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Phase Diversity Electro-optic Sampling: A new approach to single-shot terahertz waveform recording

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THz spectroscopy is an emerging tool for detection of microorganisms and harmful compounds in the food industry, the study of proteins in biomedicine and the development of electron-beam X-ray sources for molecular imaging and lithography. Recording of THz electric field evolution in single-shot is crucially needed in terahertz spectroscopy of irreversible processes in such applications as well as for data communication in the THz portion of the spectrum where there is an abundance of untapped bandwidth. However, achieving sub-picosecond resolution over a long time window has been an open problem for electro-optic sampling – the standard technique for recording terahertz waveforms. We introduce a new conceptual framework for this open problem that is inspired by time-stretch theory. The novel framework unveils a solution to this 20 year-old problem leading to a dramatic enhancement of the achievable temporal resolution. We validate this new technology in two applications. First, we present single shot recordings of long free-propagating terahertz transients with record time resolution. Second, we present recordings of ultra-short relativistic electron bunches at the European X-ray Free Electron Laser. These results show that electric signals may be now recorded with terahertz bandwidth over arbitrarily long windows, thus enabling the realization of single-shot terahertz oscilloscopes and single-shot time-domain spectroscopy systems with an arbitrary time-bandwidth product.

INTRODUCTION

Recording the complete electric field of light in single-shot (including its envelope and carrier) is considered as one of the "holy grails" of terahertz science. This type of detection is largely needed for investigating and mastering novel terahertz sources, as ultrashort quantum cascade lasers [1], ultra-wide bandwidth laser-plasma-based terahertz sources [2], and terahertz Free-Electron-Lasers [3]. Such tools are also crucial for mastering novel

"extreme photonic infrastructures", such as X-ray Free-Electron Lasers [4], and in the very active field of Laser-Plasma Acceleration (LPA) [5], which has the ambition to replace large accelerators facilities by table-top installations. Single-shot terahertz recorders are also needed in applications of terahertz sources. This includes investigations of rapidly evolving reactions in chemistry and biology [3], as well as nonlinear physics (such as nonlinear spectroscopy) which use low repetition rate high power terahertz sources. From the industrial point of view, single-shot terahertz recorders would also introduce a disruptive competition with respect to latest fast scanning terahertz spectrometers, as Asynchronous Optical Sampling (ASOPS) and Electronically Controlled Optical Sampling (ECOPS) [6, 7], in the quest for non-destructive and fast throughput inspection in, e.g., automotive and drug industries.

However, achieving electric field measurements over long durations while keeping femtosecond resolution is a largely open problem. Promising strategies consist in extending the so-called electro-optic sampling technique [8] to the single-shot case. The main principle consists in imprinting the electric field onto a laser pulse, and analyzing the output light. A popular scheme uses temporal encoding onto chirped laser pulses [9, 10], as displayed in Figure 1a.

Although the principle seems simple, retrieving the input electric shape from the data has been the subject of active investigations during the last two decades. Deconvolution algorithms have only led to limited improvements [11, 12], and most research switched to alternate hardware strategies, such as encoding the information onto the transverse or angular direction [13–17], and combining electro-optic sampling with advanced laser pulse characterization techniques [18, 19]. However these complex designs still present important trade-offs between sensitivity, maximum recording duration and temporal resolution.

We show here that electro-optic sampling systems can be transformed into terahertz digitizers, without trade-off on recording length and femtosecond resolution. Key to our approach is a completely new conceptual approach of electro-optic sampling, which has its roots in the theory of the *photonic time-stretch analog-to-digital converter* [20, 21]. As we will see, this new point of view

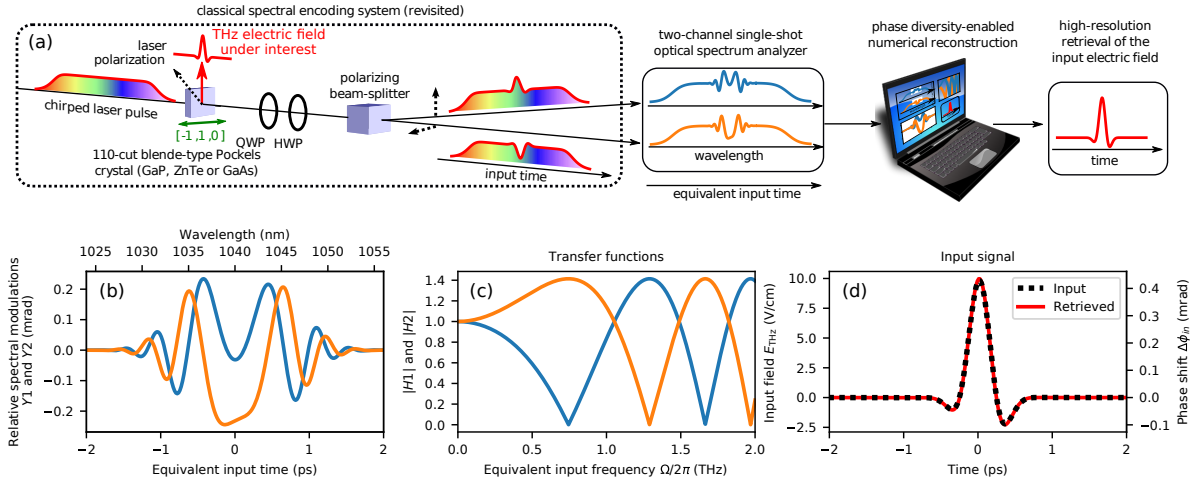


FIG. 1. **(a)** Principle of the DEOS single-shot recording method. The electric field pulse shape under interest is imprinted onto a chirped laser pulse in single-shot, using a Pockels electro-optic crystal, quarter and half-wave plates (QWP and HWP) and a polarizing beam-splitter. The first part of the optical setup (spectral encoding) is classical, except for the polarization directions and crystal axis. Here we show that from the spectra recorded on the two outputs, it is possible to retrieve without ambiguity the input electric field evolution with high resolution and arbitrarily long durations, using a novel point of view, based on *phase-diversity* (see text). **(b,c,d)** Main steps of our reconstruction method (numerical simulation). **(b)** Raw electro-optic sampling (EO) signal corresponding to the optical spectra after background subtraction. **(c)** Transfer function corresponding to the two polarization outputs. **(d)** Comparison of the retrieved (red) and actual (black squares) input signals. The compressed laser pulse is Gaussian with 40 fs FWHM and is chirped to 10 ps. The input terahertz pulse is a Gaussian with duration 500 fs FWHM. The laser polarization, QWP, and HWP axes are oriented at 45, 0 and 22.5 degrees respectively with respect to the vertical axis.

enables to obtain an extremely synthetic view of the process in Fourier domain, and to analyze electro-optic sampling in terms of *frequency domain transfer functions* (Fig. 1c). Using this approach, we show that it is possible to retrieve the input pulse (Fig. 1d) with unprecedented resolution, by using a retrieval algorithm based on the concept of *phase-diversity* [22, 23].

Then we demonstrate the resulting DEOS (Diversity Electro-Optic Sampling) strategy on two different experimental applications. First we demonstrate single-shot recordings in a table-top experiment, with free-propagating terahertz pulses, thus showing the building blocks of a novel “single-shot Time-Domain Spectroscopy” system. Second, we test the DEOS approach in an accelerator context, by probing the near-field of femtosecond electron bunches at megahertz repetition rates, at the European X-ray Free-Electron Laser [4] (EuXFEL).

RESULTS

DEOS single-shot recorder: experimental strategy and novel theoretical framework

Our experimental strategy is displayed in Figure 1a. The DEOS optical setup is extremely close to classical chirped pulse electro-optic sampling systems [9, 10, 24–29], except few – though crucial – modifications. The

electric field under interest $E(t)$ is imprinted in single-shot onto a chirped laser pulse. Then we record the optical spectra in single-shot at the two polarizer outputs. If we assume a linear chirp, the optical frequency ω_{opt} is related to the input time t by:

$$t = -\frac{\omega_{opt}}{C} \quad (1)$$

where C is the chirp rate of the laser pulse.

The optical spectra provide the input data of our retrieval algorithm. The spectra are first preprocessed in a classical way (background subtraction, amplitude scaling and wavelength to time conversion (see Fig. 1 and Methods). These signals $Y_1(t)$ and $Y_2(t)$ will be named the EO (electro-optic) signals in the following. Examples of EO signals are displayed in Figure 1b.

As a key point of the DEOS method, we use a special arrangement of the crystal, waveplates, and polarizations (see the caption of Figure 1), which leads to *different* responses on the two output channels. We will show that this information diversity will enable a numerical retrieval of the input terahertz signal.

A careful analytical analysis first shows that the measured outputs $Y_{1,2}(t)$ are related to the input field $E(t)$ by transfer functions $H_{1,2}$ in Fourier space. For a small input signal (see Supplementary Material, Sections IV-V) we can write:

$$\tilde{Y}_1(\Omega) \approx H_1(\Omega)\Delta\tilde{\phi}_{in}(\Omega) \quad (2)$$

$$\tilde{Y}_2(\Omega) \approx H_2(\Omega)\Delta\tilde{\phi}_{in}(\Omega) \quad (3)$$

where the tilde denotes the Fourier transform, and Ω is the terahertz frequency at the input. $\Delta\phi_{in}(t)$ is the phase shift corresponding to the field-induced birefringence:

$$\Delta\phi_{in}(t) = \beta E(t) \quad (4)$$

$$\text{with } \beta = \frac{\pi d}{\lambda} n_0^3 r_{41} \quad (5)$$

where n_0 is the refractive index at vanishing electric field and d the thickness of the crystal. λ is the laser wavelength in vacuum and $E(t)$ the electric field inside the crystal.

For the specific layout described in Figure 1a, the corresponding transfer functions can be written as (see Supplementary Material, Section V):

$$H_1(\Omega) = \sqrt{2} \cos\left(B\Omega^2 + \frac{\pi}{4}\right) \quad (6)$$

$$H_2(\Omega) = -\sqrt{2} \cos\left(B\Omega^2 - \frac{\pi}{4}\right) \quad (7)$$

where $B = \frac{1}{2C}$.

As a first observation, the transfer functions H_1 and H_2 present nulls at specific frequencies (Fig. 1c). This explains why it is impossible to perform a deconvolution (i.e., to retrieve the input field $E(t)$ from the recorded data) using a single channel $Y_1(t)$ or $Y_2(t)$, for broadband signals. This theoretical result on the existence of zeros is consistent with previous experimental observations [30, 31], and ours, as we will see below. Conceptually, these nulls correspond to the dispersion penalty phenomenon which is observed in the photonic time-stretch digitizer [23], as shown in Supplementary Material, Sections IVC and VB.

Using this point of view, we also remark that it should be possible to retrieve the input signal $\Delta\phi_{in}(t)$ and $E(t)$ by exploiting the information contained in both output signals, as long as the nulls of $H_1(\Omega)$ and $H_2(\Omega)$ are interleaved. In this respect, it is important to note that the relative positions of the zeros depend on the crystal orientations. For instance waveplate and polarization orientations used traditionally ([-110] axis parallel to the terahertz field) would not be acceptable, as this would lead to zeros of $H_{1,2}$ that occur at the same frequencies (see Supplementary Figure S4). In addition, the polarization arrangement considered in this article – for which the zeros are evenly interleaved – is only one of many possible choices which are compatible with the following retrieval algorithm.

Algorithm for high-resolution numerical reconstruction of the input signal

As the zeros of the transfer functions are interleaved, we can retrieve the unknown input pulse from the recorded data. The mathematical problem is well-posed, and even overdetermined, i.e., one has the freedom to choose either H_1 or H_2 for inverting the problem, except at the nulls. Several more advanced methods however exist for taking advantage of both transfer functions in the

reconstruction process. We use here the so-called Maximal Ratio Combining technique (MRC) [22, 23], which is known for optimizing the signal-to-noise ratio (SNR). The input signal can be retrieved from the measurements $Y_{1,2}$ as [23]:

$$\tilde{E}_{in}^{retr}(\Omega) = \beta \Delta\tilde{\phi}_{in}^{retr}(\Omega) \quad (8)$$

$$\text{with } \Delta\tilde{\phi}_{in}^{retr}(\Omega) = \frac{H_1(\Omega)\tilde{Y}_1(\Omega) + H_2(\Omega)\tilde{Y}_2(\Omega)}{H_1^2(\Omega) + H_2^2(\Omega)} \quad (9)$$

where $\tilde{E}_{in}^{retr}(\Omega)$ and $\Delta\tilde{\phi}_{in}^{retr}(\Omega)$ are the retrieved input electric field and crystal phase modulation, expressed in Fourier space. The input signal $E_{in}^{retr}(t)$ (or equivalently $\Delta\phi_{in}^{retr}(t)$) is then obtained by performing an inverse Fourier transform

We tested this reconstruction method numerically for various parameters, and found that it is possible to retrieve the input pulse for arbitrarily long input chirped pulses (i.e., for any duration of the analysis window), down to terahertz pulse durations of the order of the Fourier-limited pulse duration. An example of retrieved input pulse is presented in Figure 1d.

Experimental demonstration: recording free-propagating terahertz pulses with ultra-wide time-bandwidth products

In order to test the DEOS method experimentally, we first constructed the setup displayed in Figure 2a. Terahertz pulses are produced by optical rectification of 800 nm millijoule-range laser pulses in a Zinc Telluride (ZnTe) crystal. These terahertz pulses are then analyzed by an EO sampling setup based on our design.

For test purposes, we also designed the setup so that it is possible to skip or operate the grating stretcher, without changing the setup alignment. We can thus analyze the terahertz pulses using either single-shot EO sampling (i.e., using chirped pulses), or using "traditional" EO sampling by scanning the delay between the femtosecond laser pulses and the terahertz signal. We will thus test our strategy by comparing the DEOS results to the corresponding scanned EO signals.

A typical result is presented in Figure 2e, using a time window of 20 ps. The reconstructed EO signal and the reference EO signal (i.e., obtained using scanned electro-optic sampling) are found to be extremely similar in shape. The reconstruction is even able to reproduce fine details, as the small oscillations between 5 and 20 ps which are due to the water vapor absorption in air (free-induction decay), and multiple reflections on the Silicon plate. For comparison, the EO signal shapes $Y_{1,2}$ before reconstruction (i.e., corresponding to the previous state-of-art) are very different from the input, as can be seen in Figure 2c,d.

These results confirm that the DEOS method now enables investigations of terahertz sources, as well as Time-Domain Spectroscopy to be achieved in single-shot, with

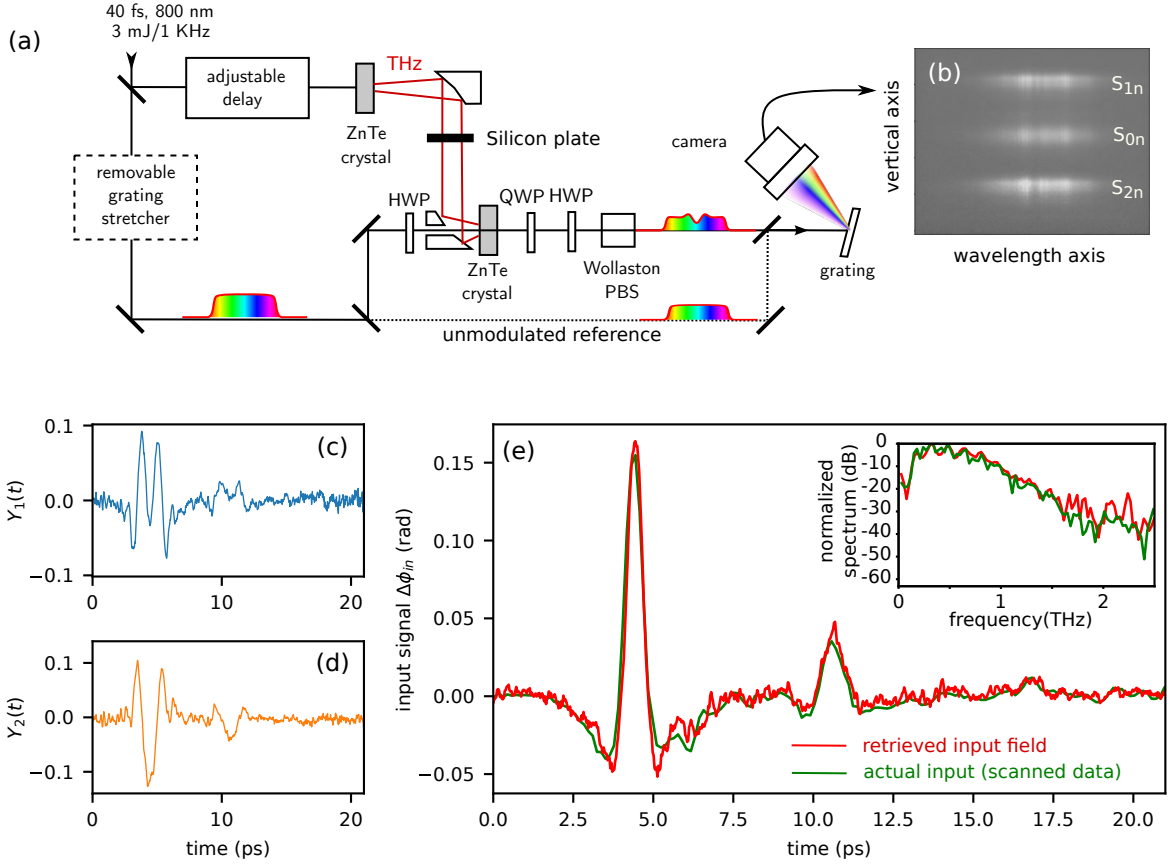


FIG. 2. Single-shot recording of free-propagating terahertz pulses over a window of the order of 20 ps. (a): DEOS experimental setup. ZnTe: 1 mm-thick, 110-cut Zinc Telluride crystal, HWP: Half-wave plates, QWP: Quarter-wave plate, PBS: Wollaston polarizing cube beam-splitter. The beams emerging from the PBS are in the plane perpendicular to the figure. (b) Raw camera image containing the single-shot spectra of the two polarization outputs $S_{1,2n}$, and the unmodulated laser spectrum S_{0n} . (c) and (d): EO signals on two polarizations channels (after background subtraction and normalization, see Methods). (e): Single-shot input signal retrieved from (c) and (d) using the DEOS phase-diversity-based algorithm (red). Green trace: actual input, obtained using scanned electro-optic sampling. Inset: Fourier spectra of the two terahertz signals.

simultaneously high temporal and spectral resolution. More precisely the time resolution and bandwidth limit of the DEOS method appear – as theoretically expected – similar to classical (and non-single-shot) ”scanned” electro-optic sampling.

Note: determination of the reconstruction parameter

As this type of reconstruction requires the knowledge of the transfer functions $H_{1,2}$, it is important to find a practically convenient strategy for determining the parameter B . We remarked that B can be determined in a very simple way, by analyzing the recorded data corresponding to the unknown signal. From the reconstructed signal $\Delta\phi_{in}^{retr}(t)$, we can simulate EO sampling signals

$$\tilde{Y}_1^{retr}(\Omega) \text{ and } \tilde{Y}_2^{retr}(\Omega):$$

$$\tilde{Y}_1^{retr}(\Omega) = H_1(\Omega)\Delta\tilde{\phi}_{in}^{retr}(\Omega) \quad (10)$$

$$\tilde{Y}_2^{retr}(\Omega) = H_2(\Omega)\Delta\tilde{\phi}_{in}^{retr}(\Omega) \quad (11)$$

where $H_{1,2}$, $\tilde{Y}_{1,2}^{retr}$ and $\Delta\tilde{\phi}_{in}^{retr}(\Omega)$ depend on B .

Then we can perform a least-square fit of \tilde{Y}_1^{retr} and \tilde{Y}_2^{retr} on \tilde{Y}_1 and \tilde{Y}_2 , using B as a free parameter. Here, we perform a classical least-square fit using the following definition for the reconstruction error ϵ :

$$\epsilon^2 = \int_{-\infty}^{+\infty} d\Omega \left(\left| \tilde{Y}_1 - \tilde{Y}_1^{retr} \right|^2 + \left| \tilde{Y}_2 - \tilde{Y}_2^{retr} \right|^2 \right) \quad (12)$$

Besides its use for checking the fit quality, it is interesting to examine the reconstructed $\tilde{Y}_{1,2}^{retr}$ and raw $\tilde{Y}_{1,2}$ EO signals (Figure 3). Those curves clearly display the nulls that stem from the zeros of the transfer function $H_{1,2}$ expressed in Eqns. (6,7).

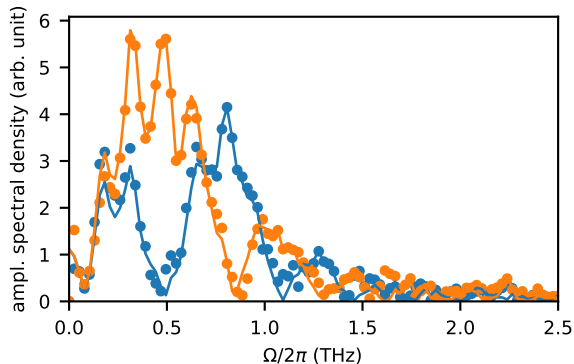


FIG. 3. Fit providing the reconstruction parameter B from a single-shot recording. Dots: Fourier spectra $|\tilde{Y}_{1,2}(\Omega)|$ of experimental data before reconstruction. Lines: spectra $|\tilde{Y}_{1,2}^{retr}(\Omega)|$ computed from the retrieved input. $\tilde{Y}_{1,2}^{retr}(\Omega)$ are fitted on $\tilde{Y}_{1,2}(\Omega)$ using B as a free parameter. Note the presence of interleaved zeros, which play a key role in the retrieval strategy. Same data and color codes as for Figure 2.

Remarkably, this result shows that the determination of the fit parameter B does not require specific experiments to be performed. The fit can be made using any unknown input signal – including the signal to be analyzed – provided its bandwidth is sufficient.

Application to near-field measurements: relativistic electron bunch shapes at the European X-ray free-electron laser

The high resolution retrieval strategy based on phase diversity is expected to find immediate applications in single-pass Free-Electron Lasers (FELs), such as EuXFEL [4]. FELs are based on self-amplification of stimulated radiation driven by ultra-short relativistic electron bunches with high peak current propagating in a periodic magnetic field (undulators). They provide femtosecond photon pulses with energies in the millijoule range, and wavelengths ranging from the ultraviolet to hard X-rays depending on the facilities. The emitted photon pulses depend directly on the properties of the electron bunches. Hence the diagnostics of their longitudinal shape has been the subject of an intense research the last years [24, 32–35]. However the resolution limit of Eq. (14) strongly hampered the application range of electro-optic sampling. The situation is also complicated by the recent trend to megahertz-rate X-ray FELs based on superconducting technology such as FLASH [36], EuXFEL [4], and the LCLS-II [37] and SHINE [38] projects.

In order to test our DEOS reconstruction strategy in this context, we realized a proof-of-principle phase-diversity setup at the EuXFEL (Figure 4a), which is the first hard X-ray FEL operating at megahertz repetition rate [4] (see also Refs. [39, 40] for examples of applica-

tions). In the present case, electron bunches are generated at 1.3 MHz rate in 600 μ s long bursts, every 100 ms (Figure 4b).

The setup, based on the single EO channel detection [35], is depicted schematically in Figure 5. The EO setup is located just after the second bunch compressor, where the electron bunches have a beam energy of 700 MeV and a typical RMS duration in the 200 fs range at a charge of 250 pC. See Methods for details.

Results are displayed in Figure 4c,d. Figure 4c depicts the raw EO signals (after background subtraction and normalization), and Figure 4d represents the reconstructed electric field. We observe a main peak, for which the duration of 218 fs (RMS) is in very good agreement with the design value of the electron bunch duration at this location and considerably shorter than measured with conventional EOSD at the same setup [35]. The smaller negative peak can be attributed to a wakefield following the electron bunch, but the precise interpretation will be subject of further investigations. Typical examples are displayed in Figures 4e,f. This measurement system can now be used to perform bunch-by-bunch high resolution measurements of the bunch shape which is crucial information for the control of the bunch compression process.

As previous electro-optic sampling systems, DEOS also simultaneously measures the arrival time of the electron bunch, which is also a crucial parameter for users of the generated X-rays. The resolution of DEOS for this measurement is expected to be similar to standard EO sampling. In the present case, the arrival time jitter (Fig. 4g) is measured to be 58 fs over a bunch train, which is much lower than the bunch duration (218 fs RMS), and consistent with Ref. [35].

From the hardware point of view, it is important to note that only few key modifications of our initial EO system [35] were needed: ensuring a proper (non-standard) orientation of the GaP crystal in the vacuum chamber, and simultaneous detection of the two EO output channels. We thus think that the implementation of a double EO output channel readout will permit to relatively easily adopt the phase diversity retrieval method in existing or planned EO diagnostics at FELs or other accelerators.

DISCUSSION

It is important to note that DEOS is fundamentally different from previous techniques aiming at retrieving the input field using numerical analysis of the EO data (deconvolution [12], and holography-inspired reconstructions [11]). The presence of zeros in the transfer functions limits the success of reconstruction methods that do not use the phase diversity technique to relatively narrow-band signals, or short analysis windows, if there is no a priori information on properties of the solution. More precisely, deconvolutions or reconstructions from a single channel is ill-posed when the signal spectrum Ω_{max} ex-

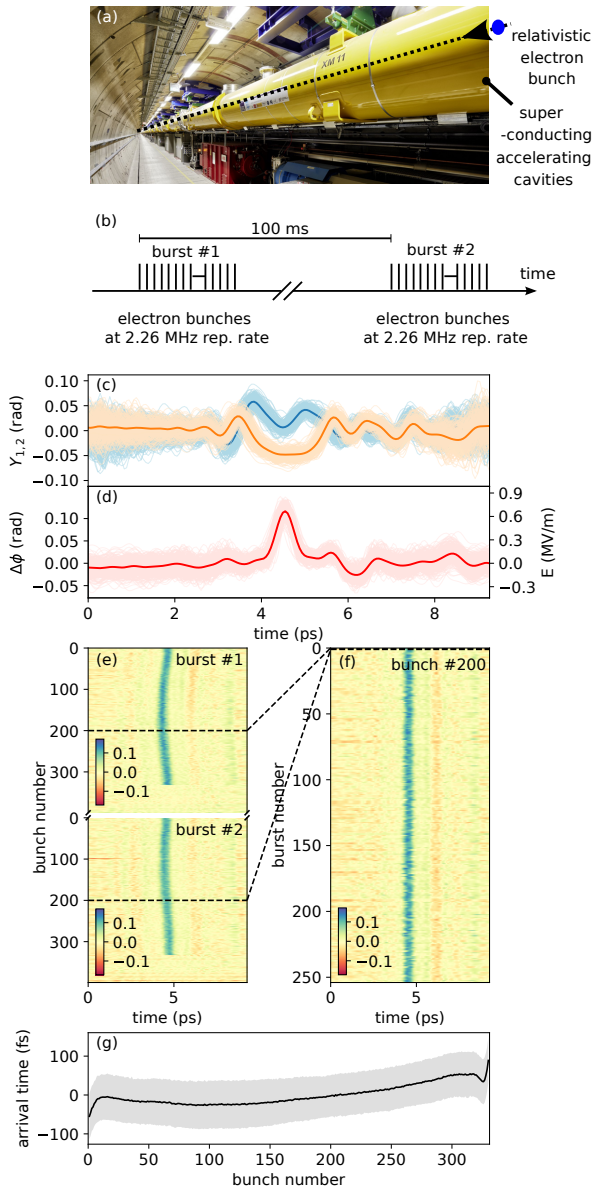


FIG. 4. Electron bunch shapes recorded at the European X-ray Free-Electron Laser (EuXFEL). **(a)** Picture from inside the 3km-long accelerator tunnel. The electro-optic sampling setup (see Fig. 5) is placed just upstream of the picture, after the first bunch compressor. **(b)** timing of the electron bunches in the conditions of the experiment. **(c)** Electro-optic signals $Y_{1,2}$ of a single bunch before reconstruction. **(d)** Reconstructed electric field. Shaded areas: superposition of single-shot curves, color curves: average over 255 bursts. **(e)** Electro-optic signal of two bursts (i.e., 800 electron bunches in total). **(f)** Shape of one bunch (with bunch number 200 within the burst) versus burst number. **(g)** Arrival time versus bunch number. Shaded areas: RMS arrival time fluctuations, color curve: average over 255 bursts. The EO data are low-pass filtered to 2.5 THz.

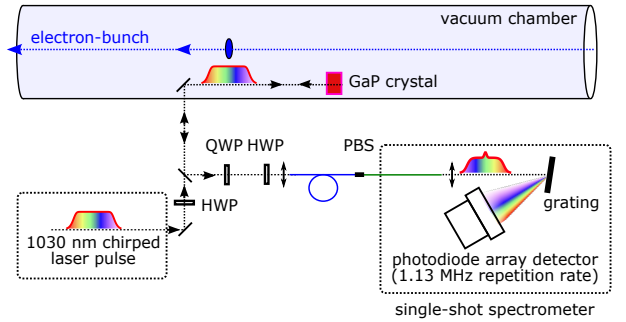


FIG. 5. Near-field electro-optic sampling setup realized at EuXFEL. HWP: half wave plate, QWP, quarter wave plate, PBS, fiber-based polarizing beam-splitter. Blue lines indicate polarization-maintaining fibers (PM980) and green lines indicate single-mode fibers (HI1060). The probe laser is reflected on a Gallium Phosphide (GaP) crystal back side. The spectrum readout is performed using a KALYPSO linear array camera operating at 1.13 MHz line rate [41, 42]. Details of laser transport and KALYPSO focusing optics are not shown – see Ref. [35] for further details.

ceeds the frequency Ω_0 of the first null: $\Omega_0 = \sqrt{C\pi/2}$ or $\sqrt{3C\pi/2}$ (depending on the channel used) for the Figure 1 setup. The limit is $\Omega_0 = \sqrt{C\pi}$ for the case of the more classical situation for which the [-110] axis is parallel to the terahertz field (see Section IV of Supplementary Material for the corresponding transfer functions).

In contrast, using phase diversity-based reconstruction allows in principle arbitrarily long windows to be recorded, while preserving the same resolution. Without taking into account crystal limitations, the ultimate resolution of DEOS is expected to be limited by the compressed width of the probe laser (as observed in simulations)

$$\tau_R \approx \tau_L \quad (13)$$

whereas previous limitations of chirped pulse electro-optic sampling were:

$$\tau_R \approx \sqrt{\tau_L \times \tau_w} \quad (14)$$

where τ_L is the compressed duration of the probe laser pulse. τ_w is the duration of the chirped pulse, which defines the temporal window of the analysis. In this case, τ_R corresponds to the shortest time scale that can be recorded without deformations.

The resolution of the DEOS scheme is theoretically expected to be similar to the classical "scanned" EO sampling (or Time-Domain-Spectrometers), but of course our technique is single-shot. In other words, the next ultimate barriers to be lifted should be the Pockels crystal limitations (due to the unavoidable phonon absorption lines (occurring at 5.3, 8.0 and 11 THz for ZnTe, GaAs and GaP [34]), and the laser pulse duration.

The maximum recording time window of DEOS is limited technically (but not fundamentally) by the spatial

resolution of the camera used for the single-shot spectrometer. Hence we believe that next work should focus on alternate spectrum recording strategies, in particular Time Stretch Dispersive Fourier Transform (TS-DFT) [21], which may be advantageous from this point of view. This would turn the present recording system to a photonic time-stretch analog-to-digital converter [20].

It is also important to note that the MRC algorithm considered here is only one possibility, and that other more complex ones should also be considered for DEOS. In particular, the MRC algorithm relies on transfer functions and is thus valid only for small phase modulations in the crystal. In conditions of high electric fields, the formal similarity with the photonic time-stretch ADC will enable back-propagation methods to be efficient [43, 44].

CONCLUSION

In conclusion, we present a novel conceptual framework for chirped pulse electro-optic sampling, that solves the "temporal resolution problem" open about 20 years ago for terahertz recorders. Technically, the key lies in the derivation of the Fourier-domain transfer function which – in turn – allows well-posed reconstruction algorithms to be possible. In practice, the resulting DEOS strategy opens the way to terahertz digitizers whose recording length and resolutions is only limited by the probe crystal speed or the laser uncompressed pulse duration. On the short term, one foreseen application of DEOS concerns a revisit of high repetition-rate Time-Domain Spectroscopy (TDS), up to megahertz rates, and consider monitoring irreversible physical and chemical processes. Another foreseen application is totally opposite, and concerns the applications of very low repetition rate terahertz sources, for which scanning strategies are impractical. This concerns nonlinear TDS using accelerator-based [3, 45, 46], as well as table-top-based high power terahertz sources [47]. Future work will include a systematic experimental study of the new limits set by the method in terms of ultimate resolution, time-window and repetition rate. In this respect, an important direction will consist in combining the present DEOS terahertz recording method with photonic time-stretch [20, 48, 49], as this should theoretically allow repetition rate to reach the hundred of megahertz range.

MATERIALS AND METHODS

Theoretical details

Proofs are given in the Supplementary Material, and we recall only main results here. The Pockels-induced phase-shift $\Delta\phi_{in}(t)$ is related to the terahertz electric field by $\Delta\phi_{in}(t) = \beta E(t)$ [see Eqns. (6,7)]. The factor β depends on the relative orientations of the crystal axes and on the polarizations of the laser and terahertz field.

For the phase-diversity scheme of Figure 1 and the setups considered in the main text:

$$\Delta\phi_{in}(t) = \beta E(t) \quad (15)$$

$$\text{with } \beta = \frac{\pi d}{\lambda} n_0^3 r_{41} \quad (16)$$

where n_0 is the refractive index at vanishing electric field and d the thickness of the crystal. λ is the laser wavelength in vacuum and $E(t)$ the electric field inside the crystal. Note that the value of β is two times smaller than for the usual layout used in classical balanced detection (see supplementary material).

Experimental recording system for the table-top experiment (Figure 2)

The laser pulses are delivered by an amplified Sapphire-Titanium laser (Coherent Astrella) with 7 mJ output, from which 3 mJ are extracted for this experiment. The emission and detection ZnTe crystals are 110-cut, with 1 mm thickness. The stretcher is a classical Treacy compressor. The Silicon filter (280 μm thick, at normal incidence) is destined to rejected the 800 nm laser light. The imaging spectrometer is composed of a reflection grating, a low-cost 1280x1024 pixels monochrome CMOS camera (UI 3240 ML NIR from IDS GmbH) equipped with a 60 mm objective (Nikkor 60 mm F2.8G ED). We also place a cylindrical lens with 100 mm focal length just before the 60 mm lens, in order to spread vertically the optical power onto the CMOS camera. The three spots are vertically binned at analysis stage, thus increasing the equivalent full-well capacity (and the signal-to-noise ratio).

At each shot n , the recorded image (see Figure 2b) provides three raw data: the spectra on the two polarization channels $S_{1n}(\lambda)$ and $S_{2n}(\lambda)$ (containing the information on the terahertz field), and the spectrum of the unmodulated laser $S_{0n}(\lambda)$. The camera is triggered by the laser, and acquires 10 images per second. For this test we did not try to achieve 1 KHz repetition rate (i.e., the repetition rate of the laser), although this type of upgrade would be straightforward using a state-of-art commercial CMOS or CCD camera (as, e.g., in Ref. [50]).

Data analysis in the table-top experiment

Main data consist of single-shot camera images taken in presence of the electric field. This image contains the three spectra $S_{1n}(\lambda)$, $S_{2n}(\lambda)$ and $S_{0n}(\lambda)$. In addition we also record beforehand the same data in absence of electric field, which we will write S_1^{ref} and S_2^{ref} and S_0^{ref} .

We obtain the EO sampling signals Y_{1n} and Y_{2n} at each shot n (displayed in Fig. 1b) in the following way:

$$Y_{1n} = \frac{S_{1n}}{\sigma_1 S_{0n}} - 1 \quad (17)$$

$$Y_{2n} = \frac{S_{2n}}{\sigma_2 S_{0n}} - 1 \quad (18)$$

where $\sigma_{1,2} = \frac{S_{1,2}^{ref}}{S_0^{ref}}$.

This preprocessing strategy also allows us to suppress the effect of shot-to-shot fluctuations of the laser spectrum shape. In other words, $Y_{1,2n}$ do not depend on the shape of S_{0n} . This can be shown easily, if we assume a fixed relation between the three spectra: $S_{0,1,2n}(\lambda) = f_{0,1,2}(\lambda)S_{Ln}(\lambda)$, with $S_{Ln}(\lambda)$ the laser spectrum at shot n , and assume that $f_{0,1,2}(\lambda)$ are functions that do not change with time (i.e., they only are determined by the optics transmissions, and adjustment). This preprocessing should theoretically allow the measurements to reach the shot-noise limit.

The obtained $Y_{1,2n}$ are then used in the MRC retrieval algorithm. We first apply a window in order to select the part of $Y_{1,2n}(t)$ for which the laser spectrum intensity is well above the detection noise (in practice we select the part where the signal is larger than 10% of the maximum). Then:

- We apply a Fast Fourier Transform (FFT) to the windowed $Y_{1,2n}(t)$ data.
- Apply the MRC formula displayed in Eq. (9).
- Finally apply and inverse FFT for obtaining the reconstructed data $\Delta\phi_{in}^{retr}(t)$ and $E_{in}^{retr}(t)$.

Experimental setup at EuXFEL (Figure 5)

The probe pulses are delivered by an amplified Ytterbium fiber laser operating at 1030 nm. The EO effect is achieved in a Gallium Phosphide (GaP) crystal, which is placed inside the vacuum chamber of the accelerator, near the electron beam [35]. The current setup permits to detect only one EO channel output at a time. The spectra are recorded in single-shot, using a grating spectrometer based on the KALYPSO fast linear array detector [41, 42] operated at a line rate of 1.13 MHz. A first series of electron bunch trains for one polarization is recorded and then the last half waveplate before the polarizing beam splitter is rotated by $\pi/8$ to record the complementary polarization. Hence, the two EO channel outputs can be used to reconstruct the individual electron bunch shapes within the burst using the phase-diversity technique. Note that a straightforward upgrade is also planned, with the aim to achieve simultaneous recording of both polarisations.

Data analysis in the EuXFEL experiment

With each burst, we record single-shot spectra on one of the two polarization directions $S_{1n}(\lambda)$, $S_{2n}(\lambda)$ and additionally unmodulated spectra with laser and no electron bunch $S_{1,2}^{no\ bunch}$, and the spectra without laser $S_{1,2}^{dark}$.

However as we do not record the unmodulated laser spectra $S_{0n}(\lambda)$ of the same laser pulses in the XFEL experiment, we cannot compensate for the shot-to-shot fluctuations of the laser faster than about 2kHz. The data analysis is then similar to the 800 nm experiment case. We thus define the EO signals (before applying the MRC reconstruction) as:

$$Y_{1n} = \frac{S_{1n} - S_1^{dark}}{S_{1}^{no\ bunch} - S_1^{dark}} - 1 \quad (19)$$

$$Y_{2n} = \frac{S_{2n} - S_2^{dark}}{S_{2}^{no\ bunch} - S_2^{dark}} - 1 \quad (20)$$

Then we apply the MRC algorithm in exactly the same way than for the previously described table-top (800 nm) experiment.

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CONFLICT OF INTERESTS

The authors declare that they have no conflict of interest

CONTRIBUTIONS

The PhLAM team realized the theoretical and numerical investigations: establishment of transfer function versus crystal arrangement, analysis algorithm based on MRC, and simulation codes. These investigations have been based on the knowledge established by UCLA (BJ) on transfer functions, phase diversity and MRC, in the framework of photonic time-stretch. Theoretical and numerical calculations have been performed by SB and ER. Table-top experiments have been designed and realized and operated by CS, ER, CE and SB. The EuXFEL electro-optic sampling system has been designed and realized by BS and CG. The setup modifications for phase-diversity studies at EuXFEL have been designed by the PhLAM and DESY teams, and realized by BS. Data

analysis has been performed by ER and SB (table-top experiments), and ER, BS, CS, and SB (FEL experiments). All authors participated to the manuscript redaction.

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SUPPLEMENTARY INFORMATION

accompanies the manuscript on the Light: Sciences and Applications website (<http://nature.com/lisa>).

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