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Single-shot measurement of a laser waveform using plasma fluorescence in ambient air

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Abstract: The temporal characterization of a laser pulse is an important task in studying ultrafast laser-matter interactions. It is ideal to measure the temporal profile of the laser pulse with a single laser shot when the repetition rate is low or its interaction with matter is unstable. Here we report a new approach for the single-shot temporal characterization of a laser pulse, based on the TIPTOE (tunneling ionization with a perturbation of the time-domain observation of an electric field) method. The waveform of the laser pulse is reconstructed from the intensity modulation of plasma fluorescence emission. The result of the single-shot measurement is compared with the time-delay scanning TIPTOE measurements, supporting the validity of the single-shot measurement.

1. Introduction

Ultrafast laser-matter interactions have been extensively investigated across diverse fields [1]. Since the temporal shape of the laser waveform plays a crucial role in these interactions, its temporal characterization has become an essential prerequisite in many ultrafast optics experiments. Several metrologies have been developed such as auto-correlator [2,3], FROG [4,5], SPIDER [6], D-scan [7,8], and others [9–13]. Some of these techniques rely on the cross-correlation of a nonlinear processes using two replica pulses that are measured as a function of the time delay between the two pulses [2–6]. The others utilize the variation of the second harmonic spectrum for different dispersion conditions of the laser pulse [7,8]. Consequently, these techniques require many laser shots for the temporal characterization.

There are many cases where the multi-shot techniques cannot easily be applied. The high-power laser may work at extremely low repetition rate. The laser pulse itself or the interaction to be studied are inherently unstable. In these situations, it is ideal to measure a laser pulse with a single laser shot. Most of the temporal characterization methods have their own single-shot version [3,5,6,8,9,13]. In these techniques, the time delay or the different dispersion conditions are spatially mapped, eliminating the necessity of the multi shot measurement. The temporal characterization of the laser pulse could be achieved with a single laser shot.

All the techniques mentioned above are frequency domain techniques in which the spectrum of the nonlinear interaction is measured. Therefore, the bandwidth of the laser pulse that can be characterized is determined by the bandwidth of the nonlinear interaction (typically, second harmonic generation), which is often limited due to the phase matching bandwidth of the nonlinear crystal [14,15]. Also, a specific nonlinear crystal is used at difference wavelengths.

Recently, a new approach called tunneling ionization with a perturbation for the time-domain observation of an electric field (TIPTOE) has been successfully demonstrated [16–22]. It is a

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time domain approach in which strong field ionization is used to sample a laser field. Since the strong field ionization occurs within a sub-cycle duration, it can be used to measure the laser field with sub-cycle temporal resolution. Also, the technique can be universally applied for a broad spectral range from UV to IR thanks to the extremely high nonlinearity of the ionization [17]. A single-shot TIPTOE measurement for 3.1–3.8 μm central wavelength has been implemented using the 3-4 photon excitation of photocurrents in a silicon-based image sensor chip [20]. However, the nonlinear excitation bandwidth of the chip is not high enough for UV or visible wavelengths.

Here, we demonstrate a single-shot measurement of the laser waveform based on the TIPTOE method. We measure the fluorescence light generated due to ionization in air using two laser pulses superposed with a small angle. The modulation of the fluorescence light is captured using an ordinary CMOS camera. The femtosecond laser pulses are measured under various chirp conditions. The comparison made with the results obtained scanning type TIPTOE confirm the validity of the single-shot measurement. Since our approach rely on tunneling ionization, it can be applicable for a broad spectral range. Thus, it will become a useful tool for the single-shot pulse characterization.

2. Experimental setup and working principle

In the TIPTOE method, sub-cycle ionization plays a role of a temporal gate for the pulse characterization. The total ionization yield is the integration of the instantaneous ionization rate over time as $N_{tot} \propto \int_{-\infty}^{\infty} w(t)dt$, where $w(t)$ is the instantaneous ionization rate that can be approximated as $|E(t)|^{2n}$ [16]. Here, n is the nonlinearity of the ionization. With the superposition of the weak signal field rE on the strong fundamental field E, the total ionization yield N_{tot} varies as $N_{tot}(\tau) \propto \int_{-\infty}^{\infty} |E(t) + rE(t-\tau)|^{2n}dt$. Here, τ is the time delay between the two laser fields and r is the amplitude ratio of signal field. Therefore, the laser field $E(t)$ can be found from the ionization yield modulation.

For the experimental demonstration of the single-shot measurement, we used a Ti:sapphire laser which produces 30 fs laser pulses at the center wavelength of 800 nm at the repetition rate of 1 kHz. The laser beam energy of 3.7 mJ was measured before the experimental setup. The laser beam was split in space using two rectangular mirrors, as shown in Fig. 1(a). These mirrors allowed us to control the intensity ratio of the two beams. The position of the input beam is adjusted so that the amplitude ratio at the focus is $r = 0.071$. The larger portion of the beam becomes the fundamental field $E(t)$, and the small portion becomes the signal field $rE(t)$.

We generated a line-shaped plasma fluorescence using two cylindrical mirrors. The first cylindrical mirror (V-CCM) has a long focal length (f = 200 mm), reducing the size of the beam along the horizontal (x-axis) direction. This was required to obtain a sufficiently high intensity for the generation of fluorescence emission at the focus. The second cylindrical mirror (H-CCM) has a short focal length (f = 25 mm), which tightly focus the laser beam. As a result, a long (4 mm) focused beam is obtained along the horizontal (x-axis) direction.

The focused laser beams produce a fluorescence emission from a plasma in air. It is assumed that the intensity of the fluorescence emission is proportional to the ionization yield [23]. To obtain the ionization yield modulation with a single laser shot, the two beams are superposed in a small angle θ as shown in Fig. 1(a). The angle θ can be controlled by the separation (d in Fig. 1(a)) of the two mirrors and the distance from the two mirrors and the focus. The delay between the two beams can be obtained as $\tau = (1/c)[x\sin(\theta) - 2y\sin^2(\theta/2)]$. It can be simplified as $\tau = (x/c)\sin\theta$ along the x-axis where $y = 0$. The interference pattern is slightly tilted because the time delay is a function of both x and y. For example, the time delay is zero ($\tau = 0$) along the white arrow where $x/y = \theta/2$ as shown in Fig. 1(b). With this experimental setup, the time delay τ can be mapped in a space. Finally, the intensity of the fluorescence emission $N_{tot}(x)$ could be measured with a single laser shot that represents the ionization yield modulation, $N_{tot}(x) \propto \int_{-\infty}^{\infty} |E(t) + rE(t-(x/c)\sin\theta)|^{2n}dt$.

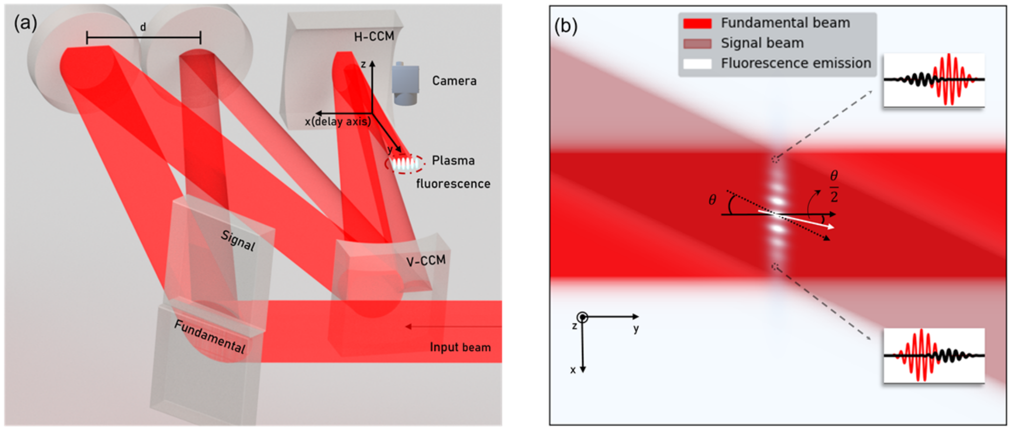

Fig. 1. Experiment setup for single-shot TIPTOE measurement. (a) Arrangement of optical elements in the experiment setup. An input laser beam is spatially separated using two rectangular mirrors. The top mirror reflects the signal beam and the bottom mirror reflects the fundamental beam. Two cylindrical mirrors (H-CCM and V-CCM) were used to produce a long focused beam along the transverse (x-axis) direction. V-CCM is the vertical-axis cylindrical mirror that controls the length of the focus. H-CCM is the horizontal-axis cylindrical mirror that focus the beam for ionization. (b) The top view of the superposition of the two beams at the focus. The two beams are superposed with an angle θ which can be controlled by the separation d in (a). The insets show the fundamental (red) and the signal (black) pulses at different positions.

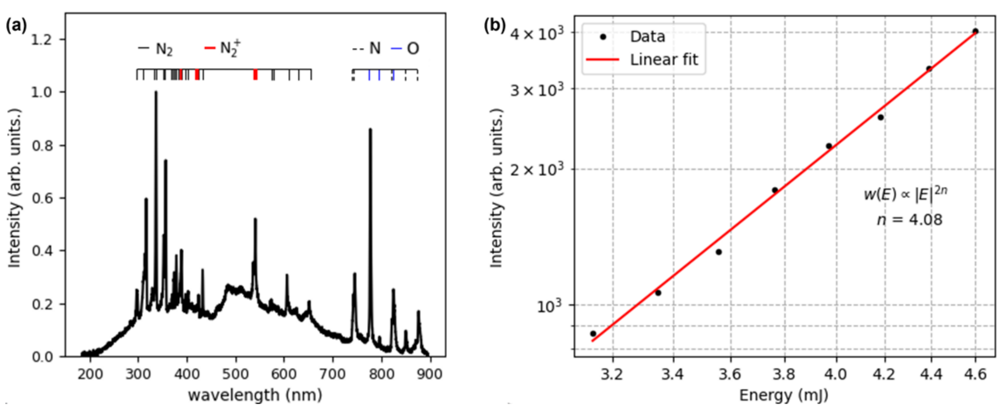

Fig. 2. Nonlinearity measurement. (a) Spectrum of plasma fluorescence emission measured using a grating spectrometer (b) Intensities of the fluorescence emission measured at different laser energies. The red line shows the linear fit of the data in a log-log scale.

In ambient air, ionized molecules and atoms generate line spectra and continuum emission [24–28]. The spectrum of the fluorescence emission is shown in Fig. 2(a). The nitrogen molecule N_2 lines (second positive band, $C^3\Pi_u$-$B^3\Pi_g$) are obtained as various peaks on the UV edge of the continuum. Next to the N_2 molecule lines, N_2 cation lines (first negative band, $B^3\Sigma_u$-$X^3\Sigma_g$) are lied on 360-400 nm range. On the range of visible red to near infrared, atomic lines of nitrogen and oxygen are detected [25]. These emissions are recorded using an imaging system arranged along the z-axis in Fig. 1(a) in which an ordinary CMOS camera (Basler, ACE2, 5320 × 3032 pixels, 2.74 µm pixel size) is used with an objective lens (Mitutoyo, M Plan Apo NIR

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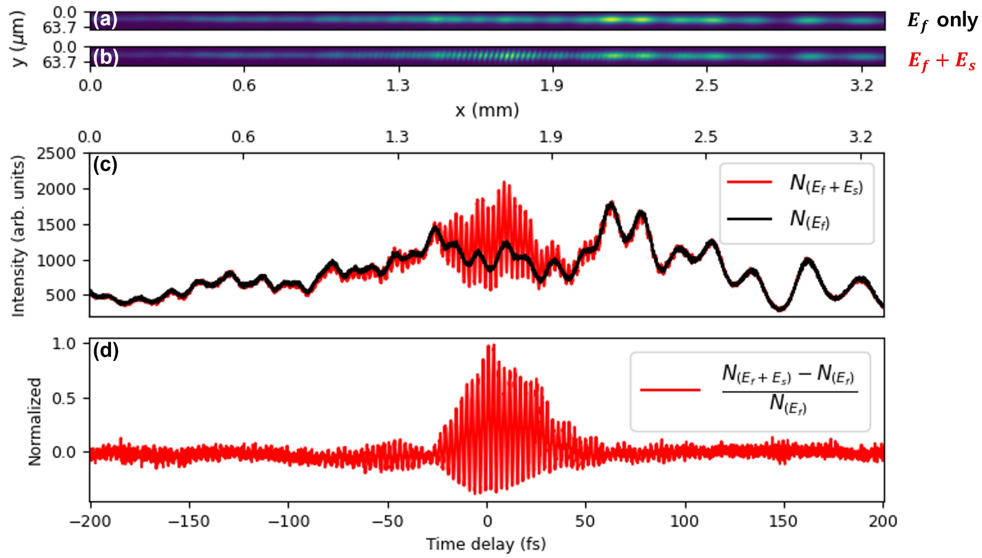

Fig. 3. Single-shot TIPTOE measurement. (a-b) Fluorescence emission captured by a CMOS camera (a) with and (b) without the signal pulse (averaged for 60 ms). (c) The ionization yield modulations obtained from the fluorescence emissions obtained with (red) and without (black) signal pulses. (d) Normalized fluorescence emission. The angle θ was 2.1 deg.

5x, 0.14 NA, 14 µm depth of field). The intensity nonlinearity of the fluorescence emission was measured as shown in Fig. 2(b). The nonlinearity n is estimated to be 4.08. We used $n = 4$ for the reconstruction algorithm.

The single-shot laser waveform was obtained from the captured ionization yield modulation. We first measured the fluorescence emission from the fundamental laser field only (ionization yield measured without a signal pulse, N_0) as a reference as shown in Fig. 3(a). Then, we have added the signal pulse to obtain the ionization yield modulation, N_{tot}. A clear modulation was observed as shown in Fig. 3(b). The ionization yield modulations obtained from the fluorescence emissions for both cases are shown in Fig. 3(c). In order to extract the signal laser field, the ionization yield modulation is normalized as $N_{tot}/N_0\text{-}1$, as shown in Fig. 3(d). Therefore, the ionization yield modulation is not affected by the intensity variation of the original beam. Then we applied the gradient descent optimization algorithm to find E_s from $N_{tot}/N_0 - 1$ [18]. In this way, the temporal profile of the laser field could be obtained with a single laser shot.

3. Comparison between single-shot and scanning TIPTOE

To verify the results obtained with a single-shot TIPTOE measurement, we performed the pulse characterization using two different approaches. We first measured laser waveforms with three different dispersion conditions. Meanwhile, the same measurements were performed using a scanning type TIPTOE [18]. It should be noted that the two approaches work at different intensity ranges. The scanning type TIPTOE works at the intensity around 10^{13} W/cm^2 in which the ionization yield modulation is directly measured using metallic electrodes. In this case, the total amount of ionization or air molecules is below a few percent [23]. The nonlinearity of the ionization is $n\sim 6$.

For the single-shot measurement, we had to increase the laser intensity to $\sim 10^{15}$ W/cm^2 to clearly see the fluorescence emission in air. This causes the reduction of the nonlinearity because

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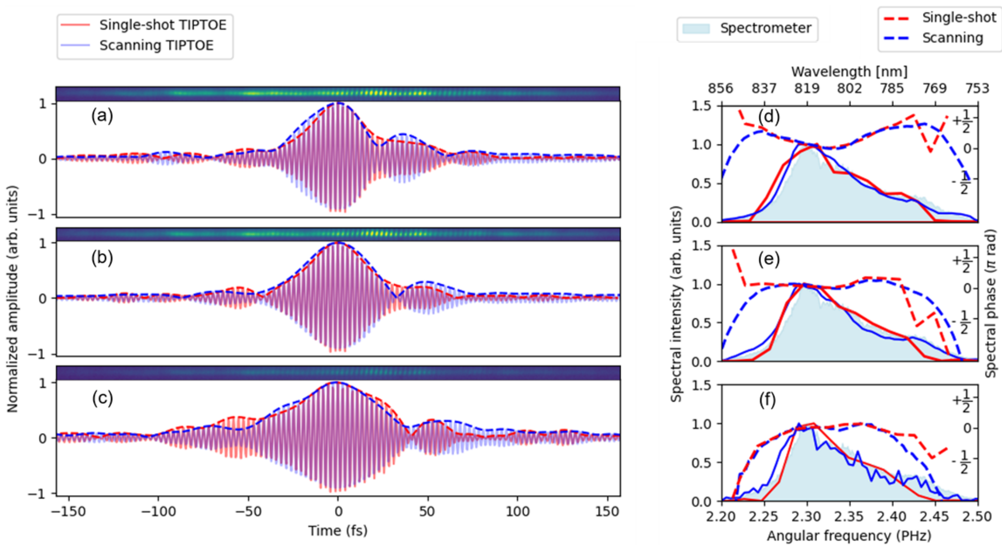

Fig. 4. Comparison of the reconstructed waveforms measured using the single-shot TIPTOE and the scanning type TIPTOE. The laser fields are measured with different dispersion conditions. The reconstructed waveforms and their spectrums are shown for (a, d) the positive GDD (+300 fs^2), (b, e) no GDD (0 fs^2), and (c, f) negative GDD (-300 fs^2) conditions. In (d), (e) and (f), The reconstructed spectral phases (dashed) and spectrum (solid) obtained from the single-shot (red lines) and scanning (blue lines) TIPTOE cases are compared with the spectrum measured using a grating spectrometer (blue shadow). The raw images of the fluorescence emission are shown above the reconstructed waveform in (a)-(c). The time range of the single-shot measurement is shorter than the scanning TIPTOE measurement because the time range of the single-shot TIPTOE measurement is limited by the size of the imaging sensor. Therefore, their spectral resolutions are different in (d)-(f). The angle θ was 1.6 deg.

most of the air molecules are ionized at this intensity. We obtained the nonlinearity of $n \sim 4$ as shown in Fig. 2. The nonlinearity n sets a limit on the duration of the fundamental pulse. For an accurate TIPTOE measurement, the duration of the fundamental pulse should be shorter than $\tau_{TL}\sqrt{2n-1}$ [18,23]. Here, τ_{TL} is the transform limited pulse duration, which is τ_{TL}=27.5 fs. This restriction is not applied to the signal pulse. In this work, we used identical pulses for the fundamental and signal pulses because the pulse duration shorter than this limit. The intensity of the laser beam was attenuated using a random pinhole attenuator that allows us to reduce the laser intensity without changing the dispersion conditions [29].

The reconstructed waveforms obtained from the two measurements for different group delay dispersion (GDD) conditions are shown in Fig. 4(a-c). The GDD was controlled using an acousto-optic programmable dispersive filter (AOPDF) [30]. The Fourier transform spectra and their phases are shown in Fig. 4(d-f). The temporal profiles of the laser fields show a reasonably good agreement. The discrepancy observed in the two measurements can be attributed to the spatio-temporal effect. The pulse is spatially superposed along the long focus. Nevertheless, the positive and negative quadratic phases are successfully reconstructed in both measurements including the inherent fourth order dispersion. The GDD values estimated from the reconstructed pulses show a very good agreement as shown in Fig. 5, supporting the validity of the single-shot measurement.

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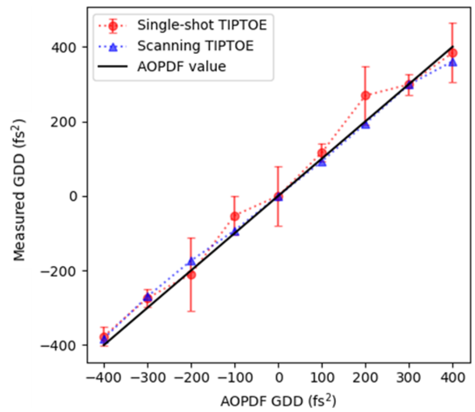

Fig. 5. The GDD values obtained from the single-shot (circles) and scanning-type (triangles) TIPTOE measurements are compared. The error bar for the single-shot TIPTOE measurement represents the standard deviation of four measurements. The GDD values set by the AOPDF is also shown with a black line.

4. Discussion

We demonstrated the single-shot TIPTOE measurement in which the laser waveform is characterized using the fluorescence emission generated using a non-collinear experiment setup. To confirm the validity of this measurement, the waveforms were measured using the scanning-type TIPTOE and the single-shot TIPTOE. The pulses measured at different dispersion conditions show a good agreement. The amount of dispersion estimated from the reconstructed waveforms show a very good agreement with the theoretically expected values, showing the validity of the single-shot measurement. We expect the single-shot TIPTOE measurement will be highly useful for the high-power lasers whose repetition rate is very low.

We measured the ionization yield modulation using the fluorescence emission in ambient air. This allowed us to use an ordinary CMOS camera without having a complicated setup for the measurement. Since we measure the fluorescence emission generated from air plasma, there is no damage issues. The fluorescence emission can be produced for a broad wavelength range and measured using an ordinary imaging device. Thus, our approach can be applied for a broad wavelength range.

The fluorescence emission would be produced more efficiently in different molecules which needs to be tested in the future work. Also, it would be ideal to measure the ionization yield directly by the current measurement. This would require the development of the current measurement device with a good spatial resolution. These improvements will reduce the required intensity for the single-shot measurement which will increase the nonlinearity of the ionization. Therefore, the dispersion range of the measurement can be broadened. Also, the laser waveform can be converted directly to the electronic signal which can be useful. Since we use one dimensional data, it can be efficiently processes for high repetition laser systems.

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

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