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Temporal characterization of a two-color laser field using tunneling ionization

J. Shin,[1,2] I. Ivanov,[1] W. Cho,[1] R. Shrestha,[1,2] and K. T. Kim[1,2,*]

[1]Center for Relativistic Laser Science, Institute for Basic Science, Gwangju 61005, Republic of Korea
[2]Dept. of Physics and Photon Science, Gwangju Institute of Science and Technology, Gwangju 61005, Republic of Korea
*kyungtaec@gist.ac.kr

Abstract: The superposition of a fundamental laser pulse and its second harmonic can form an asymmetric laser field that is useful in many applications. The temporal characterization of the two-color laser field becomes necessary. However, the temporal characterization of the two-color laser pulse is a challenging task due to its broad bandwidth and a spectral gap between the two frequency components. Here we demonstrate the temporal characterization of the two-color laser field using multiple ionization yield measurements near the laser focus. This new approach enables the complete temporal characterization of the two-color laser field, including the relative phase between the two frequency components.

1. Introduction

The superposition of a fundamental laser field and its second harmonic field forms a two-color laser field. The two-color laser field can be asymmetric depending on the relative phase between the fundamental and its second harmonic fields. The asymmetry of the two-color laser field has been exploited in many applications such as terahertz generation [1], molecular alignment [2], and the coherent control of photoemission from a nanotip [3]. The two-color laser field can also be used to control ultrafast electron dynamics [4], producing an intense high harmonic radiation [5]. Since the temporal shape of the two-color laser field greatly varies depending on the relative phase between the two fields, the temporal characterization of the two-color laser field that includes the relative phase is essential.

Although there are many temporal characterization techniques developed for femtosecond laser pulses, it is difficult to apply them directly to the two-color laser field. Some of these techniques (such as FROG, SPIDER, SRSI, and so on. . .) measure the spectrum of the response of a nonlinear medium [6–9]. However, the limited spectral bandwidth of the nonlinear response does not cover the entire spectral range of the two-color laser field. In addition, a wide spectral gap presented in the two-color laser field causes a common problem in finding the relative phase of the two-color components in these frequency-domain approaches [10]. The temporal characterization of the two-color laser fields has been implemented only for a limited case in which the spectral gap is less than the spectral width of the two frequency components [11,12]. The temporal characterization of the two-color laser field has remained a difficult problem.

There are other temporal characterization techniques in which the waveform of the laser field is directly sampled in the time domain. These techniques utilize a fast temporal gate such as attosecond extreme ultraviolet (XUV) pulses [13], electron responses [14], ionization [15–18], light-field-induced currents in solids [19], transient absorption in solids [20,21]. However, the first two techniques require an attosecond XUV pulse in a complex vacuum environment [13,14]. Among the time-domain techniques, the temporal characterization technique called the tunneling ionization with a perturbation for the time-domain observation of an electric field (TIPTOE) [22] can be used to measure a two-color laser pulse. The TIPTOE technique can be applied for a

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broadband pulse in ambient air. The TIPTOE method is successfully applied for a single-cycle laser field [23], and a few-cycle laser field [15], including their carrier-envelope phase (CEP) for a broad spectral range from UV to IR [22].

An ionization yield is measured using two replica pulses of an input laser pulse in the TIPTOE method. One laser pulse is called the main pulse, and the other pulse is called the signal pulse. The main pulse is intense enough to ionize air molecules (mostly O_2) [24]. It is this ionization that plays the role of a temporal gate. The signal field is relatively weak. It does not trigger ionization alone. However, the superposition of the main field and the signal field can significantly modulate the ionization yield. The modulation of the ionization yield provides information on the temporal profile of the signal field.

It is ideal to use a single isolated temporal gate in the time domain techniques. When ionization occurs in a single half optical cycle, the modulation of the ionization yield indeed directly represents the electric field of the signal laser pulse. Therefore, the electric field of the laser field can be completely determined, including the CEP and the relative phase of the two-color laser field. However, this condition can be satisfied only for an extremely short laser pulse (i.e., a near-single cycle laser pulse). In general, ionization occurs in multiple half-cycles for laser pulses longer than a single-cycle pulse. Therefore, the convolution effect of the multiple ionizations should be taken into account [25]. As we show below, the CEP and the relative phase information can be washed out due to the convolution effect when ionization occurs in the multi-cycle laser pulse. Therefore, additional information is required to determine the relative phase.

In this work, we show the temporal characterization of the two-color multi-cycle laser field including the relative phase of the two-color laser field, using a multiple ionization yield measurement. The relative phase of the two-color laser field is determined by the value of ionization yields measured at different relative phases. This information is combined with the ionization yield modulation, enabling the complete temporal characterization of the two-color laser field.

2. Basic principle of two-color field measurement

In the TIPTOE technique, an incident laser field is split into two replicas with different intensities. The ionization yield obtained using the two two-color laser pulses can be written as [25]

$$N(\tau) \propto \int_{t=-\infty}^{+\infty} [E_m(t-\tau) + rE_m(t)]^{2n} dt. \tag{1}$$

Here $E_m(t)$ is the main two-color field, r is the field strength ratio, τ is the time delay, and n is a nonlinear coefficient of ionization in the air [24]. The temporal profile of the signal two-color pulse, which is a weak replica of the main pulse, is $rE_m(t)$. Since the temporal profile of the laser pulse is determined using the ionization yield modulation, it is useful to define the normalized ionization yield (NIY) as

$$\delta_N(\tau) = \frac{N(\tau)}{N^{(0)}} - 1. \tag{2}$$

Here, $N^{(0)}$ is the ionization yield obtained with the main pulse only. The temporal profile of the signal laser field can be found from the NIY using a suitable reconstruction process [20].

Additional attention should be paid to the two-color laser pulses because the relative phase between the two-color components may not be correctly represented by the NIY. For example, we calculated ionization yields at two different relative phases, 0 (solid red line) and 0.5π (black dotted line), as shown in Fig. 1(a). In this calculation, an 800-nm 41-fs fundamental laser field and a 400-nm 85-fs second-harmonic field were used with the nonlinearity of $n = 6$, and the intensity ratio of $r^2 = 0.001$, which is similar to the experimental condition. While the temporal profiles of the two-color laser fields at the relative phases of 0 and 0.5π are different, their NIYs are almost identical, as shown in Fig. 1(b), showing the ambiguity of the relative phase.

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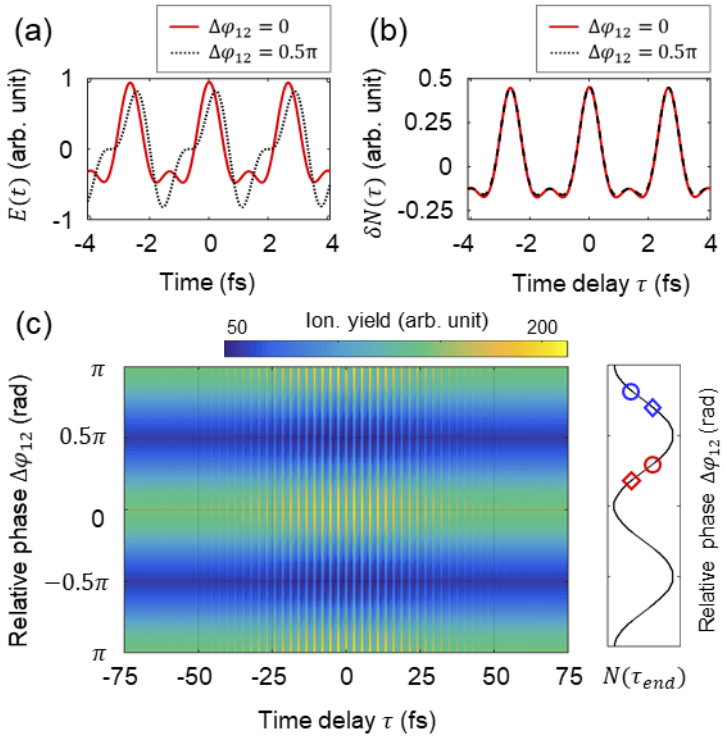

Fig. 1. Ionization yields calculated by using Eq. (**2**). The pulse duration of the fundamental field and the second harmonic field is 41-fs and 85-fs. The field strength ratio of the fundamental field to the second harmonic field is 2. We set the nonlinearity to 6 and the field strength ratio to 0.001. (a) Two-color laser fields at two relative phases 0 (red solid line) and 0.5π (black dotted line). (b) NIY obtained using the two-color laser field at the two relative phases. (c) ionization yield modulation calculated as a function of the time delay and the relative phase. The graph on the right side depicts the ionization yield at the time delay of $\tau = 75fs$.

The ambiguity of the relative phase in the NIY appears due to the convolution effect. One can show that it appears when the bandwidths of the two-color laser field are significantly narrower than the spectral gap of the two-color laser field [22]. This is the case that occurs when the two-color laser field is produced by the superposition of the multi-cycle fundamental laser pulse and its second harmonic pulse. Therefore, a special treatment is required for the temporal characterization of the two-color laser field.

The amount of the ionization yield is calculated using a two-color laser field as a function of time delay at different relative phases as shown in Fig. 1(c). Although the NIY does not correctly represents the relative phase as shown in Fig. 1(a) and 1(b), the ionization yield itself (i.e., $N^{(0)}$) greatly varies depending on the relative phase as shown in Fig. 1(c). The sinusoidal modulation of the ionization yield is clearly shown as the relative phase changes. The ionization yield is modulated because the peak intensity of the two-color laser field of the main pulse is changed as the relative phase changes. Therefore, the relative phase can be determined using the ionization yield measured separately without the signal laser field, or the ionization yield measured at a large time delay.

In order to measure the relative phase using the ionization yield, the relation between the relative phase and the ionization yield should be tested. The ionization yield calculated using

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Eq. (2) shows the maximum value when the relative phase is zero. However, it is an overly simplified equation. Therefore, we performed TDSE calculations in 3D using the model potential of O_2 [26], as shown in Fig. 2. The results indeed confirm that the relative phase and the ionization yield have the simple relation (the maximum ionization yield when the relative phase is zero) for the broad intensity range ($\sim 10^{13} W/cm^2$) at which the TIPTOE measurement is performed. Thus, the relative phase can be determined if the ionization yield is measured for different relative phases.

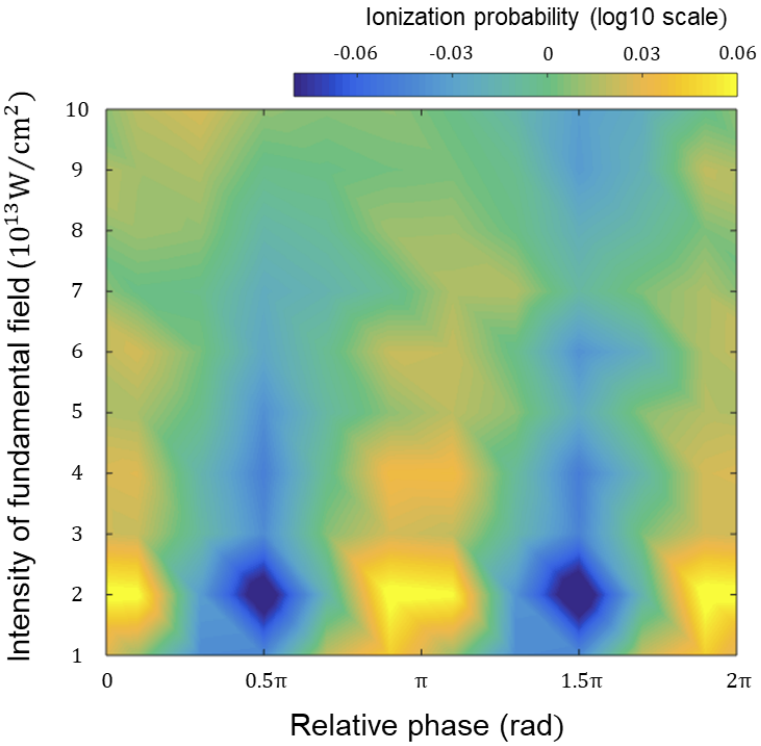

Fig. 2. Ionization probability calculated by solving TDSE. The probability is divided by the mean value which is calculated over the relative phase. The intensity ratio of the fundamental field to the second-harmonic field is 4:1.

3. Measurement results

For an experimental demonstration, an 800-nm 30-fs multi-cycle laser pulse generated from a Ti:sapphire laser system (Femtolasers, Femtopower X CEP4) was used. The second harmonic pulse was generated using a 100-um type-I beta barium borate (BBO) crystal. In order to control the polarization and time delay of the two beam, we used a Mach-Zehnder interferometer. The two beams are separated using a harmonic separator. The polarization of one beam is rotated by 90 deg. to make their polarization parallel. Then, the relative phase of the two beams is controlled by changing the length of the second harmonic beam path with a piezo stage. The two beams are recombined using the other harmonic separator.

The intensity ratio of the two color laser pulse that is required for a robust relative phase measurement is related to the power fluctuation of the two color laser pulse. If the power fluctuation gets higher, the ellipsoid (Fig. 4) becomes thicker, and the relative phase cannot be uniquely determined. When the power fluctuation is 4.1%, the minimum required intensity ratio

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is 0.03. At the entrance of the TIPTOE setup shown in Fig. 2, the intensity of the fundamental beam and the second harmonic beam were $1.2 \times 10^{13}\ W/cm^2$ and $0.32 \times 10^{13}\ W/cm^2$. The intensity ratio was 0.27. The diameter of the two-color beam at the entrance was 6.5 mm.

We used a simple in-line setup for the TIPTOE measurement using an annular mirror and a small inner mirror; the main two-color beam is reflected on the annular outer mirror, and the signal two-color beam is reflected on the small inner mirror, as shown in Fig. 3. The time delay between the two two-color beams is controlled by the inner mirror attached to a piezo transducer. The two beams are focused with a focal length of 1 m. Three electrodes are arranged near the focus. The relative phases at those three positions vary due to the difference in group velocity and the Gouy phase of the two-color components. In order to determine the relative phase using the phase map shown in Fig. 4, the position of the electrode should be set so that their relative phase differ by $\pi/2$ for the adjacent electrode. We placed the electrodes with a separation of 15.5-mm which is the minimum distance that can be set due to the electrode size. At this separation, the difference of the relative phase is estimated to be 1.18π. We observed a reasonably good phase map with this arrangement as shown in Fig. 4. In this way, the ionization yields can be measured with three different relative phases at the same time.

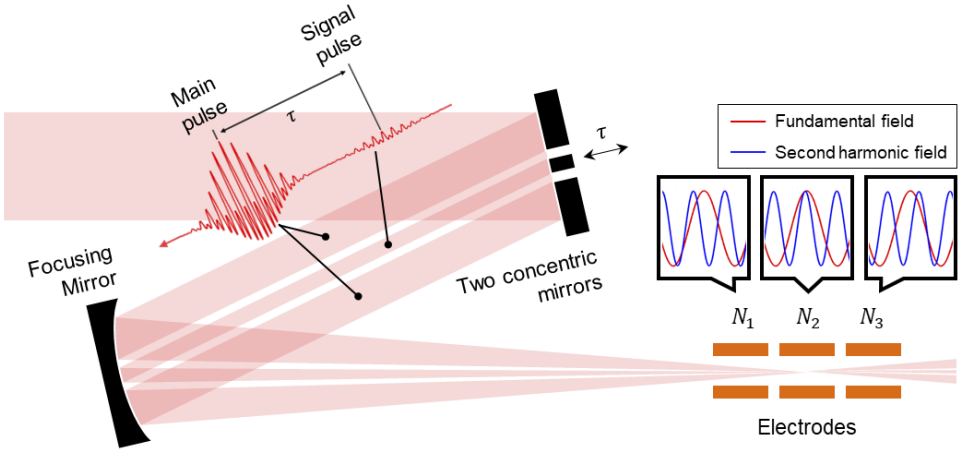

Fig. 3. Schematic diagram of the TIPTOE experiment setup. An incident two-color laser beam is separated into the outer main beam and the inner signal beam using the outer annular mirror and the small inner mirror. The main pulse and the signal pulse with a time delay τ are focused between the three metal electrodes in a row. Ionization yields (N_1, N_2 & N_3) produced by the two-color laser field at different positions are measured. The second electrode (N_2) is positioned at the focus. (Inset) The relative phase between the second harmonic field (blue line) and the fundamental field (red line) varies depending on the position along the propagation axis.

In order to determine the relative phase with a single laser shot, two electrodes would be sufficient. However, we used three electrodes to cancel out the ionization yield modulation caused by the power fluctuation of the laser. As shown in Fig. 1(c), the ionization yield is modulated as the relative phase changes. The ionization yield obtained with a two-color laser field can be written as $N_i^{(0)}(T) \equiv B(T)\{C_i + A_i \cos(2\omega_2 T + \theta_i)\}$ where C_i and A_i are arbitrary positive constants of the i th electrode, $B(T)$ is a noise caused by the power fluctuation of the electric field, T is a relative delay, and θ_i is an additional phase that is the sum of Gouy phase difference and the phase difference made by air dispersion. Note that the ionization yield of all three electrodes has the same noise factor that varies shot-to-shot.

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To eliminate the effect of the shot-to-shot noise and also determine the relative phase, we define the difference of the ionization yields as $N_x \equiv N_2^{(0)} - N_1^{(0)}$ and $N_y \equiv N_2^{(0)} - N_3^{(0)}$. Then, an elliptical relation, $(\frac{X+x_0}{b_x})^2 + (\frac{Y+y_0}{b_y})^2 = 1$ can be obtained. Here, $X \equiv \frac{dN_x+dN_y}{2A_2B(T)}$, $Y \equiv \frac{dN_x-dN_y}{2A_2B(T)}$, ${b_x}^2 \equiv (1 - \frac{A_1}{A_2}\cos\theta)^2$, and ${b_y}^2 \equiv (\frac{A_3}{A_2}\sin\theta)^2$. Also, x_0 and y_0 are arbitrary constants related to the centroid of the ellipsoid. This ellipsoid expression can be used as a fitting function of the experimental data in the relative phase map shown in Fig. 3. The result of the fitting with a least-square method is plotted as a solid black line in Fig. 3, showing a good consistency with the data. From this map, we obtained the relative phase $2\omega_2 T$. The polar angle of the point represents the relative phase of the two-color pulse. Therefore, the relative phase can be determined in this map.

The temporal profile of the two-color laser field except the relative phase can still be measured using the ionization yield modulation obtained from one of the three electrodes [25]. Then, the relative phase of the two-color laser field that is determined in Fig. 4 is taken into account in the reconstruction process. In this way, the temporal profile of the two-color laser field can be completely measured, as shown in Fig. 5(b).

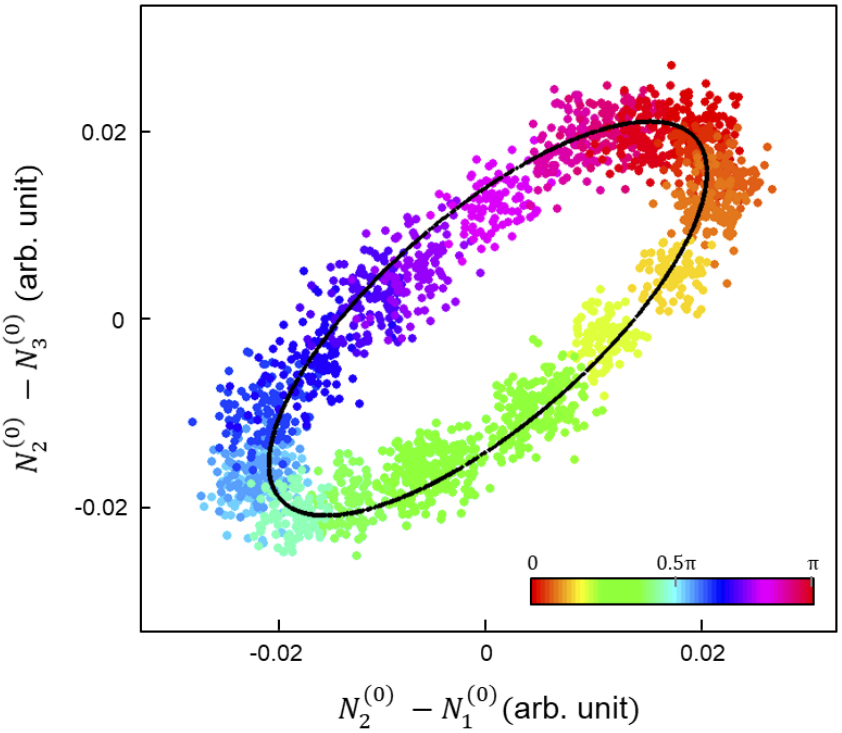

Fig. 4. A relative phase map (dots) with an elliptical fit result (solid black line). The difference of the ionization yields measured at two electrodes is used on each axis. The relative phase varies in a 2π range. Every color uniquely corresponds to the specific relative phase. The standard deviation of the relative phase measurement can be estimated from the distribution of the data taken for a specific relative phase, which is approximately 0.13 rad. We excluded the offset value of the ionization yield in this calculation to make the origin of the ellipsoid be at the center.

To check the validity of the measurement process, we compared the experimental ionization yield modulation and the reconstructed one in Fig. 5(a) and 5(b). The oscillation of the ionization yield at the end of the time delay, and the ionization yield modulation near-zero time delay

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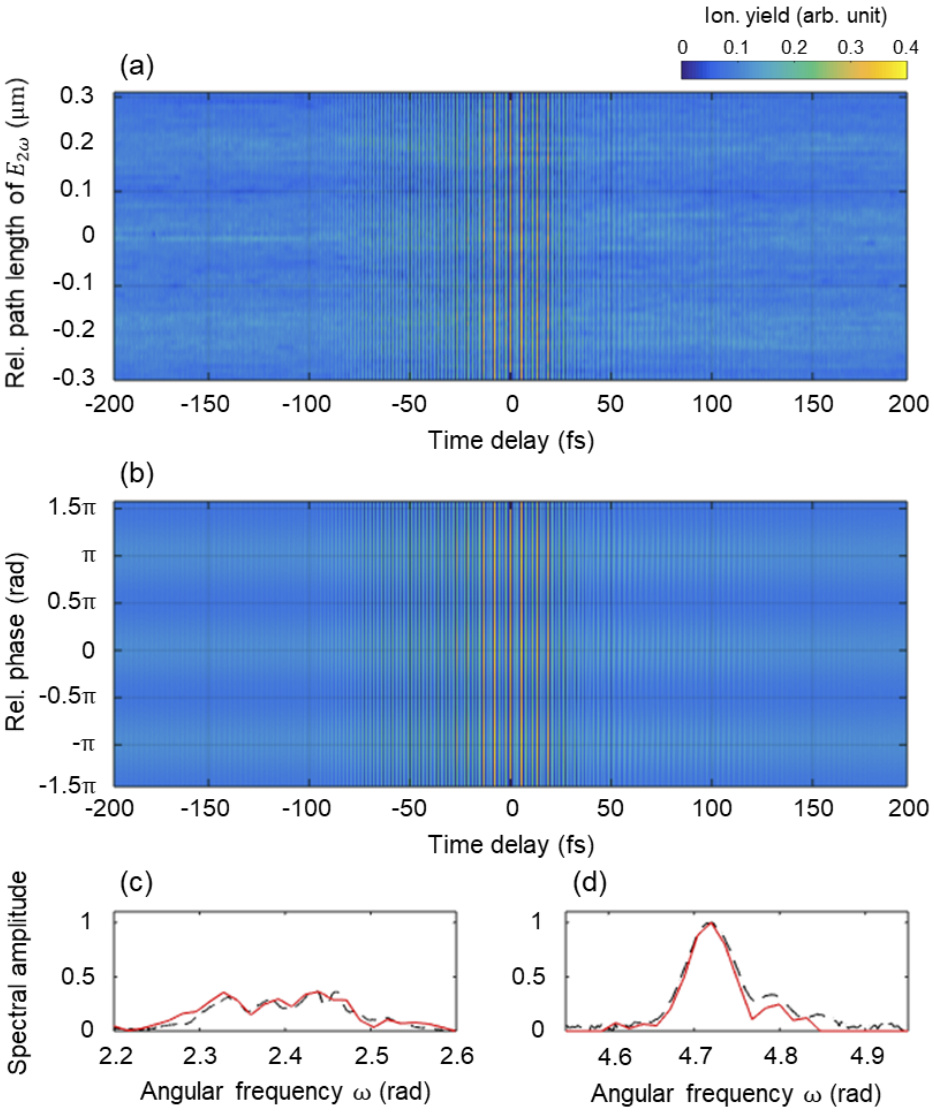

Fig. 5. (a) Experimental ionization yield modulation measured at the focus while scanning the time delay from -200fs to 200fs and the path length of the second harmonic beam from -0.3μm to 0.3μm. (b) Reconstructed ionization yield modulation. (c, d) **S**pectral amplitude of the reconstructed electric field (red solid line) and that measured by the spectrometer (black dotted line).

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are nearly identical in both data. Furthermore, we examined the spectral amplitude of the reconstructed electric field in Fig. 5(c) and 5(d). The reconstructed spectrum (red solid line) showed good consistency with the spectrum measured by the grating-based spectrometer (black dotted line, Ocean Optics, USB4000). These results support the accuracy of the TIPTOE measurement and the validity of the reconstruction process.

The temporal profile of the reconstructed two-color laser field is plotted in Fig. 6(a) for the relative phase of 0.02 rad. The full width at half maximum (FWHM) of the two-color laser field is 49 fs. Also, the temporal profiles are shown for four different relative phases (-1.62 rad, -0.65 rad, 0.02 rad, 0.80 rad) in Fig. 6(b-e). These waveforms show nearly identical waveforms predicted with a calculation (red solid line). These measurements indicate that the two-color laser field is successfully characterized by using the TIPTOE method.

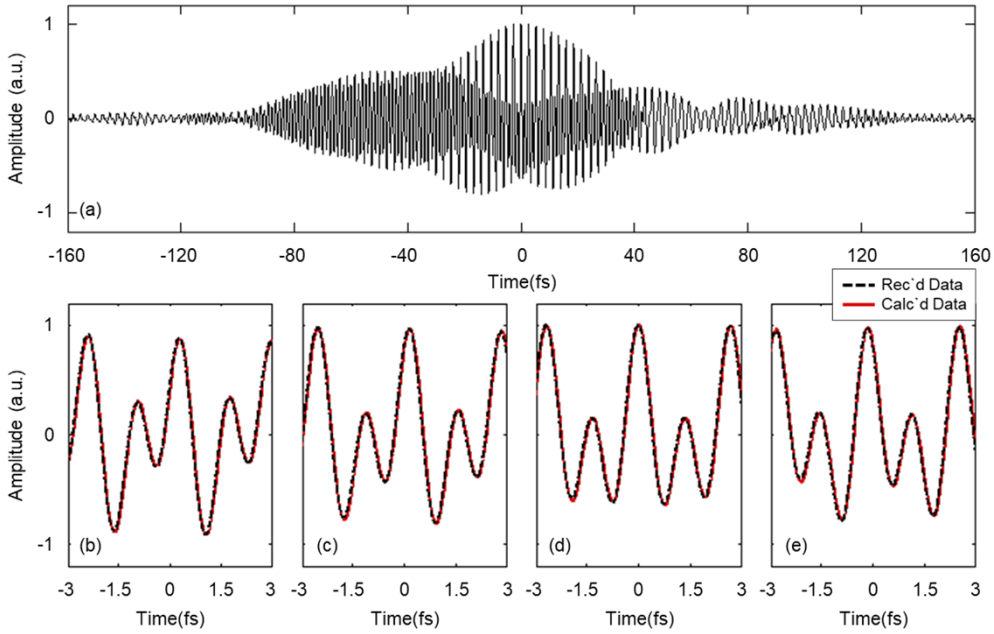

Fig. 6. (a) Temporal profile of the reconstructed two-color laser field. (b-e) A detail**ed** waveform of the reconstructed laser field (black dotted line) compared with the waveform of the calculated laser field (red solid line) at four different relative phases: (b) -0.81 rad, (c) -0.33 rad, (d) -0.01 rad, and (e) 0.40 rad. It is almost identical to the waveform predicted with a calculation. In the calculation, we determined the optical parameters from the measurement and applied those with different relative phases to define the Gaussian pulses. Then, we summed the pulses.

4. Conclusion

We demonstrated the temporal characterization of the two-color laser field using tunneling ionization. We showed that the relative phase of the two-color laser field can be determined by mapping the difference of the ionization yields measured at three different places near the focus. The reconstructed ionization yield shows a good agreement with the theoretical calculations, supporting the accuracy of the reconstruction. The reconstructed spectrum is also consistent with a spectrum measured by a spectrometer. These results support the accuracy and the validity of the TIPTOE measurement on the two-color laser fields.

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The two-color laser fields have been used in many applications [1–5]. It is critical to know the waveform of the two-color laser fields in these applications. The temporal characterization of the two-color laser fields using the TIPTOE technique reveals detailed information about the two-color field including the group delay, chirp structure, and relative phase of the two-color components. Therefore, the accurate temporal characterization of the two-color laser fields will lead to the precise measurement and control of the light-matter interactions.

We have investigated the temporal characterization of the two-color laser field that is consists of the fundamental and its second harmonics. The method that we presented in this work can be generally applied for more general cases in which two color fields has no harmonic relation.

Since we used multi-cycle laser pulses, our measurement is not sensitive to the carrier-envelop-phase of the laser pulse. For few-cycle laser pulses, however, the ionization yield is also dependent on the CEP of the laser pulse [16]. Therefore, our approach can also be used to determine the CEP of the few-cycle laser pulse with a single laser shot.

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Disclosures. The authors declare no potential conflicts of interest.

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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