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# **Sections of a Stage 1 Registered Report**

# ***Scientific Reports***

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**Possibility of multi-millijoule-level single and multicycle compact terahertz sources pumping by Co2 laser**

György Tóth1,2\*, Gabit Nazymbekov1, Gergő Illés1, Nelson Mbithi1, János Hebling1,2,3\*

1 Institute of Physics, University of Pécs, Pécs 7624, Hungary

2 Szentágothai Research Centre, University of Pécs, 7624 Pécs, Hungary

3 ELKH-PTE High-Field Terahertz Research Group, Pécs, Hungary

\* Corresponding authors: György Tóth (tothgy@fizika.ttk.pte.hu); János Hebling (hebling@fizika.ttk.pte.hu).

# **Abstract**

# **Introduction**

THz pulses generated by optical rectification can be used in spectroscopy1 and imaging2, electron3,4 and proton5 acceleration, molecule orientation6 and others7. Optical rectification of pulse front tilted short laser pulse in lithium niobate (LN) crystal is one of the most effective methods to generate THz pulses with high pulse energy and high peak intensity8-10. However, there are some limiting factors to significantly increase the THz energy further using LN crystal11,12. These limiting factors are mainly due to the shape of the non-linear prism.13 However in the conventional TPF setup the ~63° angle of the prism is mandatory to achieve phase matching. There are some new techniques14-16 – which are using micro-machined crystals – and these can reduce the aforementioned limiting factors, however the process to manufacture these crystals is immature and they not provided any breakthrougs as of today.17,18.

Semiconductor crystals are also applicable for generating good quality THz pulses with high efficiency. This requires high pumping wavelength to eliminate low order multiphoton absorption.19. With high wavelength pumping the phase matching condition is no longer met in a co-linear case, so pulse front tilting must be used, in a similar fashion as with LN. The longer pumping wavelength makes it easier to adopt the new techniques and by that we can get a well scaling THz source that operates with great efficiency22. The efficiency of the THz generation in the crystal can reach 1% (the quantum yield is well over 100% by then), but the generation of the typically needed 1.7-3.0 μm wavelength pump is usually done in a OPA, which has a low efficiency on high wavelength signals.23. A solution would be a laser which has the desired wavelength and does not have to be converted. Using a carbon-dioxide could be a solution since few picosecond pulses became avalible24, which pulses can be further compressed in a CO2 cells to few hundred femtoseconds25.

In this manuscript we will show that CO2 lasers are potentially suitable for pumping semiconductor crystals in order to generate THz. The pulse length is assumed to be in the range of 0.5-2.5 ps; multipass26-28, or hollow core fiber29-31 can be used to further compress the pump pulse.

We also assume GaAs semiconductor crystal, which is an isotropic cubic zinc-blende semiconductor crystal with a class symmetry of 3. It has a direct bandgap of 1.43 eV , a refractive index of about 3.6 in the optical region, a large nonlinear coefficient, at 10.6 m32, and a broad transmission range from 0.97 to 17m32. The damage threshold of 2 GW/cm2 was observed in GaAs at a pulse length of 250 ps33 and wavelength of 10 m. According to the scaling law of Ref.34, the damage threshold in the case of 2.5 ps could be 100 GW/cm2 and higher in the case of shorter pulse duration.

GaAs is used to generate second harmonics of CO235 for a long time, that is why in the numeric calculations we took account of the second harmonic generation.The numeric model also calculates the absorption, dispersion for the pump and the THz signal and the self-phase modulation of the pump.

# **Methods**

The following differential-equation system was solved for determine the THz-generation efficiency and the pulse shape of the THz pulse

The first equation describes the linear absorption of the generated THz pulse and the THz generation by optical rectification of the pump pulse.

The second differential equation describes the cascaded up- and down-conversion – causes the THz pulse –, and the second-harmonic generation. The last differential-equation describes the second-harmonic generation process of the pump pulse.

,where Their model took into account cascading effects caused by up-and down-conversion, pump pulse length fluctuation with propagation distance due to dispersion, and THz absorption in the THz frequency range caused by a complex dielectric function. In contrast to the Ravi et al. 37 model, our model considered the effects of free carriers on the THz field due to the high intensity optical field. The instantaneous refractive indices and absorption of the gallium arsenide and zinc selenide semiconductor crystals at the THz range were calculated from equation (1).

|  |  |
| --- | --- |
|  | (1) |

where is the dielectric permittivity of the semiconductor material in the absence of free carriers, is the permittivity attributed to the free carriers 37, it’s given by equation (2)

|  |  |
| --- | --- |
|  | (2) |

Here, q is the charge of the electron, is the vacuum permittivity, is the electron effective mass, is the THz angular frequency, is the electron scattering time, is the free carrier density-weighted average in the position z of the crystal, which was calculated from equation (3):

|  |  |
| --- | --- |
|  | (3) |

where is the nth photon absorption coefficient, h is the Planck constant, is the pump central frequency and is the temporal intensity of the pump at position z of the crystal. However, we did not consider the multiphoton absorption in our calculations due to the lack of data on photon absorption in the wavelength of interest.

## **Ethics information**

If your protocol describes research with **human participants**, the Methods section must start with a statement confirming that the research complies with all relevant ethical regulations; naming the board and institution that approved the study protocol; and confirming that informed consent will be obtained from all human participants. Information on participant compensation must also be included.

If your manuscript reports research with **non-human animals**, the Methods section starts with a statement confirming that the research complies with all relevant ethical regulations; naming the board and institution that approved the study protocol; and confirming that the ARRIVE guidelines were used to report the research.

If your manuscript reports a **clinical trial**, the Ethics information section also includes the trial registration number from ClinicalTrials.gov or an equivalent approved trials registry.

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## **Pilot data**

You may include pilot data, for example to demonstrate the feasibility of your approach. Your pilot studies and results should be described briefly in the main manuscript and reported in full in Supplementary Information.

Pilot data and custom analyses code should be made available and referred to in the Data Availability statement and Code availability statement. You may also include simulated data, for example to support your power analysis. This should also be made available.

If you report analyses of pilot data using NHST (either in the main text or in Supplementary Information), you must report statistics **in full**: statistic(degrees of freedom) = value, p = value, effect size statistic = value, % Confidence Intervals = values

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## **Design**

Your Methods section must include a description of experimental procedures in sufficient detail to allow another researcher to repeat the methodology exactly, without requiring further information other than that included in the protocol, your Supplementary Information file (if used) and, if applicable, the linked Code and Data (please refer to the **Code Availability** and **Data availability** statements below).

Provide full descriptions of any outcome-neutral criteria and positive controls. These quality checks might include the absence of floor or ceiling effects in data distributions, positive controls, or other quality checks that are orthogonal to the experimental hypotheses.

# You must have a statement on **randomization** in the Methods, if applicable.

# For experimental studies, make it clear whether the design is within-subjects, between-subjects, mixed, or other.

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# You must have a statement indicating whether **blinding** will be used in the Methods, if applicable. If there will be no blinding, this must be clearly stated in the manuscript, as follows: "Data collection and analysis will not be performed blind to the conditions of the experiments.”

If your manuscript reports the results of a **Phase 2 or 3 randomized controlled trial**, you should also attach the CONSORT checklist with your submission.

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### **Sampling plan**

Studies involving Neyman-Pearson inference must include a statistical **power analysis**. Estimated effect sizes should be justified with reference to the existing literature. Since publication bias overinflates published estimates of effect size, power analysis must be based on the **lowest** available or meaningful estimate of the effect size. For frequentist analysis plans, the a priori power must be **0.95 or higher** for all proposed hypothesis tests. In the case of highly uncertain effect sizes, a variable sample size and interim data analysis is permissible but with inspection points stated in advance, appropriate Type I error correction for ‘peeking’ employed, and a final stopping rule for data collection outlined.

Methods involving Bayesian hypothesis testing are encouraged. For studies involving analyses with Bayes factors, the predictions of the theory must be specified so that a Bayes factor can be calculated. Authors should indicate what distribution will be used to represent the predictions of the theory and how its parameters will be specified. For inference by Bayes factors, authors must be able to guarantee data collection until theBayes factor is at least 10 times in favour of the experimental hypothesis over the null hypothesis (or vice versa). Authors with resource limitations are permitted to specify a maximum feasible sample size at which data collection must cease regardless of the Bayes factor; however to be eligible for advance acceptance this number must be sufficiently large that inconclusive results at this sample size would nevertheless be an important message for the field.

Regardless of sampling method, you must list all criteria for **data inclusion** and/or **data exclusion** and how this affects your sampling strategy. This includes a full description of proposed sample characteristics. Procedures for objectively defining exclusion criteria due to technical errors or for any other reasons must be specified, including details of how and under what conditions data would be replaced. These details must be summarized in the mandatory **Design table** (Table 1).

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## **Analysis Plan**

Your proposed analysis pipeline must include all pre-processing steps, and a precise description of all planned analyses (including appropriate correction for multiple comparisons if applicable). Any covariates or regressors must be stated. Where analysis decisions are contingent on the outcome of prior analyses, these contingencies must be specified and adhered to.

Do not include exploratory analyses in the Stage 1 protocol. These should be reported in the Stage 2 manuscript, under the heading **Exploratory Analyses**.

**Should you need to deviate in any way from the description of your work in the Methods after acceptance in principle, you must seek editorial feedback first (before implementing these changes).**

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# **Data availability**

For Registered Reports, **public sharing of data and materials upon acceptance** for publication of the Stage 2 manuscript **is mandatory**. Please include a statement committing to sharing your raw data and materials on acceptance of your Stage 2 manuscript. Please deposit any pilot data that you may have already collected. Pilot data should be made accessible for peer-review, but can be placed under embargo until Stage 2 acceptance.

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# **Code availability**

For Registered Reports, **public sharing of all code upon acceptance** for publication of the Stage 2 manuscript **is mandatory**. Please include a statement committing to sharing all code on acceptance of your Stage 2 manuscript. The Code availability statement must be included separately from the Data availability statement. Please provide a link (e.g. GitHub, osf) to a live version of your code. Code used to simulate data, conduct power analyses, and analyse pilot data should be made accessible in the same location. The code must be made available for peer-review, but can be placed under public embargo until Stage 2 acceptance.

# **Results**

Do **not** include a **Results** section.

# **Discussion**

Do **not** include a **Discussion** section.

# **Acknowledgements**

Please ensure that you acknowledge all funding sources that supported the work reported in your manuscript and provide grant or contribution numbers in an Acknowledgments section after the references. Indicate what role the funder(s) had in the conceptualization, design, data collection, analysis, decision to publish, or preparation of the manuscript. If any of this information could be perceived as a competing interest, ensure that it is also included in your competing interests statement. If the funder(s) have/had no role, please include the following statement: “**The funders have/had no role in study design, data collection and analysis, decision to publish or preparation of the manuscript.**” If no specific funding supported the work, include the following statement: “**The authors received no specific funding for this work.**” Keep other acknowledgements brief and do not include effusive comments.

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# **Author contributions**

We require authors to include an author contributions statement of their individual contributions to the paper -- such as experimental work, project planning, data analysis, etc (see the CRediT taxonomy for relevant contributor roles:<https://casrai.org/credit/>). The statement should be short, and refer to authors by their initials. For details please see the Authorship section of our joint Nature Research Editorial policies at <http://www.nature.com/authors/editorial_policies/authorship.html>

**Competing interests**

We ask authors to declare both financial and non-financial competing interests. For more details, see <https://www.nature.com/srep/journal-policies/editorial-policies#competing>. If you have no financial or non-financial competing interests, please state so: “The authors declare no competing interests.”

**Figures**

You are encouraged to include Figures in the text or at the end of the protocol. Keep in mind that a total of 8 display elements (i.e., combination of Tables and Figures) is permitted in the final, Stage 2, submission. However, to enable typesetting of papers, we advise making the number of display items commensurate with your overall word length (that is, for a shorter paper the number of display items should be lower, for a longer manuscript a higher number may be allowed). Figures/Tables that are not essential should be included in your Supplementary Information file.

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# **Figure Legends**

**Figure 1. Guidelines for the preparation of figure captions.** Figure captions should be concise. Begin with a brief title and then describe what is presented in the figure and detail all relevant statistical information. If you show pilot data, list the N of each plot and report full statistics. Aim not to exceed 350 words per legend.

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# **Table 1. Design Table**

You must include this mandatory **Design table**. The columns are prescribed; the number of rows will depend on the number of research questions you will address in your Registered Report.

* Ensure that there is an **exact** correspondence between each scientific hypothesis and each statistical test. For example, it is not appropriate to write: Condition A will affect performance differently from Condition B. Instead, you must define the performance measure (e.g. Reaction Time) and the predicted direction of the difference. This would translate to, e.g.: Reaction times will be significantly higher in Condition A than Condition B.
* If your analysis strategy will depend on the results (e.g. normal vs. non-normal distribution) then specify the contingencies for making different choices, i.e. IF-THEN statements.
* You cannot interpret lack of evidence for the existence of an effect in NHST (e.g. a p>0.05 in a t-test) as evidence for the absence of an effect. To be able to interpret null results, you must commit to using Bayes Factors or equivalence testing.

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| --- | --- | --- | --- | --- |
| **Question** | **Hypothesis (if applicable)** | **Sampling plan (e.g. power analysis)** | **Analysis Plan** | **Interpretation given to different outcomes** |
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# **Supplementary information**

Please report pilot data in detail here and include any other material that provides background information.

# **References**

1 Schmuttenmaer, C. A. Exploring Dynamics in the Far-Infrared with Terahertz Spectroscopy. *Chemical Reviews* **104**, 1759-1780, doi:10.1021/cr020685g (2004).

2 Locatelli, M. *et al.* Real-time terahertz digital holography with a quantum cascade laser. *Scientific Reports* **5**, 13566, doi:10.1038/srep13566 (2015).

3 Nanni, E. A. *et al.* Terahertz-driven linear electron acceleration. *Nature Communications* **6**, 8486, doi:10.1038/ncomms9486 (2015).

4 Xu, H. *et al.* Cascaded high-gradient terahertz-driven acceleration of relativistic electron beams. *Nature Photonics* **15**, 426-430, doi:10.1038/s41566-021-00779-x (2021).

5 Pálfalvi, L., Fülöp, J. A., Tóth, G. & Hebling, J. Evanescent-wave proton postaccelerator driven by intense THz pulse. *Physical Review Special Topics - Accelerators and Beams* **17**, 031301, doi:10.1103/PhysRevSTAB.17.031301 (2014).

6 Fleischer, S., Zhou, Y., Field, R. W. & Nelson, K. A. Molecular Orientation and Alignment by Intense Single-Cycle THz Pulses. *Physical Review Letters* **107**, 163603, doi:10.1103/PhysRevLett.107.163603 (2011).

7 Salén, P. *et al.* Matter manipulation with extreme terahertz light: Progress in the enabling THz technology. *Physics Reports* **836-837**, 1-74, doi:<https://doi.org/10.1016/j.physrep.2019.09.002> (2019).

8 Hebling, J., Almási, G., Kozma, I. Z. & Kuhl, J. Velocity matching by pulse front tilting for large-area THz-pulse generation. *Opt. Express* **10**, 1161-1166, doi:10.1364/OE.10.001161 (2002).

9 Hebling, J., Yeh, K.-L., Hoffmann, M. C., Bartal, B. & Nelson, K. A. Generation of high-power terahertz pulses by tilted-pulse-front excitation and their application possibilities. *J. Opt. Soc. Am. B* **25**, B6-B19, doi:10.1364/JOSAB.25.0000B6 (2008).

10 Zhang, B. *et al.* 1.4-mJ High Energy Terahertz Radiation from Lithium Niobates. *Laser & Photonics Reviews* **15**, 2000295, doi:<https://doi.org/10.1002/lpor.202000295> (2021).

11 Ravi, K., Huang, W. R., Carbajo, S., Wu, X. & Kärtner, F. Limitations to THz generation by optical rectification using tilted pulse fronts. *Opt. Express* **22**, 20239-20251, doi:10.1364/OE.22.020239 (2014).

12 Wang, L., Kroh, T., Matlis, N. H. & Kärtner, F. Full 3D + 1 modeling of tilted-pulse-front setups for single-cycle terahertz generation. *J. Opt. Soc. Am. B* **37**, 1000-1007, doi:10.1364/JOSAB.379142 (2020).

13 Tóth, G. *et al.* Performance comparison of lithium-niobate-based extremely high-field single-cycle terahertz sources [Invited]. *Chin. Opt. Lett.* **19**, 111902 (2021).

14 Pálfalvi, L. *et al.* Numerical investigation of a scalable setup for efficient terahertz generation using a segmented tilted-pulse-front excitation. *Opt. Express* **25**, 29560-29573, doi:10.1364/OE.25.029560 (2017).

15 Tóth, G. *et al.* Numerical investigation of imaging-free terahertz generation setup using segmented tilted-pulse-front excitation. *Opt. Express* **27**, 7762-7775, doi:10.1364/OE.27.007762 (2019).

16 Tóth, G. *et al.* Single-cycle scalable terahertz pulse source in reflection geometry. *Opt. Express* **27**, 30681-30691, doi:10.1364/OE.27.030681 (2019).

17 Nugraha, P. S. *et al.* Demonstration of a tilted-pulse-front pumped plane-parallel slab terahertz source. *Opt. Lett.* **44**, 1023-1026, doi:10.1364/OL.44.001023 (2019).

18 Krizsán, G. *et al.* in *2021 Conference on Lasers and Electro-Optics Europe and European Quantum Electronics Conference.* cc\_3\_2 (Optica Publishing Group).

19 Polónyi, G. *et al.* High-energy terahertz pulses from semiconductors pumped beyond the three-photon absorption edge. *Opt. Express* **24**, 23872-23882, doi:10.1364/OE.24.023872 (2016).

20 Rice, A. *et al.* Terahertz optical rectification from 〈110〉 zinc‐blende crystals. *Applied Physics Letters* **64**, 1324-1326, doi:10.1063/1.111922 (1994).

21 Blanchard, F. *et al.* Generation of 1.5 μJ single-cycle terahertz pulses by optical rectification from a large aperture ZnTe crystal. *Opt. Express* **15**, 13212-13220, doi:10.1364/OE.15.013212 (2007).

22 Fülöp, J. A. *et al.* Highly efficient scalable monolithic semiconductor terahertz pulse source. *Optica* **3**, 1075-1078, doi:10.1364/OPTICA.3.001075 (2016).

23 Mbithi, N. M. *et al.* Investigation of terahertz pulse generation in semiconductors pumped at long infrared wavelengths. *J. Opt. Soc. Am. B* **39**, 2684-2691, doi:10.1364/JOSAB.469552 (2022).

24 Polyanskiy, M. N., Pogorelsky, I. V., Babzien, M. & Palmer, M. A. Demonstration of a 2 ps, 5 TW peak power, long-wave infrared laser based on chirped-pulse amplification with mixed-isotope CO2 amplifiers. *OSA Continuum* **3**, 459-472, doi:10.1364/OSAC.381467 (2020).

25 Panagiotopoulos, P., Hastings, M. G., Kolesik, M., Tochitsky, S. & Moloney, J. V. Multi-terawatt femtosecond 10 &#x00B5;m laser pulses by self-compression in a CO2 cell. *OSA Continuum* **3**, 3040-3047, doi:10.1364/OSAC.399992 (2020).

26 Hanna, M. *et al.* Nonlinear temporal compression in multipass cells: theory. *J. Opt. Soc. Am. B* **34**, 1340-1347, doi:10.1364/JOSAB.34.001340 (2017).

27 Kaumanns, M. *et al.* Multipass spectral broadening of 18&#x2009;&#x2009;mJ pulses compressible from 1.3&#x2009;&#x2009;ps to 41&#x2009;&#x2009;fs. *Opt. Lett.* **43**, 5877-5880, doi:10.1364/OL.43.005877 (2018).

28 Lavenu, L. *et al.* Nonlinear pulse compression based on a gas-filled multipass cell. *Opt. Lett.* **43**, 2252-2255, doi:10.1364/OL.43.002252 (2018).

29 Krebs, N., Probst, R. A. & Riedle, E. Sub-20 fs pulses shaped directly in the UV by an acousto-optic programmable dispersive filter. *Opt. Express* **18**, 6164-6171, doi:10.1364/OE.18.006164 (2010).

30 Shumakova, V. *et al.* Multi-millijoule few-cycle mid-infrared pulses through nonlinear self-compression in bulk. *Nature Communications* **7**, 12877, doi:10.1038/ncomms12877 (2016).

31 Beetar, J. E., Rivas, F., Gholam-Mirzaei, S., Liu, Y. & Chini, M. Hollow-core fiber compression of a commercial Yb:KGW laser amplifier. *J. Opt. Soc. Am. B* **36**, A33-A37, doi:10.1364/JOSAB.36.000A33 (2019).

32 Weber, M. J. *Handbook of optical materials*. (CRC Press, 2002).

33 Tochitsky, S. Y., Ralph, J. E., Sung, C. & Joshi, C. Generation of megawatt-power terahertz pulses by noncollinear difference-frequency mixing in GaAs. *Journal of Applied Physics* **98**, 026101, doi:10.1063/1.1957123 (2005).

34 Gattass, R. R. & Mazur, E. Femtosecond laser micromachining in transparent materials. *Nature Photonics* **2**, 219-225, doi:10.1038/nphoton.2008.47 (2008).

35 Thompson, D. E., McMullen, J. D. & Anderson, D. B. Second‐harmonic generation in GaAs ’’stack of plates’’ using high‐power CO2 laser radiation. *Applied Physics Letters* **29**, 113-115, doi:10.1063/1.88989 (1976).

36 Ravi, K. *et al.* Theory of terahertz generation by optical rectification using tilted-pulse-fronts. *Opt. Express* **23**, 5253-5276, doi:10.1364/OE.23.005253 (2015).

37 Ravi, K., Huang, W. R., Carbajo, S., Wu, X. J. & Kartner, F. Limitations to THz generation by optical rectification using tilted pulse fronts. *Optics Express* **22**, 20239-20251, doi:10.1364/Oe.22.020239 (2014).