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Prospects of THz Pulse Generation with mJ-Level Energy and 100 MV/cm Electric Field

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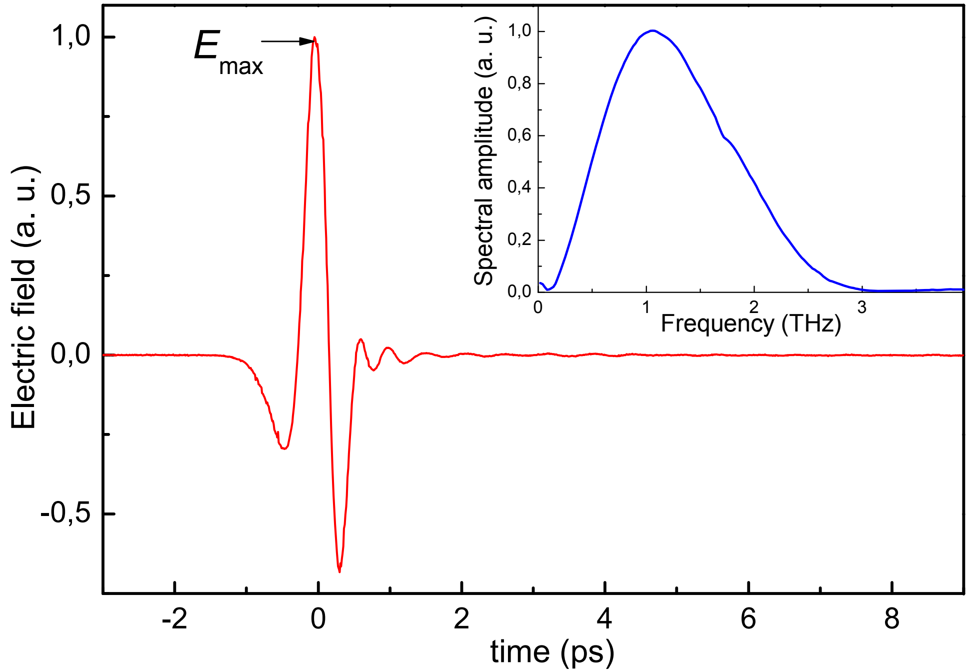
Workshop on THz Sources for Time Resolved Studies of Matter, ANL, Argonne, July 30-31, 2012

Applications of High-Energy THz Pulses



- Linear THz spectroscopy (E_{max} ≈ 100 V/cm → 10 fJ pulse energy)
  graphene, nanotubes, molecular magnets, etc.

- Nonlinear THz spectroscopy (E_{max} ≈ 100 kV/cm → µJ pulse energy)
  THz pump – THz probe measurements of dynamics (Hoffmann et al., PRB, 2009)

- Manipulation and acceleration of charged particles
  (E_{max} ≈ 100 MV/cm → 10 mJ pulse energy)
  VUV–XUV pulse generation by Thomson-scattering, X-ray free electron laser, …

Optical Rectification

Conversion efficiency depends on:

$$\eta_{\text{THz}} = \frac{2\omega_{\text{THz}}^2 d_{\text{eff}}^2 L^2 I}{\varepsilon_0 n_v^2 n_{\text{THz}} c^3} \cdot e^{-\alpha_{\text{THz}} L/2} \cdot \frac{\sinh^2[\alpha_{\text{THz}} L/4]}{[\alpha_{\text{THz}} L/4]^2}$$

- **THz frequency:** $\eta_{\text{THz}} \propto \omega_{\text{THz}}^2$
- **Material parameters → FOM**
- **Phase matching → velocity matching:** $v_{vis}^{gr} = v_{THz}^{ph}$

Figure of merit (FOM):

$\alpha_{THz} L << 1$

$$\eta_{THz} = \frac{2\omega^2 d_{eff}^2 L^2 I}{\varepsilon_0 n_v^2 n_{THz} c^3} \qquad FOM_{NA} = \frac{d_{eff}^2 L^2}{n_v^2 n_{THz}}$$

$\alpha_{THz} L >> 1$

$$\eta_{THz} = \frac{8\omega^2 d_{eff}^2 I}{\varepsilon_0 n_v^2 n_{THz} \alpha_{THz}^2 c^3} \qquad FOM_A = \frac{4 d_{eff}^2}{n_v^2 n_{THz} \alpha_{THz}^2}$$

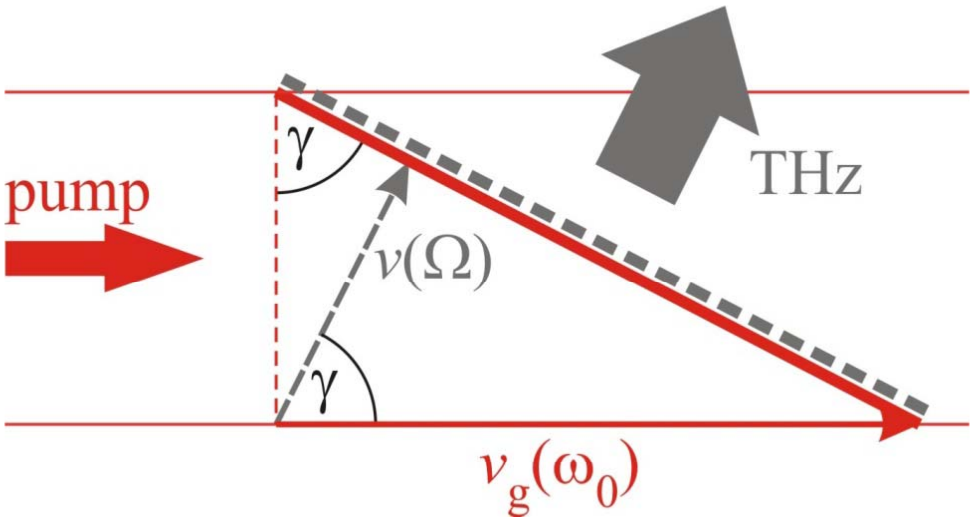
Materials for Optical Rectification

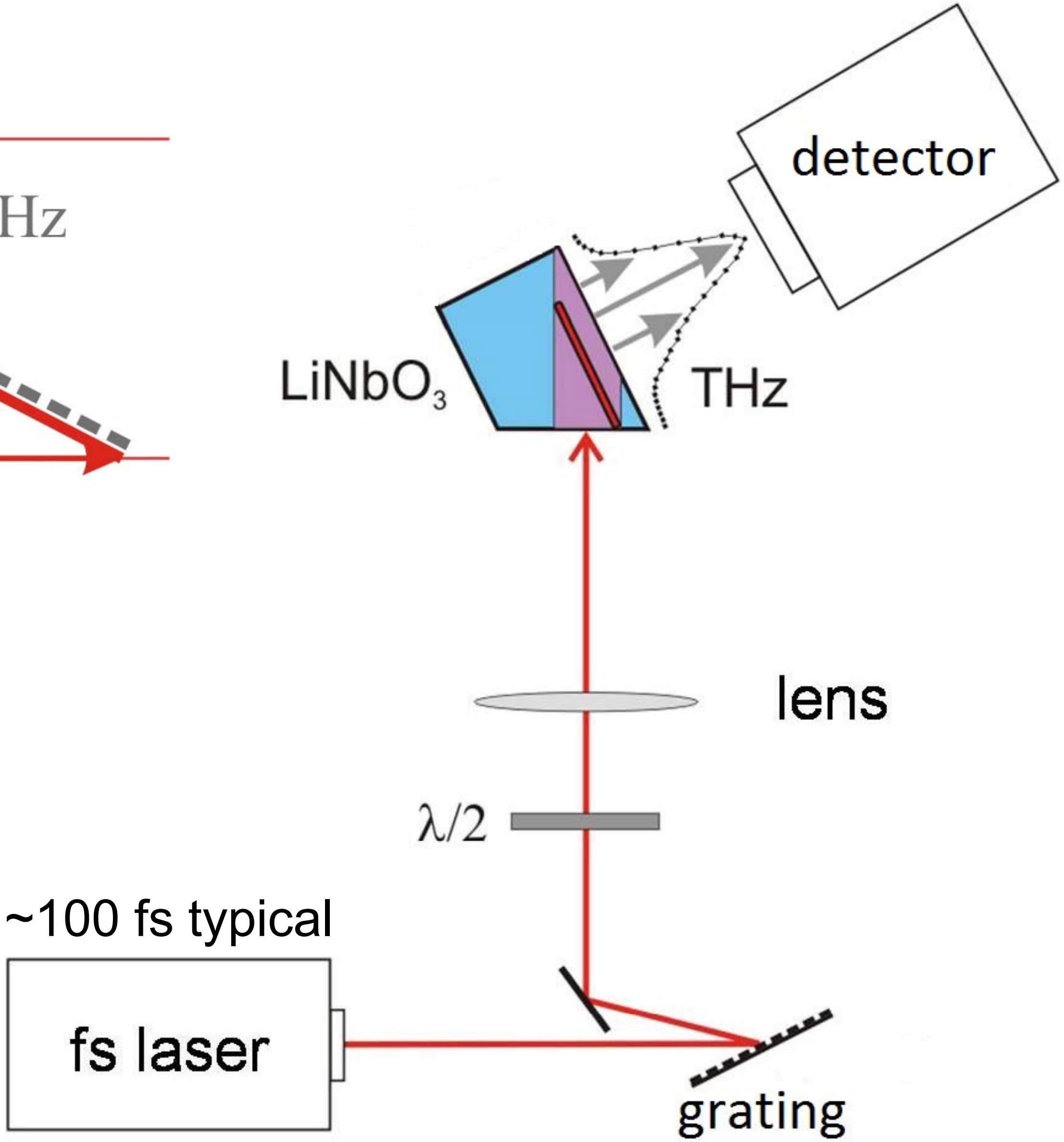
Material	d_{eff} (pm/V)	$n^{gr}_{800\,nm}$	n_{THz}	$n^{gr}_{1.55\,\mu m}$	α_{THz} (cm^{-1})	FOM* (pm^{2}cm^2/V^2)
CdTe	81.8		3.24	2.81	4.8	11.0
GaAs	65.6	4.18	3.59	3.56	0.5	4.21
GaP	24.8	3.67	3.34	3.16	0.2	0.72
ZnTe	68.5	3.13	3.17	2.81	1.3	7.27
GaSe	28.0	3.13	3.27	2.82	0.5	1.18
$sLiNbO_3$ sLN 100K	168	2.25	4.96	2.18	17 4.8	18.2 48.6
DAST	615	3.39	2.58	2.25	50	41.5

Velocity matching condition: $v^{gr}_{NIR} = v^{ph}_{THz} \Rightarrow n^{gr}_{NIR} = n_{THz}$

* for L = 2 mm

Tilted-Pulse-Front Pumping (TPFP)



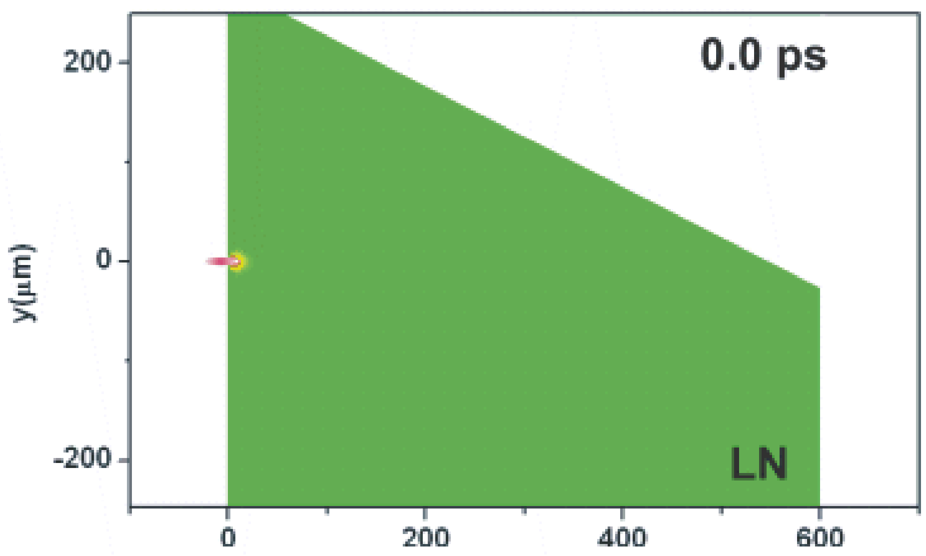


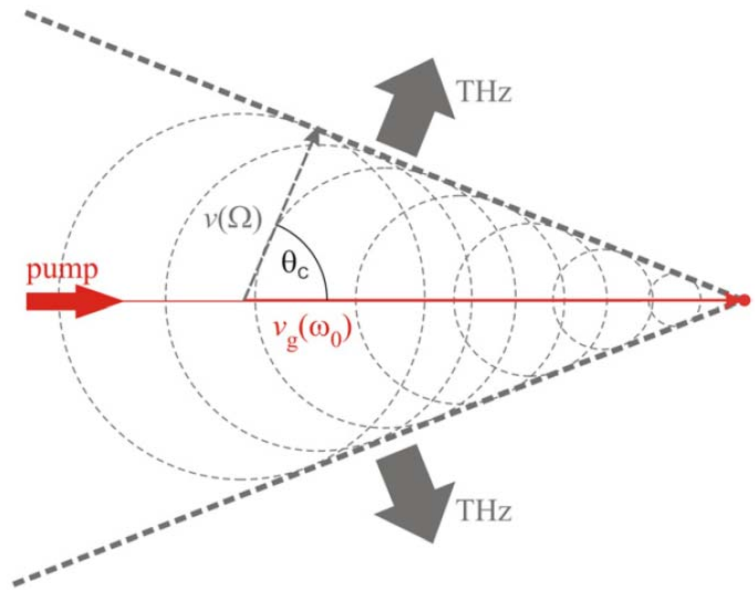
$$v_{vis}^{gr} \cdot \cos\gamma = v_{THz}^{ph}$$

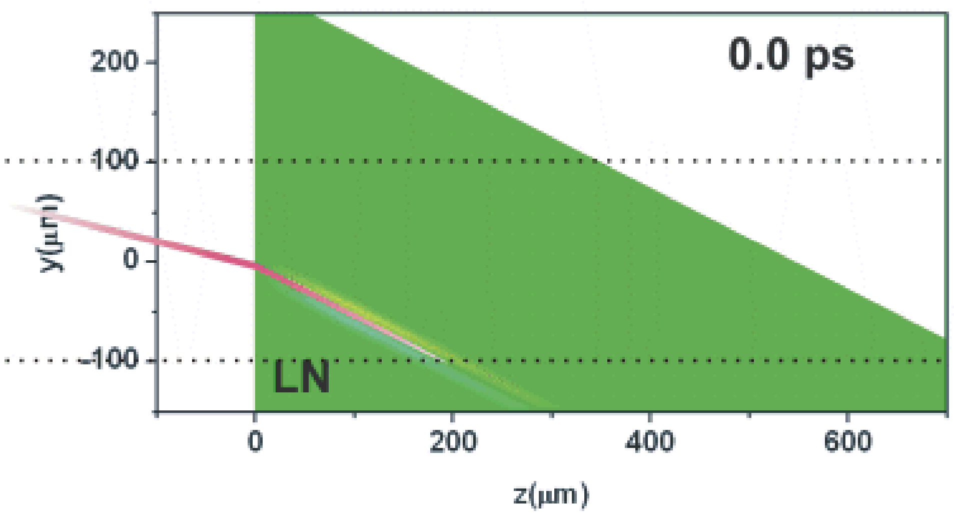
~100 fs typical

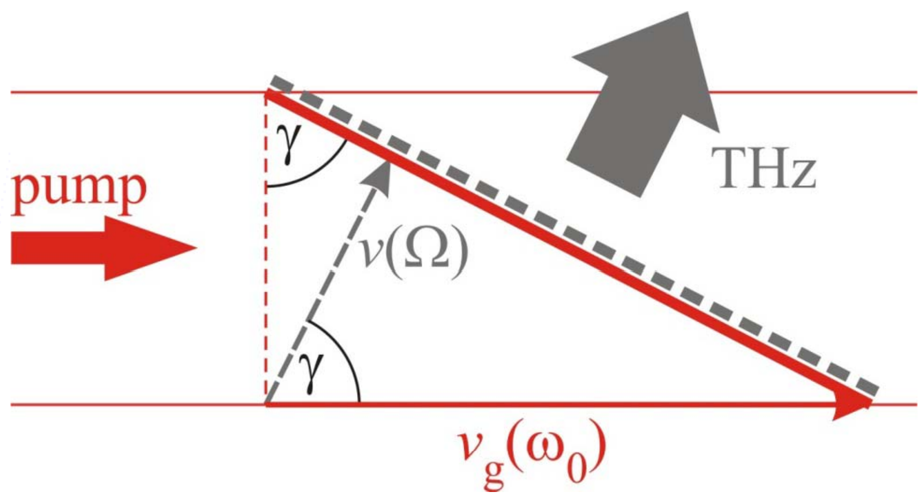
Hebling et al.,
Opt. Express, 2002

Tilted-Pulse-Front Pumping vs. Line Focusing









Stepanov et al., Opt. Express, 2005

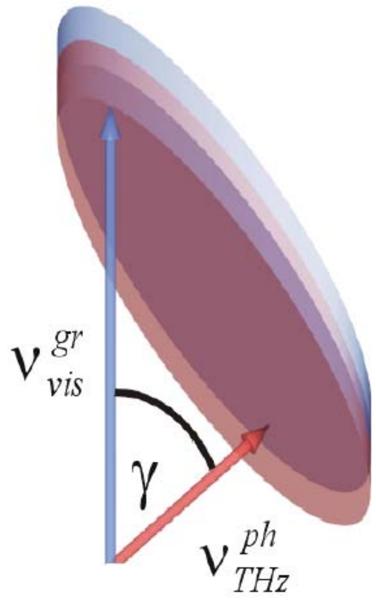
Hoffmann & Fülöp, J. Phys. D, 2011

THz Pulse Generation by Optical Rectification in $LiNbO_3$ Using Tilted Pulse Front Pumping

- Large nonlinear coefficient (d_{eff} = 168 pm/V)
- Velocity matching by tilting the pump pulse front

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

Hebling et al., Opt. Express, 2002

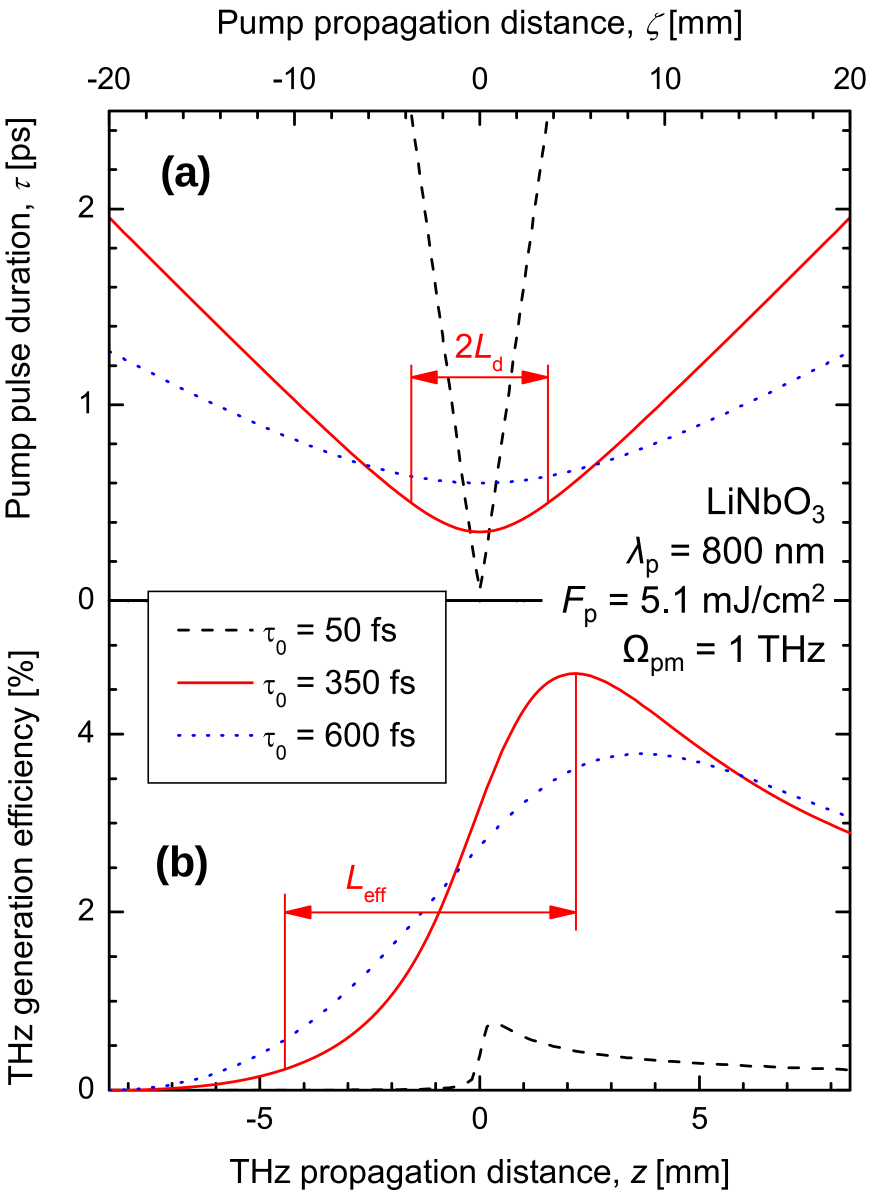


- Highest THz pulse energy from a table-top source

0.25 µJ	Stepanov et al., Opt. Express, 2005
10 µJ	Yeh et al., Appl. Phys. Lett., 2007
50 µJ	Stepanov et al., Appl. Phys. B, 2010
125 µJ	Fülöp et al., Opt. Lett., 2012

- Further increase of THz energy is in sight
  - optimal conditions (pump pulse duration & wavelength, crystal temperature & length)
    Fülöp et al., Opt. Express, 2011
  - by increasing the pumped area in optimized setups
    Pálfalvi et al., Appl. Phys. Lett., 2008
    Fülöp et al., Opt. Express, 2010

Effective Interaction Length



Pulse front tilt:

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

angular dispersion

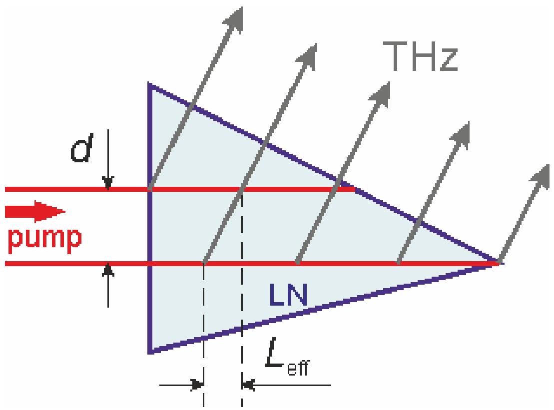
material dispersion

GVD parameter:

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

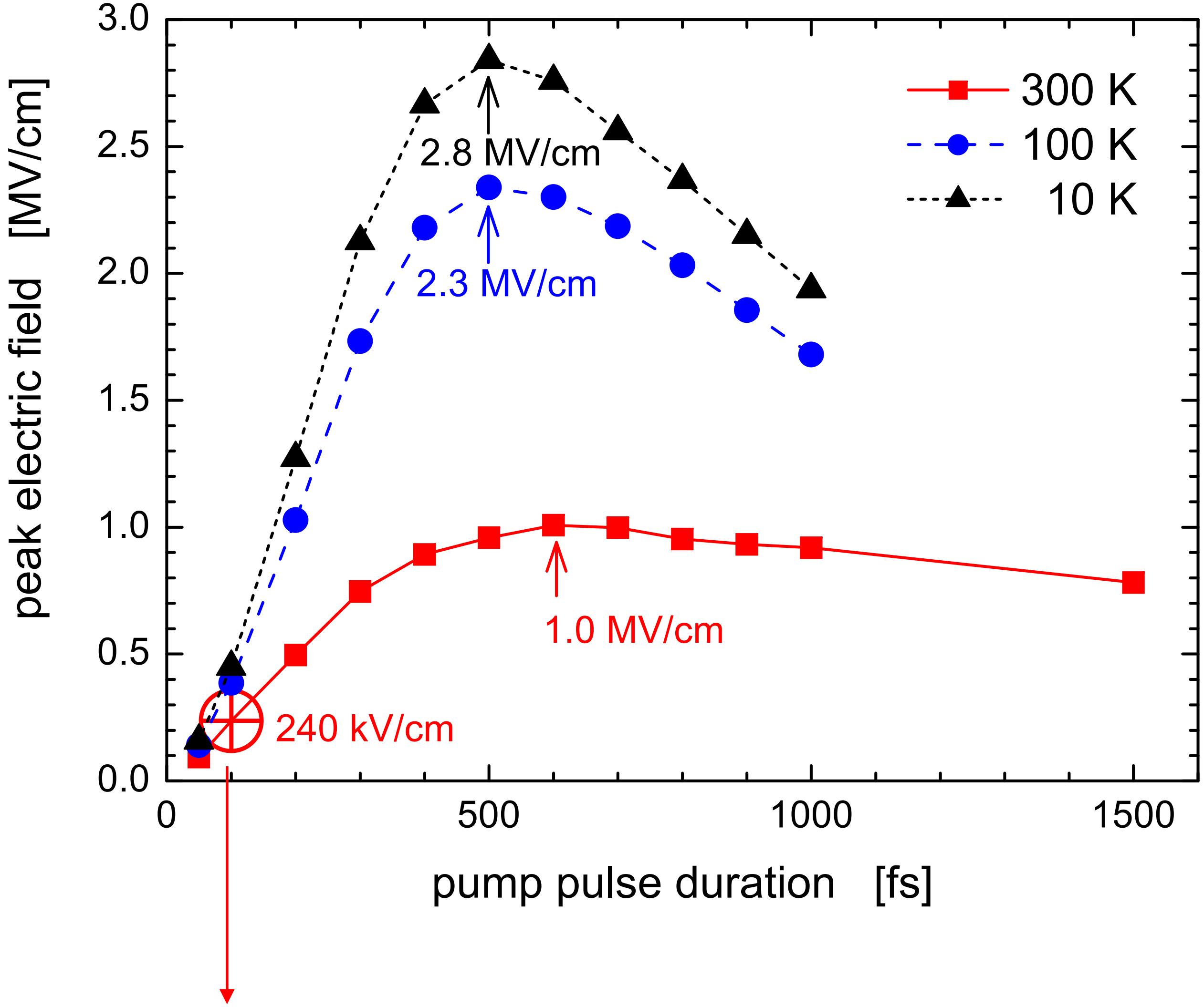
Martínez et al., JOSA A, 1984
Hebling, Opt. Quantum Electron., 1996
Fülöp et al., Opt. Express, 2010

Effective Interaction Length

Limiting effect	Possible solution
Change of pump pulse duration inside the medium	▪ Use longer Fourier-limited pump pulses ▪ Use materials requiring smaller pulse-front tilt
Absorption at THz frequencies ▪ Absorption coefficient α ▪ Multi-photon absorption (MPA) of the pump	▪ Cool the crystal to reduce α ▪ Use longer pump wavelength to suppress low-order MPA
Walk-off 	▪ Use materials requiring smaller pulse-front tilt ▪ Use large pump beam (and energy): → Optimized imiganig system → Contatc grating (no imaging)

Optimization of the Pump Pulse Length

Fülöp et al., Opt. Express, 2011



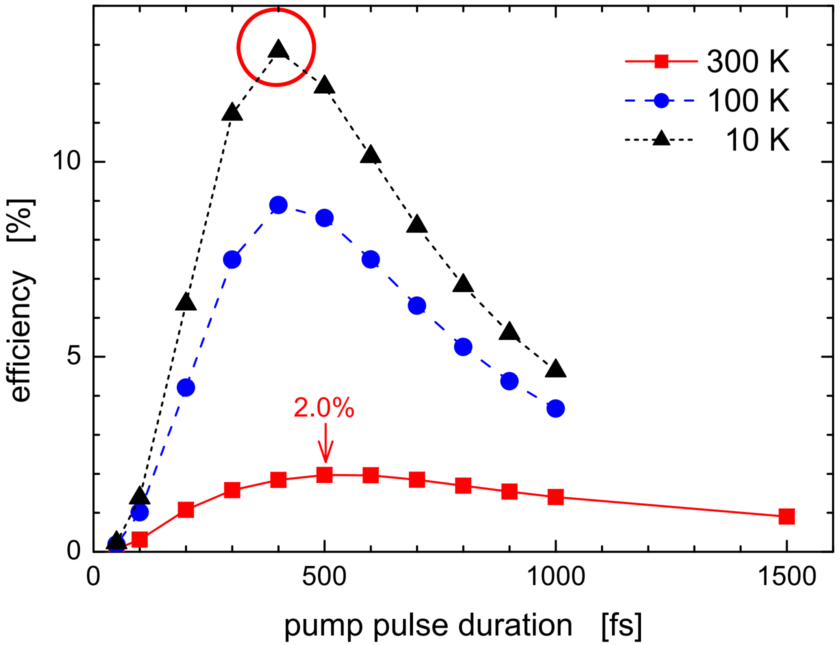
$LiNbO_3$
$L \leq 10$ mm
$I_{p,\,max} = 40$ GW/cm^2
$\lambda_p = 1064$ nm

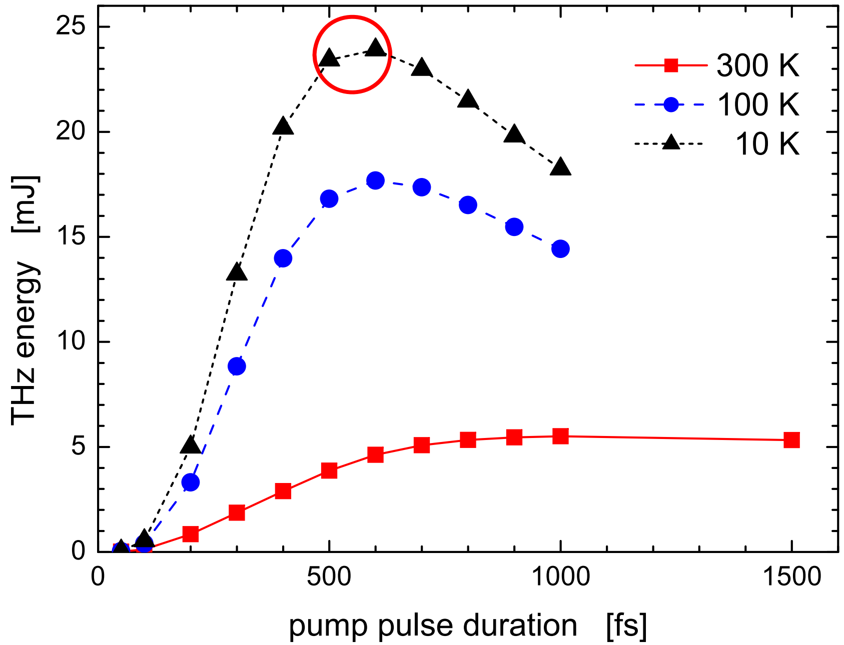
absorption coefficient of $LiNbO_3$:

T [K]	*α* [1/cm]
10	0.35
100	2.1
300	16

experimental value: 110 kV/cm Hoffmann et. al., Phys. Rev. B., 2009

Optimization of the Pump Pulse Length





τ_p = 500 fs

E_p = 200 mJ

$I_{p,\,max}$ = 40 GW/cm^2

THz energy ≈ 25 mJ

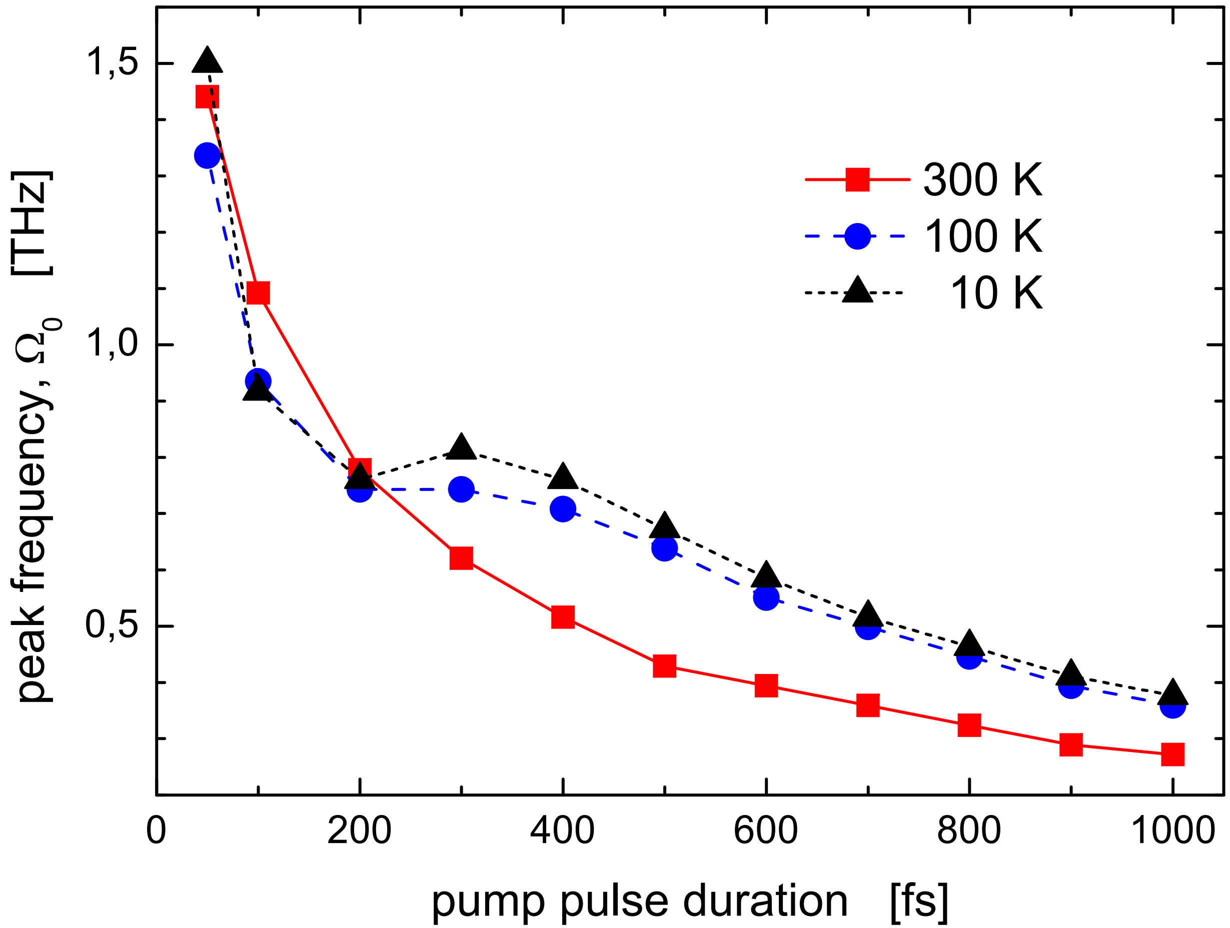
THz field = 2.8 MV/cm (unfocused)

→ 10 MV/cm level using imaging

→ 100 MV/cm level using focusing

Optimization of the Pump Pulse Length

Frequency at the spectral peak



$LiNbO_3$

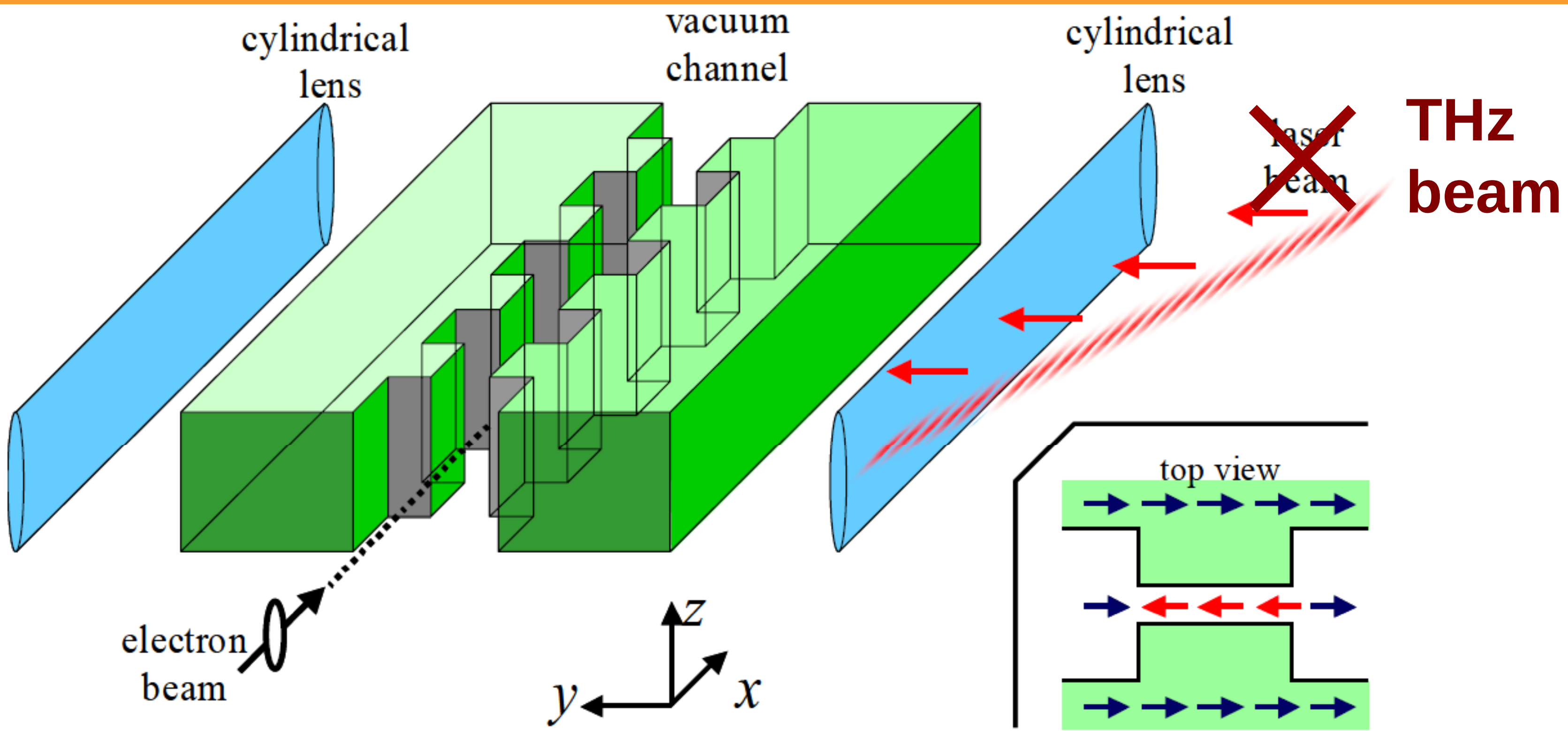
$L \leq 10$ mm

$I_{p,\,max} = 40$ GW/cm^2

$l_p = 1064$ nm

Phase matching adjusted to the spectral peak

New Appilcation Possibilities for THz Pulses with High Electric Field



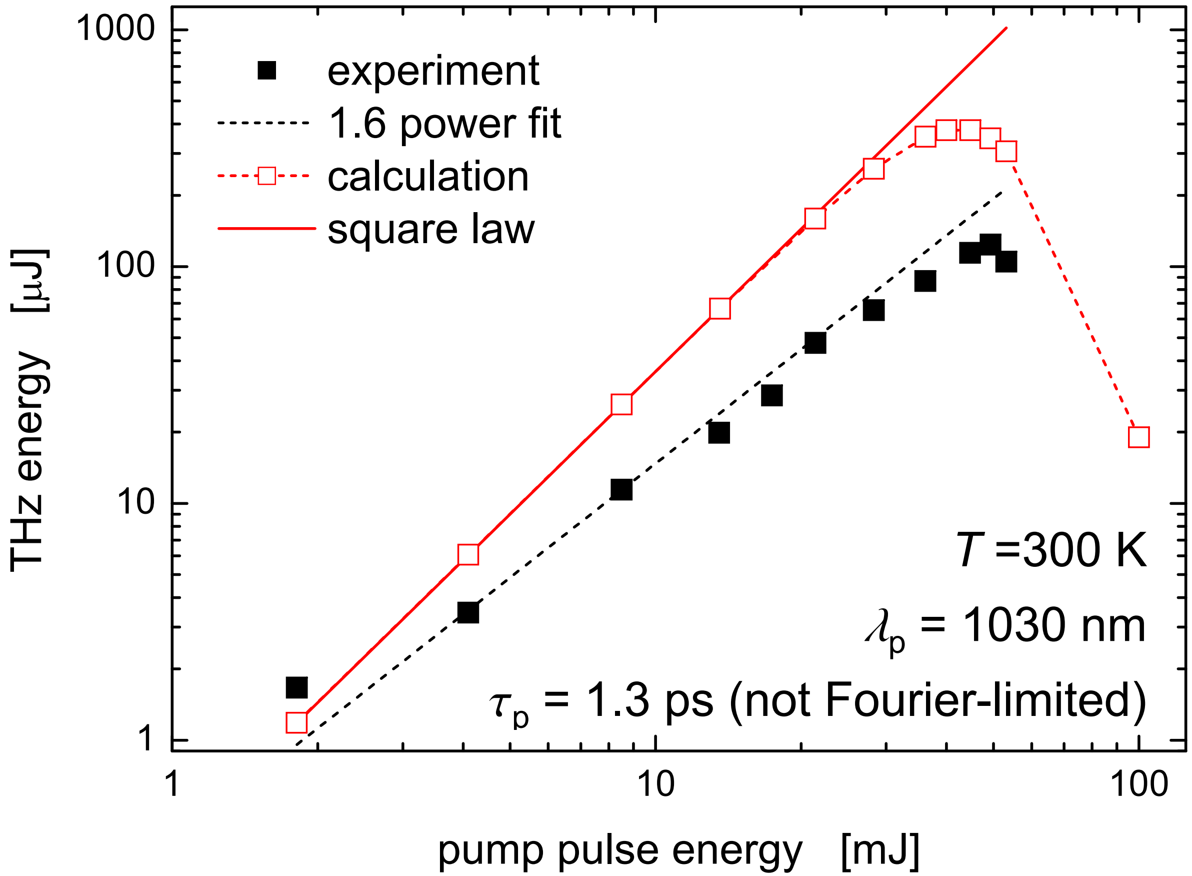
- **Acceleration**
  Plettner et al., Phys. Rev. Spec. Top. – Accel. and Beams 9, 111301 (2006)
- **Beam deflection, focusing**
  Plettner et al., Phys. Rev. Spec. Top. – Accel. and Beams 12, 101302 (2009)

1 GV/m = 10 MV/cm peak field strength is needed

New Appilcation Possibilities for THz Pulses with High Electric Field

- **Enhancement of HHG**
  E. Balogh et al., PRB, 2011
  K. Kovács et al., PRL, 2012
- **Longitudinal compression of electron bunches**
  → single-cycle MIR…XUV pulse generation
  Hebling et al., arXiv:1109.6852
  Hebling et al., presentation on Monday
- **Electron undulation**
  Hebling et al., arXiv:1109.6852
- **Proton acceleration**
  (40 → 100 MeV requires 30 mJ THz energy)
  → hadron therapy
  Pálfalvi et al., in preparation

Towards mJ Level THz Pulses: Experiment



In collaboration with

S. Klingebiel, F. Krausz, S. Karsch

MPQ, Garching

Further possibilities:

- **Optimal pump duration**
- **Cooling the $LiNbO_3$**
- **Contact grating**

Comparison with earlier results:

Stepanov et. al., Appl. Phys. B, 2010: → Fülöp et al., Opt. Lett. 2012:

E_{THz} = 50 μJ —— ×2.5 ——→ 125 μJ

η = 0.05% —— ×5 ——→ 0.25%

THz Beam Profiles

Contact grating

- No limit for lateral size
- No imaging optics

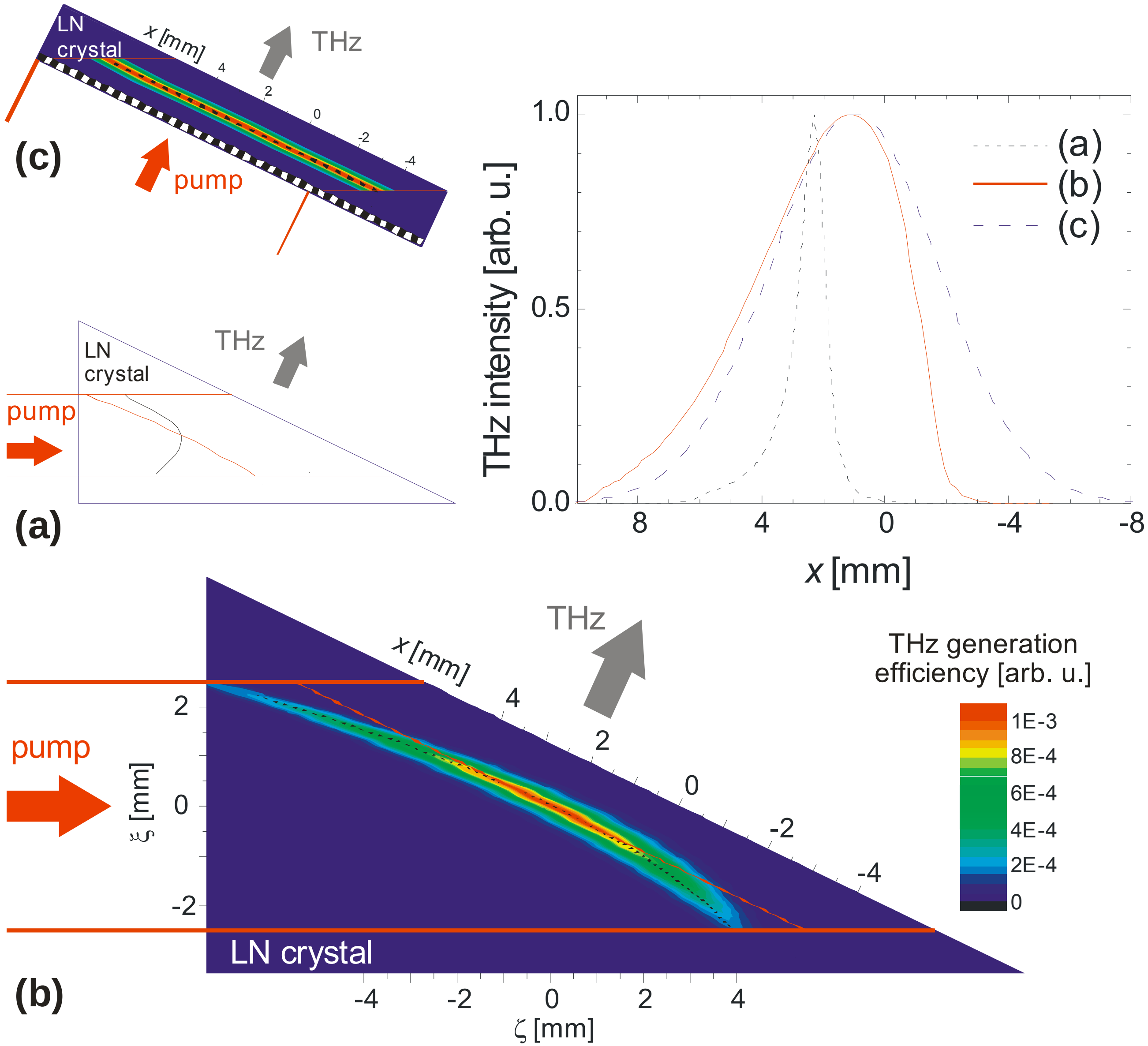
Pálfalvi et al., APL, 2008
Ollmann et al., APB, submitted

Non-optimized

Optimized

- Grating image is tangential to pulse front
- Longer focal length lens

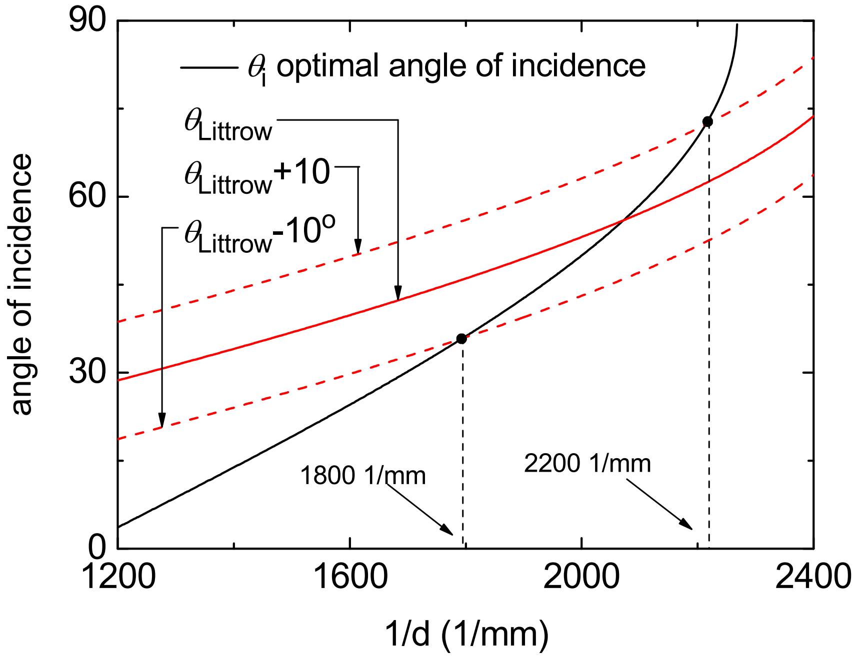
Fülöp et al.,
Opt. Express, 2010

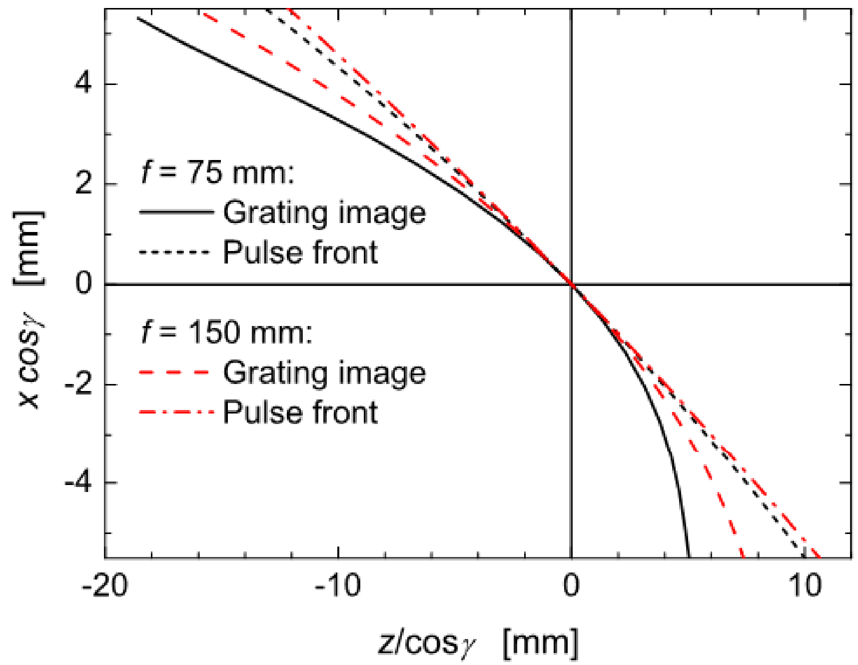


Optimizing the TPFP Setup Containing Imaging

$LiNbO_3$, λ_p = 800 nm

Fülöp et al., Opt. Express, 2010



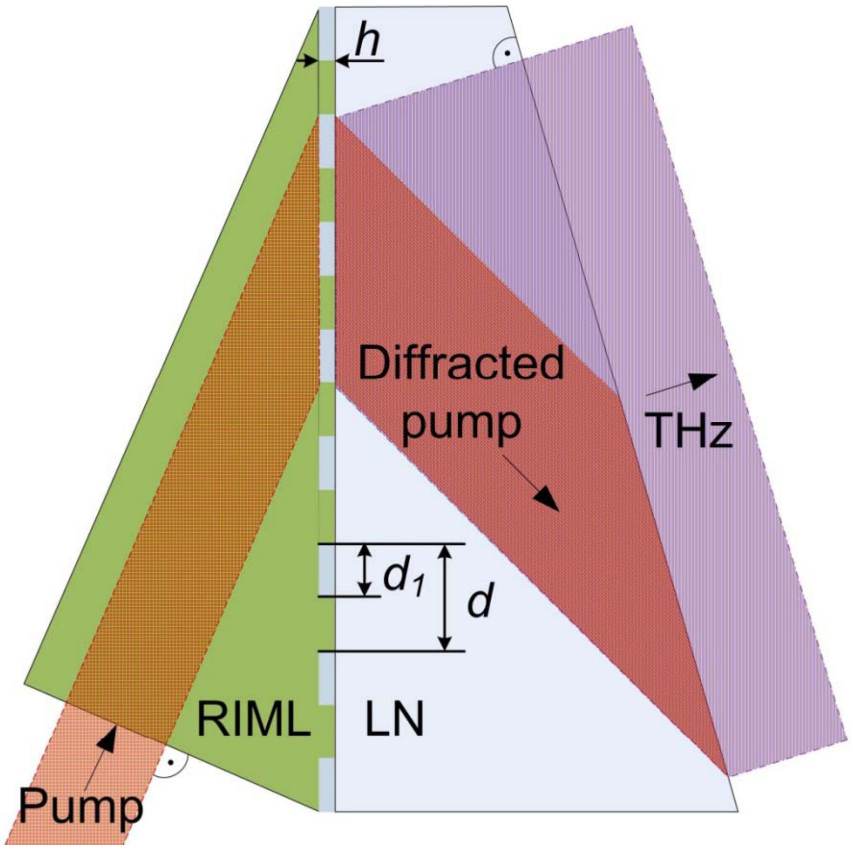


1 MV/cm field and 0.1% efficiency reached by applying our above results

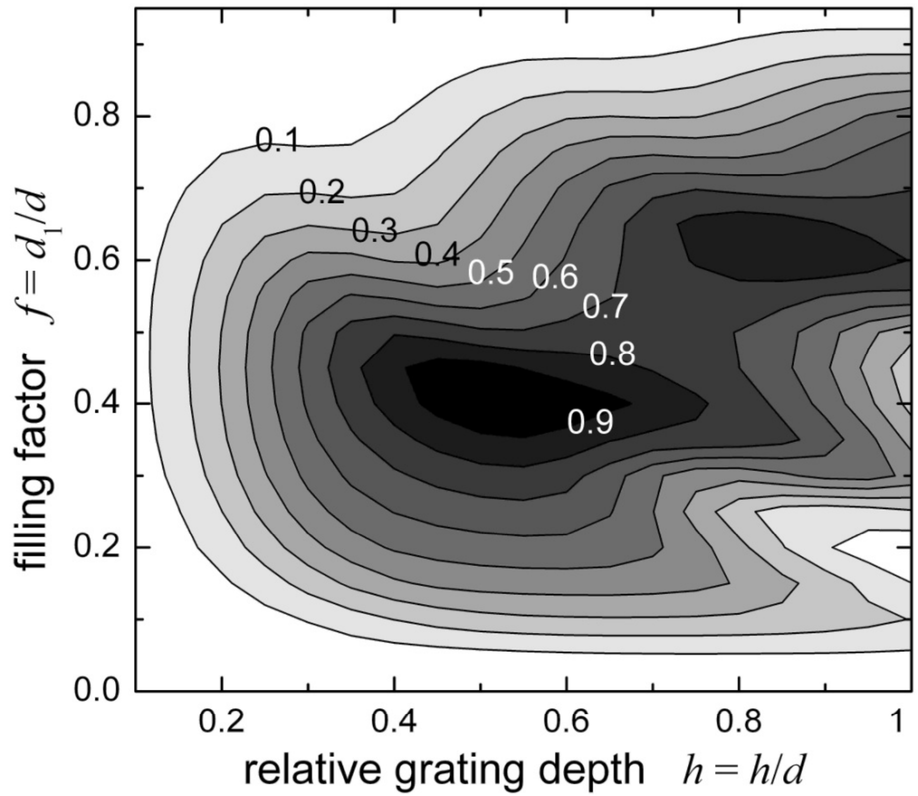
Hirori et al., Appl. Phys. Lett., 2011

Design of Contact Grating on $LiNbO_3$

- **Contact grating for THz generation proposed:** Pálfalvi et al., APL, 2008
- **Index matching by glass:** Nagashima et al., Japan J. Appl. Phys., 2010
- **Refractive index matching liquid (RIML):** Ollmann et al., APB, submitted

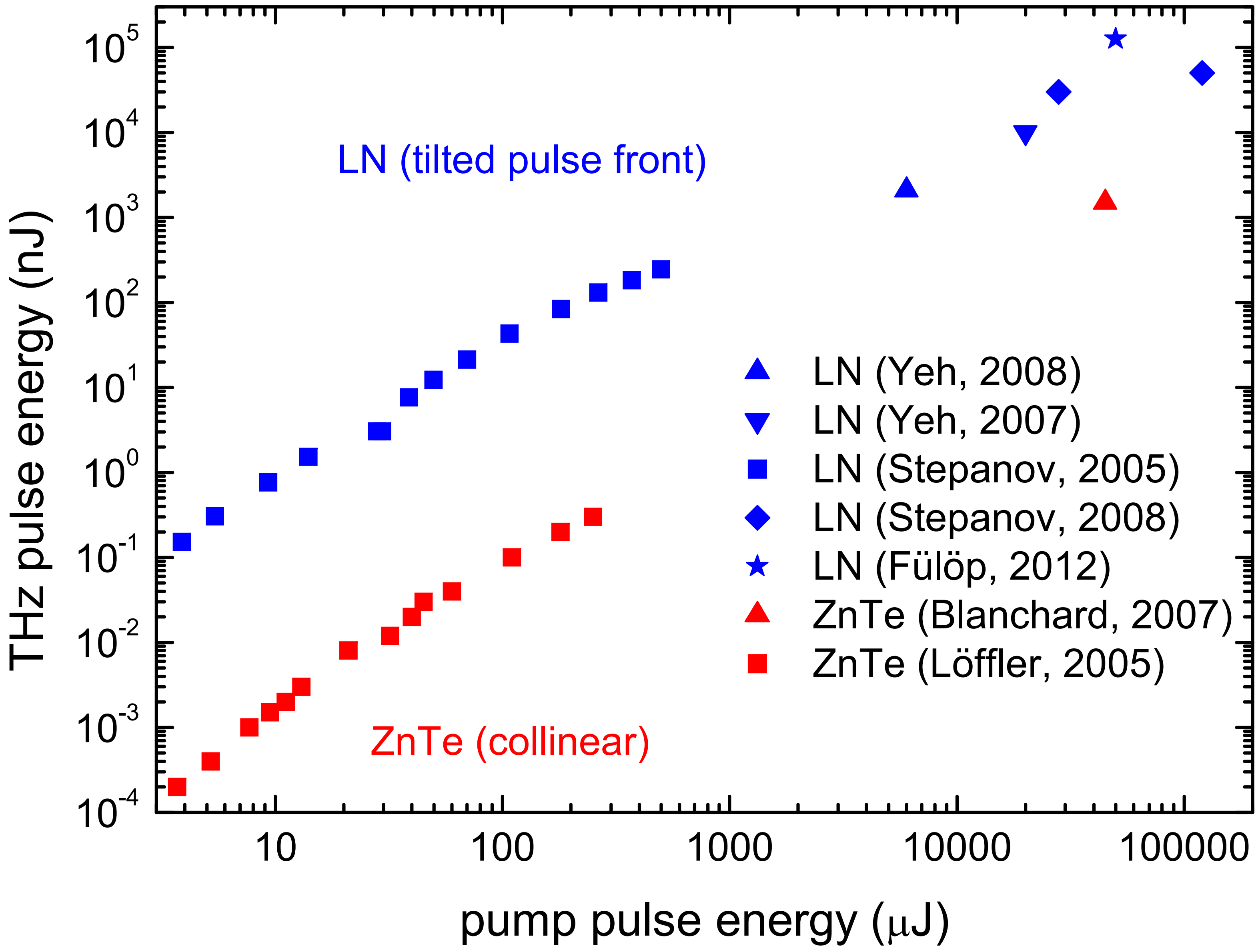


Diffraction efficiency for order -1



RIML for BK7, *d* = 350 nm pitch grating

THz Energy vs. Pump Energy



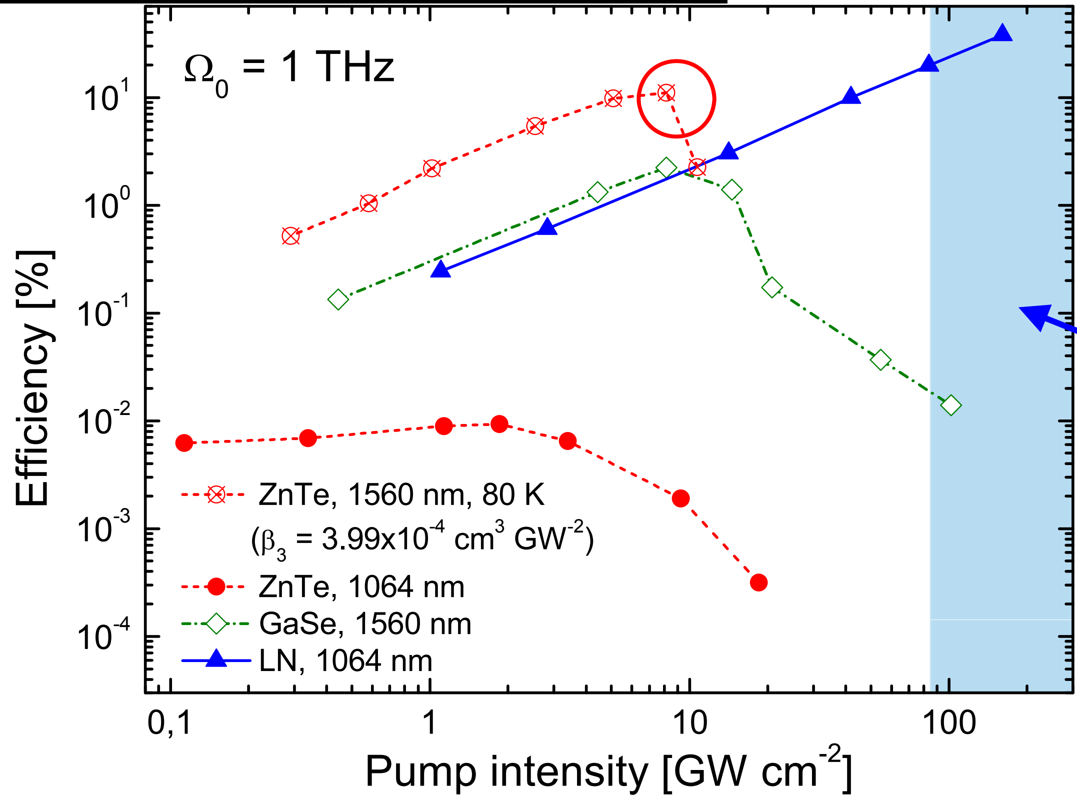
Reconsidering Semiconductors for THz Generation

Material	λ_p = 800 nm		λ_p = 1064 nm		λ_p = 1560 nm	
	MPA	γ	MPA	γ	MPA	γ
ZnTe	2PA	collinear	2PA	22.4°	3PA	28.6°
GaSe	2PA	16.5°	2PA	25.9°	3PA	30.2°
$LiNbO_3$	3PA	62.7°	4PA	63.4°	5PA	63.8°

MPA: lowest-order multi-photon pump absorption

γ: pulse-front tilt angle

Fülöp et al., Opt. Express, 2010



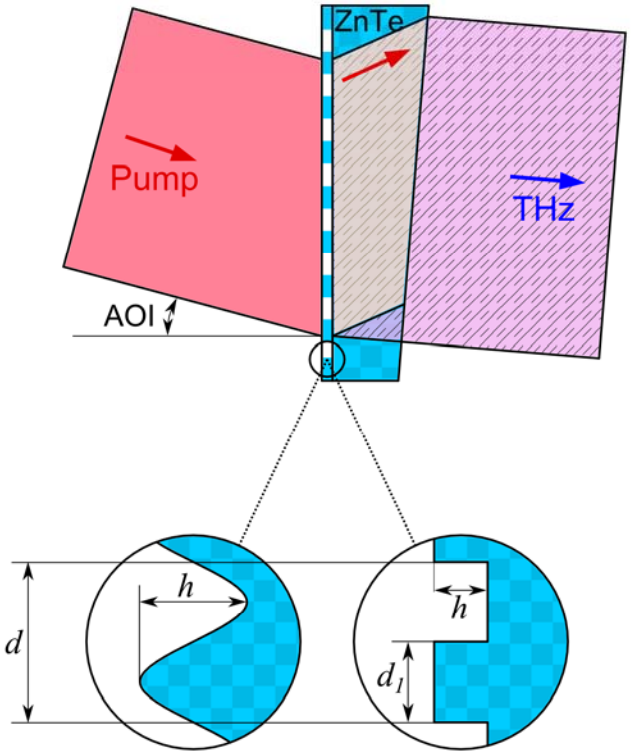
Saturation in $LiNbO_3$: Hoffmann et al., Opt. Express, 2007

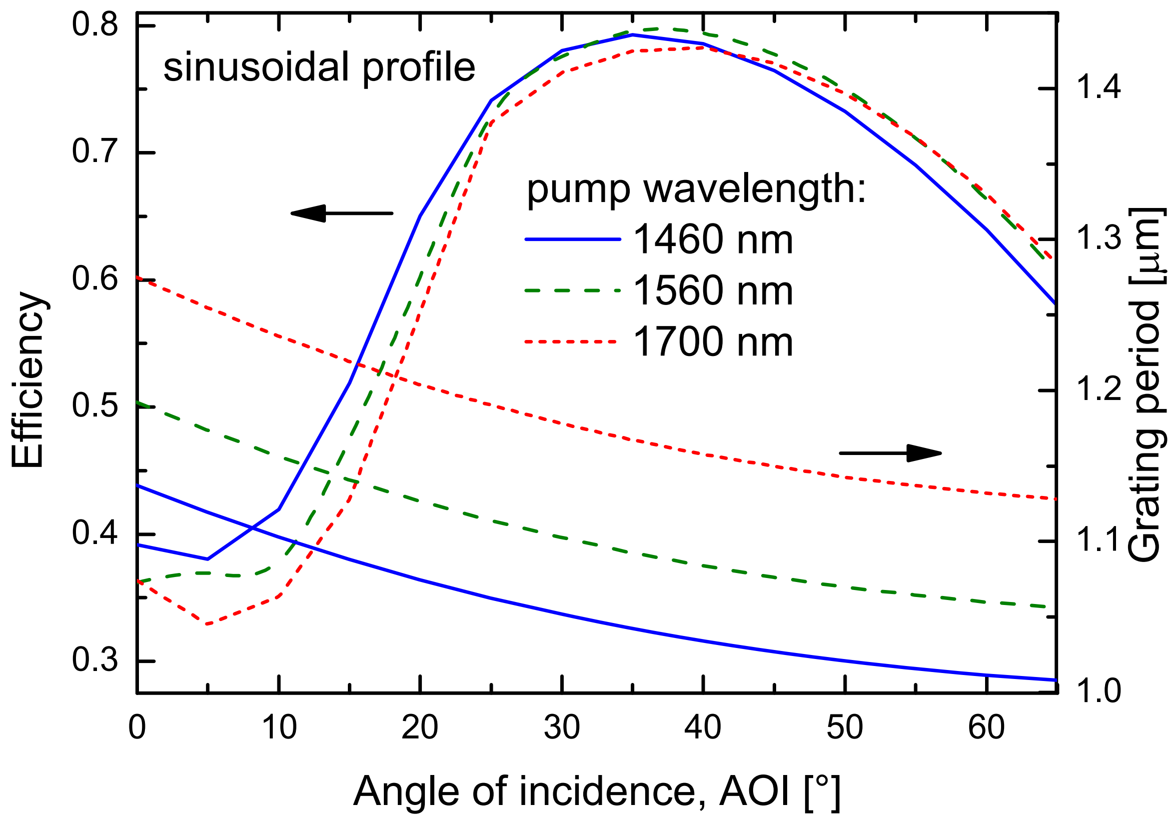
Advantages:

- **Pump spot diameter can be increased**
- **Smaller tilt angle, resulting in longer effective length**

Design of Contact Grating on ZnTe

Ollmann et al., in preparation





Pump: 1.5 µm wavelength, 5 cm beam diameter

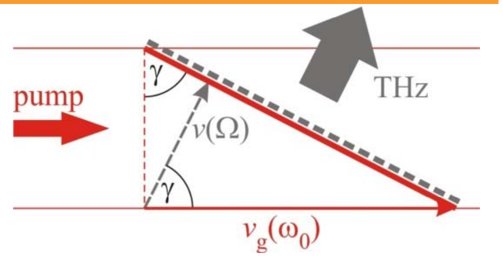
→ THz energy = 2.2 mJ

→ THz intensity = 630 GW/cm^2 (focused)

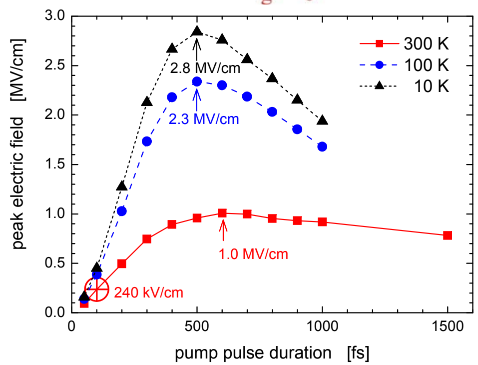
→ THz field ≈ 20 MV/cm (focused)

Summary

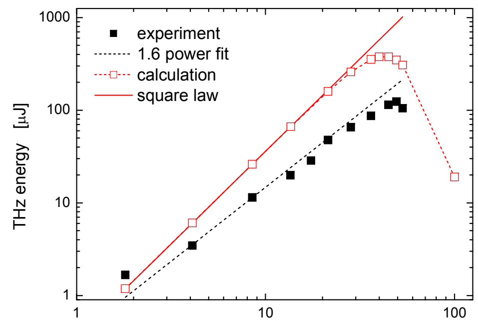
- **Single-cycle THz pulse generation with 100 MV/cm field and ~10 mJ energy is feasible around ~1 THz frequency by OR pumped by efficient DPSS lasers**



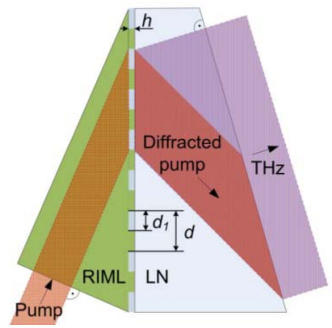
- **Optimization of the TPFP technique**
  - → **pump pulse duration: 500 fs optimal for LN**
  - → **low crystal temperature**
  - → **optimal crystal length**
  - → **contact grating setup for LN and ZnTe (extension of beam size)**



- **Preliminary experimental results:**
  125 µJ energy, 0.25% efficiency (non optimised!)



- **New application possibilities:**
  - → **nonlinear THz spectroscopy**
  - → **manipulation of electrons and ions**



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