UNAMBIGUOUS ELECTROMAGNETIC PULSE RETRIEVAL THROUGH FREQUENCY MIXING INTERFERENCE IN FREQUENCY RESOLVED OPTICAL GATING

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Abstract

We demonstrate a method for full and unambiguous temporal characterization of few-cycle electromagnetic pulses, including retrieval of the carrier envelope phase (CEP), in which the interference between non-linear frequency mixing components is spectrally resolved using Frequency Resolved Optical Gating (FROG). We term this process Real-Domain FROG (ReD-FROG) and demonstrate its capabilities through the complete measurement of the temporal profile of a single-cycle THz pulse. When applied at THz frequencies ReD-FROG overcomes the bandwidth limitations relating probe and test pulses in Electro-Optic (EO) sampling. The approach can however be extended generally to any frequency range and we provide a conceptual demonstration of the CEP retrieval of few-cycle optical field.

INTRODUCTION

Few-cycle electromagnetic pulses offer a means to both control and probe physical processes active on femtosecond timescales. State-of-the-art accelerator facilities incorporate or produce such pulses at multiple levels of operation, including: the output of 4$^{\text{th}}$ generation light sources, such as the CLARA free electron laser test facility [1]; the production of coherent transition radiation from a relativistic electron bunch [2]; and the intrinsic coulomb field of a relativistic electron bunch. In the latter two examples the radiation is directly related to the longitudinal properties of the bunch and thus can be utilized for diagnostic purposes [3] . The characterization of such ultrashort radiation is therefore often a crucial element of accelerator operation [4].

All information relating to the temporal properties of an ultrashort field can be derived from knowledge of the pulse spectrum and spectral phase: $\tilde{E}(\omega) = \tilde{A}(\omega)e^{i\tilde{\phi}}(\omega)$. The spectral phase can be mathematically described by the series expansion:

$$\tilde{\phi}(\omega) = \phi^{CE} + \tilde{\phi}^{(1)}(\omega) + \frac{1}{2}\tilde{\phi}^{(2)}(\omega) + \ldots \qquad (1)$$

The zero-order term of Eq. (1) (ϕ^{CE}) is referred to as the carrier envelope phase (CEP). For pulses in which the electric field envelope consists of many cycles this term can be identified as a time-shift of the carrier within the envelope. In few or single-cycle pulses however the distinction between the carrier and envelope components is no longer appropriate and the CEP plays a fundamental role in determining the temporal profile.

While many methods are available to measure relative changes in CEP (for example, f-2f interferometery [5]), schemes to measure the absolute value of CEP are more specific, being limited by constraints in frequency and often involve complex experimental arrangements [6]. Obtaining the full temporal field profile typically requires a separate system dedicated to the measurement of the pulse envelope and higher-order spectral phase components. Towards this latter case Frequency Resolved Optical Gating (FROG) has found common application due to its robustness and ease of experimental implementation [7]. In all forms FROG incorporates the measurement of an intensity spectrogram which is derived from a non-linear interaction between multiple pulses. As a measurement of intensity however it has been widely held that FROG techniques are incapable of determining the CEP.

In beam diagnostic applications, Electro-Optic (EO) sampling has found extensive use as a means of characterizing the complete temporal profile – including the CEP – of few and single-cycle THz pulses produced by relativistic electron sources [3]. EO sampling requires that the THz field is interrogated by a δ-like optical probe field and thus is subject to bandwidth limitations relating the probe and test fields. EO sampling has been utilized in recent work [8,9] to characterize the CEP of far-infrared pulses; as ϕ^{CE} presents as a spectral invariant, knowledge of the CEP at THz frequencies within the pulse (as can be obtained through EO sampling) is sufficient for reconstruction of the complete temporal profile following a separate FROG measurement.

In this work we demonstrate that unambiguous retrieval of an ultrashort pulse including the CEP can proceed directly from a single FROG measurement in which the interference between harmonic components is resolved. We develop a theoretical framework for this method termed Real-Domain FROG (ReD-FROG) [10], describing how it conceptually relates to EO sampling. A proof-of-principle experiment is presented in which the CEP of a single-cycle THz pulse is accurately retrieved. We finally demonstrate the conceptual application of self-referenced ReD-FROG to the recovery of an octave-spanning optical field.

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THEORY

We derive the framework of ReD-FROG for a second-order interaction mediated by the non-linear susceptibility $\chi^{(2)}$; the following approach can however be extended to higher-order non-linear interactions. The non-linear output of a second-order interaction combining fields $\tilde{E}_1(\omega)$ and $\tilde{E}_2(\omega)$ is given by:

$$I(\omega;\tau) = \left| R(\omega) \int_{-\infty}^{\infty} \mathrm{d}\Omega \tilde{E}_1(\omega-\Omega)\tilde{E}_2(\Omega)\exp(i\Omega\tau)\right|^2 \qquad (2)$$

where we measure intensity I introduce τ as the parameter expressing the relative time delay between pulses. To simplify the following discussion we neglect the frequency response of the non-linear interaction (expressed by $R(\omega)$ in Eq. (2)). To make the effect of CEP on the non-linear output, we explicitly separate the zero-order spectral phase term in the expression for electric field:

$$\tilde{E}_{\text{total}}(\omega) = \begin{cases} \tilde{E}(\omega)\exp(i\phi^{CE}) & ;\ \omega > 0 \\ \tilde{E}^*(|\omega|)\exp(-i\phi^{CE}) & ;\ \omega < 0 \end{cases} \qquad (3)$$

We impose that the electric field is a strictly real quantity and thus $\tilde{\phi}(\omega)$ must take a Heaviside-step functional form; this is key to realising a measurement of CEP. Inserting Eq. (3) into Eq. (2) yields:

$$\begin{aligned} I(\omega;\tau) \quad = \quad & |SFG(\omega;\tau)|^2 + |DFG_+(\omega;\tau)|^2 + |DFG_-(\omega;\tau)|^2 \\ & +2\Re\left\{SFG(\omega;\tau)DFG_+^*(\omega;\tau)\, \mathrm{e}^{i2\phi_2^{CE}}\right\} \\ & +2\Re\left\{SFG(\omega;\tau)DFG_-^*(\omega;\tau)\, \mathrm{e}^{i2\phi_1^{CE}}\right\} \\ & +2\Re\left\{DFG_+(\omega;\tau)DFG_-^*(\omega;\tau)\, \mathrm{e}^{i2\phi_1^{CE}-i2\phi_2^{CE}}\right\} \end{aligned} \qquad (4)$$

where

$$\begin{aligned} SFG(\omega;\tau) &\equiv \int_0^{\omega} \mathrm{d}\Omega\, \tilde{E}_1(\omega-\Omega)\tilde{E}_2(\Omega;\tau), \\ DFG_-(\omega;\tau) &\equiv \int_{\omega}^{\infty} \mathrm{d}\Omega\, \tilde{E}_1^*(\Omega-\omega)\tilde{E}_2(\Omega;\tau) \\ DFG_+(\omega;\tau) &\equiv \int_0^{+\infty} \mathrm{d}\Omega\, \tilde{E}_1(\omega+\Omega)\tilde{E}_2^*(\Omega;\tau). \end{aligned} \qquad (5)$$

Both sum-frequency (SFG) and difference-frequency (DFG) terms appear without frequency constrictions. Lines 2-4 of Eq. (4) demonstrate that measured intensity is dependent on the value of ϕ^{CE} if there is spectral overlap between SFG and DFG components. For this overlap to occur the bandwidth of the test pulse (defined Δ) is comparable to the lowest frequency components (ω_L), obeying $\Delta \geq 2\omega_L$. If we re-write Eq. (4) allowing for the harmonic field components to overlap with the fundamental fields $\tilde{E}_1(\omega)$ and $\tilde{E}_2(\omega)$, this bandwidth constraint is alleviated to $\Delta \geq \omega_L$.

The effect of pulse bandwidth on the observation of CEP in a second-order non-linear interaction obeying Eq. (2) is shown in Fig. 1, in which the input fields are at optical and THz frequencies respectively. The THz field is taken with the spectral profile $\omega^2 e^{-\omega^2}$ consistent with ~100 fs single-cycle radiation peaking at 6 THz; the optical pulse is assumed as a transform-limited Gaussian field with varying bandwidth. The absolute phase of the THz field is varied across columns ($\frac{\pi}{6}$, $\frac{\pi}{3}$ and $\frac{\pi}{2}$ from left to right). The effect of CEP variation is increasingly measurable as the spectral overlap between SFG and DFG components increases with optical bandwidth.

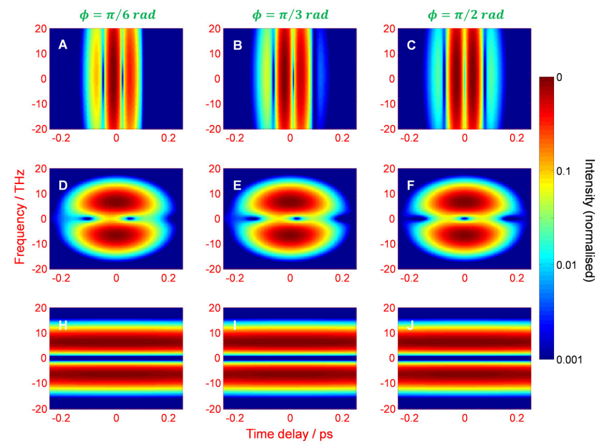

Figure 1: Influence of ϕ^{CE} on intensity spectrograms resolving the non-linear interaction of transform-limited optical and THz pulses, in which the CEP of THz (~100 fs, 6 THz) is varied across columns and the optical pulse duration varied across rows: rows A-C) optical pulse duration 10 fs, D-F) 100 fs and H-J) 1000 fs. Spectrograms are plotted with logarithmic intensity scale and are centered at the optical carrier frequency (375 THz).

The top row is representative of the bandwidth limit $\Delta\omega_{opt} \gg \omega_{THz}$, which is equivalent to the EO sampling limit in which the optical pulse being much shorter than the THz field. In this case any information relating to the optical field is lost and the intensity spectrogram is solely determined by the temporal field profile of the THz field. Retrieval of the THz field proceeds merely by integrating along the frequency axis; use of the FROG algorithm is not required. The middle row is representative of the case $\Delta\omega_{opt} \equiv \omega_{THz}$. Retrieval of *both* the optical and THz fields is possible through use of a suitable FROG algorithm (we demonstrate such a retrieval in Fig. 2 below), with information relating to the CEP encoded at the optical carrier frequency. The bottom row is representative of EO transposition, in which $\Delta\omega_{opt} \ll \omega_{THz}$ and the THz spectrum is transposed to optical frequencies. While it is clear from Fig. 1 that the THz field including CEP information cannot be directly obtained through analysis of this intensity spectrogram, we have demonstrated elsewhere that a separate self-referenced FROG measurement of the transposed field can be used to obtain the THz field [11]. Figure 1 demonstrates that the case of EO sampling can be viewed as specific case of ReD-FROG; equivalently ReD-FROG can be considered to extend THz detection beyond the bandwidth limitations of sampling.

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EXPERIMENT

As a proof-of-concept of ReD-FROG, we present the results of an experiment in which a single-cycle THz pulse produced from a large-area semi-insulating GaAs photoconductive antenna is combined with an optical pulse (~500 fs) in [$\bar{1}$10] oriented ZnTe crystal [10]. The optical probe was stretched from an initial duration of 45 fs using a grating-based zero-dispersion 4-f filter. The frequency-mixing signal (Eq. (2)) was isolated from the fundamental input using a polarizer and analyzed using a spectrometer (Jobin-Yvon iHR550) with CCD detector (PCO DiCamPRO). The relative delay between the optical and THz beams was varied using a linear translation stage.

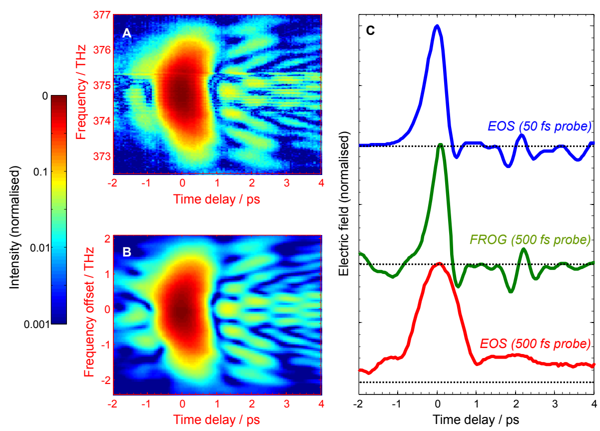

Figure 2: A) Measured and B) retrieved spectrograms resolving the interaction between ~500 fs optical probe (1 THz bandwidth) with ~1000 fs single-cycle THz pulse. C) Comparison of retrieved THz field (middle) with same field obtained with EO sampling with 45 fs (top) and ~500 fs optical probe.

The measured spectrogram is shown in Fig. 2A; this can be compared against the ReD-FROG retrieved spectrogram in Fig. 2B. Interference fringes between the SFG and DFG components can be clearly resolved at ~375 THz. A modified version of the PGCPA FROG algorithm was used to obtain the time-domain THz and optical fields from the spectrogram, in which both fields were constrained to be real in the time domain by ensuring the equivalent Hermitian property in the frequency domain. The retrieved spectrogram had a FROG error of 0.01 (512×512 grid size). The ReD-FROG retrieved time-domain field is compared against the same field inferred by EO sampling (obtained under balanced-detection conditions using the compressed 45 fs optical probe) in Fig. 2C; excellent agreement is observed. For comparison, the field obtained from EO sampling using the stretched 500 fs probe is also shown in Fig. 2C; this combination of fields does not satisfy the bandwidth criterion for EO sampling and the THz field duration is subsequently overestimated.

EXTENSION TO OPTICAL FREQUENCIES

The framework of ReD-FROG outlined above can be applied at any frequency range, assuming the bandwidth criterion $\Delta \sim \omega_L$ is met. It can therefore be applied at infra-red and optical frequencies without significant alteration; the development of octave-spanning single-shot spectrometers will greatly aid experimental implementation [2]. A conceptual demonstration of ReD-FROG at optical frequencies is given in Fig. 3A and C for the self-referenced measurement of a transform-limited Gaussian pulse (375 THz, 75 THz bandwidth) for two values of ϕ^{CE}. The pulse is combined with itself through second-order non-linear mixing and the interference between SFG and DFG components with the fundamental field resolved. The effect of CEP can be resolved in the spectrogram in interference features centered at 200 and 525 THz. The retrieved fields using the ReD-FROG algorithm incorporating the real-field constraint are shown in Fig. 3B and D, with the absolute value of CEP obtained accurately.

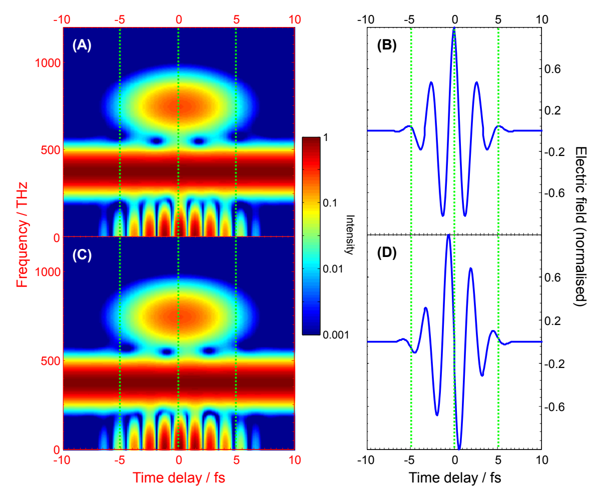

Figure 3: Spectrograms for a self-referenced measurement of a Gaussian transform-limited optical pulse (375 THz, 75 THz bandwidth), A) $\phi^{CE} = 0$ and C) $\phi^{CE} = \frac{\pi}{2}$ rad. B), D): electric field profiles recovered by ReD-FROG from spectrograms A) and C) respectively.

CONCLUSIONS

We have developed the method of ReD-FROG, in which the full temporal profile of an electromagnetic pulse including the CEP can be retrieved from a spectrogram in a single measurement. Information relating to the CEP is resolved in the interference between harmonic components obtained from non-linear frequency mixing; by introducing a real-field constraint into the FROG algorithm we unambiguously recover the temporal profile. A proof-of-concept experiment in which the temporal profile of a single-cycle THz

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pulse is detailed and demonstrates the potential of ReD-FROG to overcome bandwidth limitations inherent to EO sampling. This method opens up new possibilities in the characterization of ultrashort electromagnetic radiation and demonstrates that, contrary to long-held expectations, that FROG is capable of direct absolute phase measurement.

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