



Ultrafast lasers for THz generation using nonlinear optical techniques Part 1: ultrafast laser technology

Clara J. Saraceno

ICTP Winter School on Terahertz Optics and Photonics - February 2023

- Be interested in latest advances in ultrafast laser technology and Terahertz technology
- A running understanding of electromagnetics and Fourier transformations
- A QR-code reader for interactive questions
- A pen and piece of paper
- Don't be shy: ask questions during and after...



https://arsnova.rub.de/mobile/#id/87676337

About me

2002 - 2007	2008	2009 - 2012	2013 - 2016	2016 - 2019	2020
Studies in Lyon and	Experience	PhD degree	Postdoc	Associate Prof.	Full Prof.
, Paris, France	in Industry	Physics	Topic: High	(Tenure Track)	
'Classe	Santa	Торіс:	power ultrafast	Research:	"Photonics and
Preparatoire'	Clara,	High-power ultrafast	oscillators for	High-power	Ultrafast Laser
and	California,	thin-disk lasers	compact	ultrafast lasers,	Science"
'Grande Ecole'	USA		XUV sources	THz sources,	
specialized in optics	Topic:			time-domain	
	R&D in		Inine	spectroscopy	Photonics and
	ultrafast				Ultrafast Laser Science
1	oscillators		NEUCHÂTEL		puls 🗸
INSTITUT		NT. ETHzürich	ETH zürich		
				RUHR UNIVERSITÄ	
ParisTech				BOCHUM	

Acknowledgments



Collaborators:

Prof. Balzer, University of Duisburg Essen Prof. Preu, TU Darmstadt Prof. Globisch, Dr. Kohlhaas, Fraunhofer HHI

Prof. Havenith, RUB Prof. Keller, ETH Zürich ... etc

"THz" sub-team:

Shahwar Ahmed

Dr. Martin Saraceno

Dr. Denizhan K. Kesim Dr. Celia Millon Samira Mansourzadeh Tim Vogel Mohsen Khalili "Ultrafast Lasers" sub-team: Dr. Yicheng Wang Dr. Weichao Yao Sergei Tomilov Alan Omar



Alexander von Humboldt Stiftung/Foundation











- <u>few-cycle THz pulses</u>

 phase-stable, field-resolved
 time-resolved
 ultra broadband
- commonly accepted: low average power





Time-domain spectrometer



Time-domain spectrometer



Time-domain spectrometer





Laser light pulses with fs - ps durations

Broadband spectra with hundreds of nm

Peak powers MW – GW, intensities 10¹² – 10¹⁵ W/cm²

... and beyond



some scaling

1 Gigawatt 1 000 000 000 W



For a very short time, ultrafast lasers generate **peak powers** similar to the **power of a nuclear power plant** (some laser systems generate TeraWatts)

1 femtosecond = 0. 000 000 000 000 001 s



human hair thickness $\approx 10^{-4}$ m



distance earth sun $\approx 10^{11} \text{ m}$

Measuring **1 second** with **femtosecond precision** is like measuring the **distance between the earth and the sun** with the precision of a **hair thickness**!

ultrashort timescales



A. H. Zewail in 1994: understand transition states in chemical reactions



Prize motivation: "for his studies of the transition states of chemical reactions using femtosecond spectroscopy"

<u>access to ultrafast time-scales:</u> observe and use ultrafast dynamics

- understand atomic and molecular dynamics
- fast data communication

• ...



broad frequency combs



<u>frequency comb:</u> ruler for unknown frequencies

• optical clocks

• ...

• high-precision spectroscopy

Roy J. Glauber, John L. Hall, Theodor W. Hänsch, in 2005:



Prize motivation: "for their contributions to the development of laser-based precision spectroscopy, including the optical frequency comb technique"

of the set of the set



achieve extremely high intensities: nonlinear optics and material modification

- material processing: cold ablation
 - multi-photon biomedical imaging
 - nonlinear optics

• • • •

"almost perfect micromachining of almost any material"

Bioresorbable polymer

Polyimide Hole array drilling

Ceramics

LIGHTMOTIF

Courtesy of Lightmotif



Courtesy of Resonetics Courtesy of Dr. Kurt Weingarten, Lumentum



Courtesy of Resonetics

"... and create functional surfaces"



Notatio n	Everyday parameters	How are they linked together?	Subtleties
Ep	Pulse energy (J)		
τ _p	Pulse duration (fs)		 Definition often FWHM – can be misleading RMS pulse duration better suited but rarely used
P _{pk}	Peak power (W)	Can be calculated from E_p and τ_p	 Requires knowledge about pulse amplitude shape Simple for well-known pulse shapes (Gaussian,) Usually fixed (wanted) for a given experiment
I _{pk}	Peak intensity (W/m ²)	<i>Can be calculated from P</i> _p and <i>beam area A</i>	Requires knowledge on transverse beam profile

Light Pulse Electric Field: superposition of monochromatic waves

$$E(t) = \frac{1}{2\pi} \int \tilde{E}(\omega) e^{j\omega t} d\omega$$

$$E(t) = A(t)e^{j\omega_0 t}$$
 where $A(t) = \frac{1}{2\pi}\int \tilde{A}(\Delta\omega)e^{j\Delta\omega t}d\Delta\omega$

Pulse envelope A(t): sufficient to describe pulse entirely





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λ ₀ , ν ₀	Central Wavelength (nm), frequency (Hz)		For complex spectra the central frequency might become different to the center of mass of the spectrum
$\Delta \lambda_{p,} \Delta v_{p}$	Spectral bandwidth (nm, Hz)		 Often defined by width of spectral intensity in nm. Only relative bandwidths are the same in wavelength and frequency

Spectral domain: spectral amplitude and spectral phase

$$\tilde{E}(\omega) = \left| \tilde{E}(\omega) \right| \exp(i\varphi(\omega))$$

 \rightarrow Spectral amplitude continuous function for one pulse

 \rightarrow "short pulse – broad amplitude spectrum"



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ТВР	Time-bandwidth product (no unit)	$TBP = \tau_{p} \Delta v_{p}$	 Defined with intensity FWHM Reaches a minimum that gives us information about the shortest pulses reachable with a given spectral width Can be flawed for complex, very short pulses



Full pulse characterization needs amplitude and phase

Requires sophisticated measurement techniques (FROG, SPIDER...)









Notatio n	Everyday parameters	How are they linked together?	Subtleties
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f _{CEO}	Carrier envelope frequency		Difficult to measure
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All applies to THz but definitions become flawed because single-cycle pulse





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τ _p	Pulse duration (fs)		 Definition often FWHM – can be misleading RMS pulse duration better suited but rarely used
f _{CEO}	Carrier envelope frequency		Difficult to measure
f _{rep}	Repetition rate (Hz)		
Pav	Average power (W)	$P_{\rm av} = f_{\rm rep} E_{\rm p}$	Usually technology limited
P _{pk}	Peak power (W)	Can be calculated from E_p and τ_p	 Requires knowledge about pulse amplitude shape Simple for well-known pulse shapes (Gaussian,) Usually fixed (wanted) for a given experiment
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Question 1





A fs-pulse has an intensity FWHM duration of 100 fs an average power of 1 W and a repetition rate of 1 kHz.

What is the peak power of the pulse?

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Question 1



https://arsnova.rub.de/mobile/#id/87676337

A fs-pulse has an intensity FWHM duration of 100 fs an average power of 1 W and a repetition rate of 1 kHz.

What is the peak power of the pulse?

Knowledge and control of your lasers has influence on nonlinear conversion!



Question 2



A fs-pulse centered at 1000 nm has a spectral intensity full-width half maximum bandwidth of 100 nm, what bandwidth can we potentially reach in the THz domain with optical rectification (approximations are allowed)?

Optical rectification can be seen as frequency mixing of spectral components inside your bandwidth: but short pulses are needed!

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Traditional front-end femtosecond laser for THz-TDS

Ti:Sapphire amplifiers/oscillators at 800 nm



(typical amp) $P_{av}=1 W$ $\tau_p=30 \text{ fs}$ $f_{rep}=1 \text{ kHz}$ $E_p=1 \text{ mJ}$

- large
- expensive
- scientific system
- high pulse energy

user community: mostly scientific

Erbium fiber laser at 1550 nm



(typical) P_{av} =100 mW τ_{p} =100 fs f_{rep} =100 MHz E_{p} =1 nJ

- compact
- cheap
- fiber-based robust
- low pulse energy

user community: scientific and industrial

Low average power limited to few-watts

the workhorse of ultrafast science





Typical amplifiers:

- Pulse duration ~30 fs
- Pulse energy ~mJ
- Rep Rate ~few kHz
- Peak power ~ GW

Typical oscillators:

- Pulse duration ~20 fs
- Pulse energy ~nJ
- Rep Rate ~ tens of MHz
 - **Peak power** ~ 10s kW

→ Average power limited to few watts $P_{av} = E_p \cdot f_{rep}$

the workhorse of ultrafast science Femtosecond Ti:Sa oscillator Ti:sapphire laser: 3 non-radiative Nongreen pumps transition radiative 2 Pump Laser transition **transitions:** ≈500 nm **Regenerative amplifier** ≈800 nm Large heat Ş load non-radiative transition Ű.

the workhorse of ultrafast science



Ti:sapphire laser:



- + other problems:
- small upper-state lifetime (few µs)
 → high pump intensities needed to saturate
- degradation of crystal quality when increasing doping

Bulk geometry with large thermal load → thermal aberrations

CW power: material properties + advanced cooling geometries

Yb:YAG laser:











✓ Thin disk



Additional difficulty for ultrafast operation: nonlinearities need to be kept small

Self-phase modulation

 $n(I) = n + n_2 I$

 $I(t) \rightarrow$ self-phase modulation $I(x,y) \rightarrow$ self-focusing

$$\phi(t) = -kn(I)L_{\kappa} = -k[n+n_2I(t)]L_{\kappa}$$

Nonlinear phase [mrad] Nonlinear refractive index [cm^{2/}W]

Self-phase modulation (SPM)

Self-phase modulation



Self-focusing



→ Leads to beam degradation, spatio-temporal couplings, catastrophic damage if nonlinear phase is too large


high power fiber CPA

Most commonly used: chirped pulse amplification (CPA)



further scaling: coherent combination



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Optics Letters

10.4 kW coherently combined ultrafast fiber laser

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³INRS, Centre Énergie Matériaux et Télécommunications, 1650 Blvd. Lionel-Boulet, Varennes, J3X1S2, Canada ⁴Fraunhofer Institute for Applied Optics and Precision Engineering, Albert-Einstein-Straße 7, 07745 Jena, Germany *Corresponding author: michael.mm.mueller@uni-jena.de

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Performance:

- 10.4 kW
- 254 fs pulses
- 80 MHz
- 130 µJ



slab amplifiers 🖉

December 15, 2010 / Vol. 35, No. 24 / OPTICS LETTERS 4169

Compact diode-pumped 1.1 kW Yb:YAG Innoslab femtosecond amplifier

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We demonstrate a compact diode-pumped Yb:KGW femtosecond oscillator-Yb:YAG Innoslab amplifier master oscillator power amplifier (MOPA) with nearly transform-limited 636 fs pulses at 620 W average output power, 20 MHz repetition rate, and beam quality of $M_x^2 = 1.43$ and $M_y^2 = 1.35$. By cascading two amplifiers, we attain an average output power of 1.1 kW, a peak power of 80 MW, and a 615 fs pulse width in a single linearly polarized beam. The power-scalable MOPA is operated at room temperature, and no chirped-pulse amplification technique is used. © 2010 Optical Society of America





clever geometry: CPA avoided for moderate pulse energies issues: pointing, beam quality



- \rightarrow outstanding heat removal, extremely small thermal aberrations
- \rightarrow Yb^{3+}-doped gain: diode pumped, accessible high-power diodes
- \rightarrow good pump absorption: many passes through gain required
- \rightarrow very small accumulated nonlinearities

\rightarrow ideal for ultrafast + high power

A. Giesen, et al., Appl. Phys. B 58, 365 (1994)







Courtesy of Dirk Sutter

14 kW with $\eta_{opt.}$ >70%

4 kW TEM₀₀ (2013)

 \Rightarrow Further scaling w/ multiple heads, no barriers for power scaling beyond current levels





Gottwald et al., Security and Defense 2013

thin-disk ultrafast amplifiers



thin-disk amplifier geometries

→ Thin-disk: low gain per pass (typical 10%)





Regenerative amplifier

 → moderate amplification, high extraction (booster) → large amplification (main)



Ultrafast thin-disk multi-pass amplifier system providing 1.9 kW of average output power and pulse energies in the 10 mJ range at 1 ps of pulse duration for glass-cleaving applications

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Abstract: An ultrafast Yb-doped thin-disk multi-pass laser amplifier system with flexible parameters for material processing is reported. We can generate bursts consisting of four pulses at a distance of 20 ns and a total energy of 46.7 mJ at a repetition rate of 25 kHz. In single-pulse operation, 1.5 kW of average output is achieved at 400 kHz when optimizing for a beam quality of $M^2 = 1.5$. Alignment for maximum output power provides 1.9 kW at the same repetition rate. All results are obtained without chirped-pulse amplification in the multi-pass set-up. The application potential of the system is demonstrated exploring its performance in materials processing of dielectrics. Cleaving of 3.8-mm-thick SCHOTT borofloat glass with a velocity of 1200 mm/s is demonstrated with 300 W of input power. Single-pass modification of 30 mm borosilicate glass is enabled with a Bessel beam at 1 kW of average power delivered by four-pulse bursts of an energy of 30 mJ.

- 1.9 kW
- 400 kHz
- (now up to 2.3 kW)
- 1.1 ps
- ! No CPA



Fig. 1. Schematic set-up of the amplifier system. The seed laser is a commercial TruMicro 2000, followed by two amplifier stages. Red lines indicate the laser beam. MPC: Multi-pass cell.



thin-disk regenerative amplifiers: state-of-the-art





Courtesy of Thomas Metzger



TRUMPF

"Flagship" Laser	
Energy:	200 mJ
Power:	>1.0 kW
Duration:	500 fs
Peak Power:	0.4 TW

T. Nubbemeyer et al. OL 42, 7 (2017)

New developments: 2 kW – 20 kHz – ps, etc...

thin-disk ultrafast oscillators



modelocked lasers





- Light circulates in resonator
- Losses compensated by gain in laser medium (which is externally pumped)
- Gain saturates to a level that it compensates the losses
- Light emission typically continuous
- Power: limited by thermal aberrations and pump power

modelocked lasers



- Short pulse circulates in cavity (fs-ps)
- High repetition rate pulse train at the output (MHz)
- Pulse starting
 - Semiconductor saturable absorbers
 - Kerr lensing
- Pulse formation
 - Soliton modelocking
 - Kerr lens modelocking
- Steady-state pulse parameters: interplay of gain, (saturable) loss, dispersion, Kerr nonlinearity, etc.

saturable absorbers



first modelocked thin-disk oscillator: nearly 20 years ago

June 1, 2000 / Vol. 25, No. 11 / OPTICS LETTERS 859

16.2-W average power from a diode-pumped femtosecond Yb:YAG thin disk laser

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Received February 15, 2000 We demonstrate a power-scalable concept for high-power all-solid-state femtosecond lasers, based on passive of severage output power in pulses with 720-56 duration, 0.47 μJ pulse energy, and the subprocess of the is to our known behavior as norm-long linhum theorem certain the subprocess of regime Single-pass frequency doubling throughy and Society of America power of Sine radiation. © 2000 Optical Society of America OCIS codes: 140.3480, 140.4650, 140.5680, 190.2620.

In recent years there has been great interest in pas-sively mode-locked lasers with high average output sively mode-locked lasers with night average output powers. At present, the frontiers in the picosecond regime are slightly below 30 W1⁻² whereas in the femegime are signify below 30 W, — whereas in the tem-osecond regime average output powers of a few waits ave been demonstrated.^{4,7} The need for mode-locked have been demonstrated.^{4,-7} The need for mode-locked high-power lasers is driven by many applications, par-ticularly those involving nonlinear wavelength conver-sion, which is facilitated by the high peak powers of such lasers. For example, RGB laser displays can be built if at least a few watts of average output power can be exampled at red, arean, and blue wavelength. built if at least a few watts of average output power can be generated at red, green, and blue wavelengths. The state of the state of the state of the state of the cept for femtosecond lasers with high average power. It is based on a Yb'NAG thin disk laser⁸ which we have naminaly mode looked for what is balanced to have passively mode locked for what is believed to nave passively mode locked for what is believed to be the first time, using a semiconductor saturable absorber mirror (SESAM).319 We obtained 16.2 W of absorber mirror (SESAM).²⁰⁰⁷ We obtained 16.2 W of average power in pulses with 730-fs duration, 0.47-µJ energy, and 560-kW peak power. This is by far more energy, and 560-kW peak power. This is by far more average power than ever demonstrated in the subpi-nultiwat average power have been obtained only from Tisapphire lasers.⁵ which, however, rely either on a bulke, inefficient arean-ion nump laser or on an expen-Thisapphire lasers," which, however, rey entirer on a bulky, inefficient argonion pump laser or on an expen-sive frequency-doubled diode-pumped pump laser. In the near future our concept should allow for even sig-nificantly bisher average powers.

Theantly higher average powers. The YAG is a very interesting material for diode-samped high-power formed and the absorption band cellent thermal properties, its wide absorption bandwidth. 10 June on Autor 200 for have been demonstrated in places on Autor 200 for have been demonstrated in these on Autor 200 for have been demonstrated in these on Autor 200 for have been demonstrated in these on Autor 200 for have been demonstrated in the set of the nificantly higher average powers. at 940 nm, and its broad ampuncation banaward. Pulses as short as 340 fs have been demonstrated in

a low-power laser.¹¹ However, a major drawback of

Yb YAG concerning mode locking is its small emissi to LAM concerning more locking is its small emission cross section. In a passively mode-locked laser the saturable absorber needed for mode locking introauturation autorroom needed for mode locking intro-duces a tendency of the laser toward Q-switching duces a tendency of the laser toward Q-switching instabilities. This tendency can drive the laser into the Q-switched mode locking ¹¹ (QML) regime, with mode-locked pulses under a Q-switched envelope. This exhibits incontinuate source for early models endmode-locked pulses under a gravitation enveloped This problem is particularly severe for gain media such This problem is particularly severe for gam media such as Yb-YAG, which have low laser cross sections and succeeded in suppressing QML with a combination of measures. First the thin disk laser head allows for succeeded in suppressing QML with a combination of measures. First, the thin disk laser head allows for operation with a high laser intensity and small spot size in the gain medium, i.e., a reduced gain-saturation energy. Second, we used a SESAM with relatively small modulation double (sat) 565. Third we designed analy down, we used a ODDAM with relatively anali modulation depth (=0.5%). Third, we designed small modulation depth (~0.5%). Third, we designed a laser cavity with a low repetition rate (34.6 MHz). Finally, operation in the soliton mode-locked regime¹ (with negative overall cavity dispersion) substantially increases the stability against QML¹² Basically this is because any increase in the rates energy of a white increases the stability against QML." Bancally this is because any increase in the pulse energy of a soliton increases the bandwidth and thus reduces the effective gain because of the limited gain bandwidth of the because of the limited gain bandwidth of stable

gain because of the limited gain bandwidth of the laser medium. This technique was essential for stable mode locking of our Yb-YAG thinks laser. The thin disk laser head consists of a 220 μ m thin Yb-YAG with one face on a power densities to the us to apply quite high pump-wavelength, and reflectiving the laser and pump wavelength, and the other side has an antireflection coating. As the diameter of the pump beam (=1.2 mm) is larger than the other side has an antireriection conting. As the diameter of the pump beam ($\approx 1.2 \text{ mm}$) is larger than the thickness of the disk ($\approx 220 \mu$ m), the heat flux is

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Ultrafast Laser Physics

ETH zürich

Group of Prof. U. Keller



16.2 W, 730 fs, 34.6 MHz, 0.5 µJ Aus der Au et al. Opt. Lett. 25, 11 (2000)



- 'One-box' oscillator
- femtosecond soliton pulses

Specifications:

- Emission wavelength around 1 µm (typical 1030 nm)
- Average power: up to 350 W [1]
- Pulse energy: up to 80 µJ [2]
- Repetition rate: 3-70 MHz
- Pulse duration: 30-1000 fs

1. F. Saltarelli et al., Opt. Express 27, 31465-31474 (2019) 2. C. J. Saraceno et al., Opt. Lett. 39, 9 (2014).

\rightarrow orders of magnitude higher levels than other oscillator laser technologies

why have these (and other) Yb systems taken long to be adopted? Yb:(Sc,Y,Lu)2O3 Yb:Lu2O3 Yb:SSO • . Yb:CALGO Yb:LuO Yb:YAG Yb:KLuW Yb:YCOB Yb:LuO3 / Yb:ScO3 Yb:KYW ۲ Yb:LuScO3 Yb:YAG @ 1030 nm • Yb:YAG: narrow emission bandwidth $\Delta\lambda \sim 7 \text{ nm}$ • pulse duration and average power/pulse energy 10 100 1000 Pulse width (fs)





Self-phase modulation



 \rightarrow Creates new frequency components

-- TDL

1050

pulse compression using SPM

C. V. Shank et al., "Compression of femtosecond optical pulses", Appl. Phys. Lett. 40, 761 (1982)

Hollow capillaries



Nisoli et al, Appl. Phys. Lett. 68, 2793 (1996)

- Fibers
 - Solid-core fibers



Hollow-core fibers



- Losses increase at moderate to small core sizes, typical 70% transmission
- Suited only for very high energies (mJ and above)
- Real guiding
- Solid-core: limited by self-focusing (4 MW for linear polarization and glass), damage threshold and bending loss at large mode areas
- Hollow-core: limited by difficulties in bending and damage

T. Südmeyer *et al.*, "Nonlinear femtosecond pulse compression at high average power levels by use of a large-mode-area holey fiber", Opt. Lett. 28 (20), 1951 (2003)

K. F. Mak *et al.*, "Two techniques for temporal pulse compression in gas-filled hollow-core kagomé photonic crystal fiber", Opt. Lett. 38 (18), 3592 (2013)

Compression in multi-pass cell





Figures courtesy J. Weitenberg Fraunhofer ILT Aachen **First realization (ILT Aachen):** Schulte et al. "Nonlinear pulse compression in a multi-pass cell," Opt. Lett. 41, 4511-4514 (2016)

Example Multi-pass cell compression SESAM modelocked Yb:YAG $P_{avg} = 112W$ $f_{rep} = 13.4 \text{ MHz}$ E_P $t_p = 88 \text{ fs}$ $P_{peak} = 80 \text{ MW}$ λ = 1030 nm = 8.4 µJ $P_{\rm avg} = 123 \, \text{W} \quad f_{\rm rep} = 13.4 \, \text{MHz}$ $P_{avg} = 112 \text{ W}$ t_p = 88 fs $t_{\rm n}$ = 534 fs λ = 1030 nm 130 cm 10 - Retrieved Retrieved Measured Spectrum [a.u.] 1.0 - TL Intensity [a.u.] 9.0 8.0 thin disk − N Phase [rad] Phase [rad] Yb:YAG 88 fs 24x -550 fs² 60 cm := 300mm = -350fs² 300 = 300 mm FS -5 § 0.25 0.2 0.00 0.0 -10ROC :: GDD :: -300 -200 -100 0 100 200 300 1010 1020 1030 1040 1050 12 mm Time [fs] Wavelength [nm] 42 passes 35 mbar

- Herriott type multi-pass cell¹ + fused silica + negative dispersive mirror pair
- Generated spectrum agrees well with 3D pulse propagation model
- $M^2 < 1.15$
- Excellent efficiency: 91%

Tsai et al. "Efficient nonlinear compression of a mode-locked thin-disk oscillator to 27 fs at 98 W average power," Opt. Lett. 44, 4115-4118 (2019)

Works for an extremely large variety of parameters



Letter



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6250

Kilowatt-average-power compression of millijoule pulses in a gas-filled multi-pass cell

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Ultrafast lasers for THz generation using nonlinear optical techniques Part 2: Terahertz generation at high-power

Clara J. Saraceno

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We have kilowatt, fs lasers, so now what?



Trend: high-power from THz to XUV

spectral coverage



We have kilowatt, fs lasers, so now what?



THz lab-sources: low average power



- state-of-the-art THz power: mW level
- repetition rate or pulse strength: compromise necessary
- origin of limitations: low driving power and efficiency
- power-hungry applications: traditionally accelerators







why do we care?



- Same measurement time -- more samples per second, higher dynamic range (and bandwidth!)
 OR
- same dynamic range at 10 times shorter measurement time

Performance of a TDS?

 $SNR = \frac{\text{mean magnitude of amplitude}}{\text{standard deviation of amplitude}}$

 $DR = \frac{\text{maximum magnitude of amplitude}}{\text{rms of noise floor}}$

→ Stability from trace to trace

→ Fluctuations within one EOS trace

M. Naftaly and R. Dudley, "Methodologies for determining the dynamic ranges and signal-to noise ratios of terahertz time-domain spectrometers," Optics Letters, vol. 34, p. 1213, Apr. 2009.

why do we care?



 Same measurement time -- more samples per second, higher DR (and bandwidth!)

OR

 same DR at 10 times shorter measurement time

Average power advantage strongly depends on the experiments specifics and detection setup

our THz research in a nutshell

High average power ultrafast lasers



High average power THz-TDS



our THz research in a nutshell





source "dream":

- short, phase-stable THz pulses covering [1-10 THz] or more
- high repetition rate (MHz): high signal-to-noise ratio, short measurement times
- high energy (µJ): nonlinear spectroscopies

high average power (watts)



- 200 scientists in the Ruhr area
- solvent molecular details in the focus

selection of THz generation methods



Typically min. required driving pulse energy for efficient conversion

nJ

- 1. Burford, N. M. & El-Shenawee, M. O. Opt. Eng. 56, 010901 (2017).
- 2. Seifert, T., Jaiswal, S., Martens, U. et al. Nature Photon 10, 483–488 (2016).
- 3. Matsubara, E., Nagai, M. & Ashida, M. Appl. Phys. Lett. 101, (2012).

Photonics and Ultrafast Laser Science

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selection of THz generation methods



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All generation methods have interesting and unexplored high average power regimes of operation!

selection of THz generation methods **MHz repetition rates** thin-disk oscillators Two-color ionized photocurrent in $\chi^{(2)}$ in non-centrosymmetric crystals spintronic emitters gas plasma semiconductor Photoconductive Difference Optical rectification **Photo-currents** Spin Hall effect switch frequency mixing

~6 THz

3-10 THz

Typically min. required driving pulse energy for efficient conversion

 $\sim 30 \text{ THz}^2$

nJ

 $\sim 6 \text{ THz}^1$

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>30 THz³

mJ

Workhorse thin-disk oscillator



Workhorse thin-disk oscillator



Flexible pulse compression



- Herriott type multi-pass cell[#] + fused silica + dispersive mirror pair
- Generated spectrum agrees well with 3D pulse propagation model
- M² < 1.15
- Excellent efficiency: 91%

 $P_{avg} = 112 W$ $f_{rep} = 13.4 MHz$ $E_{p} = 8.4 \mu J$ $t_{p} = 88 \text{ fs}$ $P_{peak} = 80 MW$ $\lambda = 1030 \text{ nm}$

1. J. Schulte et al., "Nonlinear pulse compression in a multi-pass cell," Opt. Lett. 41, 4511-4514 (2016)

Collinear optical rectification



- Herriott type multi-pass cell[#] + fused silica + dispersive mirror pair
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Choice of nonlinear crystal?

velocity matching

Coherence length = measure of walk-off between pump pulses and the generated THz pulses

$$l_{\rm c} = \frac{1}{2f_{\rm T} \left| \frac{1}{v_{\rm p}^{\rm gr}} - \frac{1}{v_{\rm T}^{\rm ph}} \right|}$$

• strength of nonlinear coefficient improves conversion efficiency

• THz absorption

strongly affects bandwidth, typically phonon absorption lines

damage threshold

nonlinear process scales with intensity, limited by detrimental processes (multiphoton absorption, free-carrier absorption, damage, etc.)



Material	$E_{\rm g}~[{\rm eV}]$	$d_{\rm eff} [{\rm pm/V}]$	$n_g^{1030\mathrm{nm}}$	n _T	α _T [1/cm]	FOM [pm ² cm ² /V ²]
 GaSe	2.02	28.0	2.96 [105]	3.27	0.5	1.1
GaP	2.48	24.8	3.33 [106]	3.34	0.2	0.8
ZnTe	2.26	68.5	2.98 [107]	3.17	1.3	4.5
LN	3.8	168.0	2.22 [108]	4.96	17.0	16.9
LN 100 K		168.0	2.22 [108]	4.96	4.8	48.8
DAST		615.0	2.59 [109]	~2.25[101]	~30.0[101]	151.6
HMQ-TMS		>190.0 [110]	2.14 [111]	~2.17[111]	~50.0[111]	>5.8

etc.....

Broadband THz generation in GaP



- 1.35 mW for OR in GaP and broadband THz spectra (up to 6 THz)
- Limitations at high power level remained unknown
- Resistance to high-average power and broad bandwidth prompted further study

	P _{THz}	1.35 mW	
	E _{peak}	~ 7.5 kV/cm	
THZ	P _{peak}	~ 300 W	
	η	1.2·10 ⁻⁵	
	f _{rep}	13.3 MHz	

F. Meyer et al., "Milliwatt-class broadband THz source driven by a 112 W, sub-100 fs thin-disk laser," Opt. Express **27**, 30340-30349 (2019) N. Hekmat, et al. "Cryogenically cooled GaP for optical rectification at high excitation average powers," Opt. Mat. Express, vol. 10, (2020)



- Only small decrease of refractive index and velocity matching conditions, minor increase in conversion efficiency, broader bandwidth
- Multi-photon absorption (MPA) still the main limiting factor for efficiency in our excitation regime



c) 112 W 77 K

N. Hekmat, et al. "Cryogenically cooled GaP for optical rectification at high excitation average powers," Opt. Mat. Express, vol. 10, (2020)

tilted pulse front in Lithium Niobate

- Lithium Niobate (LN): high nonlinearity and • little multi-photon absorption
- But: phase matching requires tilting the ٠ pulse $v_{NIR}^{gr} \cos \gamma = v_{THz}$ $(n_{ar, NIR} \approx 2.2)$ $(n_{THz} \approx 5)$
- Increased experimental complexity, more • complex generation process due to spatiotemporal couplings and less bandwidth
- **Conversion efficiencies on the 1%** • level demonstrated (at lower repetition rate)





- Higher conversion efficiency at the expense of bandwidth
- Record-high average powers
- Limitations identified: combination of thermal effects and walk-off/depletion due to small spot sizes
- Scaling to watt-level: technically possible

	i		
	P _{THz}	66 mW	
N	E _{peak}	~16.7 kV/cm	
THz	P _{peak}	~18 kW	
	η	6·10 ⁻⁴	
	f _{rep}	13.3 MHz	

F. Meyer et al., "Single-cycle, MHz repetition rate THz source with 66 mW of average power," Opt. Lett. 45, 2494-2497 (2020)
F. Wulf, et al., "Analysis of THz generation using the tilted pulse front geometry in the limit of small pulse energies and beam sizes" Opt. Express 29(12), 18889-18904 (2021)

Benchmarking



A "mini-accelerator"

High-Field High-Repetition-Rate Terahertz facility @ ELBE (TELBE)



TELBE: single-cycle @100 kHz, 0.25 μ J \rightarrow 25 mW TELBE (target): single-cycle @ 1 MHz \rightarrow 0.2 mW Our oscillator-driven source @13 MHz \rightarrow 66 mW

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THz imaging





characterization and identification in the THz range..."





In collaboration with Prof. Jan Balzer, University of Duisburg Essen Lensless THz imaging of 3D printed sample



S. Mansourzadeh, D. Damyanov, T. Vogel, F. Wulf, R. Kohlhaas, B. Globisch, T. Schultze, M. Hoffmann, J. C. Balzer, and C. J. Saraceno "High-power Lensless THz Imaging of Hidden Objects," in IEEE Access, doi: 10.1109/ACCESS.2020.3048781 (2021)

THz imaging with LN source



- Same detector and acquisition time
- Contrast enhancement
- Difference in material recognizable

	Commercial system (TeraK15)	High-power TDL
Power	200 µW	20 mW
Pulse duration	100 fs	580 fs
Wavelength Laser	1550 nm (receiver optimized for this λ)	1030 nm
Repetition rate	100 MHz	13.4 MHz

Potential for improvement very large:

- Measurement acceleration
- 3D images
- Concealed objects

- ...

S. Mansourzadeh, D. Damyanov, T. Vogel, F. Wulf, R. Kohlhaas, B. Globisch, T. Schultze, M. Hoffmann, J. C. Balzer, and C. J. Saraceno "High-power Lensless THz Imaging of Hidden Objects," in IEEE Access, doi: 10.1109/ACCESS.2020.3048781 (2021)

Organic crystals:

- Combine high conversion efficiency and broad THz bandwidth in collinear scheme
- Most results limited to low repetition rate due to poor thermal properties
- BNA (N-benzyl-2-methyl-4-nitroaniline): collinear phase-matching for 1030 nm



S. Mansourzadeh, T. Vogel, M. Shalaby, F. Wulf, and C. J. Saraceno "Milliwatt average power, MHz-repetition rate, broadband THz generation in organic crystal BNA with diamond substrate" Optics Express Vol. 29, Issue 24, pp. 38946-38957 (2021)

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Back to the state-of-the-art



10 MHz – 100 MHz Moderate pulse energies (10s nJ) Promising for linear spectroscopy, imaging, etc. Back to the state-of-the-art



Lithium niobate - slab amplifier



- 144 mW (344 W input)
- 100 kHz repetition rate
- Lithium Niobate: up to 2 THz
- 150 kV/cm estimated peak E-field

P. Kramer, M. Windeler, K. Mecseki, E. G. Champenois, M. C. Hoffmann, and F. Tavella, "Enabling high repetition rate nonlinear THz science with a kilowattclass sub-100 fs laser source," Opt. Express 28, 16951-16967 (2020)

IAP, University of Jena, Germany





- 640 mW (633 W input)
 500 kHz repetition rate
- Very broadband up to 30
 THz performance limited by detection
- Estimated peak E-field 1 MV/cm

Joachim Buldt, Henning Stark, Michael Müller, Christian Grebing, César Jauregui, and Jens Limpert, "Gas-plasmabased generation of broadband terahertz radiation with 640 mW average power," Opt. Lett. 46, 5256-5259 (2021)



- 400 kHz

- 25 mW of average power (19 W input)
- Simple turn-key commercial laser
- Repetition rate variable, allows to study accumulation effects
- Estimated peak E-field 60 kV/cm

C. Millon, et al. Optics Express, accepted for publication (2023)





- 540 kHz rep rate
- 5.6 mW THz power (6 W input)
- Very broad bandwidth up to 7.8 THz
- Conversion efficiency similar to low repetition rate
- Estimated peak E-field 29 kV/cm

S. Mansourzadeh et al, APL Photonics, accepted (2023)

Back to the state-of-the-art



100 kHz – 1 MHz Very high pulse energies (mJ THz energies) Promising for nonlinear spectroscopy

Higher THz energies

Next step: Thin-disk amplifier 500 W @ 10 kHz \rightarrow 50 mJ, 700 fs



- First step: 40 kHz rep rate

- 643 mW THz power
- peak E-field --- not measured yet! should approach MV/cm

T. Vogel – in preparation

Conclusion



- Few-cycle THz broadband sources with average powers in the tens to hundreds of mW can be generated using new high-power laser technology with hundreds of watts
- Watt-level and beyond seems to be near future goals

Outlook:

- Keep improving the sources, both driving lasers and THz generation techniques
- Work on suitable detectors
- · Find other areas of application where the sources can shine







