Temporal Characterization of Ultrashort Pulse via Reflected Four-Wave Mixing with Perturbation on Solid Surface

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Abstract:

Time-domain characterization of ultrashort pulses is essential for studying interactions between light and matter. Here, we propose and demonstrate an all-optical pulse sampling technique based on reflected four-wave mixing with perturbation on a solid surface. In this method, a weak perturbation pulse perturbs the four-wave mixing signal generated by a strong fundamental pulse. The modulation signal of the four-wave mixing, which is detected in the reflection geometry to ensure a perfect phase-matching condition, directly reflects the temporal profile of the perturbation pulse. We successfully characterized multi-cycle and few-cycle pulses using this method. The reliability of our approach was verified by comparing to the widely employed frequency-resolved optical gating (FROG) method. This technique provides a simple and robust method for characterizing ultrashort laser pulses.

Keywords: pulse sampling technique; four-wave mixing; solid surface

1. Introduction

Laser pulses with ultrashort duration are crucial tools for various research fields, including ultrafast spectroscopy^[1], attosecond science^[2-4], nonlinear frequency conversion^[5-7], THz generation^[8, 9], laser-plasma acceleration^[10, 11], etc. Accurate temporal characterization of the ultrashort pulse is indispensable for carrying out such

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applications. Currently, pulse characterization techniques such as frequency-resolved optical gating (FROG)^[12] and D-scan^[13], which are indirect reconstruction method, are widely

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10.1017/hpl.2024.93

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employed for characterizing the pulses based on various nonlinear processes with simple configurations. Alternatively, directly pulse sampling in the time domain with sub-cycle gates formed by tunnel ionization, multiphoton absorption, high-order harmonic generation et.al., allows for the direct capture of the waveforms of the optical pulses. Techniques like the attosecond streak camera^[14], petahertz oscilloscope^[15], photoconductive sampling^[16] and tunneling ionization with a perturbation for the timedomain observation of an electric field (TIPTOE)[17, 18] have been successfully used for pulse measurement. However, these approaches require either complex vacuum system or sophisticated photocurrent measuring devices.

Recently, two novel all-optical pulse sampling techniques have been proposed and successfully demonstrated. These techniques involve the introduction of a perturbation to modulate third-order nonlinear processes, specifically transient grating (TG) in solid plates^[19] and third harmonic generation (THG) in ambient air^[20]. In these approaches, a weak perturbation pulse modulates the output signal of the third-order nonlinear processes initially generated by a strong fundamental pulse. The output modulation signal of these nonlinear processes directly reflects the temporal profile of the perturbation pulse. These methods have been verified by measuring few-cycle pulses across the near-infrared to mid-infrared range. However, the generation of perturbation and fundamental pulses for these two methods are based on Mach-Zehnder interferometer, which require careful alignment. In addition, because the pulses need to pass through several media, which introduce dispersion to the pulses, correction is needed to obtain the spectral phase of the pulses before the measurement setup.

In this study, we present a novel alloptical sampling method for characterizing ultrashort pulses in temporal domain, which employs reflected four-wave mixing (RFWM) with perturbation on the solid surface. Because the RFWM occurs only at the interface between the air and solid plate, this method can characterize ultra-broadband pulses. Additionally, the generation of perturbation and fundamental pulses for this method is realized by division of the wavefront of the input pulse. Thus, the experimental setup is compact, and no correction of spectral phase is required.

2. Theoretical analysis

When an optical pulse incident on a solid plate, third-order nonlinear processes such as third harmonic generation and four-wave mixing occur at sufficient high pulse intensity. The third-order nonlinear signals can be detected in both transmission and reflection geometries. By employing a weaker pulse to perturb the third-order nonlinear process, the measured signal can be calculated by^[21]

$$S(\tau) \propto \int [E_f(t) + E_p(t - \tau)]^6 dt$$

$$= \int [E_f^{-6}(t)dt + \int 6E_f^{-5}(t)E_p(t - \tau)dt + (1)$$

$$\int 15E_f^{-4}(t)E_p^{-2}(t - \tau)dt \cdots$$

 $E_f(t)$ and $E_n(t)$ The are the perturbation fundamental and pulses, respectively. The τ is the relative delay between the two pulses. Here, the $E_p(t)$ has same spectrum as $E_f(t)$, but with substantially lower intensity. Thus, the first term on the right of Eq. (1) is constant, and the other terms except the second term are negligible. Then the modulation signal caused by the weak perturbation pulse can be written as

$$\delta(\tau) \propto 6 \int E_f^{5}(t) E_p(t-\tau) dt$$
 (2)

It has been demonstrated that the $E_f^5(t)$ can be employed as an ultrafast temporal gate to sample the $E_p(t)$ when the $E_f(t)$ is nearly chirp-free^[19,20]. To analyze the influence of the

dispersion of $E_f(t)$ on the measurement, the Eq. (2) is Fourier transformed as

$$\delta(\omega) \propto \mathcal{F}\left[E_f^{5}(t)\right]^* E_p(\omega)$$
 (3)

where $\delta(\omega)$ and $E_p(\omega)$ are the spectra of $\delta(\tau)$ and $E_p(t)$, respectively. The $\mathcal{F}[E_{\ell}^5(t)]$

is the Fourier Transform of $E_f^5(t)$. Because of the finite bandwidth of $E_f^5(t)$, the bandwidth of $\delta(\omega)$ is slightly narrower than the $E_p(\omega)$. Meanwhile, the spectral phase of $E_f^5(t)$ can also introduce error to the retrieved spectral phase of the $E_p(\omega)$ if $E_f(t)$ is chirped. Thus, there is discrepancy between $\delta(\omega)$ and

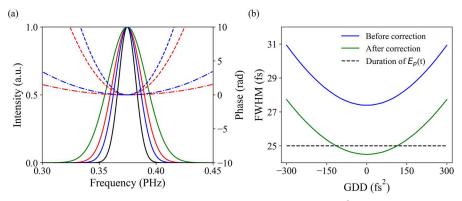


Figure 1. (a) The spectra (solid lines) and spectral phases (dash-dot lines) of E_f^5 (t) with different dispersions: GDD = 200 fs² (red) and GDD = 300 fs² (blue). For comparison, the spectral phases (dash lines) of E_f (t) were also plotted for GDD = 200 fs² (red) and 300 fs² (blue). The black and green solid lines correspond to the spectra of E_f (t) and E_f^5 (t) without chirp. (b) Measured pulse duration before (blue) and after (green) correction of spectrum for different GDD.

 $E_p(\omega)$. Nevertheless, the narrowing of the bandwidth can be partially corrected in a simple way. We can assume the $E_f(t)$ is chirp-free and the spectrum shape of $E_f(t)$ is identical to $\delta(\tau)$. Thus, the $\mathcal{F}[E_f^5(t)]$ can be easily calculated and then the $E_p(\omega)$ is approximated by

$$E_p(\omega) \approx \frac{\delta(\omega)}{\mathcal{F}[E_f^{5}(t)]^*}$$
 (4)

Fig. 1(a) shows the influence of GDD on the $E_f^5(t)$. The femtosecond pulses centered at 800 nm are taken as an example. The Transform limited (TL) pulse duration is 25 fs, which is typical for Ti:sapphire laser pulse^[22]. Compared to $E_f(t)$, the spectra of $E_f^5(t)$ are wider and the spectral phases of $E_f^5(t)$ are flatter. With bigger GDD, the bandwidth of $E_f^5(t)$ is smaller, and the dispersion of $E_f^5(t)$ is bigger, which is not preferrable for the measurement. The measured pulse durations of

25 fs pulse with different GDD for $E_f(t)$ is showed in Fig. 1(b). It is obvious that the error of the measurement is bigger with more chirped $E_f(t)$ if the bandwidth of $E_n(t)$ is not corrected. After correction of the spectra, the measurement error is clearly mitigated, and the error is less than 1 fs (4% of the original pulse duration) when the absolute value of GDD for $E_p(t)$ is not more than 200 fs². The $E_p(t)$ is stretched to ~33 fs with GDD of 200 fs², which is about 1.32 times of the TL pulse duration. To ensure accurate measurement in the experiment presented here, we tuned the fundamental pulse to near chirp-free by observing the filamentation in the air after tight focusing.

In this work, we choose to measure the modulation signal of the four-wave mixing on a solid surface in the reflection geometry for temporally sampling the ultrashort pulses. The phase-matching is ensured in the reflection geometry, which allows to characterize pulses

with broad bandwidth. The RFWM signal from the surface can be simply expressed as:

$$P_{RFWM}(t,\tau) \propto \left| E_f(t) \right|^2 E_f(t) + 3 \left| E_f(t) \right|^2 E_p(t-\tau) + 3 E_f^2(t) E_p^*(t-\tau) + 3 E_f(t) \left| E_p(t-\tau) \right|^2 + (5)$$

$$3 E_f(t)^* E_p^2(t-\tau) + \left| E_p(t-\tau) \right|^2 E_p(t-\tau)$$

The first three terms are detected to obtain the modulation signal of the four-wave mixing, while the last three terms are negligible. The first three terms are non-collinear according to the phase-matching condition [Fig. 2(a)]. Because the third-order nonlinear processes are high intensity dependent, the beam spots of

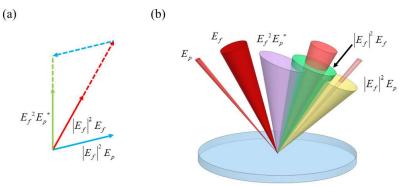


Figure 2. (a) The wave-vectors of the first three terms of Eq. (5). (b) RFWM with perturbation on the surface of a solid plate. The red and light red beams represent the fundamental pulse and perturbation pulse, respectively. Three beams generated by four-wave mixing have larger divergence angles, which are green, yellow and purple corresponding to the first three terms on the right of Eq. (5). The incident angle between fundamental and perturbation pulses is exaggerated for clarity.

the four-wave mixing are much smaller than the $E_f(t)$ and $E_p(t)$ on the surface. Thus, they are more divergent as showed by the cones in Fig. 2(b).

3. Experimental setup and results analysis

The ultrashort pulse is split into two pulses by mask1, i.e., the fundamental pulse and perturbation pulse. The diameters of the two holes in mask1 are 6 mm and 1 mm, respectively. The distance between the centers of the holes is around 4.5 mm. Because the beam profile was close to flat-top. We assumed that the energy ratio between the fundamental pulse and the perturbation pulse is about 36. It

well-known that second-harmonic generation (SHG) can occur when a laser pulse is obliquely incident on the surface of a fused silica plate^[23]. In our experiment, to eliminate the influence of surface SHG, two pulses were focused and overlapped on the front surface of a thin fused silica plate (100 µm thick) nearnormal incidence using a concave mirror with a focal length of 200 mm. Under these conditions, no SHG was detected. The peak intensity of the perturbation pulse is about 0.077% of the fundamental pulse. A lens is used for imaging the RFWM signal to a detector for collecting the RFWM, which can either be a photodiode, a camera, or a spectrometer. In this work, the spectrometers were used in the following experiments.

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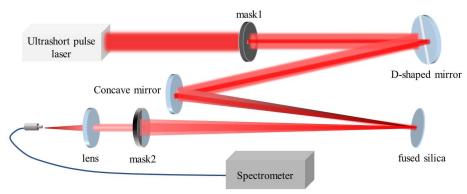


Figure 3. Schematic of the experimental setup. Mask1 is used to split the input pulse into fundamental pulse and perturbation pulse. The two pulses are focused by a concave mirror onto the fused silica surface to generate RFWM signal. The angle between the fundamental and perturbation pulses is exaggerated for clarity. The relative delay between the two pulses is controlled by the D-shape mirror. The RFWM signal is coupled into the detector by a lens after passing through mask2.

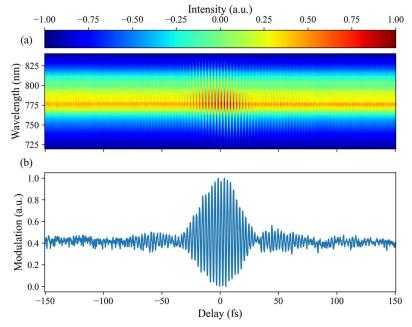


Figure 4. (a) Evolution of the RFWM signal measured by a spectrometer with different relative delays between fundamental and perturbation pulses. (b) The modulation signal of the RFWM obtained by integrating (a) along the wavelength axis.

Before the lens, a second mask (mask2) is employed to block the $E_f(t)$ and $E_p(t)$. The delay between the fundamental pulse and the perturbation pulse was tuned with a D-shaped mirror mounted on a piezo-linear-stage (Physik Instrumente P-621.1) with step size of 20 nm. The scanning range was set to 100 µm.

Firstly, we measured the ultrashort pulse with center wavelength of 800 nm delivered

from a Ti:sapphire laser. The pulse energy before the mask1 was $\sim 7.6~\mu J$. The RFWM signal with different delay was measured by a spectrometer (Ocean Insight Maya2000pro) and showed in Fig. 4(a). It is obvious that the fringes emerge only when the two pulses are overlapped temporally. Then we integrate and normalize the RFWM signal along the wavelength dimension to obtain the

modulation signal of the RFWM, as shown in Fig. 4(b).

The noise in Fig. 4(b), which is visible before -100 fs and after 100 fs, is resulted from the fluctuation of pulse energy and beam pointing. It can be removed by filtering in the frequency domain. By employing a rectangular window ranging from 700 nm to 900 nm, the noise can be effectively suppressed [see Fig. 5(a, c, e)]. In the following work, we have employed the same way to remove such noise. As shown in Fig. 5(a), the measured result shows that the pulse duration is ~27 fs. The spectrum and spectral phase of the ultrashort pulse were obtained after the Fourier transform of the waveform, as shown in Fig. 5(b). To

verify the reliability of this technique, the retrieved spectrum and spectral phase are compared to that obtain with TG-FROG. The results of RFWM-sampling agree well to the TG-FROG measurement. To further verify the reliability of the method, we introduced dispersion to the pulses by inserting or removing a piece of 1.3-mm-thick SF11 plate in the perturbation beam path. The measured results through RFWM-sampling of positive chirped and negative chirped pulses are showed in Fig. 5(c, e), respectively. The spectra and spectral phases can be seen in Fig. 5(d, f). The chirped pulses were also characterized by TG-FROG and it confirms the

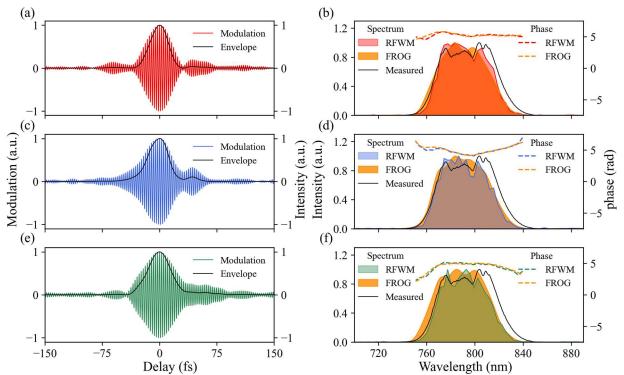


Figure 5. Temporal sampling of 800 nm pulses by RFWM with perturbation. (a), (c), (e) Measured RFWM modulation signal and the envelopes of chirp-free, positively chirped and negatively chirped pulses, respectively. (b), (d), (f) Comparison of the spectra and spectral phases retrieved by this method and TG-FROG. The black solid lines illustrate the spectra measured by spectrometer.

reliability of RFWM-sampling technique for measuring near-infrared ultrashort pulses [see Fig. 5(d, f)]. In addition, the spectra measured with spectrometer [solid black line in Fig. 5(b, d, f)] are presented for reference as well.

Afterwards, the RFWM-sampling technique was also applied to pulses generated by a TOPAS from Light Conversion with the center wavelength of 1700 nm. After filtering in the frequency domain, the modulation signal

is showed in Fig. 6(a), and the pulse duration is ~48 fs. We measured the chirped pulses with RFWM-sampling method and the RFWM modulation signal and envelopes are showed in Fig. 6(c, e). The positive chirp and negative chirp were introduced by inserting and removing a piece of 1-mm-thick ZnSe plate in the perturbation beam path, respectively. After Fourier transform of the modulation signal, the spectra and spectral phases are retrieved and presented in Fig. 6(d, f) accordingly. The spectra and spectral phases were also measured by SHG-FROG. Meanwhile, the spectra were independently measured by the spectrometer as shown in solid black lines of Fig. 6(b, d, f). The retrieved spectra by RFWM-sampling are

consistent with the spectra obtained by SHG-FROG and spectrometer. The retrieved phases by RFWM-sampling are almost identical to the phases obtained by SHG-FROG.

Finally, we employed the RFWM-sampling method to measure even shorter pulses of few optical cycles, which was obtained by the multiple-thin-plate compression technique to further compress the output pulse from a Ti:sapphire laser. The measured pulse is showed in Fig. 7(a). The pulse duration was around 12 fs (FWHM), which was longer than its TL duration of ~9 fs (FWHM) as showed by the black solid line in Fig. 7(a). The retrieved spectrum (blue solid line) and spectral phase (red solid line) are

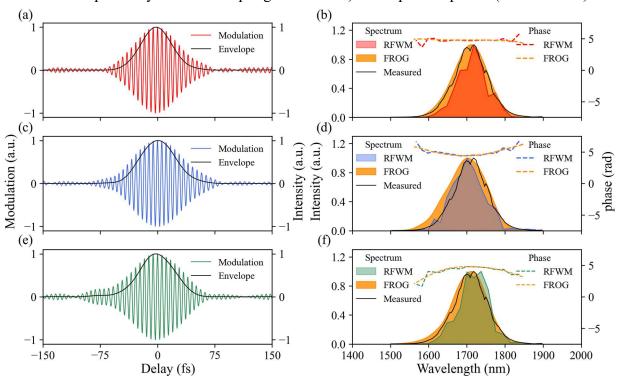


Figure 6. Temporal sampling of pulses at 1700 nm by RFWM with perturbation. (a), (c), (e) Measured RFWM modulation signal and the envelopes of chirp-free, positively chirped and negatively chirped pulses. (b), (d), (f) Comparison of the spectra and spectral phases retrieved by the RFWM with perturbation method and SHG-FROG. The black solid lines illustrate the spectra measured by spectrometer.

illustrated in Fig. 7(b). It is visible that there was residual dispersion as showed by the spectral phase, which was responsible for the expansion of the pulse duration. The spectrum was simultaneously measured by the spectrometer (orange solid line), which is

consistent with RFWM-sampling method. Because the RFWM occurs at the air—dielectric interface, the requirement of phase-matching is readily fulfilled^[24]. Thus, this method can be used to characterize ultra-broadband pulses.

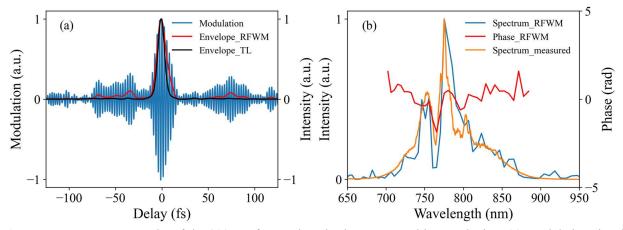


Figure 7. Measurement results of the 800 nm few-cycle pulse by RFWM with perturbation. (a) Modulation signal (blue solid line) and envelope (red solid line) of the few-cycle pulse. The envelope of the TL pulse is showed by black solid line. (b) Retrieved spectrum (blue solid line) and spectral phase (red solid line). The orange solid line illustrates the spectrum measured by spectrometer.

4. Conclusions

In conclusion, we propose a novel all-optical temporal sampling method for ultrashort pulses measurement. This method utilizes the four-wave mixing process with perturbation on solid surfaces. By capturing the RFWM signal from the surface of a 100 µm thin fused silica plate, we have successfully characterized femtosecond pulses with central wavelengths of 800 nm and 1700 nm. The reliability of this method was verified by comparing to the FROG measurement. The use of a reflection geometry in our measurement scheme allows for measurement of ultrashort pulses as the phase-matching condition is easily satisfied. As a result, the measurement of few-cycle pulse at 800 nm was demonstrated as well. It is noted that this technique is not suitable for characterization of pulses with severe spatiotemporal coupling. It is necessary to correct the spatiotemporal coupling, instance, with spatial filters, before the application of this technique. In this work, only 800 nm and 1700 nm pulses are measured, but based on the current silicon and InGaAs detectors [Thorlabs DET10D2], this method can measure the pulses spanning from 200 nm to 2600 nm with pulse energy of microjoule level. Because this method relies on the scanning of the delay between two pulses, stable pulse energy is required to obtain In measurement. addition. demonstrated in the experiment, the influence of energy fluctuation can be mitigated by filtering in the frequency domain. characterize pulses with wavelength longer than 2600 nm, the FWM signal cannot be measured with conventional detectors. Thus, other third-order nonlinear processes, i.e. third-order harmonic generation (THG) can be employed. For instance, to measure the pulse with wavelength as long as 7800 nm, the wavelength of THG signal is 2600 nm, which can be covered by InGaAs detectors. We believe this method can serve as a reliable tool for the characterization of ultrashort pulses.

Acknowledgments

This work was supported by National Key R&D Program of China (2022YFE0111500); National Natural Science Foundation of China (NSFC) (Nos. 62175256, 12104501, 92050107, 92250303); Natural Science Basic Research Program of Shaanxi (No. 2019JCW-03); Science and Technology Program of Xi'an (No. 202005YK01); The Young Scientist Basic Research Program of the Chinese

Academy of Sciences (YSBR-091); Key Deployment Research Program of XIOPM (No. S19-020-III); Innovation team in Shaanxi Province (No. J21-029-III); Youth Innovation Promotion Association XIOPM-CAS (No. WCH2021002).

Disclosures

The authors declare no conflicts of interest.

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