

Accurate determination of the absolute phase and temporal-pulse phase of few-cycle laser pulses*

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A Fourier analysis method is used to accurately determine not only the absolute phase but also the temporal-pulse phase of an isolated few-cycle (chirped) laser pulse. This method is independent of the pulse shape and can fully characterize the light wave even though only a few samples per optical cycle are available. It paves the way for investigating the absolute phase-dependent extreme nonlinear optics, and the evolutions of the absolute phase and the temporal-pulse phase of few-cycle laser pulses.

Keywords: absolute phase, temporal-pulse phase, few-cycle laser pulse, Fourier analysis

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1. Introduction

For a few-cycle laser pulse, the carrier-envelope phase Φ_{CE} (defined as the phase of the optical carrier with respect to the pulse peak^[1,2]), i.e. the absolute phase, plays an important role in a large number of extreme nonlinear optical techniques, such as high-order harmonic generation,^[1,3] photoionization,^[2,4,5] photo emission,^[6] synthesizing of pulses^[7] and nonperturbative resonant nonlinear process in a semiconductor,^[8] etc. On the other hand, there has been aroused much interest in the study of the evolutions of the temporal-pulse phase^[9] and the absolute phase^[10] in extreme nonlinear processes. Nielsen *et al*^[9] have experimentally investigated the temporal phase evolution of ultrashort solitonlike optical pulses in a semiconductor by using a fast-scan cross-correlation frequency-resolved optical gating (XFROG) method and also numerically calculated the above based on the combined semiconductor Maxwell-Bloch equations in the slowly varying envelope approximation (SVEA) and the rotating-wave approximation (RWA). In consequence, the temporal phase shows a phase jump of almost π between both pulse components breaking from the excitation-dependent temporal pulse due to Rabi flopping of the carrier density. Very recently, we^[10]

have found that the absolute phase of few-cycle laser pulse dramatically changes due to the near dipole-dipole interactions during propagation in a dense two-level medium.

Since phase stabilization approaches were presented to the generation of high-intensity few-cycle optical pulses with precisely reproducible electric and magnetic fields,^[11,12] a great number of methods of determining the carrier-envelope phase and pulse-to-pulse phase $\Delta\theta$ have been put forward.^[2,3,5,6,8,12-24] By fitting the fringe peaks of the interferogram to a correlation function, with a special pulse envelope profile assumed, Jones *et al*^[12] have presented an f -to- $2f$ scheme to find the pulse-to-pulse phase $\Delta\theta$. However, the results show an offset of 0.7 ± 0.35 rad from the theoretically expected relation $\Delta\theta = 2\pi\delta/f_{rep}$, where f_{rep} is the repetition rate of the pulse train. Stereo above-threshold ionization (Stereo-ATI) apparatus is suitable for measuring the absolute phase. In this apparatus, atoms are ionized by an ultrashort laser pulse, and the photoelectrons are recorded with two opposite detectors in a plane perpendicular to the laser beam. The shot-to-shot analysis of the electron yield shows an anticorrelation.^[2] By exploring the directional photoelectron asymmetries, both numerical calculation^[15,16] and experimental investigation^[17]

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have been conducted to measure the absolute phase. A complex Fourier transform method has been presented, which can be extended to the determination of the temporal-pulse phase subject to neglecting the value of the absolute phase, to make analysis of special fringe-pattern for topography and interferometry in hopes of retrieving the relative phase distribution, with the initial phase value set to zero.^[25] Moreover, the frequency-resolved optical gating (FROG) has been well documented for retrieving the spectral phase^[26,27] and the phase variation of ultrashort pulse with an ambiguity of the initial phase of the pulse.^[28] Numerical investigation of characterizing femtosecond optical pulses based on the spectral phase interferometry for direct electric-field reconstruction (SPIDER) has been reported by Chai *et al.*^[29] Recently, the technologies of directly measuring the electric field of few-cycle laser pulses have been developed.^[16,17,30] These technologies open the door to using the present method to retrieve the absolute phase and the temporal-pulse phase.

Here, based on Fourier analysis of some discrete samples in the duration of full-wave pulse, we present a new method of retrieving the absolute phase and the temporal-pulse phase of an isolated few-cycle pulse. To our best knowledge, it is the first time this method has been used to accurately determine not only the absolute phase but also the temporal-pulse phase and then the chirp of few-cycle laser pulse. In this method it is not necessary to assume the envelope to be a certain profile, e.g. the hyperbolic-secant (h.s.) function, etc. The retrieved field fits the sample data better if the discrete envelope retrieved from samples is employed. Furthermore, if the sampling frequency is larger than twice the cutoff frequency of the pulse, i.e. the sampling frequency satisfies the Nyquist sampling theorem,^[31] the absolute phase can be determined. So with that, only a few samples are required to be taken from the experiment. This method breaks a new promising path for investigating the phase-dependent extreme nonlinear optics and determining not only the absolute phase but also the temporal-pulse phase of few-cycle laser pulse numerically and experimentally.

The rest of this paper is organized as follows. In Section 2, we present the principle of a Fourier analysis method of retrieving the absolute phase and the temporal-pulse phase. Then, in Section 3, two examples are given: one is for two ideal chirped pulses and the other is for one pulse with a much weaker daughter pulse obtained in experiment.^[30] A summary is given

in finally Section 4.

2. Principle and algorithm

In order to demonstrate how to determine the absolute phase Φ_{CE} and the temporal-pulse phase from some samples in the duration of the field in principle, we employ an ideal pulse $E(t) = A(t - \tau) \cos[\omega_c(t - \tau) + \Phi(t)]$, where $A(t - \tau)$ is the real and positive envelope as usual, ω_c is the carrier frequency, τ is the group delay time and $\Phi(t)$ the temporal-pulse phase. In general, $\Phi(t)$ varies slowly in comparison with the variation induced by the carrier frequency ω_c . We arbitrarily sample the field $E(t)$ in a constant time step and then show how we can reconstruct the field and determine the absolute phase just from these samples. The discrete fast Fourier transform (FFT) of these samples gives two parts $\tilde{E}_+(\omega)$ and $\tilde{E}_-(\omega)$ corresponding to the positive and negative frequency components of the spectrum, respectively. If there is more than one complete optical cycle in the envelope, the spectral overlap between $\tilde{E}_+(\omega)$ and $\tilde{E}_-(\omega)$ is negligible. Thus a complex field $\tilde{E}_+(t)$, which, in fact, is a set of discrete values of $\tilde{E}_+(i)$ with i being the index, can be retrieved from a few samples by the inverse discrete fast Fourier transform of $\tilde{E}_+(\omega)$. Obviously, the retrieved envelope is $2|\tilde{E}_+(t)|$. The absolute phase depends on the determination of the central frequency and the group delay time. Following Diels *et al.*^[32] the central frequency is evaluated by

$$\tilde{\omega}_p = \frac{\int_{-\infty}^{+\infty} \omega \left| \tilde{E}_+(\omega) \right|^2 d\omega}{\int_{-\infty}^{+\infty} \left| \tilde{E}_+(\omega) \right|^2 d\omega}. \quad (1)$$

Similarly, the group delay time is determined by the first moment as follows:

$$\tilde{\tau} = \frac{\int_{-\infty}^{+\infty} t \left| \tilde{E}_+(t) \right|^2 dt}{\int_{-\infty}^{+\infty} \left| \tilde{E}_+(t) \right|^2 dt}. \quad (2)$$

The values of $\tilde{\omega}_p$ and $\tilde{\tau}$ are achieved by using the discrete FFT. The peak of envelope is $2 \left| \tilde{E}_+(i_{\tilde{\tau}}) \right|$, where $i_{\tilde{\tau}}$ is the index corresponding to the predicted delay time $\tilde{\tau}$. $4 \left| \tilde{E}_+(i_{\tilde{\tau}}) \right|^2$ is considered as the maximum of the intensity and the duration $\tilde{\tau}_p$ is the FWHM (full width at half-maximum) of $4 \left| \tilde{E}_+(i) \right|^2$. The key point

is to retrieve the absolute phase and the temporal-pulse phase from $\tilde{E}_+(i)$. As defined, the absolute phase $\tilde{\Phi}_{CE}$ is the value of $\tilde{\Phi}(t)$ at the instant of $t = \tau$.

$$\tilde{\Phi}_{CE} = [\text{unwrap}(\text{angle}(\tilde{E}_+(i))) - \tilde{\omega}_p(i\Delta t - \tilde{\tau})]_{i=i_{\tilde{\tau}}} + 2m\pi, \quad (3)$$

$$\tilde{\Phi}_{\text{off}} = [\text{unwrap}(\text{angle}(\tilde{E}_+(i))) - \tilde{\omega}_p(i\Delta t - \tilde{\tau})]_{i=i_{\tilde{\tau}}} - \tilde{\Phi}_{CE}, \quad (4)$$

$$\tilde{\Phi}(i) = \text{unwrap}(\text{angle}(\tilde{E}_+(i))) - \tilde{\omega}_p(i\Delta t - \tilde{\tau}) - \tilde{\Phi}_{\text{off}}, \quad (5)$$

where Δt is the sampling time step and m denotes any integer. The function *angle* returns to the phase angles in a range of $(-\pi, \pi]$ for each element of complex field $\tilde{E}_+(i)$; *unwrap* corrects the radian phase angle of a series of complex values of $\tilde{E}_+(i)$ by adding multiples of $\pm 2\pi$ when the absolute jumps between consecutive elements of $\tilde{E}_+(i)$ are greater than the default jump tolerances of π radian. The absolute phase $\tilde{\Phi}_{CE}$ is adjusted into the range $(-\pi, \pi]$ by adding multiples of $\pm 2\pi$. In addition, the absolute phase $\tilde{\Phi}_{CE}$ can be retrieved just based on a few samples per optical cycle even if no sample is available exactly at the instant of $t = \tilde{\tau}$ because the expression (3) diminishes the error $\tilde{\omega}_p(i_{\tilde{\tau}}\Delta t - \tilde{\tau})$ caused by the deviation of $i_{\tilde{\tau}}\Delta t$ from $\tilde{\tau}$. The temporal-pulse phase $\tilde{\Phi}(i)$ should be adjusted by an offset $\tilde{\Phi}_{\text{off}}$ because the instant of the first element $\tilde{E}_+(0)$ is not $t = \tilde{\tau}$. On condition that there is more than one complete optical cycle in the envelope, the overlap between the spectra $\tilde{E}_+(\omega)$ and $\tilde{E}_-(\omega)$ normalized by the maximum of $|\tilde{E}_+(\omega)|$ is less than 0.01, thus it is negligible. Then the complex field $\tilde{E}_+(i)$ is very close to the positive component of the field $E(t)$, and the $\text{unwrap}(\text{angle}(\tilde{E}_+(i))) - \tilde{\Phi}_{\text{off}}$ gives the time-dependent angle $\theta(i\Delta t)$ of the complex field $\tilde{E}_+(i)$ as a good approximation of the phase $\omega_c(i\Delta t - \tau) + \Phi(i\Delta t)$. According to the expressions (3)-(5), the retrieved temporal-pulse phase $\tilde{\Phi}(i)$ is the angle $\theta(i\Delta t) - \omega_p(i\Delta t - \tilde{\tau})$, which approximately equals the phase $\Phi(i\Delta t)$, and $\tilde{\Phi}_{CE}$ is close to the angle $\Phi(\tilde{\tau})$, i.e. the value of the temporal-pulse phase $\Phi(t)$ at the instant of $t = \tilde{\tau}$. Though there exist small errors, in principle, the temporal-pulse phase $\tilde{\Phi}(i)$ and the absolute phase $\tilde{\Phi}_{CE}$ retrieved by the expressions (3)-(5) are the values of $\Phi(t)$ at $t = i\Delta t$ and Φ_{CE} , respectively. It can be seen from the above analysis that we can fully characterize a few-cycle laser pulse from some samples in the duration of the field even though it is a chirped pulse.

For a chirped pulse without noise, the error in the

Here, we retrieve the absolute phase $\tilde{\Phi}_{CE}$ and the temporal-pulse phase $\tilde{\Phi}(t)$, which takes the form of a series of discrete values of $\tilde{\Phi}(i)$ given by

determined absolute phase induced by three factors: the error in the retrieved group delay, the deviation of the retrieved central frequency from the carrier frequency, and the spectral overlap between $\tilde{E}_+(\omega)$ and $\tilde{E}_-(\omega)$. As for the central frequency $\tilde{\omega}_p$, it equals the value $\omega_c + \langle \dot{\Phi} \rangle$, where $\langle \dot{\Phi} \rangle$ is the average of the chirp $\dot{\Phi}(t)$ over the pulse.^[32] According to the expressions (3)-(5), the error in the temporal-pulse phase follows $\Delta\tilde{\Phi}(i) = \left| \omega_c(\tilde{\tau} - \tau) - \langle \dot{\Phi} \rangle (i\Delta t - \tilde{\tau}) \right| \approx \left| \langle \dot{\Phi} \rangle (i\Delta t - \tilde{\tau}) \right|$. It is less than $\left| \langle \dot{\Phi} \rangle \right| \tau_p$ in the double duration of the pulse $2\tau_p$. However, the absolute phase, i.e. the temporal-pulse phase at $i\Delta t \approx \tilde{\tau}$, can be accurately determined. Further investigation shows that the deviation of the absolute phase is mainly determined by the spectral overlap normalized by the maximum of $|\tilde{E}_+(\omega)|$.

3. Numerical calculation and experimental investigation

In the present example, two chirped few-cycle pulses with different envelopes $E_1(t) = E_0 \text{sech}[1.76(t - \tau)/\tau_p] \cos[\omega_c(t - \tau) + 0.5\chi(t - \tau)^2 + \Phi_0]$ and $E_2(t) = E_0 \text{sech}[-1.39(t - \tau)^2/\tau_p^2] \cos[\omega_c(t - \tau) + 0.5\chi(t - \tau)^2 + \Phi_0]$, where $E_0 = 1 \text{ V/m}$, $\tau = 10 \text{ fs}$, $\omega_c = 2.3 \text{ fs}^{-1}$, $\chi = 0.03 \text{ fs}^{-2}$, $\Phi_0 = 1.0 \text{ rad}$ and $\tau_p = 5 \text{ fs}$ (the FWHM of the intensity), are randomly sampled in an identical time step. The sampling frequency is near five times the central frequency, i.e. the sampling time step Δt is 0.546 fs. Only based on samples in the duration of the fields can we well determine all parameters and retrieve the two envelopes, which take the same form $2|\tilde{E}_+(i)|$ but different values. According to the expression (3), the retrieved absolute phases, which are both 1.0 rad, agree very well with the initial phase Φ_0 , i.e. the absolute phase. Other retrieved parameters are $\tilde{\tau} = 10.0 \text{ fs}$, $\tilde{\tau}_p = 4.9 \text{ fs}$ and $\tilde{\omega}_p = 2.3 \text{ fs}^{-1}$ for both cases. Though the envelopes

of the two pulses are different, we can well fit the two pulses by $E(i) = 2 \left| \tilde{E}_+(i) \right| \cos[2.3(i\Delta t - 10.0) + \tilde{\Phi}(i)]$ just based on the retrieved envelopes and parameters. Figure 1(a) shows the fitted curves and the data curves plotted according to the samples. The thick black line corresponds to the h.s. envelope case, while the thick dashed one is for the Gaussian envelope case. Figure 1(b) indicates that the temporal-pulse phases $\tilde{\Phi}(i)$ retrieved by the expression (5) for both cases agree well with the phase $0.5\chi(t - \tau)^2 + \Phi_0$ over the pulses. It

can be clearly seen from the inset plot in Fig.1(b) that the chirp rates are both 0.03 fs^{-2} . Evidently, this method is independent of the pulse shape and does retrieve the absolute phase and the temporal-pulse phase. Moreover, the absolute phases of few-cycle laser pulses with noise, which are a series of discrete real values obtained by numerical simulation, have been determined.^[10] So this method is insensitive to noise.

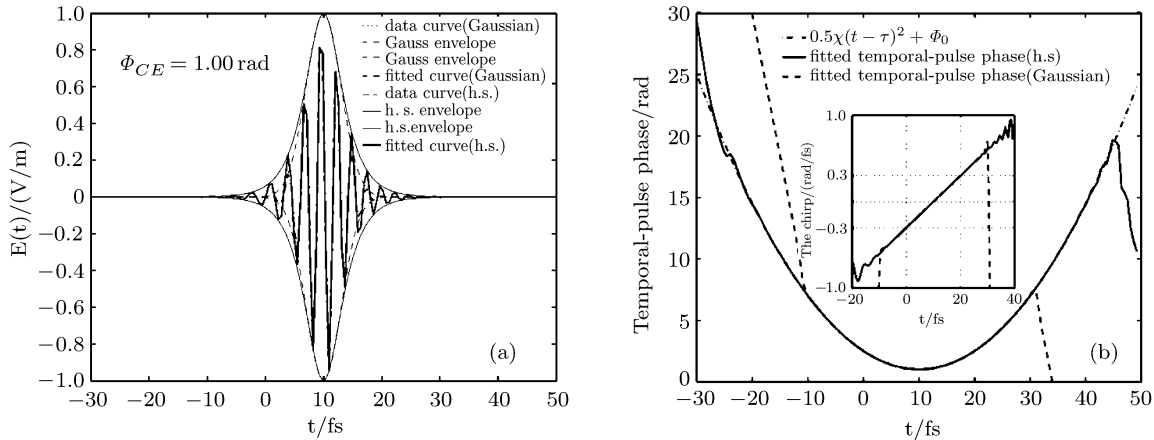


Fig.1. Determination of the absolute phase and the chirp of an ideal 5 fs chirped few-cycle pulse. (a) The fitted curves and the data curve. (b) The temporal-pulse phases. The inset lines in (b) are for the chirp of the pulse.

The fewer the samples are required to be taken from the experiment, the easier the work. Goulielmakis *et al.*^[30] have directly measured the few-cycle laser pulse in time domain in experiment. Then they reconstructed the light wave (red line in Fig.3 in Ref.[30]) from tens (more than ten per optical cycle) of discrete data of kinetic energy spectra of electrons and marked three samples per optical cycle by short bars. Moreover, from the measured spectrum of few-cycle pulse, they have fitted the field (the thin grey line in Fig.3 in Ref.[30]) by Fourier transform under the assumption of the absence of spectral phase variations, with the amplitude and the absolute phase chosen to yield the best match to the measured values. However, it is valid only if the group delay time is negligible. On the other hand, it is hard to directly indicate the absolute phase even in the case where the shape of the field has been obtained, especially for the case of only a few samples. By using the present method, although there are only three samples per optical cycle marked by Goulielmakis *et al.* in Fig.3 in Ref.[30], we still can completely retrieve all parameters and, in particular,

accurately determine the absolute phase based on the expression (3). When we keep all samples, there appears a phase jump of about π between the two components whose boundary is the point of the eighteenth sample. The phase jump of π is equivalent to a change in sign of the field. Further study shows that the phase jump depends on the shift in time, i.e. the difference between two pulses' group delay times, and the difference in absolute phase between the two successive pulses if their chirps are both negligible. According to Nielsen *et al.*^[9] after carefully observing the red line in Fig.3 in Ref.[30], it is reasonable to suggest that the part behind the eighteenth sample is another weaker daughter pulse. So, we employ the first eighteen samples to retrieve the main pulse. Since the data is discrete and the sampling frequency is just about triple the carrier frequency, the retrieved duration (4.45 fs) of the main pulse slightly deviates from the value 4.3 fs determined by Goulielmakis *et al.* When we replace the duration by 4.3 fs, we find a deviation of 150 as, which makes little difference from the light wave. So, the retrieved duration is accurate enough. The pre-

dicted central frequency 2.59 rad/fs is very close to the frequency corresponding to the wavelength 750 nm. The peak of the envelope is 7.38×10^9 V/m. It can be seen that the chirp over the main pulse is negligible from the temporal-pulse phase retrieved by expression (5) (the dotted line in Fig.2). Moreover, the group delay time is 0.14 fs but not zero and the absolute phase is about 20° . It is clear that the red line in Fig.3 in Ref.[30] is not symmetric and possesses a nonzero group delay time. If we neglect the delay time in the envelope, the absolute phase is nearly zero and agrees well with that in Ref.[30]. Thus, we fully characterize the main pulse; in particular, we can accurately determine the absolute phase. In order to gain a smooth fit plot, we assume that the envelope profile takes a common shape: h.s. function. As shown by the dashed line in Fig.2, we well fit the main pulse by $E(t) = 7.38 \times 10^9 \text{sech}[1.76(t - 0.14)/4.45] \cos[2.59(t - 0.14) + 0.35]$ V/m. In addition, similarly, the daughter pulse, which is another weaker pulse but not a part of the main pulse, is also well fitted by $E(t) = 1.13 \times 10^9 \text{sech}[1.76(t - 8.22)/3.7] \cos[2.32(t - 8.22) - 1.43]$ V/m after we have retrieved all parameters. We have filtered the lower frequency noises when we retrieved the daughter pulse. The shift in time between the main pulse and the daughter pulse accounts for the fact that the difference in absolute phase between the two pulses is not the phase jump π . Obviously, it is possible to accurately retrieve the absolute phase of few-cycle laser pulse obtained in experiment even only three samples per optical cycle are available. Furthermore, it is still valid to determine the absolute phases one by one for a few-cycle pulse train.

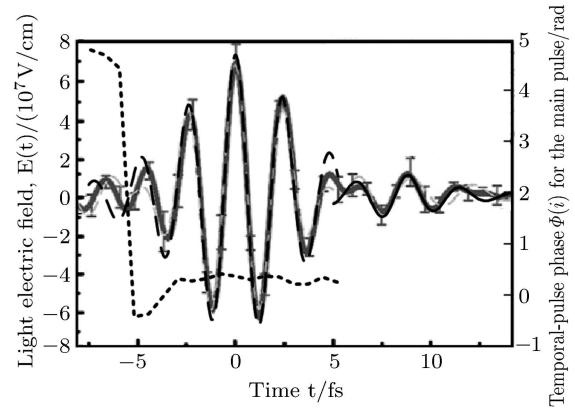


Fig.2. The fitted light wave in Fig.3 in Ref.[30]. The dashed line plot is for the fitted main pulse, the black line for the daughter pulse. The dotted line is for the temporal-pulse phase over the main pulse.

4. Summary

In summary, we have presented a Fourier analysis method, which only requires one isolated light wave, to accurately determine the absolute phase and the temporal-pulse phase of few-cycle pulse even though it is chirped. In particular, only a few samples per optical cycle are required in experiment to fully characterize the light wave. Thus, it significantly reduces the number of samples to be taken and then makes experiments easier. This method opens up attractive possibilities for phase-sensitive applications in extreme nonlinear optics, and opens the door for investigating the evolutions of the absolute phase and the temporal-pulse phase both numerically and experimentally.

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