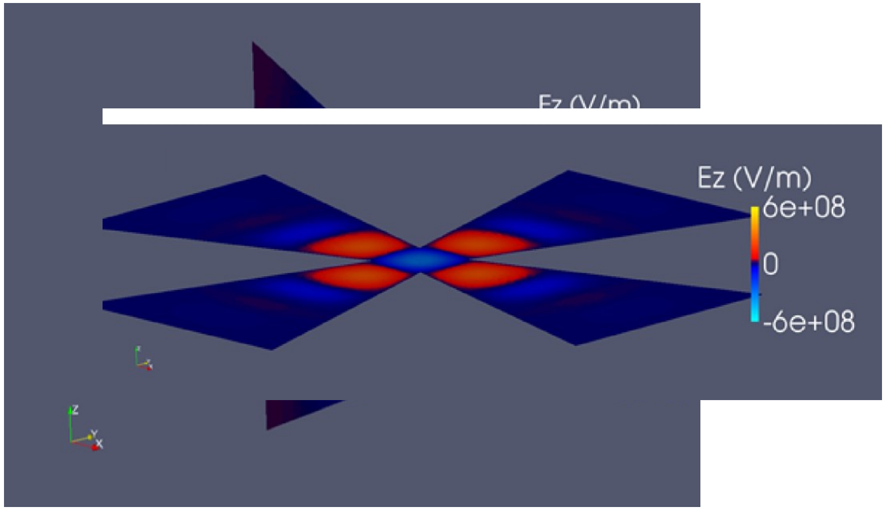
Focusing Structures for GV/m gradients







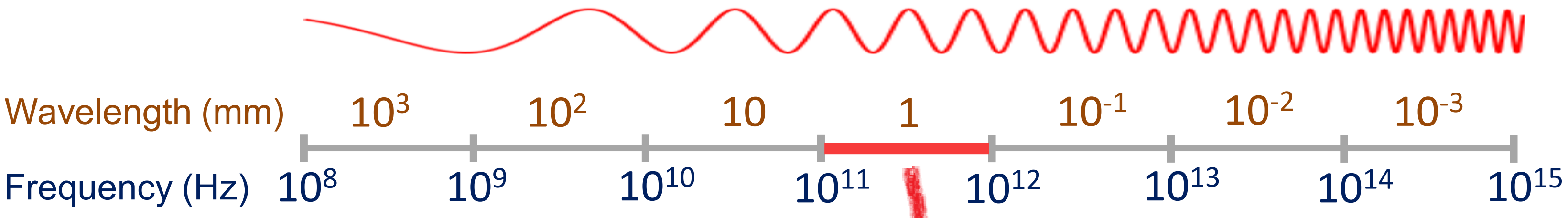


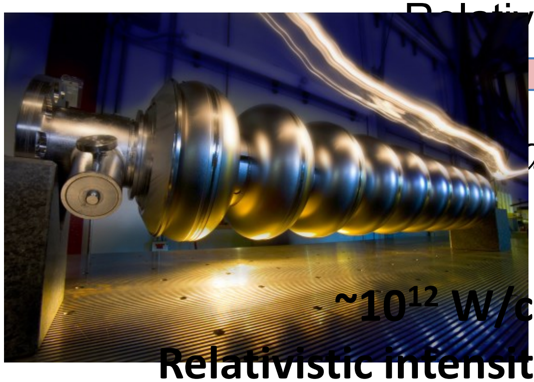


Motivation for Relativistic mm-wavelength Transients

- **Ultrafast and strong-field physical sciences:** structural dynamics, nonlinear terahertz spectroscopy, charged-particle acceleration, among others.

- **mm-wavelength relativistic transients** for particle acceleration





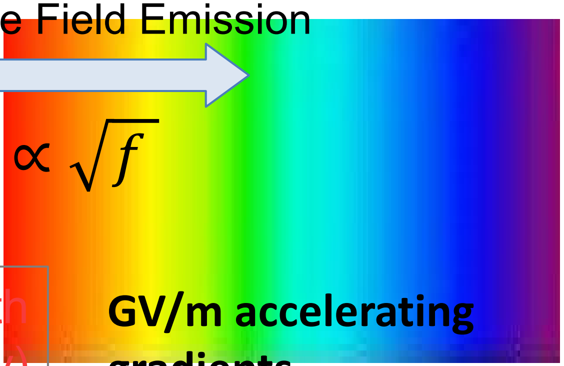
Relativistic Intensities

$a_0 \propto \lambda\sqrt{I}$

~10^{12} W/cm²
Relativistic intensities

0.3-3 mm wavelength
(0.1-1 THz frequency)

Surface Field Emission

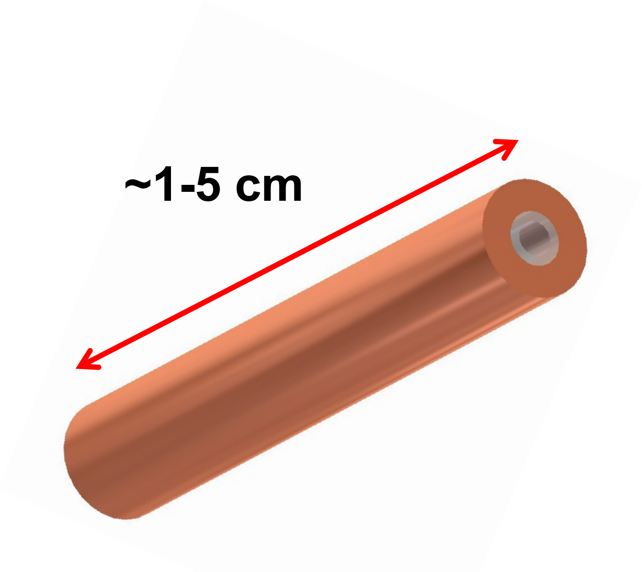


$E_{th} \propto \sqrt{f}$

GV/m accelerating gradients

Terahertz Accelerator in Dielectrically Loaded Metallic Waveguide

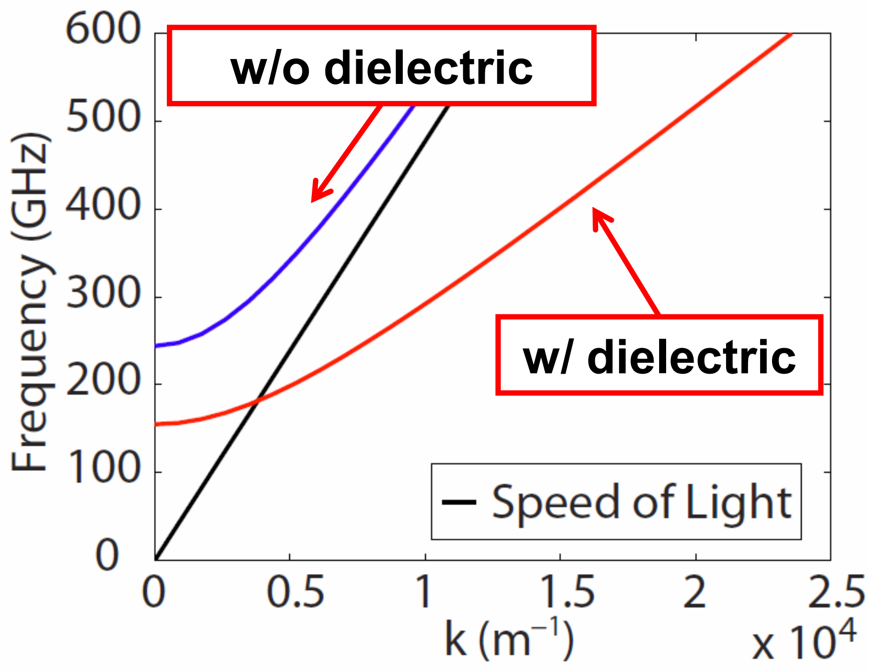
- First THz accelerator with **5 MV/m** on-axis gradients
- **Traveling way structure**: broad-band single cycle pulses
- **Phase-group-velocity matching**: THz-phase to electron velocity with thickness of dielectric



Copper Inner Diameter = 940 µm

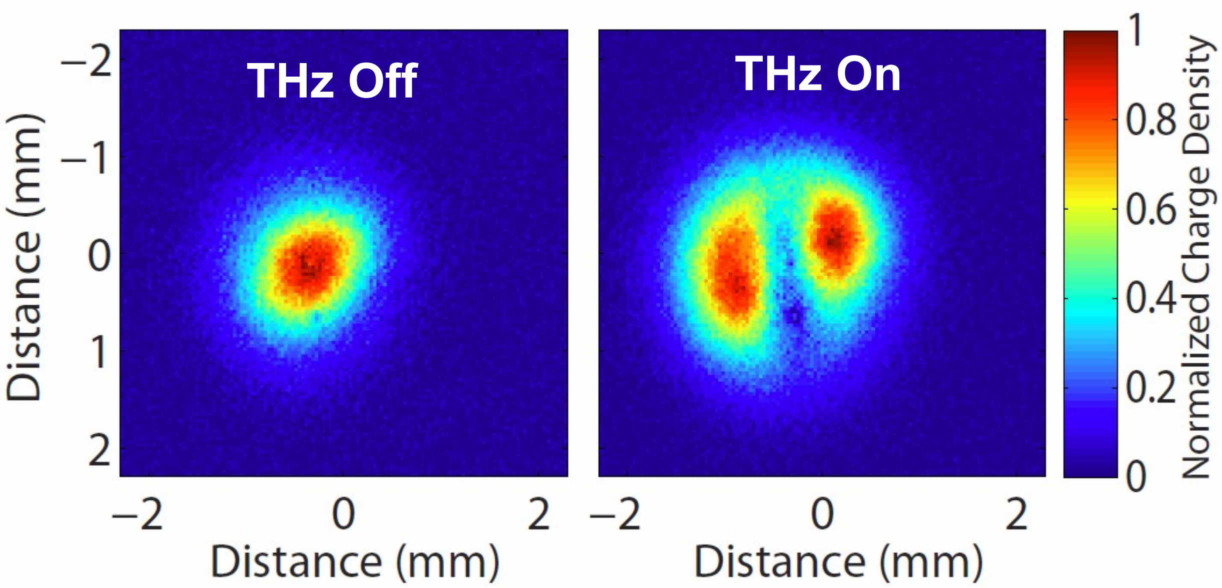
Fused Silica Inner Diameter = 400 µm

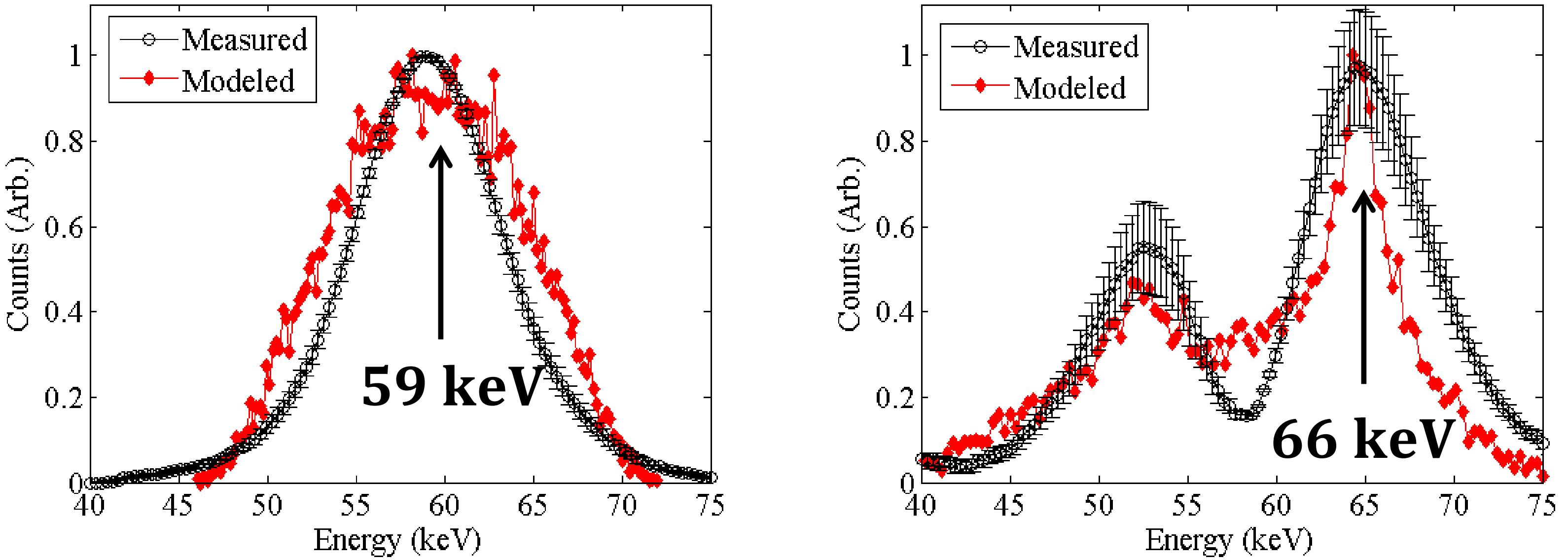
Dispersion Relation



E. Nanni et al., arXiv:1411.4709 (2014)

Terahertz Accelerator in Dielectrically Loaded Metallic Waveguide





E. Nanni et al., arXiv:1411.4709 (2014)

Saturation Mechanisms: Angular Dispersion and Cascading

Input parameters

- Fixed fluence 15 mJ/cm^2
- Temperature = 100 K
- $n_2 = 2 \cdot 10^{-15}$ cm^2/W
- $\chi^{(2)}{}_{\text{eff}} = 366$ pm/V

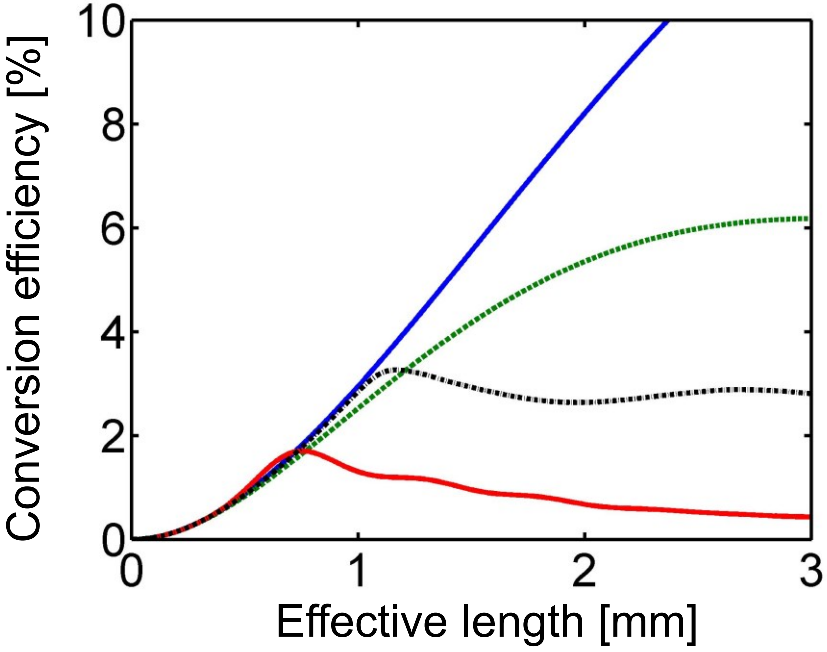
—— Absorption + Dispersion (AD)

- - - - AD + Self-phase modulation (SPM)

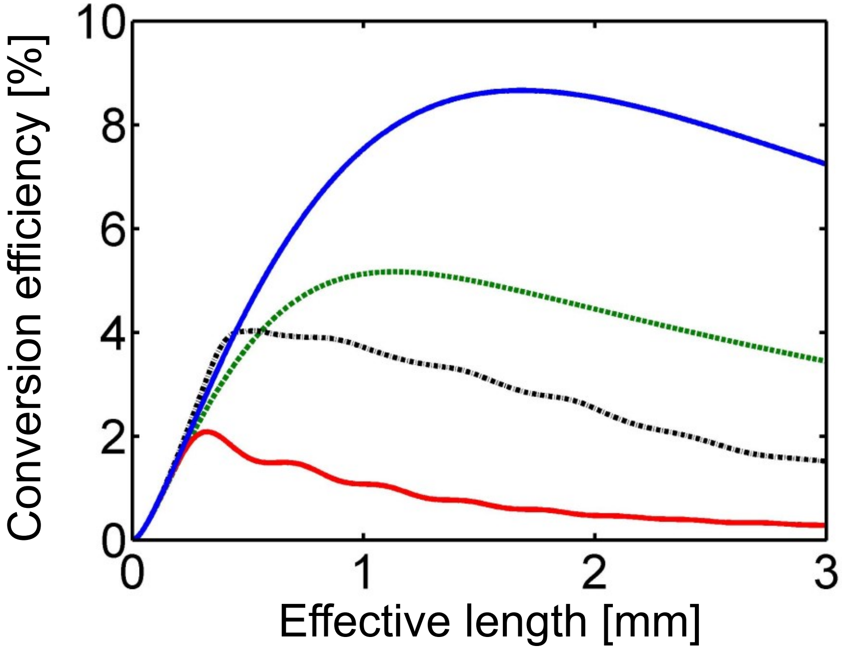
-·-·-·- AD + Cascading

—— AD + SPM + Cascading

500 fs TL duration



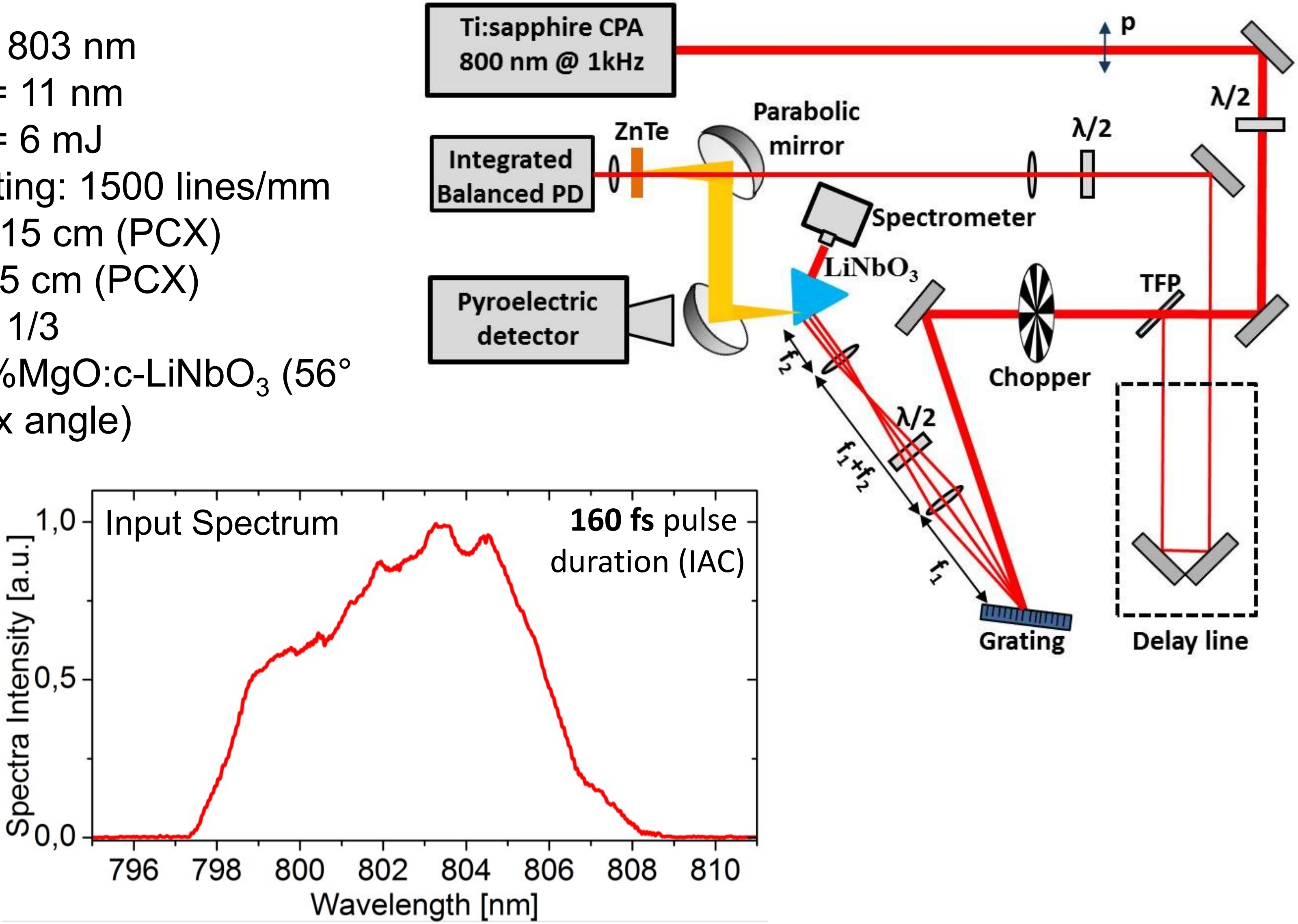
150 fs TL duration



K. Ravi et al., *“Limitations to THz generation by optical rectification using tilted pulse fronts”* (arXiv: 1406.1439)

800 nm-Pump Optical Rectification at Room Temperature

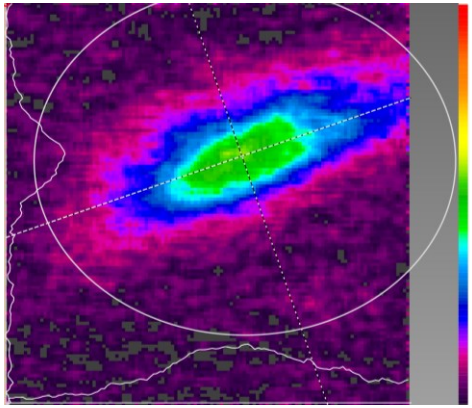
- λ_0 = 803 nm
- $\Delta\lambda$ = 11 nm
- E_{in} = 6 mJ
- Grating: 1500 lines/mm
- f_1 = 15 cm (PCX)
- f_2 = 5 cm (PCX)
- M = 1/3
- 5.6%MgO:c-$LiNbO_3$ (56° apex angle)



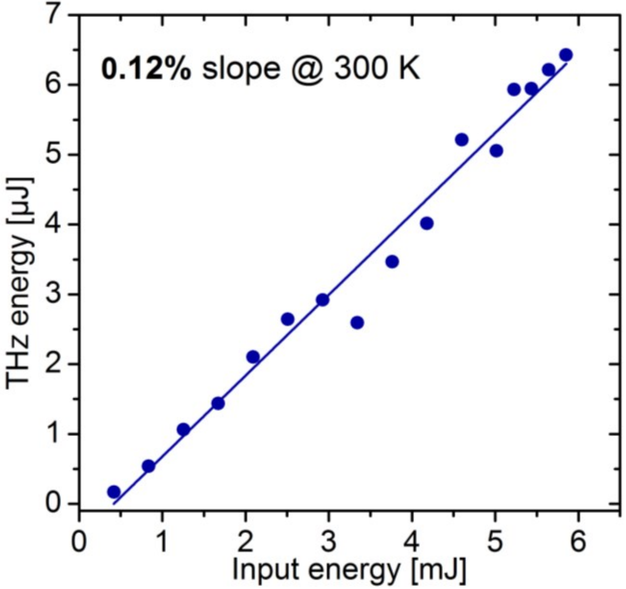
Converted Energy, Waveform, and Frequency

- E_{out} = 6.5 µJ (on the detector)
- P_{out} = 6.5 mW
- Conversion efficiency @ 300 K ~ 0.12%
- Sub-cycle electric-field waveform
- Broadband output. Peak at 0.35 THz

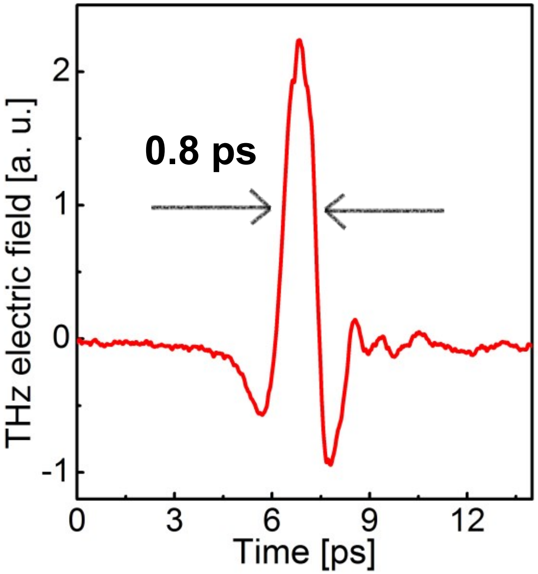
THz Beam Profile



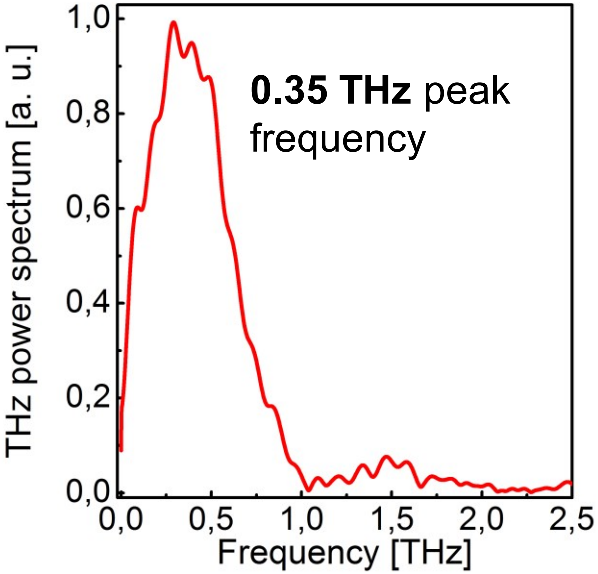
Detected Output Energy



Temporal Waveform



Terahertz Spectrum

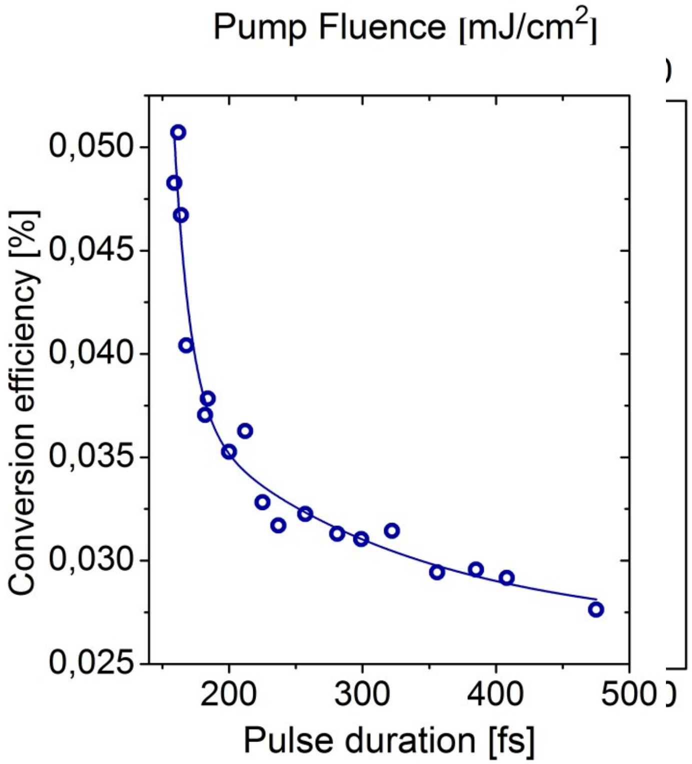


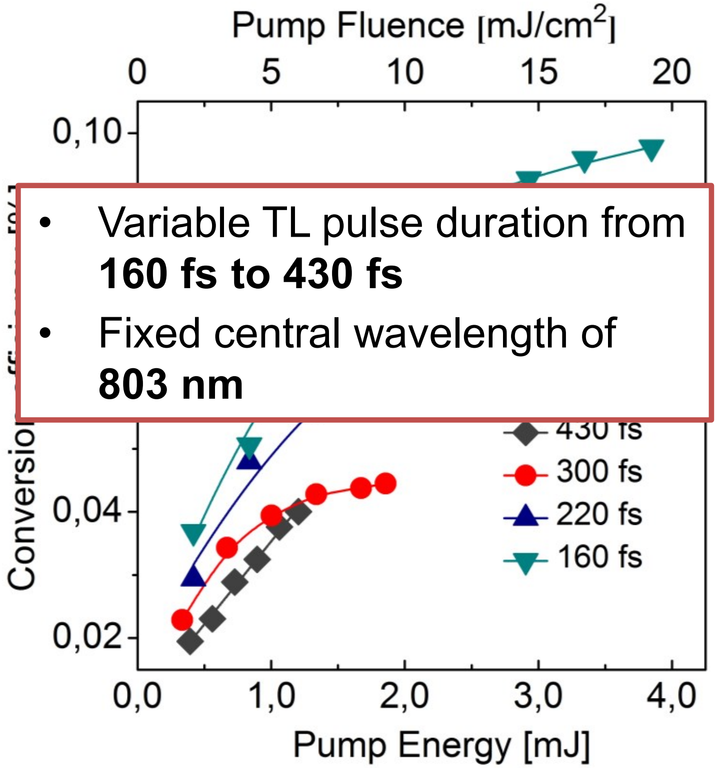
Conversion Efficiency as a Function of TL Pulse Duration

Analytical:

$$\eta_{THz} = \underbrace{\left(\frac{2\, d_{eff}^2}{\varepsilon_0 n_{opt}^2 n_{THz} c^3}\right)}_{\text{material}} \underset{\text{THz freq.}}{\Omega^2} \cdot \overset{\text{pump intensity}}{I} \cdot \underbrace{\left[L^2 \exp(-\alpha L/2)\, \frac{\sinh^2(\alpha L/4)}{(\alpha L/4)^2}\right]}_{\text{effective length}}$$

Experimental:





- Variable TL pulse duration from **160 fs to 430 fs**
- Fixed central wavelength of **803 nm**

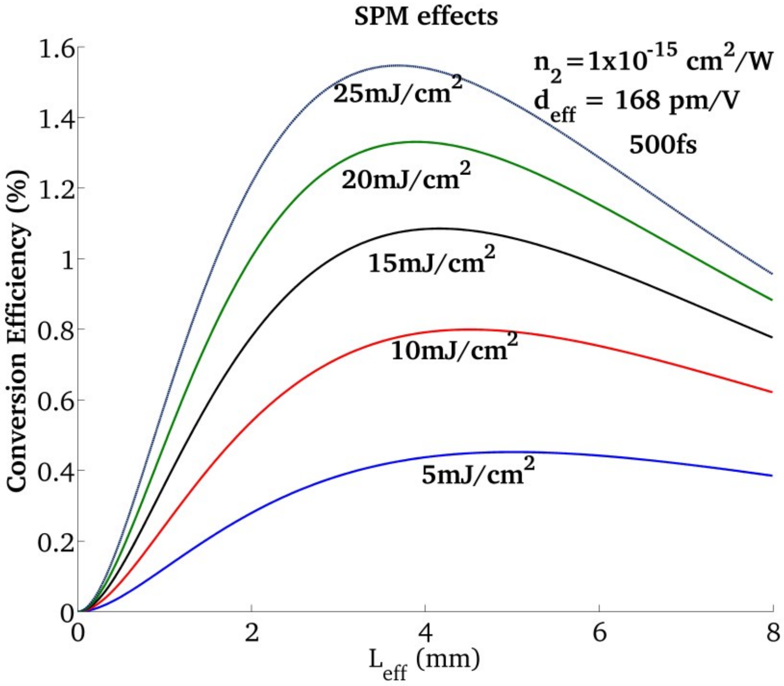
Conversion Efficiency including SPM and Cascading

Analytical:

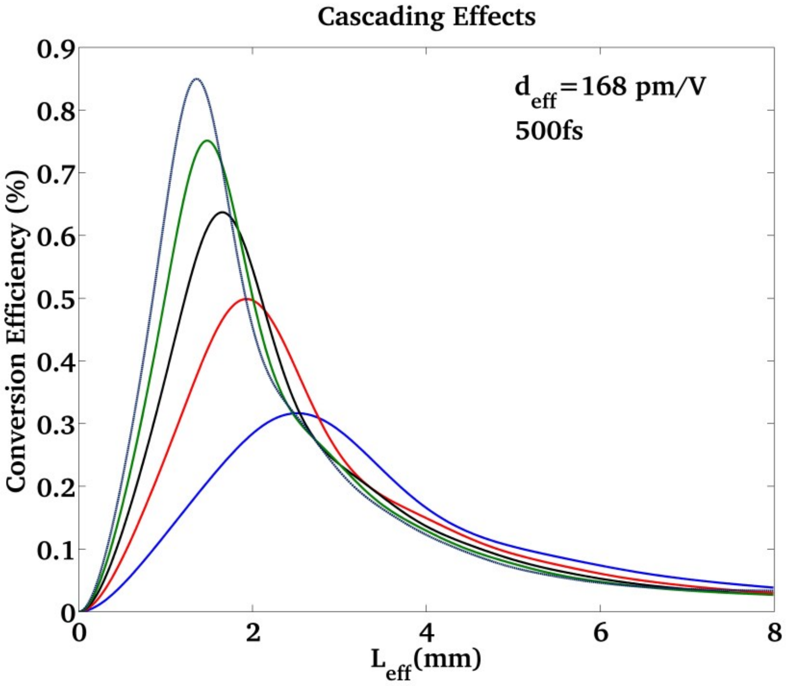
$$\eta_{THz} = \left(\frac{2\, d_{eff}^2}{\varepsilon_0 n_{opt}^2 n_{THz} c^3}\right)\Omega^2 \cdot I \cdot \left[L^2 \exp(-\alpha L/2)\, \frac{\sinh^2(\alpha L/4)}{(\alpha L/4)^2}\right]$$

Numerical:

Dispersion + Absorption **+ SPM**



Dispersion + Absorption **+ Cascading**

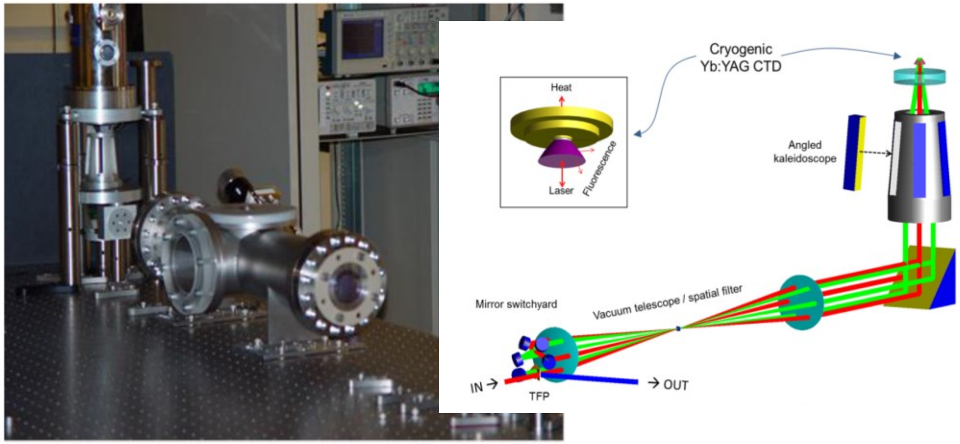


Future Work

- **Extending THz acceleration to GeV/m and relativistic particles**
  - Improvements to IR laser pulse energy (100 mJ – 1 J) with cryo-YAG or cryo-YILF multi-pass amplifiers
  - High energy accelerator development underway using single and multi-cycle pulses

Demonstrated cryo-YAG amplifier

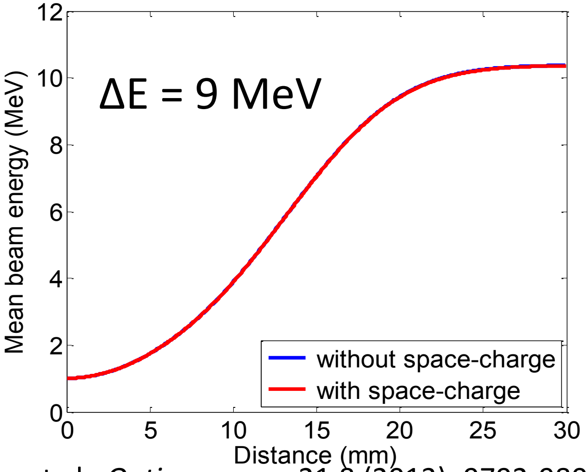
60 mJ IR pulse, uncompressed



Zapata, L., et al., CLEO, SM1F.1, 2014.

Modeling THz Acceleration

10 cycle, 10 mJ pulse, 0.74GeV/m

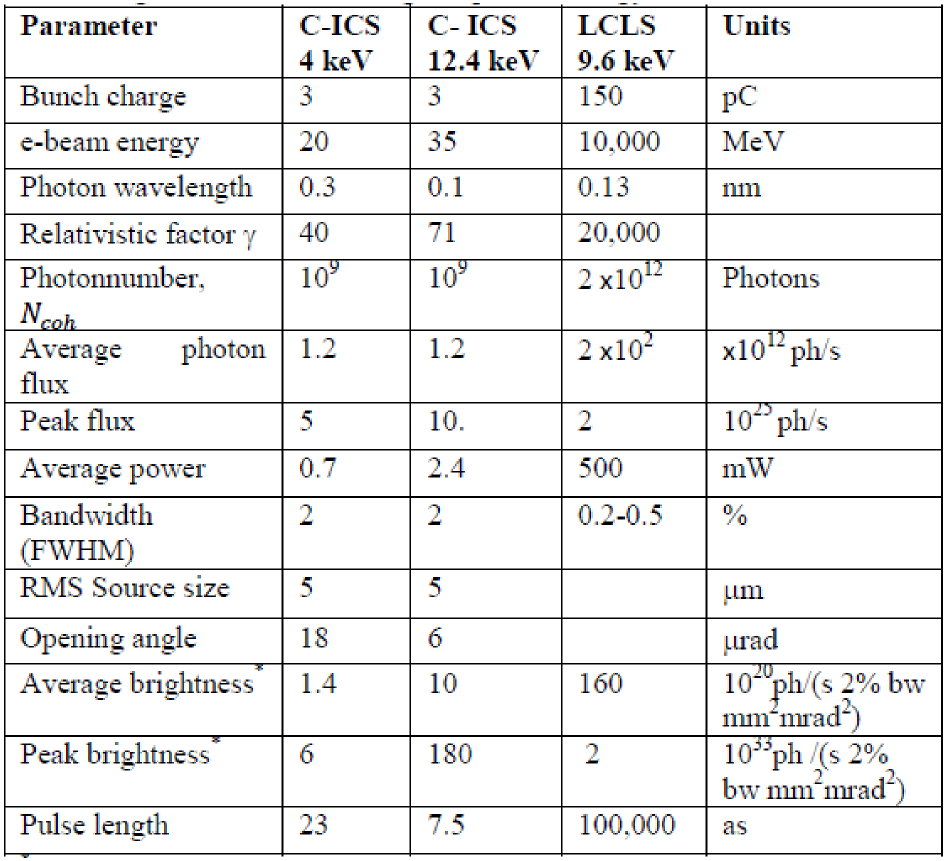


Wong, Liang Jie, et al., *Optics express* 21.8 (2013): 9792-9806.

Comparison to LCLS

$$\lambda_x = \frac{\lambda_L}{4\gamma^2}\left(1 + a_0^2 + \gamma^2\Delta\theta^2\right), \quad \text{with} \quad \Delta\theta_{rel} = \sqrt{\frac{\lambda_x}{\lambda_L(N_L+N_x)}} \approx \frac{1}{\gamma\sqrt{N_L+N_x}} \quad \text{and} \quad \frac{\Delta\omega}{\omega_x} = \frac{1}{N_L+N_x},$$

$$N_{coh} = \alpha|a_o|^2\left(\frac{N_L}{N_L+N_x}\right)^2 N_e{}^2|B_o|^2$$

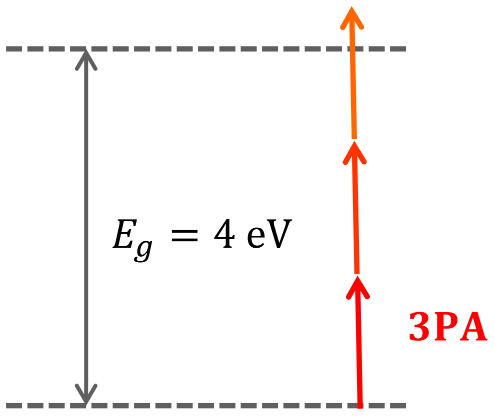


Parameter	**C-ICS 4 keV**	**C- ICS 12.4 keV**	**LCLS 9.6 keV**	**Units**
Bunch charge	3	3	150	pC
e-beam energy	20	35	10,000	MeV
Photon wavelength	0.3	0.1	0.13	nm
Relativistic factor γ	40	71	20,000	
Photonnumber, N_{coh}	10^9	10^9	2 x10^{12}	Photons
Average photon flux	1.2	1.2	2 x10^2	x10^{12} ph/s
Peak flux	5	10.	2	10^{25} ph/s
Average power	0.7	2.4	500	mW
Bandwidth (FWHM)	2	2	0.2-0.5	%
RMS Source size	5	5		µm
Opening angle	18	6		µrad
Average brightness*	1.4	10	160	10^{20}ph/(s 2% bw mm^{2}mrad^2)
Peak brightness*	6	180	2	10^{33}ph /(s 2% bw mm^{2}mrad^2)
Pulse length	23	7.5	100,000	as

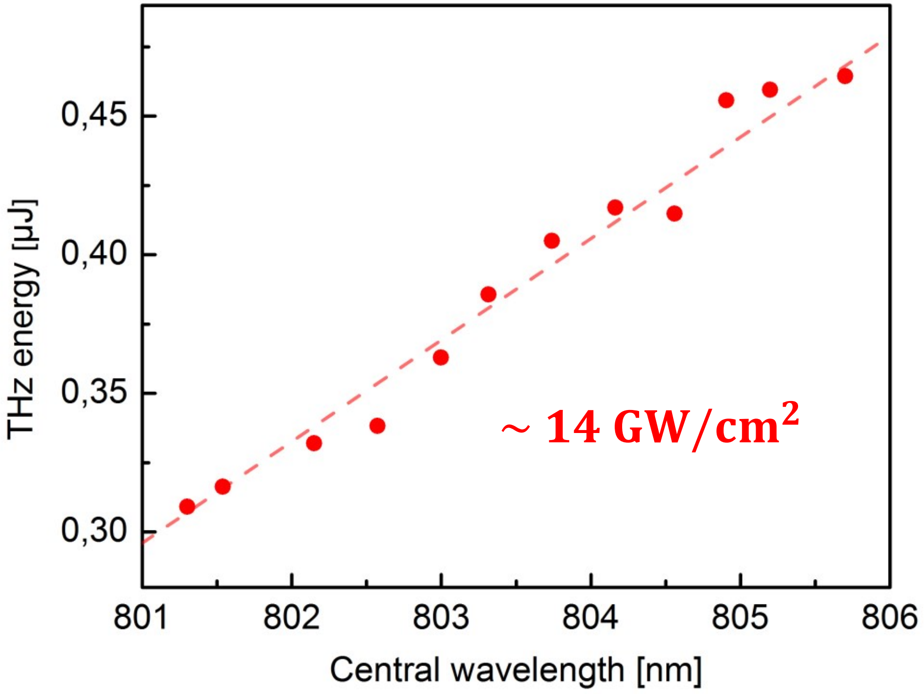
Prospects: multi-photon & THz absorption

- $LiNbO_3$ band gap = 4 eV
- 3 Photon-absorption

λ [nm]	E_p [eV]	E_{3PA} [eV]
795	1.5596	**4.68 eV**
800	1.5498	**4.65 eV**
805	1.5402	**4.62 eV**
810	1.5307	**4.59 eV**

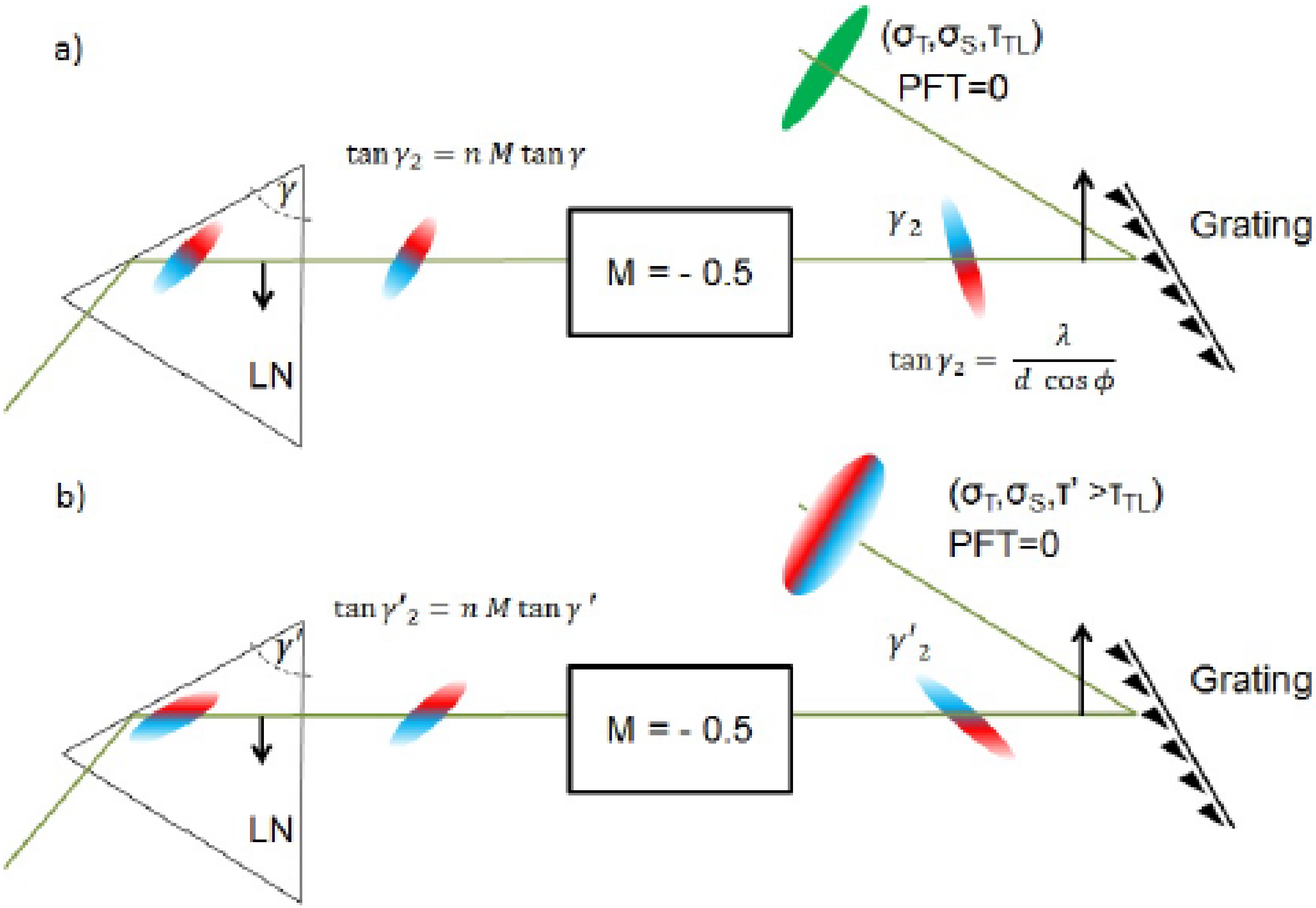


THz output energy as a function of pump central wavelength



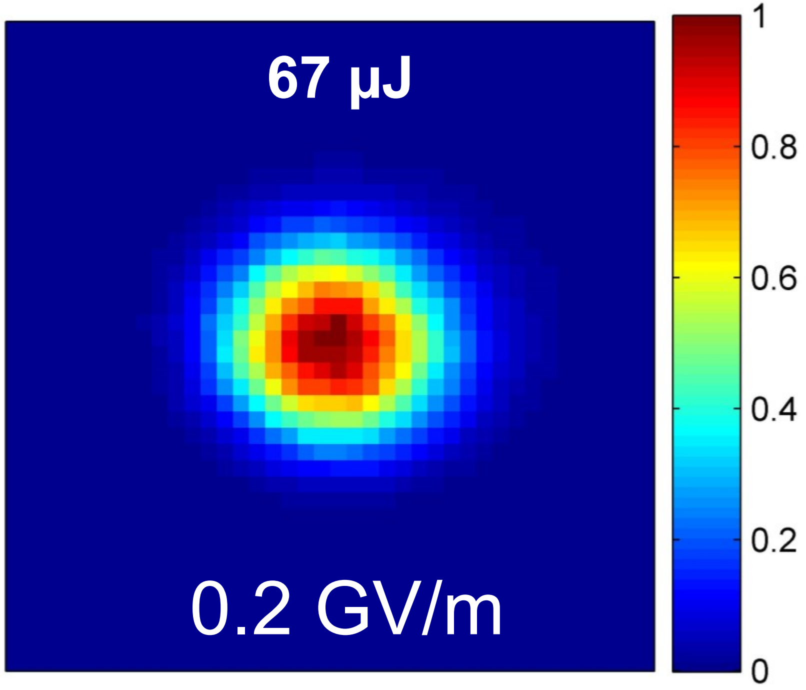
- Fixed bandwidth = 5 nm
- Fixed fluence = 5 mJ/cm^2

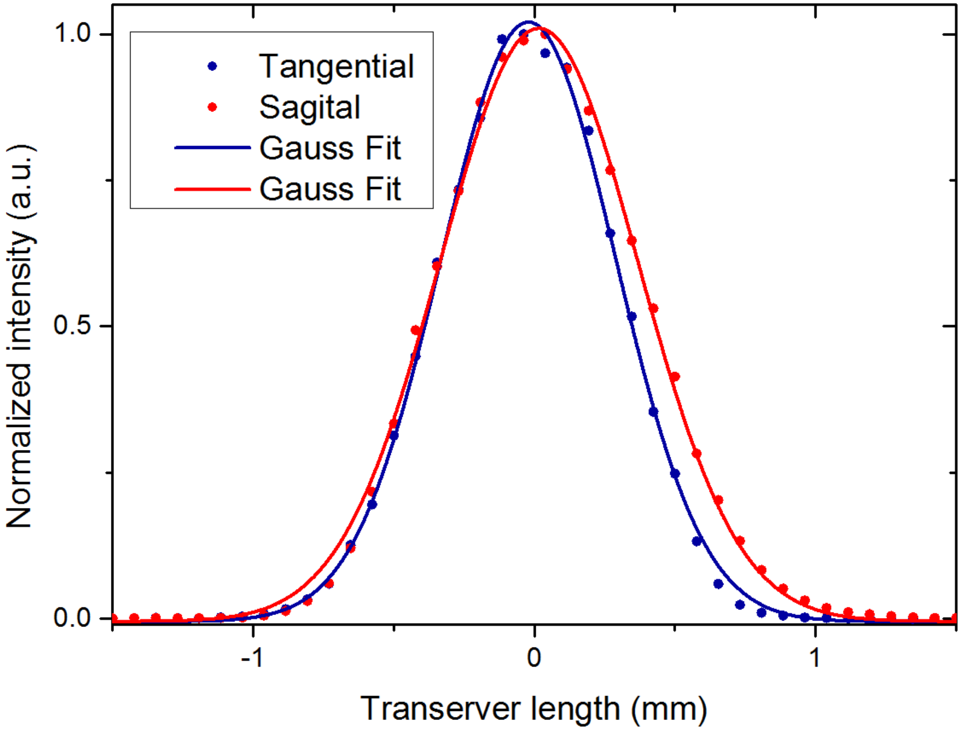
Controlling Temporal Chirp to Fine-tune the PFT angle



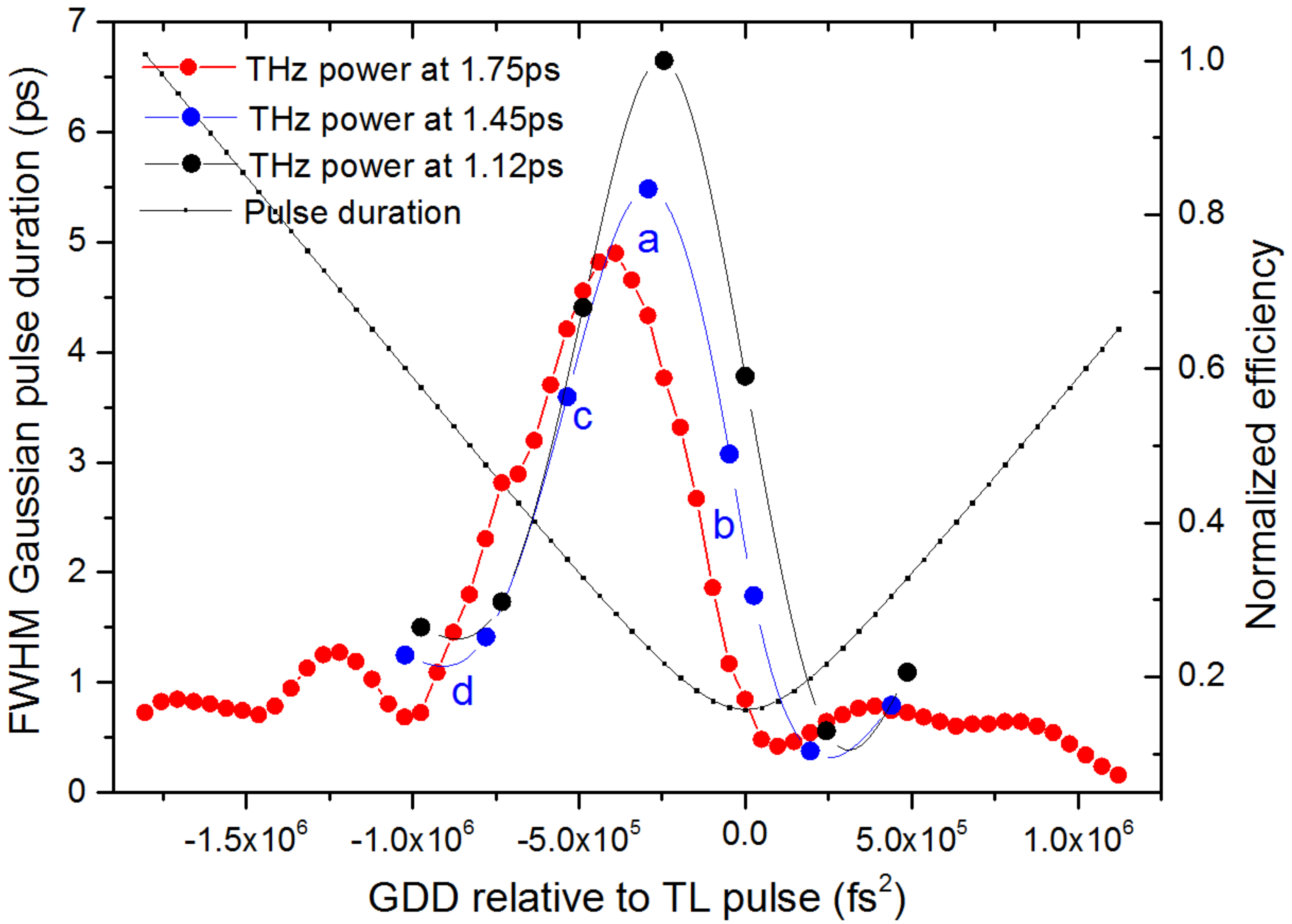
Maximum Field Strength is ~0.2 GV/m

	Tangential diameter (mm)	Sagital diameter (mm)	Divergence (mrad)	Peak THz Frequency
FWHM	0.73	0.83	5	~300 GHz
$1/e^2$	1.24	1.41		



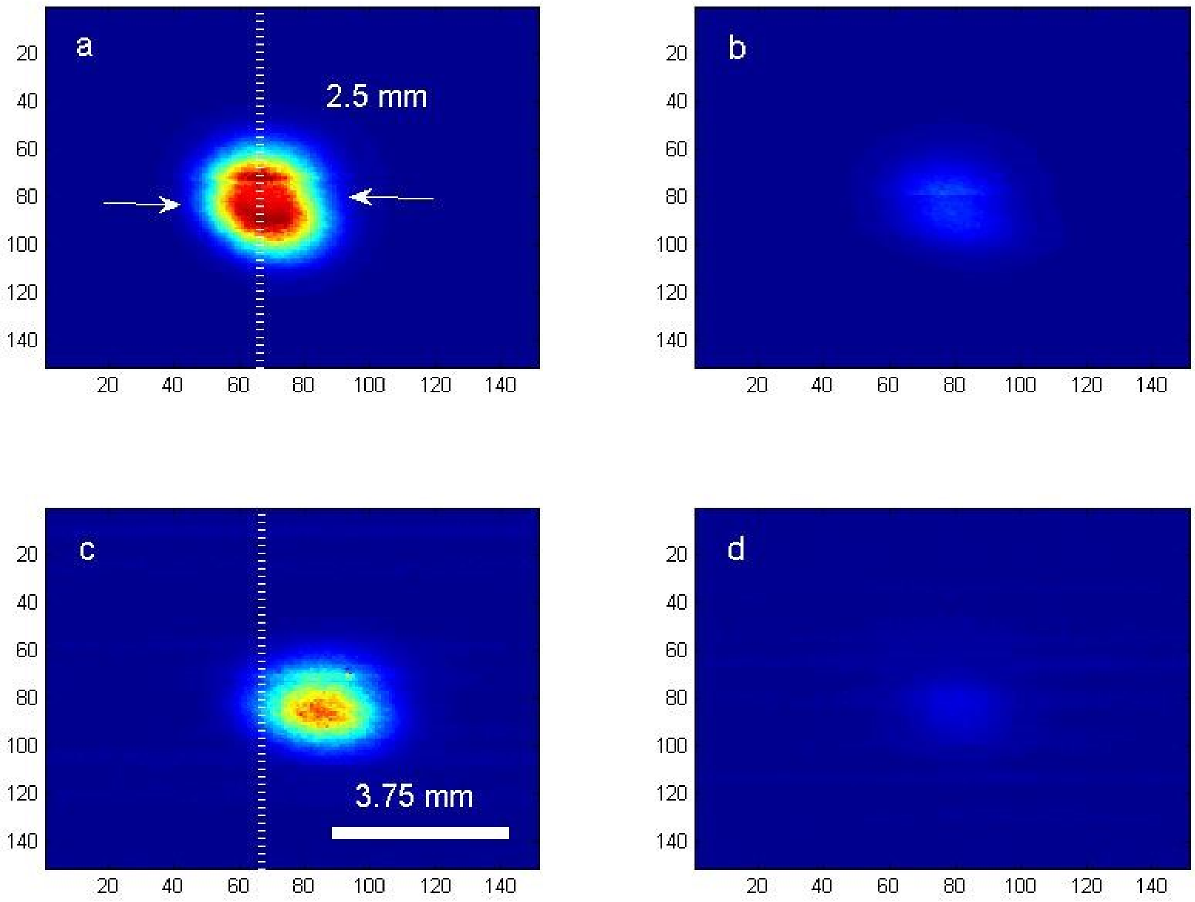


Normalized Efficiency w.r.t. TL pulse and GDD



Off of PFT Angle Higher THz frequencies still phase-match

Deviating from optimum PFT results in THz energy loss and only higher frequency spectral content



Other materials for single-cycle THz generation

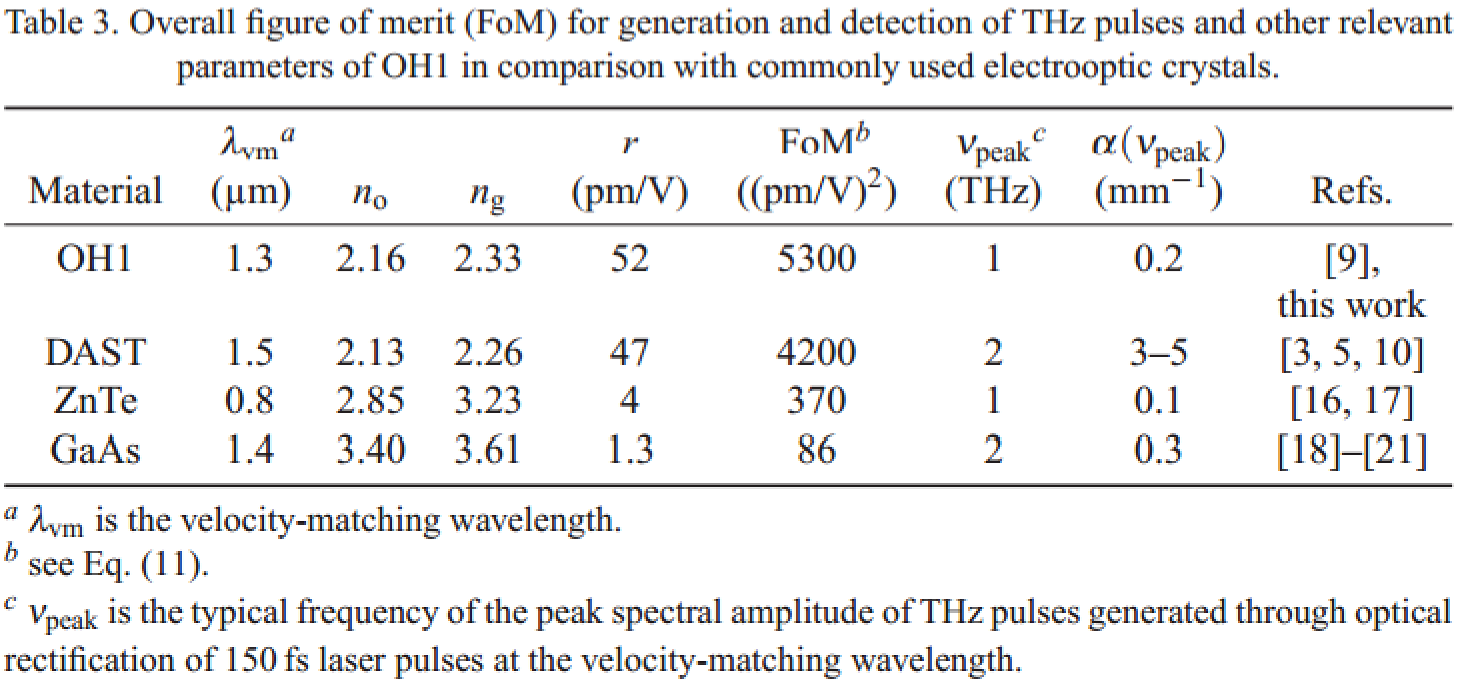


Table 3. Overall figure of merit (FoM) for generation and detection of THz pulses and other relevant parameters of OH1 in comparison with commonly used electrooptic crystals.

Material	$\lambda_{\mathrm{vm}}{}^{a}$ (µm)	n_{o}	n_{g}	r (pm/V)	FoM^{b} ((pm/V)^2)	$\nu_{\mathrm{peak}}{}^{c}$ (THz)	$\alpha(\nu_{\mathrm{peak}})$ (mm^{-1})	Refs.
OH1	1.3	2.16	2.33	52	5300	1	0.2	[9], this work
DAST	1.5	2.13	2.26	47	4200	2	3–5	[3, 5, 10]
ZnTe	0.8	2.85	3.23	4	370	1	0.1	[16, 17]
GaAs	1.4	3.40	3.61	1.3	86	2	0.3	[18]–[21]

[a] λ_{vm} is the velocity-matching wavelength.

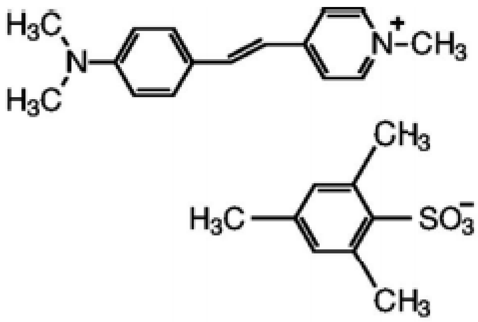
[b] see Eq. (11).

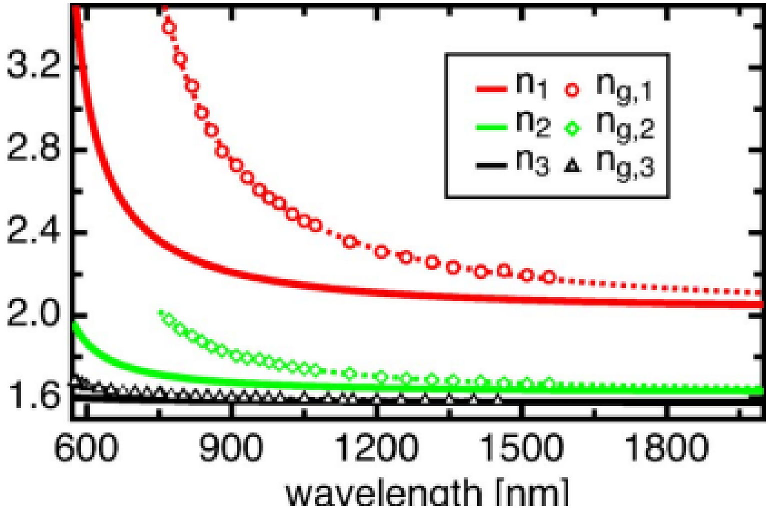
[c] ν_{peak} is the typical frequency of the peak spectral amplitude of THz pulses generated through optical rectification of 150 fs laser pulses at the velocity-matching wavelength.

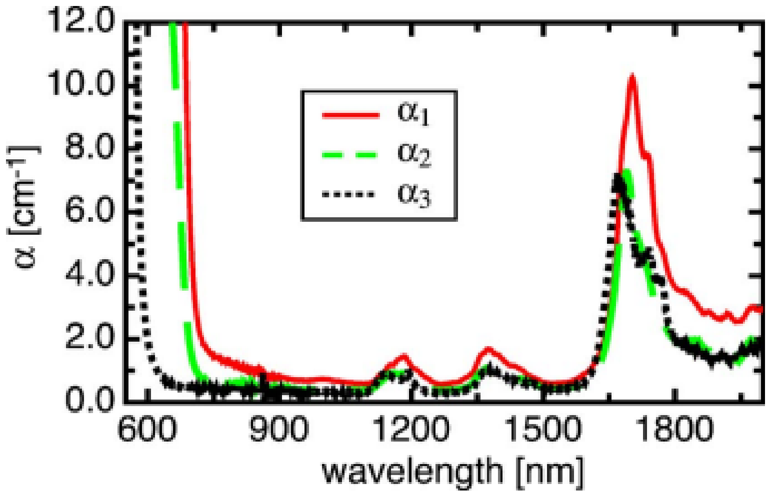
DSTMS lambda ~1.5 um

Kwon, Adv. Fun. Mat. 2008

DSTMS

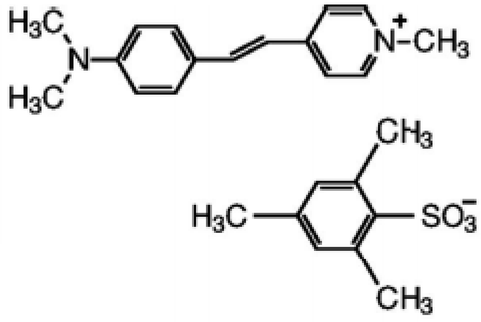


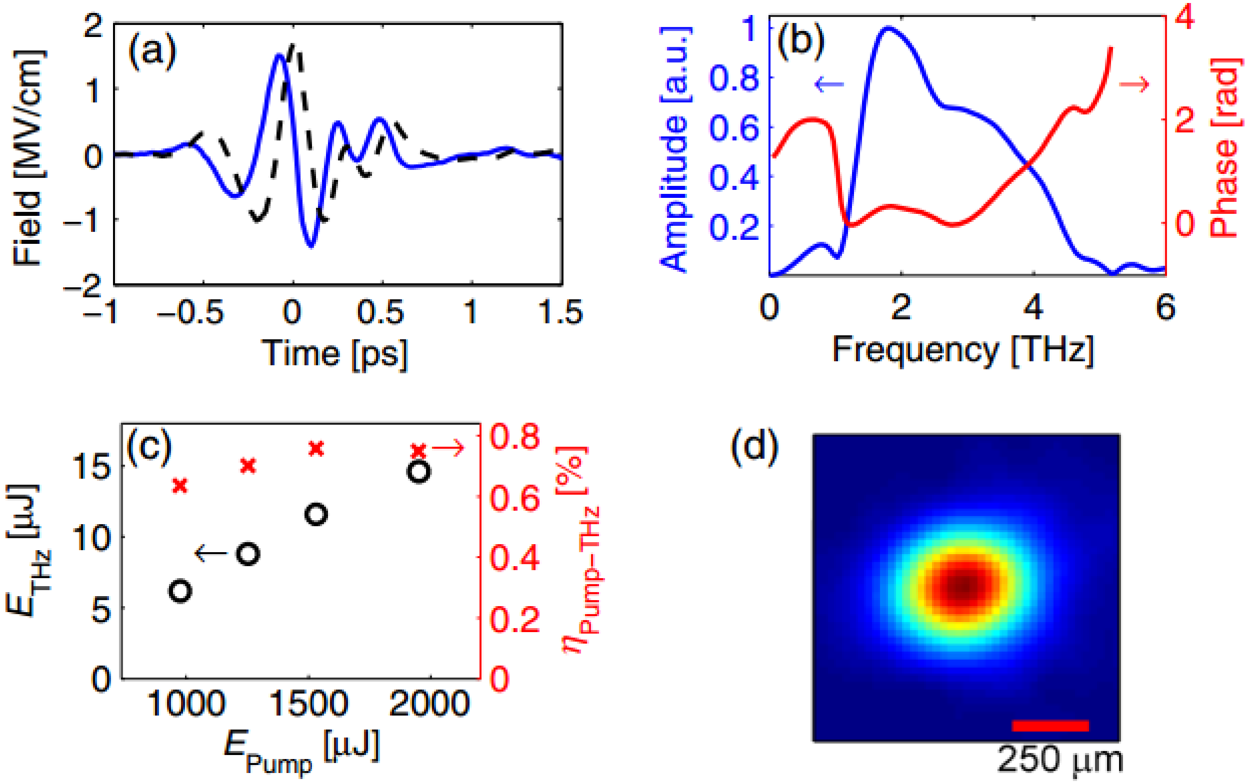




Kwon, Adv. Fun. Mat. 2008

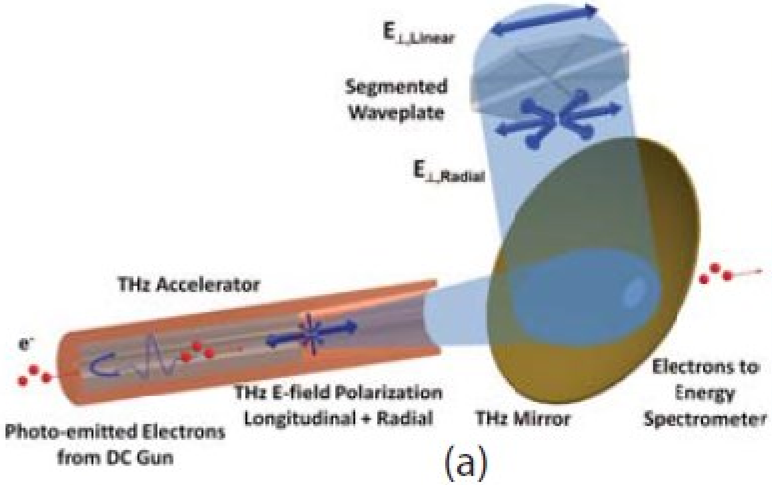
DSTMS



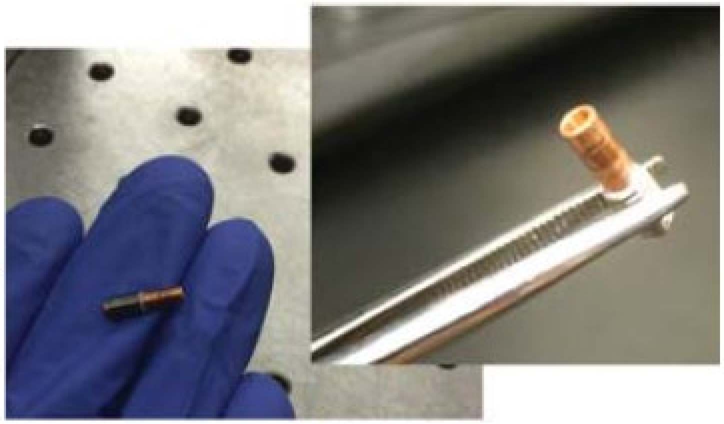


Kwon, Adv. Fun. Mat. 2008

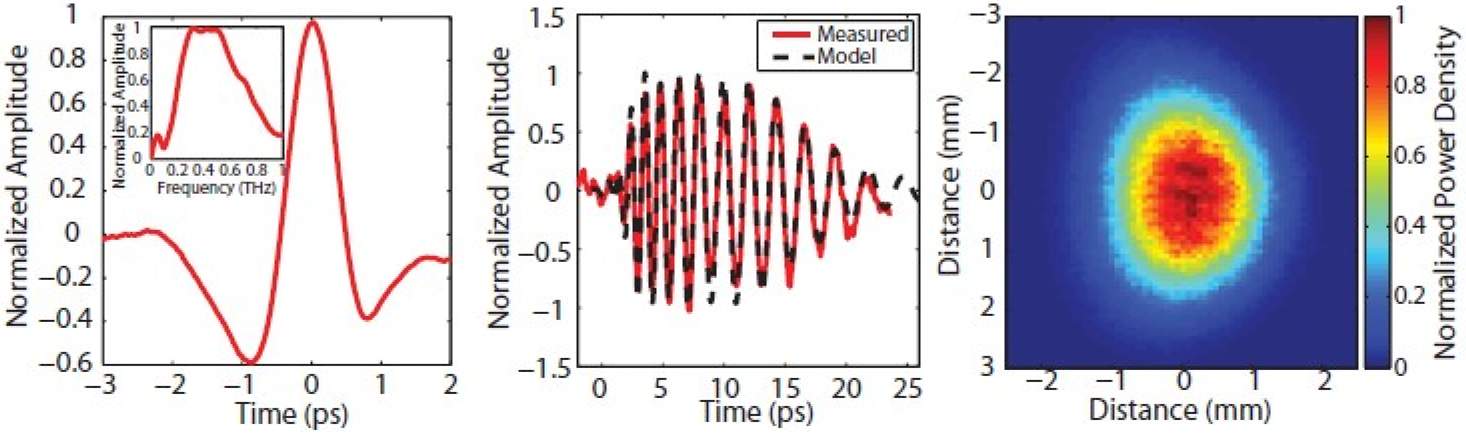
Preliminary Experiments: THz LINAC



(a)

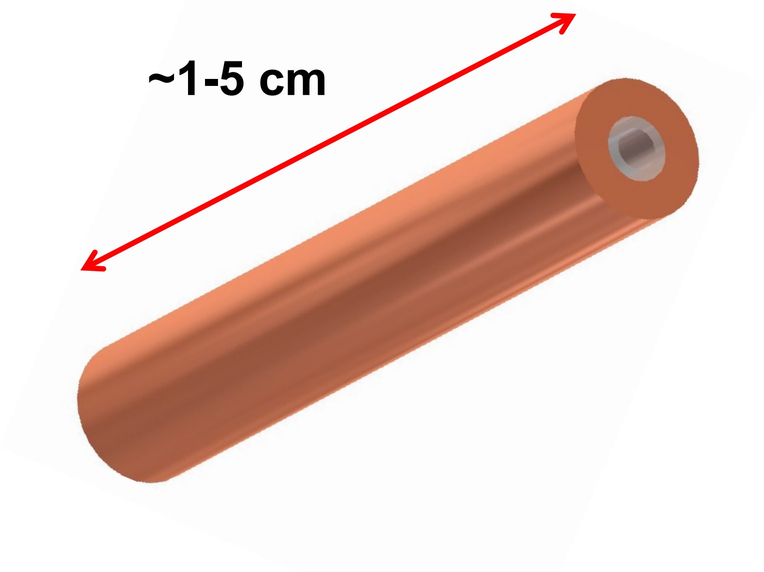


(b)



Dielectrically Loaded Metallic Waveguide

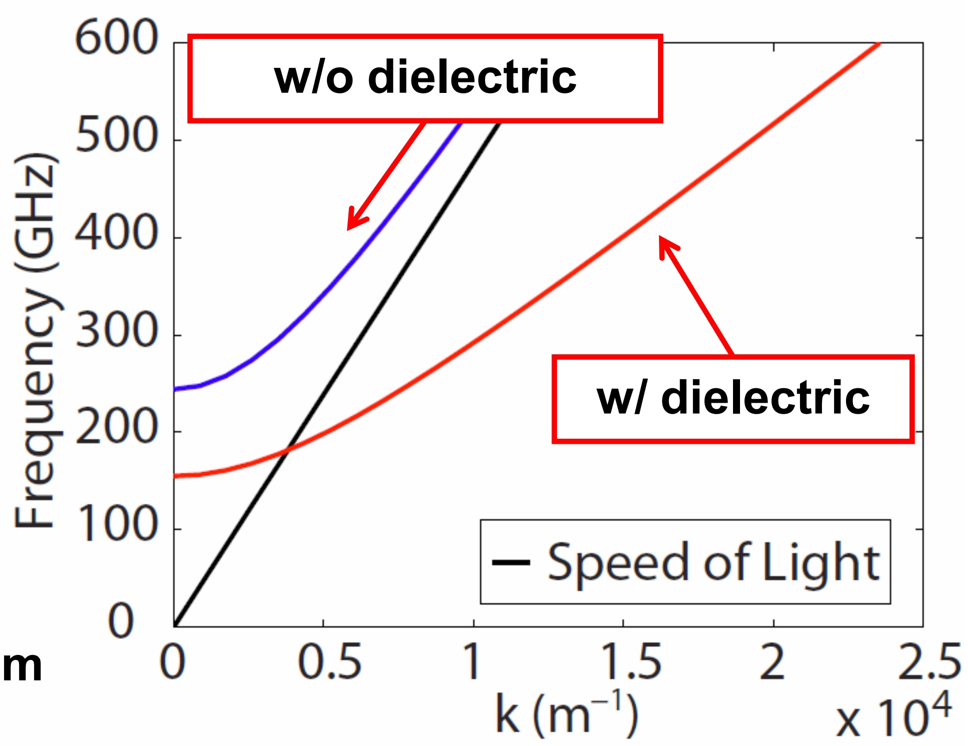
- A traveling way structure is best for coupling broad-band single cycle pulses
- Phase-group-velocity matching: THz-phase to electron velocity with thickness of dielectric



Copper Inner Diameter = 940 µm

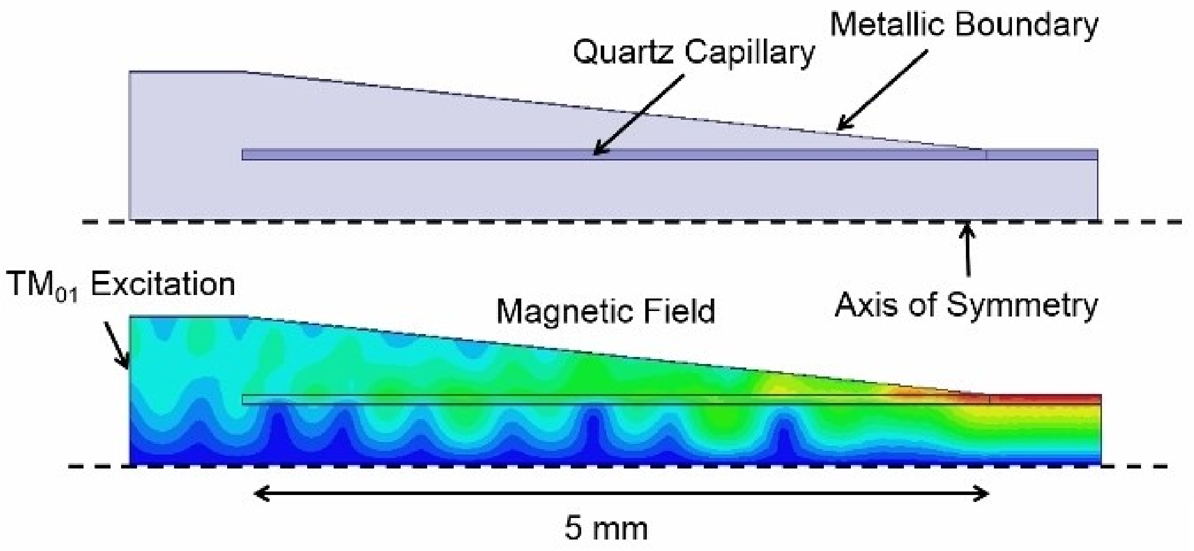
Fused Silica Inner Diameter = 400 µm

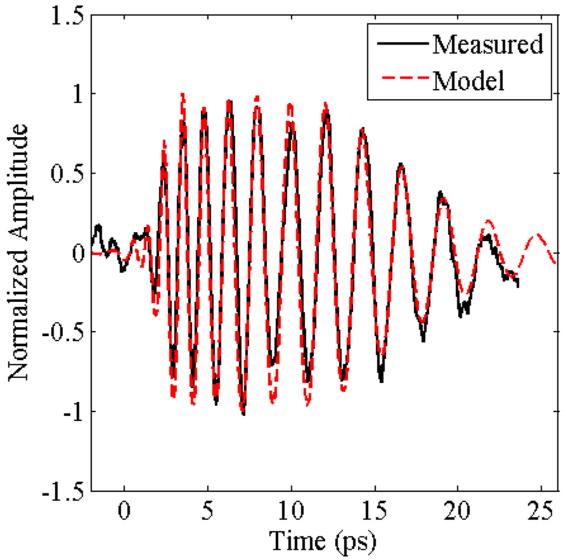
Dispersion Relation



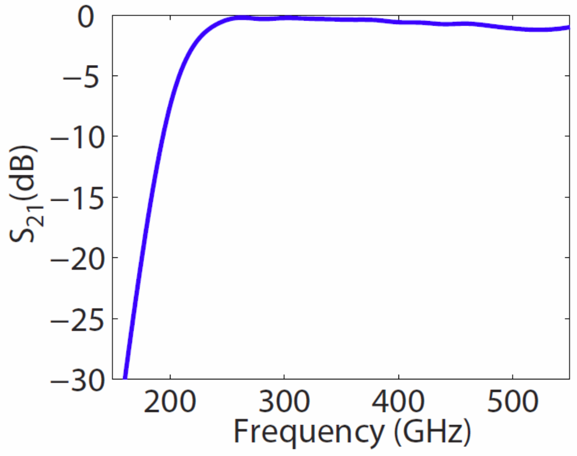
Accelerating Structures

Dielectrically Loaded Horn





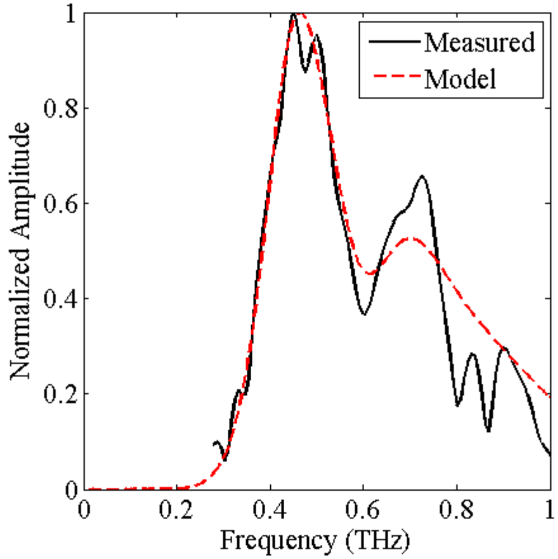
Coupling into TM_{01} - HFSS



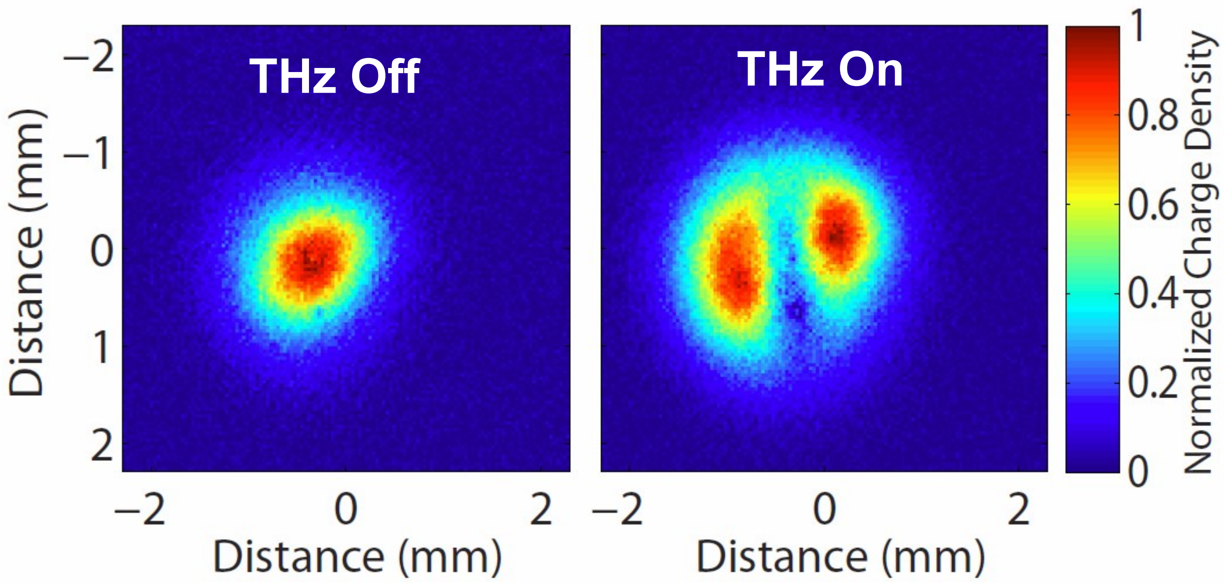
Waveguide Parameters

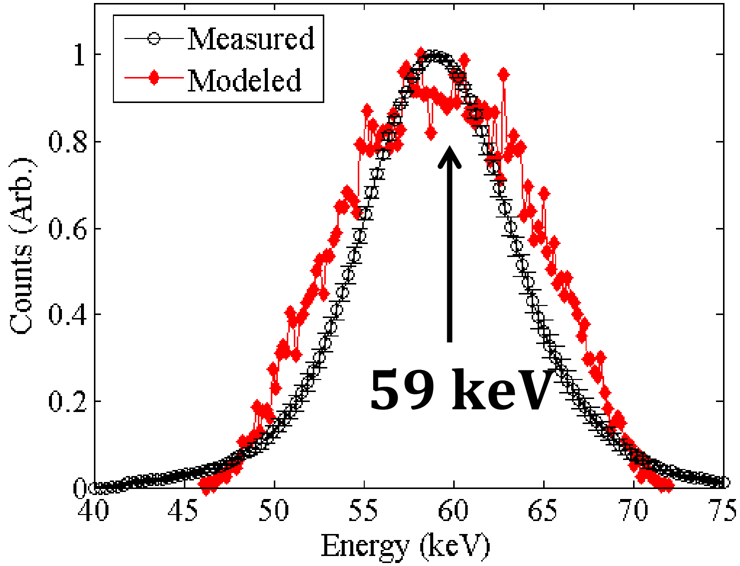
L = 4 cm

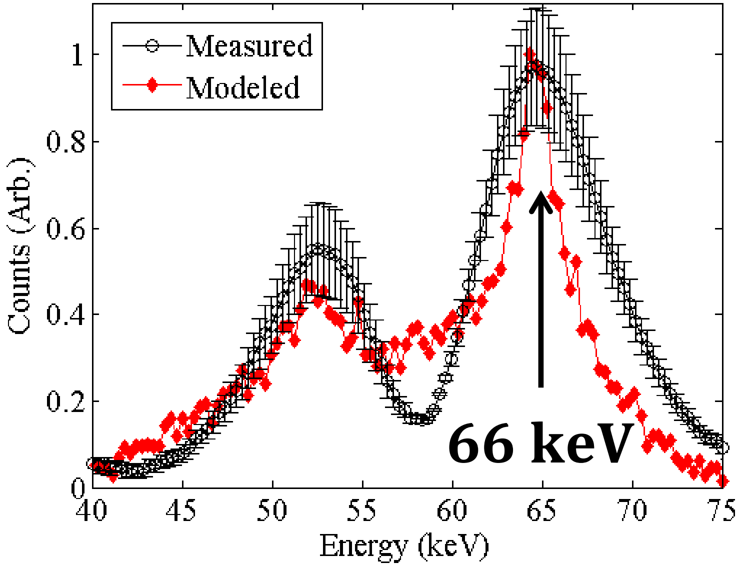
r_w = 400 μm



Terahertz-driven Linear Electron Acceleration

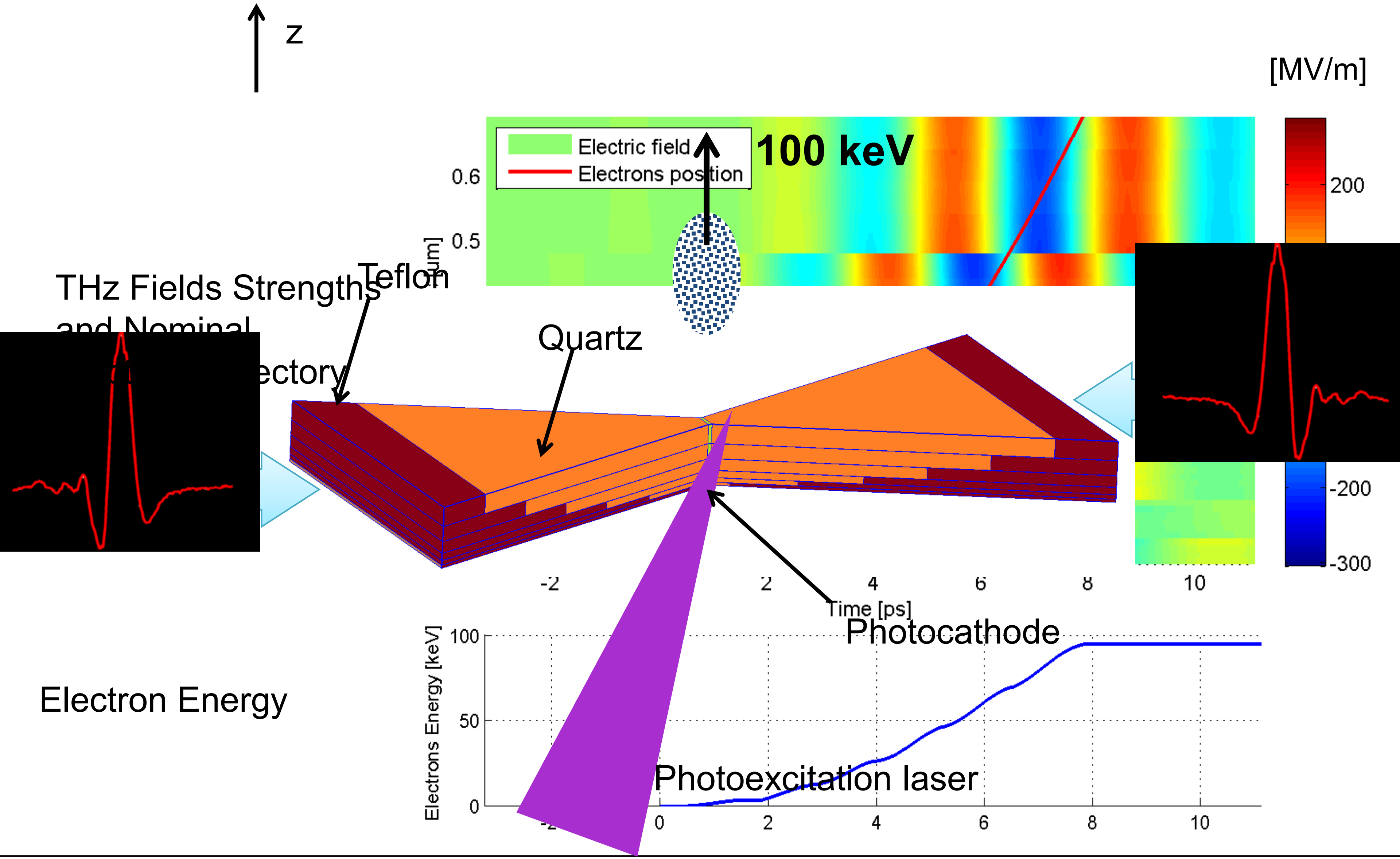






E. Nanni et al., arXiv:1411.4709 (2014)

Prospects for THz Guns



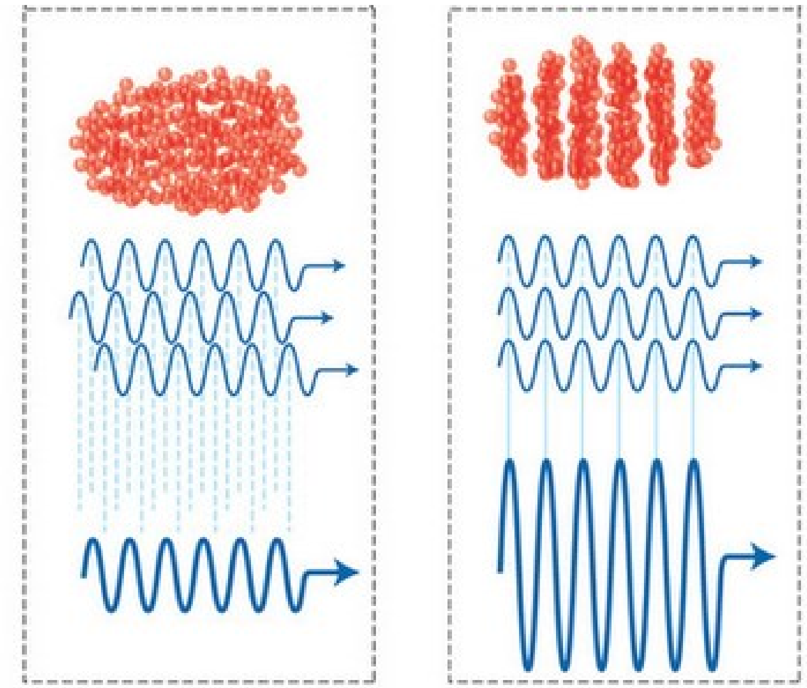
THz Fields Strengths

and Nominal

ectory

Electron Energy

Potential Advantages of THz Acceleration and Optical ICS



Incoherent emission: electrons randomly phased

Coherent emission: electrons bunched at radiation wavelength

Employing an optical undulator may translate into:

- Significantly smaller electron bunch energy due to reduced undulator period

Magnet	Optical
$\lambda_x = 1nm$	$\lambda_x = 1nm$
$\lambda_u = 3cm$	$\lambda_u = 1\mu m$
$\mathrm{E} \approx 1.5\ GeV$	$\mathrm{E} \approx 8\ MeV$

$$\lambda_x = \frac{\lambda_L}{4\gamma^2}\left(1 + a_0^2 + \gamma^2\theta^2\right)$$

- Shorter undulator length (mm)

- FEL gain occurs with micro-bunched electron beams: high brilliance coherent emission.

W. S. Graves PRL 108 (2012)
J. Kim et al, FEL (2004) McNeil, Nat. Phot. 4 (2010)