V-FROG—single-scan vectorial FROG

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Abstract

We propose and experimentally demonstrate single-scan vectorial frequency-resolved optical gating (FROG) which characterizes the amplitude, phase and polarization of ultrashort laser pulses using a single measured spectrogram. It is carried out by rotating the polarization of the incoming pulse (using a half-wavelength waveplate), in parallel to scanning the delay between the pulse and its replica in an otherwise ordinary FROG apparatus. A ptychography-based phase retrieval algorithm extracts the full pulse information from the recorded spectrogram. We numerically show that this method is reliable and use it to experimentally reconstruct a pulse with intricate time-dependent polarization. We also show that this method can be used to remove time-reversal ambiguity of second harmonic generation FROG.

1. Introduction

Ultrafast pulses with time-varying polarization are used for various applications [1], e.g. exploring optical chirality [2], multi-photon ionization [3], molecular chiral dynamics [4] etc. Such experiments usually require accurate characterization of the pulse’s temporal polarization state, in addition to its amplitude and phase characterization. In the femtosecond regime, these diagnostics are often performed using dual-channel spectral interferometry, particularly its variant polarization labeled interference versus wavelength of only a glint [5–7], or by time-resolved ellipsometry [8–10]. However, both methods require a synchronized reference pulse that is separately characterized. A third method [11] is based on recording three separate spectrograms using an established technique for measuring linearly polarized pulses (e.g. frequency-resolved optical gating (FROG) or dispersion-scan (d-scan) [12]—each one for a different linear polarization projection of the pulse (often horizontal, vertical and 45°). Here we propose a method to characterize pulses with time-varying polarization from a single spectrogram of an ordinary FROG device.

FROG is a popular method for reconstruction of linearly polarized pulses. It relies on recording a spectrogram (also termed a FROG trace) produced from the probed pulse interacting with its delayed replica in a nonlinear medium at various delays [5]. FROG is a robust technique, which stems from its redundancy—a ‘complete’ spectrogram contains \( N \times N \) measurements which are used to reconstruct the \( N \) parameters of the complex-valued time-domain pulse representation. Recently, ptychographic and other optimization based FROG reconstruction methods were developed, enabling reconstruction from ‘incomplete’ spectrograms [13–16]. Later, it was proven analytically that for the noiseless case, even spectra consisted of only three time-delays can be sufficient to uniquely determine a pulse [17]. Furthermore, this redundancy also enables reconstruction from other types of spectrally or temporally truncated spectrograms which may result from experimental considerations [15]. Additional utilizations to FROGs redundancy include multiplexed FROG where several pulses of a pulse burst are retrieved simultaneously from a single measured trace which corresponds to the incoherent sum of the FROG traces of the individual pulses [18], and reconstruction of an unstable pulse-train’s characteristics [19].

Characterizing a vectorial pulse requires accurate retrieval of both its polarization components—essentially reverting the problem to the reconstruction of two linearly polarized pulses and finding their relative timing and phase. However, since ordinary FROG is insensitive to both these
parameters, additional measurements are required (e.g., a third spectrogram at 45° or an interferometric measurement etc).

In this work we propose and demonstrate, numerically and experimentally, vectorial FROG (V-FROG), which utilizes FROG’s redundancy to enable reconstruction of a vectorial pulse with ultrafast evolving polarization from a single spectrogram. V-FROG introduces polarization-state information in the recorded spectrogram by rotating a waveplate (WP) during a FROG scan at known angles. The reconstruction is performed using a ptychographic phase retrieval algorithm [20], adapted for the proposed measurement scheme. We numerically show that V-FROG gives reliable reconstruction results, comparable to scalar FROG under similar conditions. We demonstrate experimentally reconstruction of an ultrafast pulse with intricate time-dependent polarization. This method does not require interferometric setups and inherits all the advantages of ptychographic FROG. Finally, we show that using WP with phase retardation of $\delta \neq \pi$ (i.e., not a half-WP) removes the time-direction/conjugate ambiguity. The proposed scheme to characterize ultrafast pulses with time-varying polarization from a single spectrogram opens new directions, such as single-shot vectorial pulse reconstruction.

2. Vectorial FROG method

2.1. Concept & apparatus

While the suggested concept is general, we consider here a FROG apparatus based on second harmonic generation (SHG). To upgrade the device for measuring pulses with time-varying polarization, we add an achromatic half-wavelength WP and a fixed polarizer before the FROG apparatus (see figure 1(a)). With each scanning step of the FROG, $j = 1, 2, \ldots, J$, we rotate the WP to an angle $\theta_j$ and, as before the upgrade, introduce a delay-$\tau_j$, between the pulse and its replica. The incoming pulse after the WP, $E(\theta_j)(t)$ is given by

$$E(\theta_j)(t) = J_{WP}(\theta_j) E(t)$$

where

$$J_{WP}(\theta) = \begin{pmatrix} \cos \frac{\delta}{2} + i \sin \frac{\delta}{2} \cos 2\theta & i \sin \frac{\delta}{2} \sin 2\theta \\ i \sin \frac{\delta}{2} \sin 2\theta & \cos \frac{\delta}{2} - i \sin \frac{\delta}{2} \cos 2\theta \end{pmatrix}$$

is the Jones matrix of a WP with phase-retardance $\delta = \delta_x - \delta_y$ rotated at an angle $\theta$ with respect to the polarizer transmission axis-$\parallel$. Then, the polarizer blocks $E_{\parallel}(\theta_j)(t)$, such that the measured FROG trace is given by

$$I_{V-FROG}(\omega, j) = \left| \int E_{\parallel}(\theta_j)(t) E_{\parallel}(\theta_j)(t - \tau_j) e^{-i\omega t} dt \right|^2.$$ 

Thus, the V-FROG trace consists of the SHG autocorrelation spectra of linearly polarized pulses, where the polarization axis of the incoming pulse varies in each scanning step.

Two comments are worth mentioning. First, rotating the WP in front of the fixed polarizer is similar to rotating a polarizer. However, one cannot simply give up the WP and rotate the polarizer because the crystal
in SHG-FROG requires an incoming pulse with specific polarization axis. Thus, in principle, one can rotate all the FROG apparatus (including the polarizer) instead of rotating a WP. Additionally, in case the incoming pulse is altered by the dispersion introduced by the optical traits of the WP and polarizer, the reconstruction process can be adjusted to take their impact into account.

2.2. Reconstruction algorithm
Our algorithm is based on the ptychographic reconstruction algorithm for FROG (without soft thresholding) [9], which in turn relies on the extended ptychographic iterative engine [22]. It is convenient to rewrite equation (2) in the following form,

$$I_{V\text{-FROG}}(\omega,j) = |\mathcal{F}\{\chi_j(t)\}|^2,$$

where $$\chi_j(t) = E^{(j)}_\parallel(t) E^{(j)}_\parallel(t - \tau_j)$$ is the generated SHG signal, and $$\mathcal{F}\{\}$$ is the Fourier transform operator.

We initialize the reconstruction algorithm by a random complex-valued vector for both polarization components of the reconstructed pulse $$\tilde{E}^{\text{rec}}_{j\parallel}(t)$$. In each iteration of the algorithm, we update the reconstructed pulse using all the measured spectra in an arbitrary order. Therefore, in each iteration the reconstructed pulse is updated $$J$$ times. The update order is reshuffled at the start of each iteration by generating a permuted array of the integers $$j = 1, 2, \ldots, J$$, denoted by $$k(j)$$.

Following is a description of the steps used to obtain $$\tilde{E}^{\text{rec}}_{j\parallel}(t)$$ from $$\tilde{E}^{\text{rec}}_{j\parallel}(t)$$ and the measured spectrum at the step $$k(j)$$, $$I_{V\text{-FROG}}(\omega,k(j))$$. Notably, because only the component parallel to the polarizer is measured in each step, we can only calculate a correction for that component.

We start by calculating both components of the reconstructed pulse following the WP using equation (1),

$$\tilde{E}^{(j\parallel\text{rec})}_{j\parallel}(t) = J_{WP}(\theta_{k(j)}) \tilde{E}^{\text{rec}}_{j\parallel}(t).$$

Subsequently, we calculate $$\psi_j(t)$$, the expected SHG signal generated by the recovered pulse and its shifted replica,

$$\psi_j(t) = O(t) P(t),$$

where we denote $$O(t) \overset{\text{def}}{=} \tilde{E}^{(j\parallel\text{rec})}_{j\parallel}(t)$$ as the reconstructed ‘Object’ and its shifted replica $$P(t) \overset{\text{def}}{=} \tilde{E}^{(j\parallel\text{rec})}_{j\parallel}(t - \tau_{k(j)})$$ as the ‘Probe’. We then Fourier transform $$\psi_j(t)$$ and replace its modulo with the measured amplitude,

$$\varphi_j(\omega) = \sqrt{I_{V\text{-FROG}}(\omega,k(j)) \mathcal{F}\{\psi_j(t)\}} \mathcal{F}\{\psi_j(t)\}|.$$ (6)

Subsequently, we obtain an updated signal by inverse Fourier transforming the corrected signal, obtaining an SHG signal with the correct power spectrum,

$$\tilde{\psi}_j(t) = \mathcal{F}^{-1}\{\varphi_j(\omega)\}.$$ (7)

Next, we calculate the corrections $$\Delta_O(t)$$ for the ‘Object’ and $$\Delta_P(t)$$ for the ‘Probe’

$$\Delta_O(t) = \gamma \frac{P(t)|\tilde{\psi}_j(t) - \psi_j(t)|}{1 - \alpha|P(t)|^2 + \alpha|P(t)|^\alpha_{\text{max}}}$$

$$\Delta_P(t) = \gamma \frac{O^*(t)|\tilde{\psi}_j(t) - \psi_j(t)|}{1 - \alpha|O(t)|^2 + \alpha|O(t)|^\alpha_{\text{max}}}.$$ (8)

where $$\alpha, \gamma$$ are heuristic parameters. We used $$[\alpha, \gamma] = [0.3, 0.5]$$ which we found to give good reconstruction results. Afterwards, we update the measured component by adding the above correction terms to the reconstruction from the previous iteration ($$\Delta_P(t)$$ is shifted back by $$\tau_{k(j)}$$):

$$E^{(j\parallel\text{rec})}_{j\parallel+1}(t) = P(t) + \Delta_O(t) + \Delta_P(t + \tau_{k(j)}).$$ (9)

Finally, we update the vectorial reconstruction using the corrected component by applying the inverse operator to equation (1),

$$E^{\text{rec}}_{j+1\parallel}(t) = J_{WP}^{-1}(\theta_j) \begin{pmatrix} E^{(j\parallel\text{rec})}_{j\parallel+1}(t) \\ E^{(j\parallel\text{rec})}_{j\parallel+1}(t) \end{pmatrix}.$$ (10)
3. Results

3.1. Numerical analysis—comparison to scalar FROG

We explored the validity of V-FROG numerically by reconstructing vectorial pulses from noised spectrograms. We also compared its performance with conventional SHG-FROG. To this end, we numerically produced a set of 100 scalar pulses that all conform to a Gaussian power spectrum which is centred at 800 nm and 213 nm spectral bandwidth. Each pulse ($N = 128$ grid points) is produced by applying a random spectral chirp to the above power spectrum, conditioned that the FWHM of the pulse is smaller than 200 fs. The spectral chirp is generated by taking random white noise and low-pass filtering it, retaining its first 13 coefficients out of 128. To produce vectorial pulses, we used the above set of pulses as the $x$ axis polarization component and a random permutation of the same set as the $y$ axis polarization. The calculated V-FROG traces for each pulse are $128 \times 128$ points with equally spaced delays, $d_t = 1.57$ fs, and spanning the same frequency window (i.e. $d_t \times df = 1/N = 1/128$).

We numerically tested V-FROG with various equispaced rotation sets, i.e. $\Delta \theta = \theta_j - \theta_{j-1}$ is constant, as well as different WPs with $\delta$ ranging from $\pi/2$ to $\pi$ (i.e. from a quarter WP to a half WP). The reconstructions of the spectrograms and pulses were fine, except for $\Delta \theta \sim 0, 45^\circ, \text{and } 90^\circ$ (in these cases there is no measured data on the relative phase and timing between the S and P polarization components).

We obtained the best and most consistent performance when $\delta = \pi$ and $\Delta \theta = 60^\circ$, that is, $\theta_j = 60^\circ \times (j \mod 3) = \{0^\circ, 60^\circ, 120^\circ, \ldots\}$ ($\Delta \theta = 30^\circ$ resulted with similar results) which we used in the subsequent analysis. The reason is likely that these values enable adequate sampling of both vectorial components and their superposition.

We added Gaussian noise to the calculated spectrograms’ intensity at several signal-to-noise ratios (SNRs), in the range 0–60 dB using 10 dB steps as indicated in figure 2 and reconstructed them using our prescribed reconstruction method. We evaluated the reconstructed pulses using the SNR metric,

$$\text{SNR} = -10 \cdot \log_{10} \left( \frac{E_{\text{orig}}^{\text{rec}}}{E_{\text{orig}}^{\text{rec}} - E_{\text{orig}}^{\text{rec}}} \right).$$

The reconstructed spectrograms were normalized to have a peak value of unity and evaluated using the standard FROG error $G$ [5],

$$G = \sqrt{\frac{1}{N^2} \sum_{\omega,j} \left| I(\omega,j)^{\text{rec}} - \mu I(\omega,j)^{\text{orig}} \right|^2},$$

where $\mu$ is a real normalization constant which minimizes $G$.

For comparison, we followed a similar procedure for ten scalar pulses and no rotation of WP (i.e. conventional FROG). In all runs, we limited the algorithm to 30 iterations and the best spectrogram reconstruction was chosen out of ten different runs for each spectrogram.

Figure 2 displays the results we obtained. As shown, pulse reconstruction SNRs (RSNRs) scale with spectrogram FROG error (shown in log scale). Among the 700 runs, there were several outliers which we examined. The outliers were cases where the algorithm stagnated and a few more runs improved the results.
3.2. Experiment

In this section we experimentally demonstrate V-FROG. To prepare the measured pulse we started with a $\sim 40$ fs linearly-polarized pulse from an ultrafast Ti:sapphire laser system which we transmitted through two multi-order WPs, followed by a cube polarizer, which also adds significant chirp to the pulse, and finally through another multi-order WP.

To measure the pulse, we used a setup as depicted in figure 1, which included an achromatic $\delta = \pi$ WP as well as a SHG-FROG device manufactured by few-cycle Inc. containing a 10 $\mu$m thick, Type I BBO and an Avantes AvaSpec-Mini2048CL spectrometer with a 1.4 nm FWHM resolution.

We recorded a 256 $\times$ 256 spectrogram with three alternating rotation angles given by $[\theta_1, \theta_2, \theta_3] = [0^\circ, 15^\circ, 45^\circ]$. The delay-step was given by $\Delta \tau = 10$ fs and we interpolated the frequency step accordingly to $\Delta f \approx 39$ THz. Figure 3(a) portrays the recorded and reconstructed V-FROG spectrograms, the error is given by $G = 2.79 \times 10^{-3}$.

Figure 3(b) illustrates the reconstructed pulse electric field, where due to the pulse length we present ten experimental optical cycles as one optical cycle in the figure while colour denotes the degree of circular polarization. The reconstructed pulse is chirped, has an intricate time-varying polarization and its duration is $\sim 650$ fs long.

To verify V-FROG’s reconstruction results, we also recorded two ordinary FROG spectrograms using identical parameters at $[\theta_P, \theta_S] = [0^\circ, 45^\circ]$ (figure 3(c)).
Figure 4. Schematic diagram of single-shot vectorial d-scan. We add a transversely-dependant WP behind the prism in the setup described in [18]. This introduces polarization-dependency into the measured spectrogram, varying along with the dispersion variance, resulting with a similar measurement set to V-FROG, which should allow for a full vectorial reconstruction of the pulse.

The spectrogram reconstruction errors are $[G_P, G_S] = [3.28, 2.7] \cdot 10^{-3}$, and the pulse RSNRs compared to the V-FROG reconstruction of the same components are $[RSNR_P, RSNR_S] = [15.6, 15.3]$ dB, indicating that indeed the pulse is well-reconstructed.

4. Breaking time-reversal ambiguity

Conventional SHG-FROG suffers from several ambiguities, among them time-reversal, i.e. a pulse and its complex conjugate with reversed time direction produce identical FROG traces [5]. Interestingly, this ambiguity is removed in V-FROG. To show this feature, we examine a simple example: a pulse $\vec{E}_{RH}(t)$ with right-hand circular polarization and a WP with phase retardation $\delta = \pi/2$ (i.e. a quarter WP) Applying equation (1) we get the field after the WP:

$$E^\| (\theta_j) (t) = \left[ J_{WP} (\theta_j) \vec{E}^{RH} (t) \right]^\| \vec{E}^{RH} (t)$$

$$= \frac{1}{2} \left( 1 + i \cos 2\theta_j \ i \sin 2\theta_j \right) E(t)$$

$$= \frac{1}{2} \left( 1 + \sin 2\theta_j + i \cos 2\theta_j \right) E(t). \quad (12)$$

Substituting it into equation (2), we find that the SHG signal follows a squared Malus’s law pattern,

$$I_{V-FROG}^{RH} (\omega, j)$$

$$= \frac{1}{4} \left( 1 + \sin 2\theta_j \right)^2 \left( \int E(t) E(t - \tau_j) e^{-i\omega \tau} d\tau \right)^2$$

$$= \frac{1}{4} \left( 1 + \sin 2\theta_j \right)^2 I_{FROG} (\omega, j). \quad (13)$$

On the other hand, following the same calculation for the complex-conjugate pulse with reversed time direction leads to a different FROG trace:

$$I_{V-FROG}^{RH} (\omega, j) = \left( 1 - \sin 2\theta_j \right)^2 I_{FROG} (\omega, j) \neq I_{V-FROG}^{RH} (\omega, j). \quad (14)$$

For more complicated pulses with time-varying polarization, the calculation evidently depends on the details of the pulse. Still, we found through numerical investigations that as long as the WP phase retardation is $\delta \neq \pi$ and the pulse is not linearly-polarized, there is no time-reversal ambiguity.

5. Conclusions

We proposed and demonstrated, numerically and experimentally, V-FROG—a vectorial-pulse characterization method which is based on upgrading existing FROG devices by simply adding a rotating WP and a polarizer at their input. Unlike previous methods [5, 9, 11], which separate the vectorial-characterization problem into several scalar sub-problems, V-FROG relies on FROG method’s inherent redundancy to record a single spectrogram which contains polarization-state information in addition to temporal and spectral information.

While here we used an achromatic half-wavelength WP, other WPs can also be used—e.g. WPs with a wavelength-dependant retardance. This will not conceptually alter the measurement or reconstruction process as equations (4) and (10) can be modified to account for this by applying the appropriate...
transformation for each axis. Additionally, achromatic WPs have an axis-dependent delay which affects V-FROG measurements of high-bandwidth pulses. It is possible to take this effect into account by calculating or measuring the different delays and altering equations (4) and (10) accordingly. We verified numerically that for pulses with 200 nm bandwidth this is not mandatory.

We found that V-FROG performs almost equally to ordinary FROG under similar conditions. The proposed multiplexing of polarization, amplitude and phase information to single spectrogram should allow single-shot characterization devices (e.g. GRENOUILLE [23] and single-shot d-scan [24]) to also measure pulses with time-varying polarization (e.g. see figure 4 for proposed upgraded single-shot d-scan). Another exciting direction is blending this concept with FROG-based spatio-temporal reconstruction [25] to facilitate self-referenced vectorial spatio-temporal pulse reconstruction.

Finally, it is worth noting that while in this work we adapted a ptychography-based phase-retrieval algorithm, other reconstruction methods may be applied which may further improve the results such as employing deep neural networks (i.e. deep-learning) [26], using a maximum-likelihood estimate approach [27], or utilizing structure-based prior knowledge on the laser pulses [28].

Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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