

Quantum path analysis for arbitrary optical-waveform measurements

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An arbitrary optical waveform can be measured using a perturbative approach known as the petahertz optical oscilloscope, in which an electron trajectory in the process of high-order-harmonic generation is used as a temporal gate. Here, we report that the petahertz optical oscilloscope exhibits a significant frequency dependence for the short wavelength signal field due to the finite time span of the electron trajectory. We show that the intrinsic response of the atom to the signal field can be used to correct the frequency dependence of the measurement. It is shown that the optical waveform having the spectral components from the second harmonic to the fourth harmonic of the fundamental field can accurately be reconstructed after the correction of the frequency dependence.

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I. INTRODUCTION

A light pulse is an oscillating electromagnetic wave that is widely used as a primary tool in various fields in science and technology. The recent advances in the femtosecond laser technology [1] and the compression techniques [2–6] have reduced the pulse duration down to a few-cycle limit, which greatly enhances the temporal resolution of time-resolved studies. Since the envelope of such pulses contains only one or two oscillations, it has become very important to measure the optical waveform (i.e., the shape of the oscillating optical field) to fully understand an interaction between the light and matter [7].

The waveform of the optical pulse can be measured if there is a fast temporal gate with a subcycle resolution. One approach to measure the optical waveform is to use a photoionization process initiated by an isolated attosecond pulse obtained through high-order-harmonic generation [8,9]. The temporal resolution of the measurement is limited only by the bandwidth of the attosecond pulse which covers the ultraviolet wavelength range. However, the technique requires a complicated photoelectron measurement which can be applicable only for high-repetition-rate sources due to the low efficiency in collecting photoelectrons.

An all-optical measurement called “petahertz optical oscilloscope” is another approach which has recently been demonstrated [10]. The technique uses an electron trajectory in the process of high-order-harmonic generation as a fast temporal gate. It offers a very efficient way to measure the optical waveform because the XUV radiation is measured instead of the photoelectron spectra. However, the finite time span of the corresponding electron trajectory used as a gate would limit the temporal resolution of the measurement [11]. The purpose of this study is to investigate the frequency dependence of the optical-waveform measurement. In addition, we discuss how we can avoid the frequency dependence of the measurement.

The petahertz optical oscilloscope utilizes the electron trajectories in high-order-harmonic generation which occurs when a strong fundamental laser field is irradiated on atoms or molecules. In the strong laser field, an electron is liberated through tunneling ionization. Then, the electron is accelerated and driven back when the sign of the fundamental field is changed. High-order-harmonic radiation is emitted when the electron recombines with its parent ion [12].

Short and long trajectories, along which the ionized electron moves, are contributed to harmonic radiation [13]. The energy of the harmonic radiation increases in time for the short trajectories while it decreases for long trajectories as shown in Fig. 1. In a normal condition where phase matching does not play a significant role, short trajectories dominantly contribute to harmonic radiation [14,15]. Therefore, we will analyze the harmonic radiation generated only through short electron trajectories.

The petahertz optical oscilloscope is a perturbative approach in which two laser pulses, the fundamental and the signal, are used. The strong fundamental field is used to generate high-order-harmonic radiation. Measurement of the signal pulse is carried out by superposing it on the fundamental laser pulse. Since the intensity of the signal pulse is only $\sim 0.1\%$ of the fundamental, it only gently perturbs the electron motion in the process of high-order-harmonic generation. The accumulated action $S_e(p, t_r, \tau_e)$ and the recombination time t_r are changed in response to the addition of the signal. The phase shift of the harmonic radiation can be approximated as [16]

$$\sigma_e = \int_{t_r - \tau_e}^{t_r} \left[v_{\text{SFA}}(t) \int_{t_r - \tau_e}^t E_S(t') dt' \right] dt.$$

Here, v_{SFA} is the electron momentum without the signal field. E_S is the signal field.

It is only during the excursion time, τ_e , of the electron that the signal field is effective. Thus, the amount of harmonic phase shift is proportional to the average strength of the signal field during the excursion time of the electron. If the signal field is slowly varying during the excursion time of the electron, the average field is not much different from the instantaneous signal field. On the other hand, if the signal field rapidly

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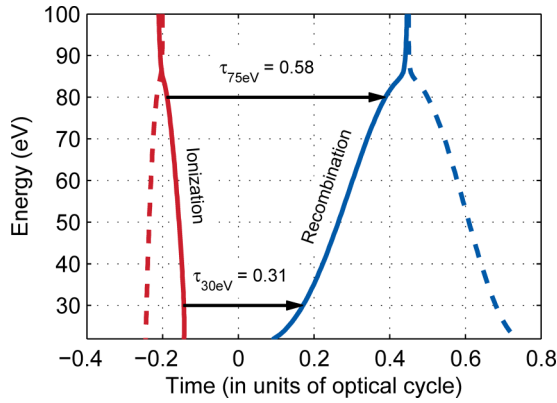


FIG. 1. Real parts of the ionization and recombination times of short (red solid line) and long (blue dashed line) quantum paths. The saddle-point approximation is used for an Ar with an intensity of 3×10^{14} W/cm² and a wavelength of 800 nm.

oscillates, the effect of the perturbation would be averaged out. Thus, the duration of the electron trajectories would set the lower limit of the signal wavelength that can be measured.

The electron trajectories in high-order-harmonic generation and the phase shift of high-order-harmonic radiation can be analyzed using the saddle-point approximation. The saddle-point solutions are complex when the ionization potential is taken into account [13]. The real parts of the ionization and recombination time obtained from the saddle-point model for a neon atom with an intensity of 3×10^{14} W/cm² and a wavelength of 800 nm are shown in Fig. 1. The excursion time of short trajectories near the highest energy (or cutoff) of the harmonic radiation is about $0.58 t_0$, comparable to the period of the second harmonics. By contrast, it is much shorter near the ionization threshold. For example, at 30 eV, the excursion time is $0.31 t_0$, comparable to that of the third harmonics.

A careful analysis is performed to show the dependence in measuring signal waveform on its frequency. The phase shifts of the harmonic radiations are calculated using a sinusoidal signal field, $E_S(t) \propto \sin[\omega_S(t + \tau_d)]$ with different signal angular frequency ω_S and the time delay τ_d . The phase shift of the harmonic radiation, σ_ε , calculated at 30 eV for a neon atom is shown in Fig. 2. If there is no frequency dependence in the measurement, the phase shift σ_ε should modulate with the same phase of the signal field when the time delay τ_d is changed. Indeed, the phase shift σ_ε oscillates with the same phase of the signal field for wavelengths longer than 400 nm. For shorter signal wavelengths, however, the amplitude of the phase shift decreases because the signal field cannot be approximated as a constant during the time span of the electron trajectory. At 300 nm, the amplitude of the phase shift is minimized. The phase of the modulation is also changed. At 200 nm, the sign of the phase modulation is changed. This phase variation over different frequencies would bring a serious consequence in a waveform measurement. While the amplitude variation for different signal wavelengths can simply be calibrated by measuring the spectrum of the signal field, the phase variation cannot easily be determined in the experiment. Instead, this intrinsic phase variation might be mistaken to be the original phase of the signal field.

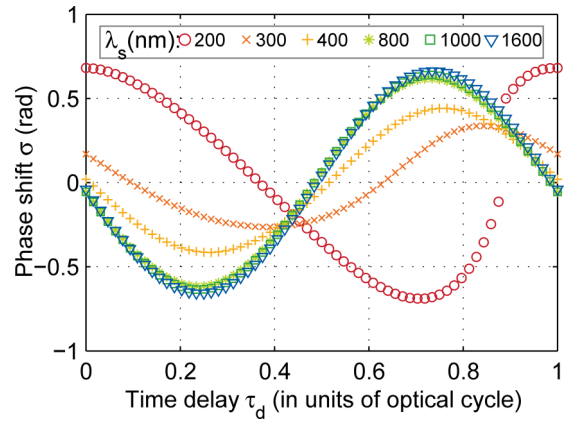


FIG. 2. The modulation of the phase shift with the sinusoidal signal field for a neon atom at the observation energy of 30 eV.

For more systematic study, we calculate the amplitude and phase modulation with different signal frequencies and observation energies. The calculation results are fitted to a sinusoidal function $\sigma_\varepsilon(\tau_d) = A(\omega_S, \varepsilon) \sin[\omega_S \tau_d + \Phi(\omega_S, \varepsilon)]$ as summarized in Fig. 3 for argon and neon. For the comparison, we also show the excursion time of the electron trajectories converted to angular frequency, $2\pi/\tau_e$, by the dashed line. The amplitude $A(\omega_S, \varepsilon)$ falls to zero at a specific signal frequency and an observation energy. The phase $\Phi(\omega_S, \varepsilon)$, at this specific point, exhibits spiral structure whose center is singular. In an argon atom, the first minimum occurs at $2.6\omega_0$ and 48 eV near

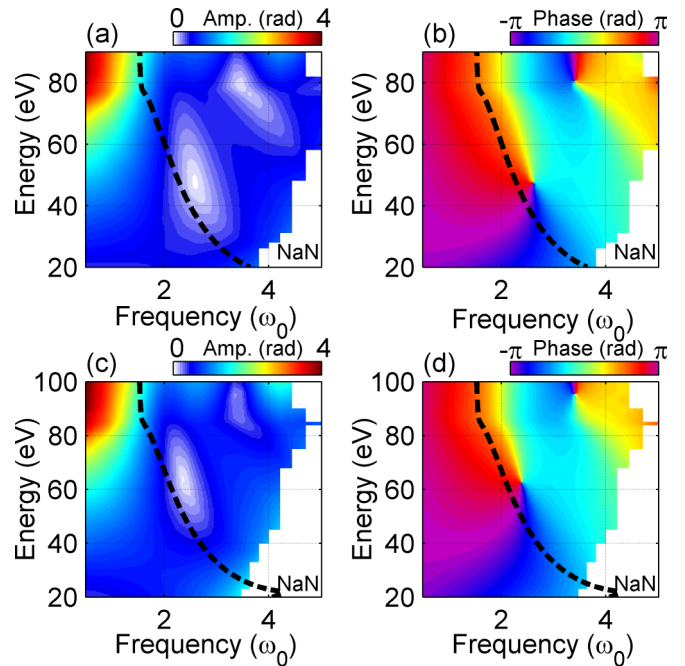


FIG. 3. Amplitude and phase of frequency response calculated by using saddle-point approximation for Ar (a,b) and Ne (c,d). The angular frequency calculated from the excursion time of the electron trajectories is shown with black dashed lines for comparison. The amplitude and phase could not be determined for the white area marked as “NaN” because the phase modulation $\sigma_\varepsilon(\tau)$ is not sinusoidal.

the angular frequency calculated from the excursion time as shown in Fig. 3(a). In a neon atom, it occurs at $2.4\omega_0$ and 64 eV as shown in Fig. 3(c). These amplitude minima are formed when the signal wavelength is comparable to the time span of the electron trajectory because the perturbation effect is averaged out.

The same calculations are achieved by solving the time-dependent Schrödinger equation in one-dimensional (1D) space with a soft-core potential [17]. The petahertz optical oscilloscope works with the harmonic radiation generated in a single half optical cycle. This can be achieved in the experiment by generating an isolated attosecond pulse. In the calculation used here, a temporal window is used to select an isolated attosecond pulse. The calculation results show the exact same behavior with the result obtained with the saddle-point approximation for the amplitude and phase of the modulation as shown in Fig. 4.

The frequency dependence of the measurement can be corrected using the amplitude and phase response obtained in the calculation. It should be noted that the waveform measurement is a linear process since the signal field superposed to the fundamental is very weak. If the frequency component of the signal field $\tilde{E}_S(\omega)$ is known, the phase shift of the harmonic radiation becomes the Fourier transform of the signal field multiplied by the frequency response of the system:

$$\sigma_\varepsilon(\tau) = \int d\omega A(\omega, \varepsilon) e^{i\Phi(\omega, \varepsilon)} \tilde{E}_S(\omega, \varepsilon) e^{-i\omega\tau} + \text{c.c.}$$

Thus, the original signal field can be found by taking the inverse Fourier transform with the amplitude $A(\omega, \varepsilon)$ and the phase $\Phi(\omega, \varepsilon)$ corrections.

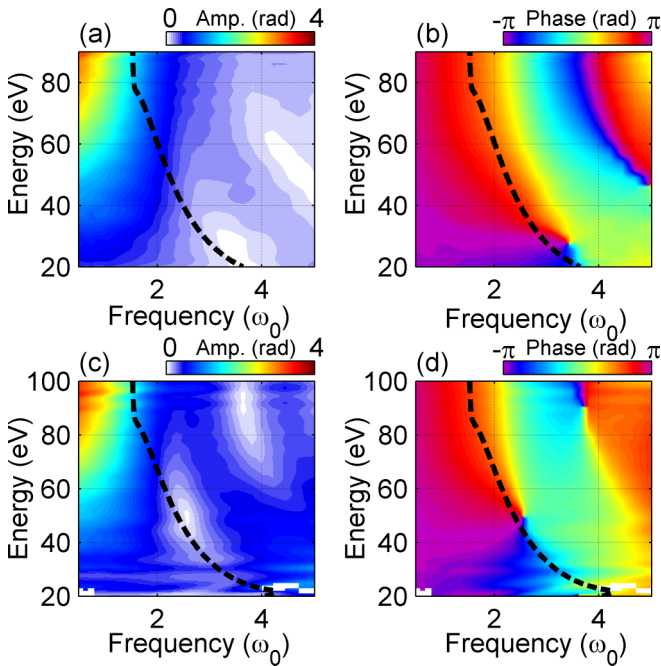


FIG. 4. Amplitude and phase of the modulation calculated by numerically solving the time-dependent Schrödinger equation for Ar (a,b) and Ne (c,d). The angular frequency calculated from the excursion time of the electron trajectories using the saddle-point approximation is shown with black dashed lines for comparison.

Since the spectrum of the signal field can be easily measured, the amplitude response $A(\omega, \varepsilon)$ can be determined in the experiment. However, it is difficult to determine the phase response $\Phi(\omega, \varepsilon)$ in the experiment because the phase response is mixed up with the original phase of the signal field. Thus, the phase response obtained in the theoretical calculations should be used for the correction. However, it is unreliable to directly use the phase response obtained from the calculation since different results are obtained depending on the ionization potential and the calculation models as shown in Figs. 3 and 4.

There is a general rule in the amplitude and phase modulation, which enables us to correct the frequency dependency of the waveform measurement. The phase response changes continuously near the amplitude-minima and phase-singular point as shown in Figs. 3 and 4. The phase jumps down by π above the energy of the singular point, and it jumps up by π below the energy of the singular point. This peculiar condition can serve as a reference to match parameters in the experiment and the simulation.

A proof-of-principle simulation is carried out. We have calculated a signal laser field with the spectrum covering from the second harmonic to the fourth harmonic of the fundamental laser field. The signal pulse has the group delay dispersion of 0.4 fs^2 and the third-order dispersion of 0.4 fs^3 . The phase shift of the harmonic radiation is calculated in Ne atom using the 1D time-dependent Schrödinger equation (TDSE) model. Two line-outs of the phase map shown in Fig. 4(c) at slightly above (50 eV) and below (46 eV) the observation energy of the amplitude minima (49.8 eV) are plotted as green and blue dotted lines, respectively, in Fig. 5. They bifurcate and make a phase jump into different directions by π . This is due to the intrinsic response of the atom to the short wavelength signal field shown in Figs. 3 and 4. Thus, the spectral phase of the signal field can be determined by the average of these two phase curves as shown with the red line in Fig. 5. The average curve can be compared with the original spectral phase of the signal field used in the calculation as shown with the black line in Fig. 5. The curvature of the spectral phase is

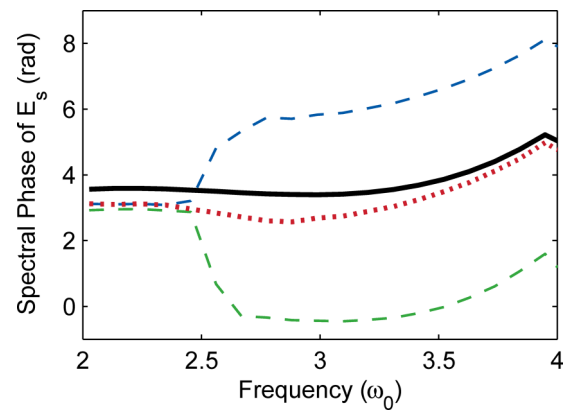


FIG. 5. Spectral phases of the signal pulse calculated at the observation energy of 50 eV (green dashed line) and 46 eV (blue dashed line). The spectral phases are extracted from Fig. 4(d). Their average (red dotted line) is shown with the original phase (black solid line).

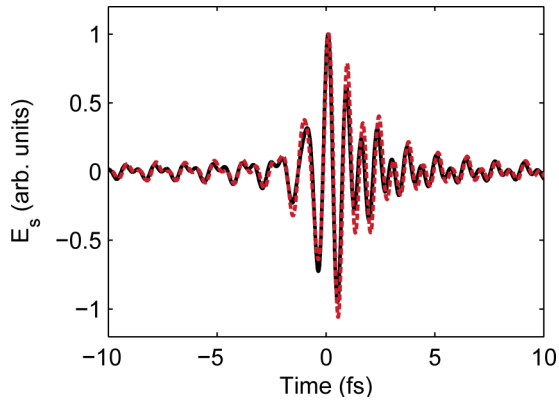


FIG. 6. The original signal field (black solid line) and the reconstructed signal field (red dotted line) after the phase correction. The group delay of the signal pulse is adjusted for better comparison.

well reconstructed. Also, the reconstructed signal is almost identical with the original signal field as shown in Fig. 6. These reconstruction results confirm that the short wavelength signal can be measured if the phase of the measured waveform is properly corrected.

Since the amplitude response becomes weaker near the singular points, it would be difficult to determine the phase near the singular points in real experiments. However, the phase can still be determined using multiple data obtained at different observation energies. In real experiments, the spectrum of harmonic radiation can be measured using a flat-field spectrometer which records the spatial distribution of the harmonic radiation as a function of observation energy. Thus, the phase of the modulation is available over the whole range of the observation energy with one data set. The phase variation near the singular point can be obtained at different observation energies, and it can be stitched with the phase obtained near the observation energy of the singular point.

In summary, the frequency dependence of the petahertz optical oscilloscope has been analyzed using two theoretical models. The optical-waveform measurement shows a significant dependence on the signal frequency due to the finite time span of the electron trajectories. It is shown that the frequency dependence of the measurement can be corrected using the intrinsic behavior of the measurement system.

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