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Jitter-free terahertz pulses from LiNbO₃

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Intense terahertz pulses are indispensable for modern science and technology, but time-critical applications require ultimate stability of the field cycles with respect to a reference clock. Here we report the nonlinear optical generation of terahertz single-cycle fields by femtosecond laser pulses under passive compensation of timing jitter. The converter is based on optical rectification in a LiNbO₃ slab with two silicon prisms for extracting and combining the emitted Cherenkov radiation from both sides into a single beam. In this way, we achieve suppression of timing jitter to <200 as/μm of beam displacement, a factor of >70 better than in conventional non-collinear geometries. © 2021 Optical Society of America under the terms of the [OSA Open Access Publishing Agreement](#)

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Single-cycle terahertz radiation is a valuable tool for understanding complex materials, accelerating and controlling particle beams, characterizing biological specimens, or detecting hidden objects. Particular advantages of laser-generated terahertz pulses are their extremely high peak values of the electric and magnetic fields in combination with their natural lock in time with respect to the generating laser system [1,2]. Consequently, ultrashort terahertz pulses have found widespread applications in molecular alignment [3,4], ultrafast manipulation of spins [5], spectroscopy of quantum dots [6], terahertz control of electrons [7], and time-domain sensing of biomolecules [8]. An additional emerging research area is the all-optical control and diagnostics of electron beams [9,10] for applications in particle acceleration [11,12], ultrafast electron microscopy [13], or nanophotonic electron diffraction [14]. Almost all of the above applications require the highest peak electric fields and average power, but also a precise lock of the terahertz field cycles to a reference clock, usually a femtosecond laser or the short electron pulses in a particle accelerator.

Optical-to-terahertz converters based on optical rectification of femtosecond laser pulses in LiNbO₃ (lithium niobate, LN) are in many cases an optimal solution for high-power and high-field terahertz generation due to LiNbO₃'s superior damage threshold and high optical nonlinearity [15]. However, the group velocity of laser pulses is about two to three times lower than the phase velocity of terahertz radiation. This velocity mismatch must be resolved, for example, by tilted-pulse-front pumping [16–20] or Cherenkov schemes [21–25] in which the terahertz radiation is emitted at an angle to the pump laser beam.

Although extremely high conversion efficiencies can be achieved, a critical problem of any non-collinear geometry is an unavoidable transfer of the beam pointing jitter to the timing of the terahertz field cycles. This limits the time resolution of pump–probe experiments, reduces the sensitivity of field-resolved molecular fingerprinting, and constrains the ability to compress electron pulses to femtosecond or attosecond duration. Although a timing jitter of less than ~5 fs has been reported [2], modern electron beam control [2,9,11,26] or time-domain spectroscopy of molecular responses [8] aims at sub-femtosecond time scales [27], and jitter control of terahertz radiation therefore becomes an emerging topic of research.

In this Letter, we propose and experimentally demonstrate a concept for passive compensation of the timing jitter in Cherenkov-type terahertz generation geometries that is intrinsically stable against pointing drifts of the generating laser beam. The compensation of the timing jitter is achieved by combining two terahertz beams generated in the form of Cherenkov quasi-plane waves by the same laser pulse and emitted in opposite directions. The beams can be combined with external mirrors or, more elegantly, using total internal reflection inside the generation structure [28]. In our experimental configuration, a thick layer of LiNbO₃ is sandwiched between two silicon prisms for output coupling [28] and pumped by intense femtosecond laser pulses with a cylindrical focus [29]. The two Cherenkov radiation fields are collected with the two prisms and internally reflected to produce a collinear beam of two terahertz waves [28]. In the case of pump laser pointing imperfections, the two terahertz waves have opposite values of time delay; therefore, a combination of both into a common focus produces terahertz single-cycle pulses with no remaining jitter in time.

In our experiments, we measure the timing properties of a conventional, non-collinear Cherenkov-type terahertz converter [30] in comparison to the properties of the geometry where we combine two terahertz beams using the double-prism geometry (see Fig. 1). In order to produce high-field pulses and to avoid the possible pump intensity jitter due to imperfect coupling, we avoid the waveguiding approach [28] and use instead a 0.5 mm thick, 20 × 20 mm² LiNbO₃ slab that is optimized for intense pump pulses and high thermal load [30]. The driving laser is an Yb:YAG amplifier system (Pharos, LightConversion) at a central wavelength of 1030 nm, a pulse duration of 265 fs, and a repetition rate of 50 kHz. In the following, we refer to the coordinate system of Fig. 1. The pump beam with a full width at half-maximum (FWHM) of ~2 mm is extended in the *y* direction by a cylindrical 1:3 telescope and then focused by a

Fig. 1. Passive compensation of the terahertz timing jitter. (a) Conventional non-collinear emission scheme with a LiNbO₃ slab (light cyan) and a silicon prism (light blue) for collection of terahertz Cherenkov radiation. A pointing jitter Δx of the pump beam (pink and blue arrows) translates to a timing jitter Δt of the generated terahertz waveforms (pink and blue waveforms). (b) Measured terahertz electric fields for different lateral translations Δx , showing the systematic shift of the emission. The waveforms are vertically displaced for clarity. (c) Measured terahertz delay as a function of Δx (points) and linear fit to the data (solid line). The space–time coupling constant is (14 ± 1) fs/ μ m. (d) Collinear geometry without timing jitter via coherent combination of two Cherenkov waves (pink and blue). (e) Measured terahertz electric fields for different lateral translations Δx . (f) Measured terahertz delay as a function of Δx (points) and linear fit to the data (solid line). The remaining drifts are smaller than 0.2 fs/ μ m.

cylindrical lens ($f = 300$ mm) in the x direction. Figure 1(a) depicts the conventional configuration that has been mostly used to date [22,23,30,31]. A slab of 1% MgO-doped LiNbO₃ with the crystallographic c axis aligned in the y direction is bonded to a high-resistivity silicon prism that is cut at an angle of 40.4° , corresponding to the half-apex angle of the Cherenkov emission after refraction into silicon [22]. The laser pulses create via optical rectification a line-like nonlinear polarization that propagates in the z direction with superluminal velocity and therefore emits terahertz single-cycle pulses in the form of a Cherenkov wedge of two quasi-plane waves in non-collinear direction. The arrival time Δt of the terahertz waveform (blue and pink) therefore depends on the lateral position Δx of the driving pulses with respect to the center of the physical structure (blue and pink arrows). The coupling coefficient is $\frac{\Delta t}{\Delta x} = \frac{n_{\text{LN}}}{c} \cos \theta_{\text{LN}}$, where $\theta_{\text{LN}} \approx 27^\circ$ is the Cherenkov emission angle in LiNbO₃. The high refractive index of LiNbO₃ in the terahertz region, $n_{\text{LN}} \approx 5$ [15], enhances any change of the optical path difference about five-fold as compared to propagation in air. Therefore, a pump-beam pointing jitter of merely 1 μ m already produces a timing jitter of more than 14 fs.

To measure this effect, we record the emitted electric field with electro-optic sampling (EOS) using the pump laser pulses as a gate and a 1 mm thick ZnTe crystal, a quarter-wave plate, a Wollaston prism, balanced photodiodes, and a lock-in amplifier for detection. For our pump parameters, the nonlinearity of ZnTe is high, and the velocity matching is sufficient to detect the expected frequencies of < 1 THz. The terahertz beam is focused by a parabolic mirror ($f = 102$ mm) and overlapped with the optical beam of 60 μ m (FWHM) focal spot size in the ZnTe crystal. Pump beam translation is implemented by displacing a

bending mirror with a micrometer translation stage, taking into account the well-defined additional beam path in air. The back reflection of the pump beam from the LiNbO₃ slab is monitored to rule out angle changes. Figure 1(b) shows the measured terahertz waveforms as a function of a lateral translation Δx of the pump beam. We see a clear shift of the waveform with increasing beam displacement. To quantify this arrival time drift, we cross-correlate the measured terahertz waveforms with the reference waveform at zero lateral translation and extract the peak value of the correlation. Figure 1(c) shows the results. We see a shift of the terahertz transients of (14 ± 1) fs/ μ m, in accordance with the value calculated above (14.8 fs/ μ m). Despite its simplicity and other advantages [22,23,30,31], such a scheme is therefore not suitable for high-resolution time–domain spectroscopy or femtosecond-level electron beam control.

In contrast, Fig. 1(d) depicts the collinear generation scheme, where total internal reflection in two silicon prisms with apex angles of 25° produces a combined terahertz beam that propagates collinearly to the pump laser [28]. We expect that a positive shift by Δx (from blue to pink) causes a delayed terahertz emission of the lower part, but an advanced emission of the upper part. The magnitudes of both effects are the same. A single lens or parabolic mirror that combines both partial beams into a common focus should therefore produce a waveform whose timing is independent of Δx .

The measured waveforms are shown in Fig. 1(e). We see that the shape of the field cycles is very similar to the one before, but the emission timing is now constant. Figure 1(f) shows the results of our cross-correlational analysis. The residual timing drift is less than 0.2 fs/ μ m. This coupling constant is by a factor of > 70 smaller than in the conventional non-collinear scheme of Fig. 1(a). Hence, a residual beam pointing jitter

Fig. 2. Spatial and spectral characterization of the collinear optical-to-terahertz converter. (a), (b) Focus profiles of the terahertz beams from either side of the structure. (c) Focus profile of the combined emission. (d) y -integrated terahertz intensity of the combined beam. The part of the beam with the highest field is indicated in blue. (e) Beam widths $w_{x,y}$ of the combined beam in the x direction (upper panel) and the y direction (lower panel) in dependency on z . The points show data, and the solid line shows a Gaussian-beam fit. (f) Peak intensity of the combined beam as a function of z (dots) in comparison to a Gaussian-beam fit (solid). (g) Spectrum of the generated terahertz pulses, together with the measured single-cycle waveform (inset). (h) Terahertz power and pulse energy in dependency on the pump power and pump pulse energy. At 6 W, the quantum efficiency is $\sim 12\%$. The solid line shows the quadratic dependency at low power levels.

of 1 μm produces a timing jitter of less than 200 as. Jitter-free terahertz radiation generated in the reported collinear Cherenkov geometry is therefore an appropriate tool for timing-sensitive applications, in particular for terahertz pump-probe techniques, electron pulse compression to femtosecond and attosecond durations [2,26], or time-domain spectroscopy of biomolecules at ultimate levels of signal-to-noise [8].

The reported passive suppression of timing fluctuations relies on the coherent temporal and spatial interference of the two terahertz beams that are emitted from either side of the structure. We assess this overlap in our converter by characterizing the terahertz focus with a pyroelectric beam camera (Pyrocam-III, Spiricon Inc.). The residual pump beam that exits the LiNbO₃ slab is deflected to a dump with a 1 mm wide mirror, and stray light is suppressed with a Teflon plate. The terahertz beam is focused by the same parabolic mirror as above. Figures 2(a) and 2(b) show the measured beam profiles of either side of the structure. We see rather wide Gaussian shapes with elliptical spot sizes that are a consequence of the cylindrical geometry of the pump beam [20,29]. When both emissions are combined into a single terahertz beam, we observe the appearance of an interference pattern along the x direction [Fig. 2(c)]. Figure 2(d) shows the intensity integrated along the y axis, where the high-intensity central part (blue) amounts to about 65% of the total energy. By comparing the peak intensities in Figs. 2(a)–2(c), we find that the peak value of the combined beam exceeds the individual intensities by a factor of ~ 4 . From this interferometric field enhancement, we conclude that the beams overlap coherently in space and time, as expected.

To characterize the depth of focus, we move the beam-profiling camera through the focus of the parabolic mirror.

The beam widths along x and y are defined by the standard deviation $\sigma_{x,y}$ of the intensity profile in the x and y directions, respectively. Figure 2(e) shows the beam widths $w_x = 2\sigma_x$ (top panel) and $w_y = 2\sigma_y$ (bottom panel) as a function of the camera position. The Rayleigh lengths are 8 and 20 mm along x and y , respectively. One can see that astigmatism is negligible, thus verifying approximately flat phase fronts at both exit facets of our structure. In most applications, only the part of the terahertz beam with the highest fields is relevant [blue part in Fig. 2(d)] and a practical value for the Rayleigh length can therefore alternatively be determined by recording the z -dependency of the peak intensity of the beam. The corresponding measurement is shown in Fig. 2(f) and yields an effective Rayleigh length of (16 ± 3) mm, in accordance with the values obtained from Fig. 2(e).

The spectrum of our terahertz pulses is plotted in Fig. 2(g). We see a continuous coverage between ~ 0.1 and ~ 1.0 THz without detectable structures or notches. Due to the flat phase fronts of the emitted Cherenkov radiation, we do not expect any spatio-spectral coupling or spatial chirp [32]. Figure 2(h) shows the pump-power dependence of the terahertz power, measured with a pyroelectric detector (Gentec-EO). After the expected quadratic power scaling (solid line), the terahertz power increases linearly and eventually saturates for pumping powers above ~ 7 W. At a pump energy of 120 μJ , the peak electric field is 5 kV/cm, which is of the same order as previously reported with thick LiNbO₃ slabs [30] and can be used for nonlinear terahertz optics [33] at high repetition rates. We obtain a maximum power efficiency of 1.7×10^{-4} at around 6 W pump power, corresponding to a quantum efficiency of 12%. Such values, taking into account the scaling behavior with

pump pulse duration and terahertz photon energy [34], are in accordance with previous experiments with non-collinear converters [28,31,35], suggesting similar mechanisms of saturation, i.e. nonlinear distortion and absorption of the pump pulse in LiNbO₃ [31]. If more pump power is available than provided by our laser system, the saturation threshold can be increased by upscaling the size of the generating structure along the y axis and adapting the cylindrical focus size [20,29,30].

In conclusion, the reported way of generating intense terahertz single-cycle pulses eliminates the timing jitter of the waveform and provides an attosecond-level stability of the arrival time of the field cycles with respect to the timing of the pump pulses. We foresee a wide range of emerging applications, for example, in laser-driven particle acceleration [11,12], coherent control of femtosecond and attosecond electron pulses [2,9–11,26,27], or field-resolved molecular fingerprinting [8].

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Data Availability. Data underlying the results presented in this paper are not publicly available at this time but may be obtained from the authors upon reasonable request.

REFERENCES

1. T. Kampfrath, K. Tanaka, and K. A. Nelson, *Nat. Photonics* **7**, 680 (2013).
2. C. Kealhofer, W. Schneider, D. Ehberger, A. Ryabov, F. Krausz, and P. Baum, *Science* **352**, 429 (2016).
3. S. Fleischer, Y. Zhou, R. W. Field, and K. A. Nelson, *Phys. Rev. Lett.* **107**, 163603 (2011).
4. M. Sajadi, M. Wolf, and T. Kampfrath, *Nat. Commun.* **8**, 14963 (2017).
5. S. Baierl, M. Hohenleutner, T. Kampfrath, A. K. Zvezdin, A. V. Kimel, R. Huber, and R. V. Mikhaylovskiy, *Nat. Photonics* **10**, 715 (2016).
6. B. C. Pein, W. Chang, H. Y. Hwang, J. Scherer, I. Coropceanu, X. Zhao, X. Zhang, V. Bulović, M. Bawendi, and K. A. Nelson, *Nano Lett.* **17**, 5375 (2017).
7. L. Wimmer, G. Herink, D. R. Solli, S. V. Yalunin, K. E. Echternkamp, and C. Ropers, *Nat. Phys.* **10**, 432 (2014).
8. I. Pupeza, M. Huber, M. Trubetskov, W. Schweinberger, S. A. Hussain, C. Hofer, K. Fritsch, M. Poetzlberger, L. Vámos, E. Fill, T. Amotchkina, K. V. Kepesidis, A. Apolonski, N. Karpowicz, V. Pervak, O. Pronin, F. Fleischmann, A. Azzeer, M. Žigman, and F. Krausz, *Nature* **577**, 52 (2020).
9. D. Ehberger, A. Ryabov, and P. Baum, *Phys. Rev. Lett.* **121**, 094801 (2018).
10. L. Zhao, Z. Wang, C. Lu, R. Wang, C. Hu, P. Wang, J. Qi, T. Jiang, S. Liu, Z. Ma, F. Qi, P. Zhu, Y. Cheng, Z. Shi, Y. Shi, W. Song, X. Zhu, J. Shi, Y. Wang, L. Yan, L. Zhu, D. Xiang, and J. Zhang, *Phys. Rev. X* **8**, 021061 (2018).
11. D. Zhang, A. Fallahi, M. Hemmer, X. Wu, M. Fakhari, Y. Hua, H. Cankaya, A.-L. Calendron, L. E. Zapata, N. H. Matlis, and F. X. Kärtner, *Nat. Photonics* **12**, 336 (2018).
12. E. Curry, S. Fabbri, J. Maxson, P. Musumeci, and A. Gover, *Phys. Rev. Lett.* **120**, 094801 (2018).
13. A. Ryabov and P. Baum, *Science* **353**, 374 (2016).
14. K. J. Mohler, D. Ehberger, I. Gronwald, C. Lange, R. Huber, and P. Baum, *Sci. Adv.* **6**, eabb8804 (2020).
15. L. Palfalvi, J. Hebling, J. Kuhl, A. Peter, and K. Polgar, *J. Appl. Phys.* **97**, 123505 (2005).
16. J. Hebling, G. Almási, I. Z. Kozma, and J. Kuhl, *Opt. Express* **10**, 1161 (2002).
17. J. A. Fülöp, L. Palfalvi, G. Almási, and J. Hebling, *Opt. Express* **18**, 12311 (2010).
18. H. Hirori, A. Doi, F. Blanchard, and K. Tanaka, *Appl. Phys. Lett.* **98**, 091106 (2011).
19. J. Hebling, K.-L. Yeh, M. C. Hoffmann, B. Bartal, and K. A. Nelson, *J. Opt. Soc. Am. B* **25**, B6 (2008).
20. A. G. Stepanov, J. Hebling, and J. Kuhl, *Appl. Phys. B* **81**, 23 (2005).
21. M. Theuer, G. Torosyan, C. Rau, R. Beigang, K. Maki, C. Otani, and K. Kawase, *Appl. Phys. Lett.* **88**, 071122 (2006).
22. S. B. Bodrov, M. I. Bakunov, and M. Hangyo, *J. Appl. Phys.* **104**, 093105 (2008).
23. K. Suizu, T. Tsutsui, T. Shibuya, T. Akiba, and K. Kawase, *Opt. Express* **17**, 7102 (2009).
24. S. Fan, H. Takeuchi, T. Ouchi, K. Takeya, and K. Kawase, *Opt. Lett.* **38**, 1654 (2013).
25. K. Takeya, T. Minami, H. Okano, S. R. Tripathi, and K. Kawase, *APL Photonics* **2**, 016102 (2017).
26. A. Ryabov, J. W. Thurner, D. Nabben, M. V. Tsarev, and P. Baum, *Sci. Adv.* **6**, eabb1393 (2020).
27. P. Baum and F. Krausz, *Chem. Phys. Lett.* **683**, 57 (2017).
28. M. I. Bakunov, E. S. Efimenko, S. D. Gorelov, N. A. Abramovsky, and S. B. Bodrov, *Opt. Lett.* **45**, 3533 (2020).
29. S. B. Bodrov, A. N. Stepanov, M. I. Bakunov, B. V. Shishkin, I. E. Ilyakov, and R. A. Akhmedzhanov, *Opt. Express* **17**, 1871 (2009).
30. M. V. Tsarev, D. Ehberger, and P. Baum, *Appl. Phys. B* **122**, 30 (2016).
31. S. B. Bodrov, I. E. Ilyakov, B. V. Shishkin, and M. I. Bakunov, *Opt. Express* **27**, 36059 (2019).
32. Q. Wu, T. D. Hewitt, and X. C. Zhang, *Appl. Phys. Lett.* **69**, 1026 (1996).
33. H. A. Hafez, S. Kovalev, K.-J. Tielrooij, M. Bonn, M. Gensch, and D. Turchinovich, *Adv. Opt. Mater.* **8**, 1900771 (2020).
34. M. I. Bakunov, S. B. Bodrov, A. V. Maslov, and M. Hangyo, *Phys. Rev. B* **76**, 085346 (2007).
35. S. B. Bodrov, I. E. Ilyakov, B. V. Shishkin, and A. N. Stepanov, *Appl. Phys. Lett.* **100**, 201114 (2012).