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Mostafa Shalaby, Carlo Vicario and Christoph P. Hauri



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Extreme nonlinear terahertz electro-optics in diamond for ultrafast pulse switching

Mostafa Shalaby,¹ Carlo Vicario,¹ and Christoph P. Hauri^{1,2,a}

¹SwissFEL, Paul Scherrer Institute, Villigen-PSI, Switzerland

²Ecole Polytechnique Federale de Lausanne, 1015 Lausanne, Switzerland

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Polarization switching of picosecond laser pulses is a fundamental concept in signal processing [C. Chen and G. Liu, *Annu. Rev. Mater. Sci.* **16**, 203 (1986); V. R. Almeida *et al.*, *Nature* **431**, 1081 (2004); and A. A. P. Pohl *et al.*, *Photonics Sens.* **3**, 1 (2013)]. Conventional switching devices rely on the electro-optical Pockels effect and work at radio frequencies. The ensuing gating time of several nanoseconds is a bottleneck for faster switches which is set by the performance of state-of-the-art high-voltage electronics. Here we show that by substituting the electric field of several kV/cm provided by modern electronics by the MV/cm field of a single-cycle THz laser pulse, the electro-optical gating process can be driven orders of magnitude faster, at THz frequencies. In this context, we introduce diamond as an exceptional electro-optical material and demonstrate a pulse gating time as fast as 100 fs using sub-cycle THz-induced Kerr nonlinearity. We show that THz-induced switching in the insulator diamond is fully governed by the THz pulse shape. The presented THz-based electro-optical approach overcomes the bandwidth and switching speed limits of conventional MHz/GHz electronics and establishes the ultrafast electro-optical gating technology for the first time in the THz frequency range. We finally show that the presented THz polarization gating technique is applicable for advanced beam diagnostics. As a first example, we demonstrate tomographic reconstruction of a THz pulse in three dimensions. © 2017 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution (CC BY) license (<http://creativecommons.org/licenses/by/4.0/>). [<http://dx.doi.org/10.1063/1.4978051>]

INTRODUCTION

Controlling and switching light pulses at ever shorter gating time are essential for the realization of next generation signal processing and optical communication systems.^{1–3} One of the key concepts for signal processing is the ultrafast polarization flip of an optical pulse by an electro-optical (EO) modulator. State-of-the-art modulators are driven by an electric gating pulse of several kilovolts per cm which induces a strong phase shift on the optical pulse due to the electric-field dependent Pockels effect. These devices operate at best at GHz frequencies, i.e., on the nanosecond time scale. The limitation in the modulation speed of the gate is primarily given by the modern electronics high-voltage drivers which are presently not able to switch at with the THz frequencies. Therefore THz electronics operating on the femtosecond time scale has remained an unexplored region in the past. Ongoing efforts towards shorter switching gates are challenged by the limited bandwidth of the multi-kilovolt electronics required for driving the EO effect.

The source of the high intensity electric field in EO modulators can also come from propagating electromagnetic waves. However, it comes with several limitations. For example, in the THz range, the most common THz Pockels materials are semiconductors with a relative small bandgap. In such materials, intense THz excitation leads to other unwanted nonlinear effects such as absorption and spectral distortions.⁴ An alternative way to achieve EO modulation is to use the nonlinear Kerr effect

^aAuthor to whom correspondence should be addressed. Electronic mail: christoph.hauri@psi.ch

which exists in all the materials. Many insulators are good Kerr materials with large bandgap and negligible dispersion. In addition, the Kerr effect scales with the electric field square. Thus, it can provide a shorter gate than the electric field-dependent Pockels effect. Yet, the Kerr effect is hard to establish in conventional EO materials as the third-order nonlinearity is substantially weaker and requires higher electric field pulses. In order to understand the challenges of the Kerr gate, it is helpful to write the Kerr-induced phase change $\Delta\varphi^{\text{Kerr}}$ as a function of the EO medium length L , the nonlinear coefficient n^{Kerr} , and the laser pulse intensity $E_{\text{THz}}^2(t)$,

$$\Delta\varphi^{\text{Kerr}} = \Delta n(\lambda_0) \times 2\pi L / \lambda_0, \text{ where } \Delta n(\lambda_0) = c\epsilon_0 n^{\text{Kerr}} E_{\text{THz}}^2(t), \quad (1)$$

where c and ϵ_0 are the vacuum speed of light and permittivity and λ_0 the laser wavelength, respectively. Switching could be achieved in a long optical medium, such as an optical fiber, but this approach is challenged by the need of polarization-maintaining fibers and the occurrence of nonlinear effects due to the intense pulses. This could lead to a significant degradation of the switching contrast due to self-phase modulation or the excitations of (hot) carriers^{5,6} as the process of thermalization takes up to nanoseconds. In the past, conventional ultrafast lasers like Ti:sapphire systems have been explored for Kerr-based polarization switching but the main hurdle has been the required high laser intensity which complicates the polarization rotation with nonlinear absorption and spectral pulse distortions.⁷ The small bandgap of most solid semiconductor Kerr media ($\Delta E \approx 1-1.5$ eV) enables linear and multi-photon laser absorption with a high probability due to the large photon energy ($h\nu_{800 \text{ nm}} \approx 1.55$ eV). Some other media, such as the liquid CS_2 , offer large n^{Kerr} but their use as ultrafast modulators is limited by the slow recovery time (\sim ps),^{8,9} their toxicity, and the liquid nature.

In principle, the use of a thin modulator medium is favorable for minimizing nonlinear pulse distortions, for reducing the linear attenuation of the signal and for down-sizing the EO device technology. However, according to the scaling law, the necessary phase shift of $\Delta\varphi = \pi/2$ (i.e., a polarization rotation of 90°) requires a stronger electric field. Typical EO modulators have an interaction length of several cm in order to match the power level available from conventional power electronics.

In this letter we present a threefold advances in order to overcome the present challenges of the Kerr-based EO modulators. First, we propose the use of an intense single THz electric field cycle to induce an ultrafast Kerr gate on the femtosecond time scale. Second, we propose a thin, transparent insulator diamond window of only 0.5 mm thickness as EO material to avoid multi-photon absorption, delayed electron effects and gate distortions due to hot carriers. Third, we show that the proposed scheme finds application beyond signal processing and introduce the THz-based polarization gating technique as a tool for powerful 3-dimensional laser beam diagnostics for an ultrashort THz pulse.

RESULTS AND DISCUSSION

Our THz-induced pulse gating setup is shown in Figure 1. As a gating field, we use the recently developed λ^3 THz bullets' source at the SwissFEL facility (Paul Scherrer Institute). We used a special scheme of wavefront control and improved THz focusing to reach extremely intense THz electric field of 83 MV/cm with spectral peak ~ 3.5 THz.¹⁰ The source employs optical rectification of a mid-infrared pulse from an optical parametric amplifier pumped by a TW-scale Ti:sapphire laser system.¹¹ The temporal THz field shape of the resulting 50 μJ THz pulse is measured by electro-optical means in a 50 μm thin GaP <110> crystal with a balanced photodiode detection scheme.^{10,12,13} The maximum field strength was calculated from the THz focus size, temporal trace, and pulse energy from a calibrated THz energy meter (Gentec THz12D-3S).¹⁰

To induce the Kerr modulation, the intense terahertz pulse co-propagates through a diamond window collinearly with the 50 fs pulse centered at 800 nm to be switched. The diamond window serving as an EO modulator has a diameter of 4.5 mm and a thickness of 0.5 mm. It was purchased from e6 (UK) as a single crystal. However, after performing the analysis, we found out that it was poly-crystalline. In order to provide an almost equal electric field strength transverse to the switched pulse, the diameter of the THz radiation has been chosen about 5 times larger than the optical probe. The THz pump was vertically polarized and the near IR was polarized at 45° before the sample. We followed two experimental schemes. In the first, we measured the THz induced birefringence on the probe using a combination of a quarter-wave plate and a Wollaston prism in a balanced

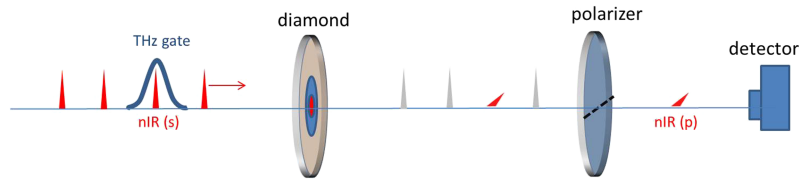


FIG. 1. Ultrafast polarization switching based on the prompt terahertz-induced Kerr effect in diamond. The intensity spot size for the vertically polarized THz gating pulse (blue) and 45°-polarized near IR pulse (red) undergoing Kerr rotation are indicated. A polarizer is used to filter out the polarization switched pulse.

photon-detection scheme. In the second, we detected the polarization switching by placing a polarizer after the diamond window. The polarizer is set to block the near IR pulse in the absence of THz radiation (Fig. 1). Our measurements show that the $\chi^{(3)}$ Kerr nonlinearity and the induced phase retardation $\Delta\varphi^{Kerr}$, though weak in diamond, go along with an intrinsically fast response proportional to $E_{THz}^2(t)$. The few oscillations of the THz electric field (Fig. 2(a)) induce the ultrafast Kerr polarization gate shown in Fig. 2(b). With a measured gating time of 125 fs (full width at half maximum), we demonstrate the fastest polarization switching and largest EO switching contrast ever reported in this frequency range. The THz electric field squared defines entirely the overall shape and the opening time of the Kerr gate as the switching occurs only during the presence of the THz field and vanishes as soon as the THz field has gone (Fig. 2(b)). We observed no signature of delayed response in diamond, which is a big advantage over the commonly used semiconductor modulators. The observed gating time is orders of magnitude shorter than the electronic modulators while nearly full (90%) polarization extinction is achieved. The results show that the THz electric gating field is able to establish a significantly faster electro-optical modulation than what is achieved by the fastest conventional electronics. The main advantage of using a THz gating pulse is that while the THz field is high the THz photon quantum energies of several meV are significantly smaller than the optical ones and consequently do not induce single or multi-photon absorption in the diamond Kerr medium, thanks to the large bandgap of 5.5 eV. Using THz as gate thus overcomes the carrier excitation problem of conventional lasers while a femtosecond gating time is achieved.

The measured phase retardation induced by the $\chi^{(3)}$ nonlinearity shows the expected linear dependence on the THz peak intensity (Fig. 2(c)). For fluence scans, the THz radiation is attenuated using two wire grid polarizers with nearly no spectral dependence in the used spectral range. The maximum phase retardation induced on the 800 nm probe beam was 2.7 rad. In Ref. 14, n^{kerr} was measured to be 3×10^{-6} cm²/W for 1 THz pump and 800 nm probe. This implies a THz peak field of 55 MV/cm (5.7 TW/cm²) in our experiment. This value agrees reasonably with the estimated peak field (51 MV/cm) from our independent calibration¹⁰ using the spot size, THz energy, and time trace as well as a corresponding measurement of air nonlinearity.¹⁵

The polarization switching scheme presented here thus offers three orders of magnitude larger phase retardation than what has been provided by the state-of-the-art THz sources in the past.¹⁴ In this configuration, the transmitted energy of the nIR pulse behind the crossed polarizer is measured to be as high as 90%, demonstrating the efficiency of this pulse picking system. We mention that the THz pulses produced by OR carry an absolute phase which is stable from shot to shot. A gate pulse with a stable absolute phase is essential for the realization of the THz-optical phase modulator to provide the same phase-shift for consecutive shots. Different spectral filters (bandpass, high pass) have been used for the THz pump beam to study the Kerr response on the 800 nm probe beam. In comparison to absorption-based modulation,^{16,17} polarization modulation gives an advantage of on/off contrast because it is easy to achieve extremely high extinction ratio through polarization control.

Tens of MV/cm field strength is required to induce the observed Kerr switching. Even though the Kerr coefficient is weaker than in many other media such as liquid,¹⁸ air,¹⁵ and solids,^{19,20} diamond has three main advantages for the realization of an ultrafast THz Kerr gate. First, diamond is an insulator with a wide bandgap ($E_{gap} = 5.5$ eV) which reduces the probability of electron-hole pair generation across the bandgap upon nonlinear excitation. Second, there is a good velocity matching between the triggering THz gate pulse and optical pulse ($n^{THz} = 2.38$, $n_{phase}^{800\text{ nm}} = 2.4$, $n_{group}^{800\text{ nm}} = 2.44$).²¹

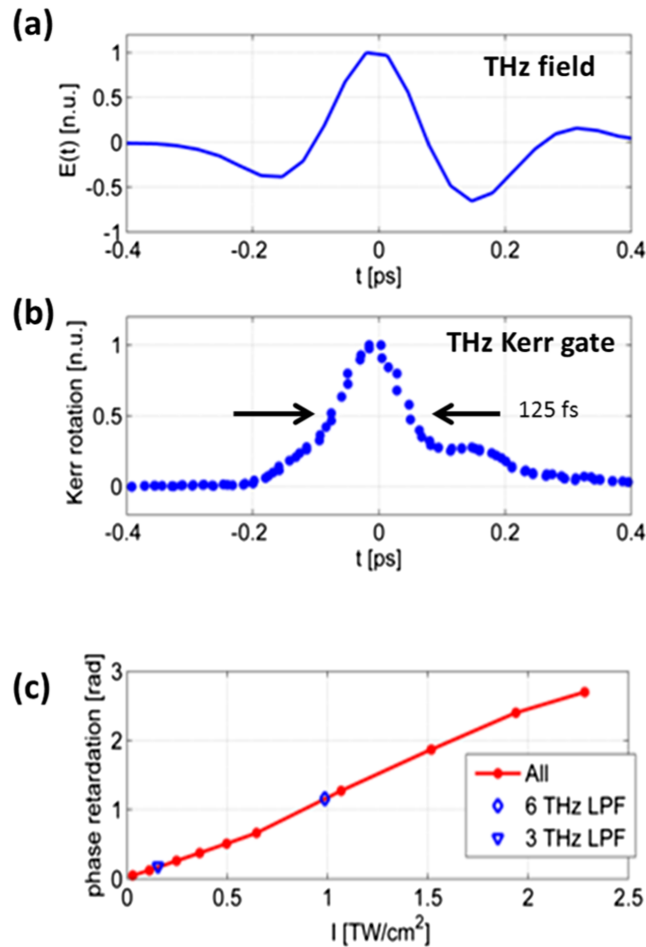


FIG. 2. (a) Temporal trace of the THz electric field oscillation recorded by electro-optical sampling. (b) The THz-induced polarization rotation on the near IR pulse bears the temporal characteristics of the THz pulse in (a). The ultrafast Kerr gate exhibits a 125 fs (FWHM) gating time. (c) THz intensity scan. Dependence of the measured phase retardation induced on the near IR pulse at the THz peak intensity. We measured it in the case where the full THz spectrum is used and when 3 and 6 THz LPF (low pass filters) are placed before the diamond window.

Third, diamond offers both negligible dispersion and high transmission over the THz bandwidth and beyond. The combination of diamond with the intense single-cycle THz pulse is thus excellently suited to serve as a high-field EO gate. Although we used an extremely intense THz source, much weaker sources (such as tilted LiNbO_3 or plasma source) may be used with longer windows benefiting from the good velocity matching and transparency whether in a single or multi-stages. We expect any diamond window to behave in a similar way. Other materials may be used too. However, the commonly used materials such as sapphire or quartz do not fill the above mentioned requirements of velocity matching and broadband response.

We have done further characterization of the diamond-based THz polarization gate. To perform measurement on the spatial homogeneity, we replaced the single-point detector behind the polarizer (Fig. 1) by a 2-dimensional CCD (Dataray, UCT, 1400×1600 pixels, $4.65 \mu\text{m}$ pixel size) to record the intensity profile of the transmitted near IR beam after the polarizer. As before, the near IR pulse focus was kept smaller than the THz spot size to ensure a homogeneous illumination by THz radiation across the probe spot. A series of ultrafast snapshots of the switched near IR beam profile at different delays have been recorded with the CCD (Fig. 3(a)). This gives access to the space-time evolution of the nIR intensity profile. Figure 3 shows the absolute (Fig. 3(b)) and normalized (Fig. 3(c)) intensity profile projected into the x - t plane as a function of the delay. The small tail extending out towards

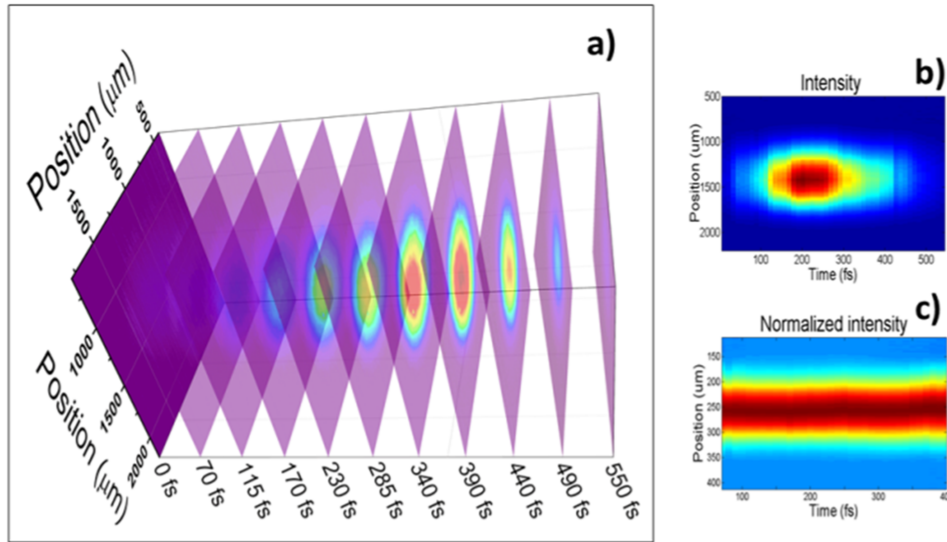


FIG. 3. Characterization of the transverse homogeneity of the THz switch. (a) A series of measurements at different delays between the THz field and probe beam have been performed using a CCD located behind the polarizer. This gives access to the space-time evolution of the nIR intensity profile. In the experiment, the laser pulse is focused tighter than the THz (see Fig. 1) and experiences uniform phase shift, as clearly observed in the measurements. The plot illustrates the gate time convoluted with the near IR pulse duration resulting in approximately 150 fs (FWHM). In (b) and (c) the laser absolute (b) and normalized (c) intensity is projected in the x-t plane as a function of the delay. The constant beam size of the normalized projection for each delay testifies that the THz homogeneously illuminates the nIR probe beam intensity profile at any time.

longer delays in Fig. 3(b) arises from the asymmetry of the THz window (Fig. 2(b)). The normalized nIR intensity plot shows that the FWHM probe beam diameter is constant for all delays. This testifies that the THz pulse homogeneously illuminates the near IR probe beam intensity profile at any time. The near IR pulse experiences uniform phase shift, as clearly observed in the measurements.

Next we show that the technique of THz polarization gating can be used for high resolution spatio-temporal profiling of the THz beam itself. The THz source used here is an example for an advanced, multi-octave-spanning THz source where the beam quality is generally challenged by spatial inhomogeneities.^{10,22,23} Assuming a maximum frequency of 10 THz, the scale of inhomogeneity is expected to be more than 30 μm (due to diffraction limits) which is not smaller than the probe spot size. However, in terms of resolution, the THz imaging technology presently available hardly satisfies the experimental needs for characterizing a tightly focused THz beam¹⁰ and does not give any information about the spatio-temporal evolution of the THz intensity profile. In the following, we show that THz Kerr gating is a powerful tool for space-time resolved mapping of the THz intensity profile along the THz bullet. The experimental configuration (Fig. 4) consists of a single-cycle THz tightly focused to its diffraction-limited spot size in the diamond plate.

For the 3-dimensional imaging of this THz bullet, the near IR beam is transversely expanded in order to overflow the THz focus by almost a factor of 20 (Fig. 4(a)). The polarization gating technique allows for imaging the 3-dimensional THz bullet in time by recording the THz-induced polarization rotation of the ultrashort probe beam on the 2-dimensional CCD sensor. Indeed, the maximum polarizer transmission coincides with near IR probe which underwent polarization rotation of $\Delta\varphi = \pi/2$ induced by the corresponding THz sub-cycle part overlapping with the probe. A sequence of different beam slices recorded along the THz bullet at different times is shown in Figure 3. The reconstructed 3-dimensional bullet unravels a cigar-like shape with a smooth intensity profile in the waist. The tomography shows the bullet to exhibit slight asymmetry at the head and the tail (at 190 fs and 80 fs, respectively). For the reconstruction, the linear dependence of the phase modulation with intensity is modulated with the sine squared transfer function of the polarizer. We note that 3-dimensional reconstruction using a CCD has been previously shown through nonlinear wave

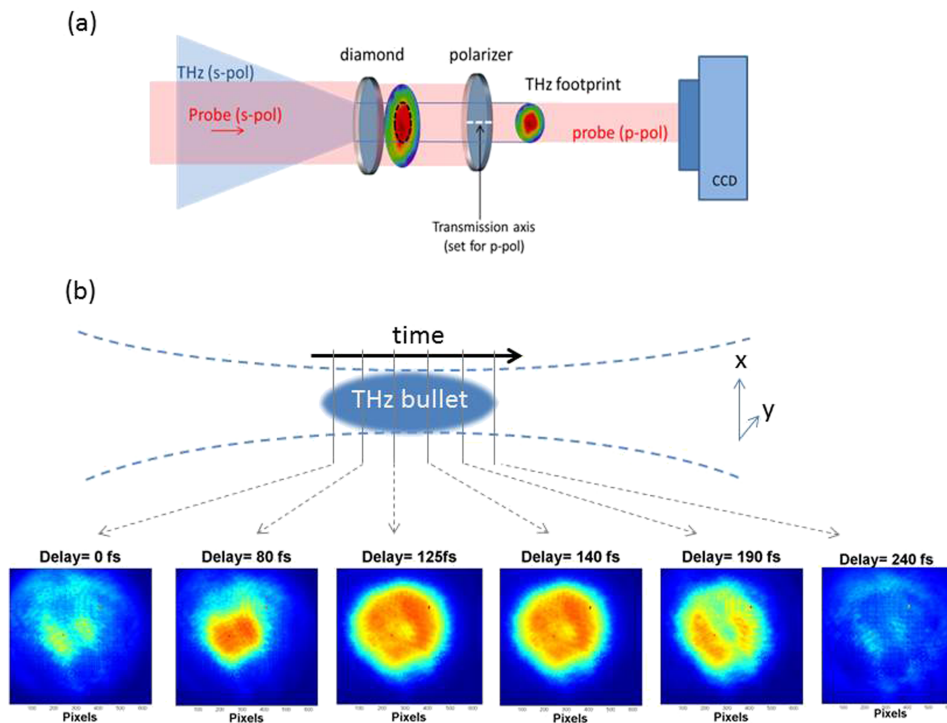


FIG. 4. Three dimensional imaging of the THz light bullet in the focus using the polarization gating technique. The tightly focused THz beam is probed by a collimated nIR beam much larger than the bullet focus. THz intensity slices along the THz bullet are recorded and imaged by the 50 fs probe beam onto a CCD sensor. The series show the 2-dimensional intensity profiles measured at different positions (times) along the longitudinal THz envelope. This allows the reconstruction of the THz bullet in time and space.

mixing.²⁴ However, the striking advantage of our polarization rotation-based technique is its linear response in comparison with nonlinear wave mixing in Ref. 24.

CONCLUSIONS

We have demonstrated an ultrafast THz-driven optical modulator for pulse switching in a thin diamond window. The EO phase modulation originates from the weak Kerr nonlinearity and is driven by the interaction with an ultra-intense 55 MV/cm THz single-cycle pulse. The THz-based Kerr switch exhibits ultrafast gating time of 125 fs and a rise/fall time of ≈ 120 fs. The gate driven at THz frequencies overcomes current electronics bandwidth limitations and undesired nonlinearities typically observed in semiconductor Kerr shutters at high laser intensities. While the presented proof-of-principle experiment has been performed at 800 nm, the exceptional broadband transmission properties of diamond suggest a broadband THz-driven Kerr switch across the spectral range from the deep-UV to the far infrared. In addition to the large optical damage threshold, diamond has exceptional heat conductivity ideal for high power all-optical modulators. We have finally shown a 3-dimensional tomographic imaging of the THz intensity bullet and probe pulse as a first application of THz-induced gating. The presented ultrafast THz-based all-optical switch opens new scientific pathways in signal processing and pulse gating at THz frequencies and opens a route towards advanced spatio-temporal beam metrology in the THz range.

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