Time-resolving tunneling ionization via strong-field photoelectron holography

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Strong-field tunneling ionization is the initial step for various ultrafast dynamics in intense laser-atom and/or molecule interactions. Time-resolving tunneling ionization is of crucial importance for accurately understanding these ultrafast processes. In our previous work [Phys. Rev. Lett. 121, 253203 (2018)], we proposed an attosecond photoelectron interferometer based on strong-field photoelectron holography to resolve the tunneling ionization. Here, we show more details about how the complex ionization time of tunneling is retrieved with our scheme. We demonstrate that the Coulomb interaction, which is a tedious and complex problem in dealing with photoelectron interference in strong-field ionization, can be safely canceled in our scheme. This is further confirmed by performing our scheme for both the model atoms with short-range and Coulomb potentials. Additionally, the validity and accuracy of our scheme are confirmed by its application in different targets and different laser parameters. Our scheme provides a reliable and versatile method to resolve the time information of strong-field tunneling ionization.

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I. INTRODUCTION

Laser-induced tunneling ionization of atoms and molecules is one of the most fundamental strong-field processes and it serves as the first step for plenty of processes in attosecond science [1–4]. When an atom or a molecule is exposed to an intense laser pulse, the bound electron can be tunneling ionized through the potential barrier. The ionized electron wave packet (EWP) is further accelerated by the oscillating electric field of the laser pulse. Part of the EWP could be driven back to scatter off or recombine with the atomic or molecular core, producing various nonlinear phenomena [5] such as above-threshold ionization [6–8], nonsequential double ionization [9–15], and high-order harmonic generation [16–20]. In past decades, these phenomena have been widely applied to the attosecond community, including attosecond pulse generation [21–24], molecular tomography [25,26], and laser-induced electron diffraction [27–30]. In these ultrafast processes and their applications, one of the most important questions concerns the temporal properties of the tunneling ionization, i.e., the time when the process is initiated.

In the past ten years, this question has draw much attention and has been hotly discussed [31]. Recently, rapidly developing attosecond technology has facilitated measurement with unprecedented temporal resolution, which opens up the opportunity to revisit this fundamentally important question [32–36]. For example, by applying the attoclock technique, the tunneling time of the electron has been experimentally measured with attosecond resolution [32]. The measured results indicate that the tunneling time is negligibly small. In the attoclock technique, the tunneling time is defined as the time delay between the maximum of the electric field and the maximal probability of electron emission. Quantum mechanically, the electron is described by wave function. In the presence of the laser field, the electron wave function continuously extends from the inner barrier to the outer barrier [37]. The different ionization instants correspond to the different measured momenta of electrons and the orders of the resultant harmonics.

In recent years, many experimental and theoretical efforts have been made to determine the ionization time related to the observables. For example, through high-order harmonic spectroscopy, the ionization time for each harmonic order in high-harmonic generation was reconstructed [38,39]. The reconstructed results agree well with the predictions of the quantum-orbit (QO) model [7]. The QO model physically originates from the application of Feynman’s path-integral approach in strong-field laser-atom interaction. In the QO model, the ionization time is complex. Its real part denotes the time when the electron exits the tunneling barrier. The imaginary part is interpreted as the tunneling time that characterizes the under-barrier electron motion [7,40,41]. This concept was first proposed in the pioneering paper of the strong-field physics by Keldysh and later was further developed and demonstrated by a wide range of theoretical literature [42–45]. During past years, it has been reported that the complex ionization time is crucially important for revealing the attosecond dynamics of the strong-field ionization process in time-resolving experiments [46–51]. It is also vital for exploring the nonadiabatic effect in the tunneling process, which has been of great concern [52–56]. Therefore, accurately retrieving this time is essential for fully understanding the strong-field processes and for further performing their applications. In Ref. [38], the real part of the ionization time is reconstructed. In Ref. [39], the imaginary part of the ionization time is reconstructed based on some assumptions about the real part.
of the ionization time and the rescattering time. Actually, it has been pointed out that these assumptions significantly affect the accuracy of the time retrieval [57].

Complementary to the measurements based on harmonic spectra, strong-field photoelectron holography (SFPH) is expected to be a powerful tool to probe the ultrafast electron dynamics in tunneling ionization. The SFPH interference derives from the interference of the near-forward rescattering EWP (which recollides with the parent ion after tunneling ionization) and the direct EWP (which drifts to the detector directly after tunneling ionization). This interference has been identified in a recent experiment with xenon [58] and then observed for other atoms [59–62] and molecules [63,64], and has been widely analyzed [65–75]. It was pointed out that the SFPH interference has the potential for probing the structural and temporal properties of atomic and molecular systems with angstrom and attosecond precision [58]. To date, this SFPH has been applied to experimentally reveal the tunneling exit point in atomic tunneling ionization [59]. The initial phase of tunneling EWP has been extracted from the SFPH pattern [63,69]. Recently, we have theoretically demonstrated that with SFPH the phase of the scattering amplitude of the atoms and molecules can be extracted [71]. More interestingly, we have shown that the charge migration in molecules can be directly visualized by analyzing the SFPH structure [75].

Very recently, we proposed that with SFPH both the ionization and rescattering times of the electron in the tunneling ionization can be accurately determined [76]. The method is closely analogous to the optical Michelson interferometer. In the traditional Michelson interferometer, a light source is split into two arms, each of which is reflected back toward the light beam-splitter mirror, as shown in Fig. 1(a). The amplitudes of these arms are combined through the superposition principle on the screen, resulting in an interferogram. By analyzing the interferogram fringes, the path characterizing the two arms could be inferred. In our scheme, the tunneling ionized EWP plays as the source of the Michelson interferometer, and the direct and near-forward rescattering EWPs act as its two arms, as shown in Fig. 1(b). Due to the coherent nature of EWPs, these two kinds of EWP can induce the SFPH pattern in the photoelectron momentum distribution (PEMD) [58]. When an orthogonally polarized perturbative pulse is added to the fundamental (FM) driving field, the paths of these two EWPs are modified and thus a displacement of the interference fringes is manifested in the PEMDs. Analyzing the response of the interference fringes to the perturbation enables us to retrieve the time information of the paths of the EWPs. This introduces us to the attosecond photoelectron interferometer.

In this paper, we follow our previous work [76] and show more details about how the complex ionization time and the associated rescattering time for strong-field tunneling ionization are retrieved. In our work, the PEMDs are calculated by solving the time-dependent Schrödinger equation, from which the SFPH structure is extracted. Note that in intense laser-atom and/or molecule interactions, the influence of the Coulomb interaction is a very tough problem and it impedes the information extraction from the PEMDs. Moreover, it has been demonstrated that the Coulomb potential significantly affects the spacing of the SFPH fringes and induces unexpected structure [65,77–83]. Therefore, dealing with the SFPH interference in strong-field ionization could be tedious and complex. In our scheme, we demonstrate that the complex influence of the Coulomb correlation on the response of the SFPH fringes to the orthogonal perturbation can be safely canceled and thus the time retrieval processes are greatly simplified. We further show that our scheme works well for both the model atoms with the Coulomb and short-range

![Fig. 1](image-url)
potentials. Our retrieved results exhibit excellent agreement with the predictions of the QO model. In addition, we confirm the validity and accuracy of our scheme by applying it to different atomic systems and different laser parameters. Our work provides a reliable and versatile way to measure the temporal properties of strong-field tunneling ionization.

II. THEORETICAL METHODS

In our scheme, the orthogonal two-color (OTC) field combined by a strong fundamental field and an orthogonally polarized perturbative field is used. The strong FM field induces the ionization of atoms. The orthogonal perturbation, which is much weaker than the FM field, does not affect the ionization of the electron, while it can affect the phase of the electron during the propagation. This is essential in the attosecond temporal properties of strong-field tunneling ionization.

A. Numerically solving TDSE

With the above OTC field, we investigate the ionization of atoms by solving the two-dimensional time-dependent Schrödinger equation (TDSE) with the single-active-electron approximation. The TDSE reads [atomic units (a.u.) are used throughout unless otherwise stated]

\[
\frac{\partial \Psi(\vec{r}, t)}{\partial t} = i[H(\vec{r}, t)]\Psi(\vec{r}, t),
\]

where \( \Psi(\vec{r}, t) \) represents the wave function and \( \vec{r} = (x, y) \) denotes the electron position in the polarization plane of the OTC field. In length gauge, the Hamiltonian \( H(\vec{r}, t) \) is written as

\[
H(\vec{r}, t) = -\frac{1}{2} \nabla^2 + V(r) + \vec{r} \cdot \vec{E}(t),
\]

where \( V(r) = -1/\sqrt{x^2 + y^2 + a} \). \( a \) is the soft-core parameter, which is selected to match the ground-state energy of atom. For xenon (\( I_p = 0.446 \) a.u.), \( a \) is set to be 0.92, and for argon (\( I_p = 0.581 \) a.u.), \( a \) is set as 0.39.

To numerically solve the TDSE, the split-operator spectral method on a Cartesian grid is employed [84]. The initial wave function is prepared by imaginary-time propagation [85]. It is further smoothly split into two parts: the inner (0 – \( R_s \)) and outer (\( R_s – R_{\text{max}} \)) regions in real-time propagation. At any given time \( \tau \), the electron wave function \( \Psi(\tau) \) is given by [86]

\[
\Psi(\tau) = \Psi(\tau)[1 - F_1(R_s)] + \Psi(\tau) F_1(R_s) = \Psi_I(\tau) + \Psi_O(\tau).
\]

Here \( F_1(R_s) = 1/[1 + e^{-(R_s-R_s)/\Delta}] \) is the splitting function. \( \Delta \) represents the width of the crossover region and \( R_s \) is the boundary of the inner space [87]. In the inner space, the wave function \( \Psi_I(\tau) \) is propagated under full Hamiltonian, while in the outer space, the wave function \( \Psi_O(\tau) \) stands for the “ionized part” analytically propagating under the Volkov Hamiltonian [86,88]. Specially, at each time step \( \tau \), the ionized EWP in the outer space is firstly transformed into momentum space \( \mathcal{C}(\vec{p}, \tau) \) and then it is propagated from time \( \tau \) to the end of the laser pulse. Finally, we obtain the PEMD related to the sum of the wave function in momentum space at \( \tau \),

\[
dP(\vec{p}) \over dE d\theta = \sqrt{2E} \left\{ \sum_\tau \mathcal{C}(\vec{p}, \tau) \right\}^2,
\]

where \( \mathcal{C}(\vec{p}, \tau) = e^{-i\int_{-\infty}^{\infty} \vec{A}(r) dt} \mathcal{C}(\vec{p}, \tau) \). \( E = \vec{p}^2 / 2 \) represents the electron final energy and \( \theta \) is the angle between the electron final momentum \( \vec{p} \) and the polarization direction of the FM field. \( \vec{A}(\tau) = -\int_{-\infty}^{\infty} \vec{E}(\tau) dt \) is the vector potential of the OTC field.

In our simulation, the Cartesian grid ranges from –700 to 700 a.u. for both \( x \) and \( y \) directions with a grid size of \( \Delta x = \Delta y = 0.34 \) a.u. The boundary of the inner space \( R_s \) is set to be 200 a.u. with the width of crossover region \( \Delta = 8 \) a.u. The propagation time step of the wave function \( \delta t \) is equal to 0.05 a.u. At the end of the pulse, the wave function is propagated for three additional optical cycles of the FM field to make sure the “slow” electrons reach the boundary \( R_s \) [67,89].

B. Quantum-orbit model

Our interferometer is based on the SFPH. The SFPH structure of the PEMD derives from the interference of the direct and near-forward rescattering EWPs, and it is given by

\[
M^2(\vec{p}) = |M_d(\vec{p}) + M_r(\vec{p})|^2 = |M_d(\vec{p})|^2 + |M_r(\vec{p})|^2 + 2|M_d(\vec{p})||M_r(\vec{p})|\cos(\delta \theta).
\]

\[
\delta \theta = \delta \theta_F + \alpha,
\]

where \( \alpha \) represents the phase for the interaction of the rescattering electron with the target ion, and \( \delta \theta_F \) accounts for the phase difference between the direct and rescattering EWPs accumulated during their propagation in the laser field.

Following Ref. [58], the phase \( \delta \theta_F \) can be calculated with the saddle-point approximation, which provides us the
quantum orbit to analyze the SFPH interference [90]. In the OTC field, $\delta \theta_F$ is written as

$$
\delta \theta_F = \frac{1}{2} \int_{0}^{t_0} [p_x + A_x(t)]^2 dt + \frac{1}{2} \int_{0}^{t_0} [p_y + A_y(t)]^2 dt
$$

$$
- \frac{1}{2} \int_{t_r}^{t_F} [k_x + A_x(t)]^2 dt - \frac{1}{2} \int_{t_r}^{t_F} [k_y + A_y(t)]^2 dt
$$

$$
+ I_p (t^R - t^D),
$$

(8)

where $A_x(t)$ and $A_y(t)$ indicate the vector potential of the FM and orthogonal components of the OTC field, respectively. $p_x$ and $p_y$ are the final parallel momentum (along the $x$ axis) and transverse momentum (along the $y$ axis) of the electron, $k_x$ and $k_y$ denote the parallel and transverse components of the canonical momentum $\vec{k}$ of the electron before rescattering. $I_p$ is the ionization potential of the atom. $t^D$ is the ionization time of the direct electron. $t^R$ and $t_r$ are the ionization and rescattering times of the rescattering electron. At the given time $t$, the momenta of the rescattering electron along the polarization directions of the FM field and the orthogonal perturbation before rescattering are $k_x + A_x(t)$ and $k_y + A_y(t)$, while those of the direct electron are $p_x + A_x(t)$ and $p_y + A_y(t)$.

In our study, the orthogonal component of the OTC field is very weak, and thus the ionization and rescattering times can be approximately determined by the saddle-point equation in the FM field. For the direct electron, the saddle-point equation is written as

$$
\frac{1}{2} [p_x + A_x(t^D)]^2 + \frac{1}{2} p_y^2 + I_p = 0,
$$

(9)

while for the rescattering electron, it reads

$$
\frac{1}{2} [k_x + A_x(t)]^2 + I_p = 0,
$$

$$
\frac{1}{2} [k_y + A_y(t)]^2 = \frac{1}{2} [p_x + A_x(t)]^2 + \frac{1}{2} p_y^2,
$$

\[
\int_{t_r}^{t_0} [k_x + A_x(t)] dt = 0,
\]

\[
\int_{t_r}^{t_0} [k_y + A_y(t)] dt = 0.
\]

(10)

Here Eq. (9) stands for the energy conservation for the direct electron at tunneling ionization. The first and second equations in Eq. (10) indicate the energy conservation for the rescattering electron during tunneling ionization and rescattering, respectively. The third and fourth equations in Eq. (10) represent the return conditions.

It should be noted that with the QO method, the obtained ionization and rescattering times are complex numbers, so all the quantities except the measured final momenta $p_x$ and $p_y$ are complex numbers, and thus $\delta \theta$ in Eq. (7) is also complex. In our scheme, the imaginary part of the phase $\delta \theta$ affects the amplitudes of the direct and rescattering electrons and influences the visibility of the interference fringes. It is the real part of $\delta \theta$ determines the position of the interference fringes that we focus on. Therefore in this paper, we consider only the real part of $\delta \theta$.

III. RESULTS AND DISCUSSION

A. Photoelectron momentum distributions obtained by solving TDSE

In Fig. 2, we show the photoelectron momentum distributions (PMDs) for strong-field tunneling ionization of xenon atoms obtained by solving the TDSE. Figure 2(a) shows the result in the 1600-nm single-color field only. Figures 2(b)–2(d) display the results in the OTC field consisting of a strong 1600-nm FM field and a weak second harmonic with relative phase $\varphi = 0, 0.5\pi, 1.5\pi$, respectively. The intensity of the 1600-nm laser field is $1.5 \times 10^{14}$ W/cm$^2$ and of the 800-nm pulse $1.0 \times 10^{12}$ W/cm$^2$. 

FIG. 2. (a) PMD for strong-field tunneling ionization of a xenon atom by a 1600-nm single-color laser field in the log scale. The black lines indicate the positions of SFPH interference fringes (from top to bottom: the first-order minimum labeled by $1_{\text{min}}$ at $p_\| > 0$, the zeroth-order maximum labeled by $0_{\text{max}}^+$, and the minus first-order minimum labeled by $-1_{\text{min}}^-$ at $p_\| < 0$). (b)–(d) PMDs for the OTC field combined by a strong 1600-nm FM field and a weak second harmonic with relative phase $\varphi = 0, 0.5\pi, 1.5\pi$, respectively. The intensity of the 1600-nm laser field is $1.5 \times 10^{14}$ W/cm$^2$ and of the 800-nm pulse $1.0 \times 10^{12}$ W/cm$^2$. 

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laser focal volume effect, these two types of interference are usually invisible in experiments. The other nearly horizontal fringes marked by the black lines in Fig. 2(a) are referred to as the SFPH pattern, which originates from the interference of the electrons reaching the detector directly after tunneling and those near-forward rescattering with the target ion [71]. This SFPH pattern has been observed in a broad range of experiments and is the most pronounced interference structure in the PEMDs for the near- and mid-infrared laser pulses [58,94]. In the present paper, we focus on this interference pattern. For the single-color field, the holographic structure is exactly symmetric about $p_x = 0$, as displayed in Fig. 2(a), while for the OTC field, the paths of tunneling ionized electrons are disturbed by the orthogonal second harmonic, resulting in the shift of the interference fringes, as shown in Figs. 2(b)–2(d). Interestingly, depending on the parallel momentum $p_x$, the fringe shift is changing with relative phase $\varphi$. As we will show below, this shift and its $p_x$ dependence provide us with information about the electron tunneling ionization.

B. Holographic fringes extracted from PEMD

To make the SFPH fringes and their shift more visible, we first wash out the vertical fringes by averaging the raw PEMD over $p_x$ with a window function [71]. Several cuts of the obtained PEMD at different $p_x$ are displayed in Figs. 3(a) and 3(b), where the effect of the orthogonal second harmonic of the OTC field can be clearly seen. For instance, for $p_x = -1.1$ a.u., the interference minima shift towards right and left in the OTC field at $\varphi = 0$ and $\varphi = \pi$ with respect to the single-color field, respectively. For $p_x = -1.6$ a.u., the shift is reversed. Then we utilize the procedure introduced in [71] to eliminate the envelop of the PEMD induced by the ionization amplitude of the electrons and extract the interference term $\cos(\delta \theta)$ between the direct and rescattering electrons. Figures 3(c) and 3(d) separately display the examples of the obtained interference term $\cos(\delta \theta)$ in the single-color field and the OTC field with $\varphi = 0.5 \pi$ as functions of transverse momentum $p_y$ for $p_x$ ranging from $-2$ to $2$ a.u. One can see two types of interference fringes. The irregular one, visible in the momentum region $|p_x| \in [0, 0.4]$ a.u., is identified as the holographic interference which formed by the multiple forward-scattering electrons [95]. The obvious regular fringes in the momentum region $|p_x| \in [0.4, 2]$ a.u. are the SFPH structure that we are interested in here. Clearly, for the single-color field this structure is exactly symmetric about $p_x = 0$ and $p_y = 0$. For the OTC field, the positions of the interference minima and maxima of this SFPH structure are also symmetric with respect to $p_x = 0$, while for $p_x = 0$, it is obviously asymmetric. For a better view, we display the cut of $\cos(\delta \theta)$ at $p_x = -1.1$ a.u. for the single-color field and for the OTC field with relative phase $\varphi = 0$, $\pi$, $0.5 \pi$, and $1.5 \pi$ in Fig. 3(e). We also show $\cos(\delta \theta)$ for another cut at $p_x = -1.6$ a.u. in Fig. 3(f). It is indicated that the positions of the interference maxima and minima which depend on $p_x$ are sensitive to the relative phase $\varphi$. At last, we monitor the positions of the interference maxima and minima for the single-color field and the OTC field in the momentum region $p_x < 0$, as shown in Fig. 3(g). It is more obvious that for the OTC field the SFPH structure shifts along the $p_x$ direction with respect to that for the single-color field, and this shift is significantly modified by relative phase.

To show the shift of the SFPH interference fringes and its $p_x$ dependence quantitatively, we seek the position of the first interference minimum ($1^\text{min}$) as a function of relative phase $\varphi$ in the OTC field and calculate its shift relative to the position in the single-color field. Figure 4(a) displays the obtained result. It clearly indicates that the shift $\Delta p_x$ oscillates with $\varphi$. The oscillation amplitude and the corresponding relative phase at which $\Delta p_x$ maximizes vary gradually with $p_x$. For
example, $\Delta p_x$ maximizes at $\varphi = 1.5\pi$ for $p_x = -1.8$ a.u. and maximizes at $\varphi = 2\pi$ for $p_x = -0.9$ a.u. For comparison, we also display the shift for the zeroth-order maximum ($0_{\text{max}}$) of the interference fringes in Fig. 4(b). The obtained result is exactly the same as that for the first-order minimum of interference fringes shown in Fig. 4(a).

We can also trace the photoelectron yield at $p_y = 0$ as a function of the relative phase of the OTC field. In the single-color field, this yield maximizes at $p_y = 0$. If an orthogonal perturbation is added, the photoelectron is blown away from $p_y = 0$ by an amount of the vector potential $A_y(t^p, \varphi)$. Thus, the position of the maximum in the PEMD locates at $p_y = 0$ only when the vector potential $A_y(t^p, \varphi) = 0$. With this relation, it has been shown that the ionization time for each parallel momentum can be extracted by monitoring the photoelectron yield in the PEMD at $p_y = 0$ [96,97]. In Fig. 5(a), we display the ionization yield of photoelectron at $p_y = 0$ in the OTC field with respect to that of the single-color field, $\Delta Y = (Y^{\text{OTC}} - Y^{SC})/Y^{SC}$, where $Y^{SC}$ and $Y^{\text{OTC}}$ are the photoelectron yields in the single-color and OTC fields at $p_y = 0$, respectively. It is shown that $\Delta Y$ periodically changes with $\varphi$. As the parallel momentum $p_y$ changes, the phase $\varphi$ where $\Delta Y$ maximizes varies gradually. This $p_y$ dependence of the relative phase provides us another intuitive observation.

Note that the transverse momentum where the photoelectron yield maximizes in PEMD is not coincident with the zeroth-order maximum of the SFPH interference fringes in $\cos(\delta\varphi)$. To illustrate this, in Figs. 5(b) and 5(c) we show the PEMD and $\cos(\delta\varphi)$ at $p_y = -1.6$ a.u. as functions of $\varphi$, respectively. There is a phase shift between the PEMD and $\cos(\delta\varphi)$. This shift is more clearly seen in Fig. 5(d), where we trace the position of the maximum in PEMD and the zeroth-order maximum in $\cos(\delta\varphi)$ at $p_y = -1.6$ a.u. as functions of $\varphi$. It is shown that the maximum in PEMD shifts most away from $p_y = 0$ at $\varphi = 1.7\pi$, while it is at $\varphi = 1.4\pi$ for the interference term $\cos(\delta\varphi)$. The reason is demonstrated as follows.

For the photoelectron yield at $p_y = 0$, the direct electron has the dominant contribution. Thus, the behavior of the relative yield $\Delta Y$ as a function of the relative phase is a representation of the response of the direct electron to the orthogonal perturbation [96,98], while for the interference term $\cos(\delta\varphi)$, it contains both contributions of the direct and rescattering electrons. Thus, the response of $\Delta p_x$ to the relative phase $\varphi$ is different from that of $\Delta Y$. We should mention that in the OTC field, the response of the direct electron to the relative phase $\varphi$ is quite different from that of the rescattering electron. Taking the photoelectron with momentum $(p_x, p_y) = (-0.9, 0)$ a.u. in the single-color field as an example, we treat it as the direct or rescattering electron and calculate its final transverse momentum in the OTC field, as shown in Fig. 6. Obviously, the final transverse momentum for the direct electron maximizes at $\varphi = 2\pi$, while for the rescattering electron it maximizes at $\varphi = 1.7\pi$.

Summarizing, from the PEMDs we can obtain three independent observations: the amplitude of the SFPH fringes shift, and the corresponding relative phase where $\Delta p_x$ peaks....
[as shown in Fig. 4(a)] and the relative phase where \( \Delta Y \) maximizes [as indicated in Fig. 5(a)]. As explained above, the observation in \( \Delta Y \) reflects the disturbance of the orthogonal second harmonic on the yield of the direct electron at \( p_x = 0 \) [96,97]. The observations in \( \Delta Y \) originate from the perturbation of the orthogonal second harmonic to the phase difference of the direct and near-forward rescattering electrons, as shown in detail in the following sections.

C. Shift of the holographic fringes and effect of the Coulomb interaction

In this section, we reveal the origin of the SFPH interference fringe shift \( \Delta \rho_Y \) in the PEMDs for the OTC field with respect to the single-color field and the effect of the Coulomb interaction on this shift. It should be mentioned that in the pioneering work on the SFPH, the effects of the Coulomb interaction on the phase difference between the direct and rescattering electrons are not taken into account, and thus the predictions of the SFPH fringes deviate from the TDSE calculations and experimental data [58,65].

This point is clearly indicated in Fig. 7, where we show the interference term \( \cos(\delta \theta) \) extracted from the TDSE results and calculated by Eq. (7) but without \( \alpha \). In our previous work [71], we pointed out the term \( \alpha \), which accounts for the interaction between the parent ion and the rescattering electron (i.e., the phase of the scattering amplitude). With this term included, the SFPH pattern given by Eq. (7) agrees excellently with the phase of the scattering amplitude. With this term included, the Coulomb effect does not matter for the observations of the SFPH fringe shift and can be canceled out safely.

First, we simplify the formula of the phase difference \( \delta \theta_F \) given in Eqs. (7) and (8). For the near-forward rescattering SFPH, the interference fringes exist at small \( p_y \), and thus we have \( p_x \approx k_x \) through the second equation of Eq. (10). Then it can be easily proved that the second, fourth, and fifth terms in the right side of Eq. (8) can be approximately canceled by

\[
\frac{1}{2} \int_{t_D}^{t_R} (p_x + A_x)^2 dt - \frac{1}{2} \int_{t_D}^{t_R} (k_x + A_x)^2 dt + I_p(t_i^R - t_i^D) \\
\frac{1}{2} \int_{t_D}^{t_R} (k_x + A_x)^2 dt - \frac{1}{2} \int_{t_D}^{t_R} (k_x + A_x)^2 dt + I_p(t_i^R - t_i^D) \\
\frac{1}{2} \int_{t_D}^{t_R} (k_x + A_x)^2 dt + I_p(t_i^R - t_i^D) \approx 0.
\]

(11)

Here, in the last line of Eq. (11), we have used Eq. (9) and the first equation of Eq. (10). This approximation is further confirmed by our numerical calculations, as shown in Fig. 8(a). Indeed, it is nearly zero for \( p_y \), close to the \( p_x \) axis, where the SFPH interference appears. Therefore, the first and third terms in Eq. (8) are the dominant terms accounting for the phase \( \delta \theta_F \). With this in mind, Eq. (7) is simplified as

\[
\delta \theta = \frac{1}{2} \int_{t_D}^{t_R} [p_y + A_y(t)]^2 dt - \frac{1}{2} \int_{t_D}^{t_R} [k_y + A_y(t)]^2 dt + \alpha.
\]

(12)

Moreover, through Eq. (9) and the first equation of Eq. (10), it can be approximated that the ionization times for
the direct electron and the rescattering electron are the same, i.e.,
\[ t_{i}^{D} = t_{i}^{R}. \] (13)

Quantitatively, the difference in the real and imaginary parts of the ionization time are separately displayed in Figs. 8(b) and 8(c). It is indicated that for the momentum range we are interesting in, the differences are well below 0.5 a.u. Thus, this approximation is reasonable. In the following, we omit the superscript in \( t_{i} \).

According to the fourth equation in Eq. (10), we have \( k_{y} = \frac{\int_{t_{i}} A_{y}(t)dt}{\int_{t_{i}} A_{y}(t)dt} \). The right side of Eq. (12) can be straightforwardly simplified further,
\[ \frac{1}{2} \int_{t_{i}}^{t_{f}} [p_{y} + A_{y}(t)]^{2} dt - \frac{1}{2} \int_{t_{i}}^{t_{f}} [k_{y} + A_{y}(t)]^{2} dt = \frac{1}{2} (t_{f} - t_{i}) (p_{y}^{2} - k_{y}^{2}) + p_{y} \int_{t_{i}}^{t_{f}} A_{y}(t) dt - k_{y} \int_{t_{i}}^{t_{f}} A_{y}(t) dt \]
\[ = \frac{1}{2} (t_{f} - t_{i}) (p_{y}^{2} - k_{y}^{2}) + \frac{1}{2} (t_{f} - t_{i}) (2k_{y}^{2} - 2p_{y}k_{y}) \]
\[ = \frac{1}{2} (p_{y} - k_{y})^{2} (t_{f} - t_{i}) + \alpha. \] (14)

Thus we have
\[ \delta \theta_{\text{OTC}} = \frac{1}{2} (p_{y} - k_{y})^{2} (t_{f} - t_{i}) + \alpha. \] (15)

For the single-color field, \( k_{y} = 0 \), and thus the phase difference in the single-color field is written as
\[ \delta \theta_{\text{SC}} = \frac{1}{2} p_{y}^{2} (t_{f} - t_{i}) + \alpha. \] (16)

From Eqs. (15) and (16), we can predict the traverse momentum \( p_{y} \) of the SFPH interference minima and maxima in the single-color field and the OTC field. Here we take the first-order minimum of the interference fringes as an example. It is located at the position \( p_{y} \), where \( \delta \theta = \pi \), and we obtain
\[ p_{y}^{\text{SC}} = \sqrt{\frac{2(\pi - \alpha_{\text{SC}})}{t_{f} - t_{i}}} \] (17)
and
\[ p_{y}^{\text{OTC}} = \sqrt{\frac{2(\pi - \alpha_{\text{OTC}})}{t_{f} - t_{i}}} + k_{y}, \] (18)

where \( \alpha_{\text{SC}} \) and \( \alpha_{\text{OTC}} \) are the phase due to the Coulomb interaction in the single-color field and the OTC field, respectively. Obviously, the position of the interference minimum depends on the phase of the scattering amplitude \( \alpha_{\text{SC}} \) and \( \alpha_{\text{OTC}} \).

When the orthogonal second harmonic of the OTC field is a weak perturbation, \( \alpha_{\text{SC}} \approx \alpha_{\text{OTC}} \). Then the shift of the interference fringes in the OTC field with respect to that of the single-color field is written as
\[ \Delta p_{y} = p_{y}^{\text{OTC}} - p_{y}^{\text{SC}} = \text{Re}[k_{y}]. \] (19)

It is shown that the shift of the SFPH fringes induced by the OTC field equals to \( k_{y} \), and it depends on the relative phase, and also on the parallel momentum through the ionization time \( t_{i} \) and the rescattering time \( t_{r} \). The Coulomb interaction between the parent ion and the rescattering electron influences the position of the interference fringes but does not affect the shift of the interference fringes. This is a key point of our scheme.

At last we check the validity of the approximations in Eqs. (11) and (13). In Fig. 8(d), we display the obtained \( \Delta p_{y} \) of the first minimum of the SFPH pattern as a function of relative phase \( \varphi \) at \( p_{x} = -0.9 \) a.u. The solid curve indicates the data calculated by Eq. (19) and the open circles are the data from Eq. (8). In Fig. 8(e), we show \( \Delta p_{y} \) for another cut at \( p_{x} = -1.3 \) a.u. The good agreement between the results of Eqs. (19) and (8) indicates the validity of our approximations. Then, with Eq. (19), we calculate \( \Delta p_{y} \) as a function of \( \varphi \) for \( p_{x} \) ranging from \(-1.95 \) to \(-0.75 \) a.u., as shown in Fig. 8(f). It is observed that the result is in good agreement with the TDSE result presented in Fig. 4(a), confirming the validation of the approximation made in Eq. (19).

D. Retrieving the complex ionization time and rescattering time

The periodic oscillation of \( \Delta p_{y} \) in Fig. 4(a) introduces us to fit its \( \varphi \) dependence by the function
\[ \Delta p_{y} = P_{m} \cos(\varphi - \Phi_{m}), \] (20)
where \( P_{m} \) describes the amplitude of the oscillation and \( \Phi_{m} \) characterizes the relative phase where \( \Delta p_{y} \) maximizes. Figures 9(a) and 9(b) display the obtained \( P_{m} \) and \( \Phi_{m} \) as functions of \( p_{x} \), respectively.

\( P_{m} \) and \( \Phi_{m} \) provide us two independent quantities to retrieve the complex ionization time and rescattering time. According to Eq. (19), we have
\[ \frac{\partial \Delta p_{y}}{\partial \varphi} \bigg|_{\varphi=\Phi_{m}} = \text{Re} \left[ \frac{\partial k_{y}(t_{i}, t_{r}; \varphi)}{\partial \varphi} \right] \bigg|_{\varphi=\Phi_{m}} = 0, \]
\[ \Delta p_{y} \bigg|_{\varphi=\Phi_{m}} = \text{Re} \left[ k_{y}(t_{i}, t_{r}; \varphi) \right] \bigg|_{\varphi=\Phi_{m}} = P_{m}. \] (21)

Note that \( t_{i} \) and \( t_{r} \) are complex numbers. For rescattering time \( t_{r} \), it has been reported that the imaginary part is negligibly small \[39,99\] and thus it is a good approximation that \( t_{r} \) is real. So there are three quantities to be retrieved. One more independent observation is needed.

The third observation can be obtained from the relative yield \( \Delta Y \), as shown in Fig. 5(a). In the OTC field, \( \Delta Y \) is modulated by relative phase, and it peaks at \( p_{y} = 0 \) when \( \varphi \) satisfies \[96,97\]
\[ \text{Re} \left[ k_{y}(t_{i}; \varphi) \right] \bigg|_{\varphi=\Phi_{m}} = 0. \] (22)

This reveals that by measuring the value of the phase \( \Phi_{m} \) for maximal \( \Delta Y \), we could get another observation and equation for our retrieval. In Fig. 9(c), we present \( \Phi_{m} \) extracted from the TDSE results in Fig. 5(a) as a function of \( p_{x} \). This provides the third observation \( \Phi_{m} \) for determining \( t_{i} \) and \( t_{r} \).

Before retrieving \( t_{i} \) and \( t_{r} \), we first compare these three observations from TDSE results with those predicted by the classical model \[5\], QO model, and QO model with artificially setting the imaginary part in the ionization time \( \text{Im}[t_{i}] = 0 \). For the quantity \( P_{m} \) [Fig. 9(a)], the QO model result agrees well with the TDSE result and \( P_{m} \) minimizes at \( p_{x} = -1.32 \) a.u., while for the classical model, the result deviates from the TDSE obviously and the minimum of
FIG. 9. (a) The amplitudes $P_m$ extracted from the TDSE result (dashed blue curve) and predicted by the QO model (solid blue curve), classical model (red triangles), and QO model with $\text{Im}[t_1]=0$ (green dots) as functions of $p_x$. (b) The same as (a) but for the phase $\Phi_m$ at which $\Delta\phi_1$ maximizes. (c) The same as (a) but for the phase $\theta_m$ where the yield $\Delta Y_p$ peaks. (d) The real parts of the ionization times ($\text{Re}[t_1]$) retrieved from the TDSE results (dots) and predicted by the QO model (solid curve) and classical model (dashed curve) as functions of $p_x$ (scaled by the amplitude of the vector potential of the FM field $A_0$). (e) The same as (d) but for the rescattering times. (f) The retrieved tunneling time ($\text{Im}[t_1]$). The dots show the data from the TDSE results. The solid curve represents the QO model result. The dashed curve indicates the Keldysh time.

$P_m$ shifts towards the right. For the QO model with $\text{Im}[t_1]=0$, the prediction of $P_m$ is smaller than the TDSE result, though the position of the minimal $P_m$ is in coincidence with the TDSE result. For $\theta_m$ [Fig. 9(c)], the classical model result also deviates from the TDSE result, while both predictions from the QO model and QO model with $\text{Im}[t_1]=0$ agree with the TDSE result excellently for $p_x$ not close to the boundary. This indicates that the imaginary part of $t_1$ does not affect $\theta_m$. For $\varphi_m$ [Fig. 9(b)], the differences between the predictions of the different models and the TDSE result are much more obvious. For example, only the prediction from the QO model is in agreement with the TDSE result, and the result of the QO model with $\text{Im}[t_1]=0$ deviates seriously from the TDSE result. This indicates that the observation $\varphi_m$ sensitively depends on the imaginary part of the ionization time.

With Eqs. (21) and (22), we retrieve the ionization time, including both the real and imaginary parts, and the associated rescattering time. The obtained results are shown in Figs. 9(d)–9(f). It is indicated that both the ionization and rescattering times agree well with the predictions of the QO model and deviate distinctly from the classical model results. It is worth noting that in the classical model the ionization time is real. In the QO model, the ionization time is complex and the imaginary part is interpreted as the time for the electron tunneling through the potential barrier. With our scheme, the retrieved imaginary ionization time agrees excellently with the prediction of the QO model, as shown in Fig. 9(f). We also show the Keldysh time which is calculated by $\sqrt{2E_p/|\vec{E}(t_1)|}$. This imaginary time decreases as $p_x$ moves towards zero. This is easy to understand. For $p_x$ more close to zero, the instantaneous electric field at tunneling increases, and thus the width and height of the potential barrier decrease, leading to the decreasing of the tunneling time. We should mention that in previous studies of retrieving the ionization and rescattering times with harmonic spectroscopy, a zero or Keldysh time for the imaginary ionization time was assumed [38,57]. In our scheme, no such assumption is made. Both the real and imaginary parts of the ionization time and the rescattering time are treated as unknown quantities.

Summarizing, we have shown in detail how to retrieve the ionization and rescattering times of the electron in tunneling ionization with our attosecond photoelectron interferometer. This is based on the approximations made in Eqs. (11) and (13), the validity of which has been carefully checked. The important merit of our method is that the Coulomb interaction does not influence the shift of the SFPH fringes and thus it can be canceled. In the following sections, we confirm this point by applying our method for the model atoms with Coulomb and short potentials. We also apply our method to different targets and different parameters of the laser field.

E. Results of short-range potential

In Sec. III C, we demonstrate that the Coulomb potential does not affect the shift of the SFPH fringes and it can

FIG. 10. (a) The amplitudes $P_m$ extracted from the QO model results (solid black curve), and the TDSE results for the model atoms with short-range potentials $b=10$ (dotted purple curve) and $b=20$ (dash-dotted green curve), and the Coulomb potential (dashed blue curve). (b) The same as (a) but for the phase $\Phi_m$. (c) The same as (a) but for the quantity $\theta_m$. 

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be safely canceled. Here, we further confirm this issue by illustrating our results for the short-range potential:

\[ V(r) = -\frac{1}{\sqrt{x^2 + y^2 + a}} e^{-\frac{x^2 + y^2}{b}} , \]  

(23)

where \( b \) denotes the screening parameter. The results are illustrated by considering two values, \( b = 10 \) and 20. The soft-core parameter \( a \) is modulated to match the ground state of the Xe atom (\( I_p = 0.446 \) a.u.). Figures 10(a)–10(c) separately show the quantities \( P_m, \Phi_m, \) and \( \theta_m \) extracted from the TDSE results as functions of parallel momentum \( p_x \). For comparison, we also present the obtained TDSE results for Xe with Coulomb potential (\( b = \infty \)) and the QO model. Clearly, the results of the short-range potentials and Coulomb potential nicely agree with the predictions of the QO model. This confirms that the Coulomb potential does not affect the shift of the SFPH fringes.

F. Results of argon

We apply our interferometer to different atomic systems. In the QO model, the ionization and rescattering times are obtained with saddle-point method. The saddle-point equations for the direct and rescattering electrons in Eqs. (9) and (10) indicate that \( t_i \) and \( t_r \) are related to the target species through ionization energy \( I_p \). In this section we take Ar atom as an example.

Figure 11 shows the obtained \( P_m, \Phi_m, \) and \( \theta_m \) from the attosecond photoelectron interferometer. The data of the dashed curves are obtained by the TDSE calculations, and the data of the solid curves, orange triangles, and black dots are predicted by the QO model, classical model, and QO model with \( \text{Im}[\pi] = 0 \), respectively. It is shown that \( P_m, \Phi_m \) from the QO model agree excellently with the TDSE results, while those from the classical model and QO model with \( \text{Im}[\pi] = 0 \) deviate seriously from the TDSE results. For \( \theta_m \), the classical model result shift down compared with the TDSE result, particularly at large \( p_x \). Both the results of the QO model and QO model with \( \text{Im}[\pi] = 0 \) are in coincidence with the TDSE result. These behaviors are the same as those of the Xe atom, as shown in Figs. 9(a)–9(c). This confirms the effectiveness of our scheme.

G. Dependence on the frequency of the orthogonal perturbation

In the attosecond photoelectron interferometer shown above, we use the second harmonic to disturb the paths of the direct and rescattering electrons. The perturbation could be chosen as the other frequencies and our interferometer also works. Here, we take the fourth harmonic as an example. The other laser parameters are the same as those in Sec. III A.

The obtained \( P_m, \Phi_m, \) and \( \theta_m \) are displayed in Fig. 12, where the data of the dashed curves are extracted from the TDSE results. The green triangles, solid curves, and blue dots are the results predicted by the classical model, QO model, and the QO model with \( \text{Im}[\pi] = 0 \), respectively. It is obvious that for the quantities \( P_m, \Phi_m, \) only the results of the QO model agree well with the TDSE results, and the other models deviate from the TDSE results distinctly. For \( \theta_m \), the TDSE result is well reproduced by both the QO model and the QO model with \( \text{Im}[\pi] = 0 \), while it is inconsistent with the classical model result. According to Eq. (19), the shift of the interference fringes is written as

\[ \Delta p_x = \text{Re} [\pi] \]

\[ = -\text{Re} \left\{ E_x [\cos(\omega_0 t_r + \phi) - \cos(\omega_0 t_i + \phi)] \right\} \frac{q^2}{q^2} (t_r - t_i) \]

(24)

where \( q \) is the harmonic order. It shows that \( P_m \) minimizes when \( \omega_0 t_r - \omega_0 t_i = 2n\pi \) (\( n = 1, 2, 3, \ldots \)), i.e., the traveling time \( \Delta t = t_r - t_i \approx \frac{q}{2} T_0 \) (\( T_0 \) is the period of the FM field). Thus for the second-harmonic perturbation \( (q = 2) \), there is one minimum in \( P_m \) for \( p_x < 0 \) [100]. For the fourth-harmonic
at various FM laser intensities $I = 0.75I_0$, $I_0$, and $1.5I_0$ ($I_0 = 1.0 \times 10^{14}$ W/cm$^2$), while the intensity of the orthogonal second harmonic stays unchanged ($1.0 \times 10^{12}$ W/cm$^2$). It is shown that at all intensities, these observables predicted by the QO model and extracted from the TDSE results agree well with each other. From our previous paper, we know that the quantity $P_m$ exhibits a minimum at $p_x = 0.6A_0$ ($A_0$ is the amplitude of the vector potential of the FM driving field), where the traveling time is $0.5T_0$ [100]. We demonstrated that with this character the laser intensity can be accurately calibrated. For the laser intensities here, the minima locate at $p_x = -0.95$ a.u., $-1.10$ a.u., and $-1.32$ a.u., respectively, exactly corresponding to $0.6A_0$ of the FM driving fields.

IV. SUMMARY

In conclusion, we have used attosecond photoelectron interferometry, which is analogous to the optical Michelson interferometer, to resolve the temporal properties of the electron tuning ionization. In this work, we numerically investigate the PEMDs of an Xe atom for the OTC field consisting of a strong 1600-nm FM field and a perturbative second harmonic, from which we extract the SFPH pattern. Our results show that the position of the SFPH pattern is sensitive to the relative phase of the OTC field, and it shifts along the direction of the transverse momentum depending on the parallel momentum. With QO model, we reveal its origin. Moreover, we show in detail that by analyzing this shift of the SFPH pattern, the ionization time, including both the real part and imaginary part, and the related rescattering time can be retrieved accurately.

The Coulomb effect is a tough problem in intense laser-atom and/or molecule interactions and it complicates the information retrieval from the PEMDs. Here we demonstrate that it does not affect our scheme and can be safely canceled. This point is further confirmed by performing interferometry for both model atoms with the Coulomb potential and short-range potential. Additionally, we demonstrate the validity and accuracy of our scheme by applying it to the Ar atomic system, the OTC field combined by a strong FM field, and a weak fourth harmonic and different intensities of the FM field. Our work provides a reliable and versatile way to measure the time information of strong-field tunneling ionization.

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