Electronic Response Time around Tunnel Exit in Strong-Field Ionization

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(Dated: November 17, 2021)

Tunneling ionization is a basic process of strong-field atomic physics. Revealing its time-resolved dynamics is one of the goals of attosecond science. Here, we show that after tunneling, a finite response time (about 100 attoseconds) is needed for the electronic state to evolve into an ionized state. We construct a semiclassical model with a compact expression to describe this response time. With this expression, a simple Coulomb-calibrated mapping relation between time and observables is obtained. Comparisons with experiments give direct evidence for our theory. Our work uncovers the transient response process around tunnel exit and provides a simple tool for quantitatively explaining and predicting experimental phenomena in attosecond measurements.

\textbf{Introduction.}—The development of ultrashort and ultrashort laser technology provides the opportunity for people to access the fast motion of the electron in atoms or molecules with attosecond time resolution.\textsuperscript{[1–3]} Such resolution for time is not achieved directly. Instead, a mapping relation between the experimental observable (e.g., the photoelectron momentum distribution (PMD)) and the time recording a specific physical event (e.g., the ionization time at which the bound electron is free) is needed. The classical or quantum electron-trajectory theory\textsuperscript{[4]} arising from the simple-man model (SM)\textsuperscript{[5–7]} or strong-field approximation (SFA)\textsuperscript{[8]} provides such mapping relations, which can be used to deduce the time information of a physical process from experiments.

However, these mapping relations are obtained with neglecting the Coulomb effect. Indeed, present studies have shown that the long-range Coulomb potential has significant influences on strong-field electronic dynamics.\textsuperscript{[4–11]} As a result, these relations cannot be directly applied to many real cases. Up to now, the lack of a Coulomb-calibrated mapping relation between the observable and the time corresponding to a definite physical event has led to a great debate on the explanation of experimental results in attosecond community, e.g., the results of attoclock experiments.\textsuperscript{[12–13]} As some experiments indicate that the distilled time in attoclock does not give evidences for the existence of tunneling time (i.e., the time the tunneling electron spends under the barrier)\textsuperscript{[12–17]}, other experiments support the opposite argument.\textsuperscript{[18–19]} These arguments have aroused widespread concern in the field of physics and naturally raised the question, that is, what is the time measured in attoclock?\textsuperscript{[20]}

Recent studies have also shown that the Coulomb effect can induce a large ionization time lag\textsuperscript{[21]} relative to SM and SFA predictions. The inclusion of the time lag into the SM mapping relation is able to qualitatively explain complex strong-field phenomena.\textsuperscript{[21–24]} However, a general theory description for this lag is not acquirable and the physical origin of this lag is unclear.

Here, based on numerical solution of the time-dependent Schrödinger equation (TDSE) and a Coulomb-modified SFA model (MSFA), we show that this lag reflects a finite response time, which is needed for the electron to change its state from the tunneling one to the ionized one after tunneling. An analytical expression for this lag is obtained, which quantitatively formulates the dependence of the response time on laser and atomic parameters (see Eq. (3)). With this expression, a simple Coulomb-calibrated mapping relation between photoelectron momentum and the ionization time is activated, which provides a new theoretical tool for studying experimental phenomena in strong-field ionization. The agreement between predictions of our theory and TDSE simulations and a series of experiments (e.g., see Fig. 3 and Fig. 4) strongly supports the following picture of tunneling ionization for atoms in strong laser fields. 1) Tunneling of the electron under the barrier is instantaneous. 2) After tunneling, the electron is not free instantly. Instead, a finite response time is needed for the electron to adapt to the ionization event. This picture bridges these contrary arguments about tunneling time.

\textbf{Theory.}—To introduce our theory, we choose ionization of atoms in strong elliptical laser fields with high ellipticity as the example, where some complex effects such as rescattering and quantum interference are less and we can focus on the effect of the response time on PMD.

Numerically, we solve the TDSE of the He atom in two-dimensional (2D) cases with the spectral method.\textsuperscript{[24]} The Coulomb potential used has the form of \( V(r) = -Z/\sqrt{x^2 + y^2 + 0.5} \) with the effective charge \( Z = 1.45 \) a.u. (in atomic units of \( \hbar = e = m_e = 1 \)). The elliptical electric field \( \mathbf{E}(t) \) has the form of \( \mathbf{E}(t) = f(t)[\hat{e}_x E_x(t) + \hat{e}_y E_y(t)] \), with \( E_x(t) = E_0 \sin(\omega t) \) and \( E_y(t) = E_1 \cos(\omega t) \). Here, \( E_0 = E_L/\sqrt{1+\epsilon^2}, \) \( E_1 = \epsilon E_L/\sqrt{1+\epsilon^2}, \) \( E_L \) is the maximal laser amplitude related to the peak intensity \( I \) and \( \epsilon \) is the ellipticity with \( \epsilon = 0.87. \) \( \omega \) is the laser frequency and \( f(t) \) is the envelope function. More numerical details can be found in [25].

According to the SM, the mapping relation between
The time \( \tau \) lag of the above expression indicates that with the offset mapping relation between the final momentum of each SFA electron trajectory \([5, 26]\), a Coulomb-modified solution of the Newton equation including both is the exit velocity of the electron at the exit position \( t_0 \) and the ionization time \( t_i \) (with \( |v_x(t_i)| > 0 \)) emerges (a). This lag reflects the response time of the electronic wave function to the ionization event. It can be evaluated with the expression of Eq. (3) and can also be read from the offset angle \( \theta \) in PMD (b) with the mapping relation of Eq. (1). With the knowledge of response time, the electron motion can be divided into these three steps of tunneling (I), response (II) and classical motion (III). These processes can be described with saddle-point, semiclassical and SM theories, respectively, raising a model which can be termed as TRCM. With TRCM, the PMD of SFA (c) can be directly transited into the Coulomb-modified one (d), in good agreement with TDSE (b) and without the need of solving Newton equation including Coulomb force. Laser parameters used are as shown.

Figure 1: Sketch of the Coulomb-induced ionization time lag for the MPR and its characterization in PMD. When the electron exits the laser-Coulomb-formed barrier at the peak time \( t_0 \) of the laser field (with the velocity \( v_x(t_0) = 0 \)), due to the existence of the Coulomb potential, it can not ionize immediately and a time lag \( \tau = t_i - t_0 \) between the exit time \( t_0 \) and the ionization time \( t_i \) (with \( |v_x(t_i)| > 0 \)) emerges (a). This lag reflects the response time of the electronic wave function to the ionization event. It can be evaluated with the expression of Eq. (3) and can also be read from the offset angle \( \theta \) in PMD (b) with the mapping relation of Eq. (1). With the knowledge of response time, the electron motion can be divided into these three steps of tunneling (I), response (II) and classical motion (III). These processes can be described with saddle-point, semiclassical and SM theories, respectively, raising a model which can be termed as TRCM. With TRCM, the PMD of SFA (c) can be directly transited into the Coulomb-modified one (d), in good agreement with TDSE (b) and without the need of solving Newton equation including Coulomb force. Laser parameters used are as shown.

The drift momentum \( p \) and the ionization time \( t_0 \) is \( p = -A(t_0) \). Here, \( A(t) \) is the vector potential of the electric field \( E(t) \). In SFA, it is \( p = v(t_0) - A(t_0) \) where \( v(t_0) \) is the exit velocity of the electron at the exit position \( r(t_0) [11] \). Based on a MSFA model [21], which is related to the solution of the Newton equation including both the electric-field force and the Coulomb force [21] for each SFA electron trajectory [3, 26], a Coulomb-modified mapping relation between the final momentum \( p_f = v(t \to \infty) \) and the ionization time \( t_i = t_0 + \tau \) is also obtained [22]. That is \( v(\infty) = v(t_i) - A(t_i) \). Here, \( \tau \) is the Coulomb-induced ionization time lag relative to \( t_0 \).

For the most probable route (MPR), which is related to the brightest part of PMD, the tunnel event occurs at the time \( t_0 \) with \( |E_x(t_0)| = E_0 \) and \( v_x(t_0) = 0 \). Then we can define the offset angle \( \theta \) related to MPR with [22]

\[
\tan \theta = \frac{v_x(\infty)}{v_y(\infty)} = A_x(t_i)/(A_y(t_i) - v_y(t_0)).
\]  

The above expression indicates that with the offset angle \( \theta \) measured in experiments, one can deduce the lag \( \tau = t_i - t_0 \). When \( \gamma_L \ll 1 \), with considering \(|v_x(t_0)/A_y(t_0)| \ll 1 \), we also have \( v(\infty) \approx -A(t_i) \). Then we have \( \tan \theta \approx A_x(t_i)/A_y(t_i) \). Below, we will call the expression that neglects \( v_x(t_0) \) “adiabatic Eq. (1)” [11]. Here, \( \gamma_L = w\sqrt{2l_F}/E_L \) is the Keldysh parameter [23]. A similar expression to the adiabatic one has been used in [22] to deduce the lag \( \tau \) and has been termed as Coulomb-calibrated attoclock (CCAC). Here, the theory description is given. A sketch of the lag \( \tau \) and the definition of the angle \( \theta \) in PMD are presented in Fig. 1.

For MPR, when the electron exits the barrier with \( |v_x(t_0)|^2/2 = 0 \) and \( V(r(t_0)) < 0 \), from the viewpoint of classic-quantum correspondence, it does not match an ionized state described in quantum mechanics. A finite response time is therefore needed for the electron to acquire some necessary kinetic energy before ionization. With considering the virial theorem, we assume \( |v_x(t_i)|^2 = [V(r(t_i))]|/n_f \approx |V(r(t_0))|/n_f \). Here, \( n_f = 2, 3 \) is the freedom degree of the system studied. Then we can obtain the analytical expression of the lag \( \tau \). That is [23]

\[
\tau \approx \sqrt{|V(r(t_0))/n_f|/E_0}.
\]  

The exit position \( r(t_0) \) can be evaluated with solving the saddle-point equation (SPE) \( p + A(t_0)]^2/2 = -I_p \). By neglecting the field \( E_x(t) \) in solving SPE, we also have \( x(t_0) \approx (E_0/\omega^2)[\sqrt{\gamma^2 + 1} - 1] \) and \( y(t_0) \approx 0 \) with \( \gamma = w\sqrt{2l_F}/E_0 \). In the single-active electron approximation, the potential \( V(r) \) for a hydrogen-like atom has the form of \( V(r) = -Z^+ / r \). Then we have [23]

\[
\tau \approx \sqrt{Z^+ \omega^2/[n_f E_0(\sqrt{\gamma^2 + 1} - 1)]}.
\]  

Figure 2: Comparisons with 2D-TDSE results of He for the offset angle and the time lag. The left (right) column shows results as a function of laser wavelength (ionization potential). The first row: predictions of 2D-TDSE and Eq. (1) for the offset angle, with \( \tau \) and \( v_x(t_0) \) in Eq. (1) evaluated using the exact solution of SPE. The second row: predictions of 2D-TDSE, CCAC and Eq. (3) for the lag.
Here, $Z^*$ is the effective charge. For real three-dimensional (3D) cases such as in experiments, $Z^*$ can be evaluated with $Z^* = \sqrt{2}p$. For 2D TDSE, the value of $Z^*$ can be chosen as that used in simulations. Equation (3) shows the dependence of $\tau$ on the laser and atomic parameters of $E_0$, $\omega$ and $I_p$. This dependency can be used to test the applicability of Eq. (3).

Once the lag $\tau$ is obtained with Eq. (2) or Eq. (3), we can also evaluate the angle $\theta$ through Eq. (1) with $\theta = \arctan(A_x(t_i)/A_y(t_i)-v_y(t_0))$ at $t_i = t_0 + \tau$. We do so with two manners, the exact one where we calculate the lag $\tau$ defined by Eq. (2) and the value of $v_y(t_0)$ both with the exact solution of SPE (Eq. (1) SPE), and the full analytical one where we calculate $\tau$ with Eq. (3) and $v_y(t_0)$ with $v_y(t_0) = [\sqrt{2p}/\text{arcsinh}(\gamma) - E_1/\omega] \sin \omega t_0$ obtained with neglecting the field $E_y(t)$ in solving SPE (Eq. (1) Analy.). Note, the analytical one is applicable for MPR and for a small $\gamma_L$. Next, we compare the theory predictions of $\tau$ and $\theta$ with TDSE and experiments.

**Comparisons with TDSE.-** We first compare our theory prediction with TDSE of 2D cases which allow us to explore a wide parameter region. In the first row of Fig. 2, we present the comparison for the offset angle. For a specific laser intensity, as increasing the laser wavelength, the offset angle of TDSE decreases. In addition, at a certain wavelength, the TDSE offset angle is larger for the case of the lower laser intensity, as seen in Fig. 2(a). The TDSE offset angle is not very sensitive to the change of $I_p$ for the present parameter region, with a small decrease as increasing $I_p$, as shown in Fig. 2(c). One can see that the theoretical predictions are very near to the TDSE ones and well reproduce the remarkable parameter-dependent phenomena.

Further comparisons for the lag $\tau$, obtained with Eq. (3), TDSE and CCAC, are presented in the second row of Fig. 2. In CCAC, we first obtain the offset angle from the PMD of TDSE simulations. Then we obtain the time $t_i$ through adiabatic Eq. (1) with $\theta = \arctan(A_x(t_i)/A_y(t_i)-v_y(t_0)) \sim \omega t_i$. In TDSE, we first find the time $t_i$ which corresponds to the maximal value of the instantaneous ionization rate $P(t) = dI(t)/dt$. Here, $I(t) = 1 - \sum_m |\langle m|\Psi(t)\rangle|^2$ is the instantaneous ionization yield, $|m\rangle$ is the bound eigenstate of $H_0 = p^2/2 + V(r)$ and $|\Psi(t)\rangle$ is the TDSE wave function. We only consider the first several bound eigenstates with $m = 0, 1, 2, ... 5$. The upper limit $m_{\text{m}}$ of $m$ is determined with the eigenenergy $E_{m_{\text{m}}+1}$ of the $(m_{\text{m}} + 1)$ th eigenstate agreeing with the semiclassical analysis in Eq. (2). That is $E_{m_{\text{m}}+1} \approx V(r(t_0)) + [v_y(t_0)]^2/2 \approx (3/4)V(r(t_0))$. Then the lag $\tau$ is obtained with $\tau = t_i - t_0$ at $|E_x(t_0)| = E_0$. One can observe for the broad parameter region, the difference for $\tau$ between results of Eq. (3) and TDSE or CCAC is near to or smaller than 10 attoseconds. This small difference between TDSE and Eq. (3) suggests the close correspondence for the definition of ionization between these two methods, and that between CCAC and Eq. (3) indicates that the lag $\tau$ (the response time) can be approximately evaluated with the relation $\tau \sim \omega$.

From Eq. (3), we also have $\tau \approx \sqrt{Z^*/(n_2 I_p E_0^4)}$ when $\gamma^2 \rightarrow 0$. For real 3D cases, with $Z^* = \sqrt{2p}$, we have $\tau \approx (1.5\sqrt{2p}E_0)^{-0.5}$ which indicates that $\tau$ is larger for smaller $I_p$. In practice, the ionization yield of the system depends strongly on $I_p$. This limits the $I_p$-dependent comparisons in Fig. 2 to a small parameter region. (For more discussions on effects of $\lambda$ and $I_p$ on $\tau$ and $\theta$ for both 2D and 3D cases, see Figs. S6-S7 in [25].)

**Comparisons with experiments.-** Deeper insights are obtained when comparing theory predictions with experi-
ments. Firstly, in Fig. 3, we compare the theoretical results of the offset angle with experimental data of He abstracted from [28]. For $I \geq 1.5 \times 10^{14}\text{W/cm}^2$, the curve of adiabatic Eq. (1), which neglects $v_0(t_0)$ and is related to the adiabatic intensity scaling in experiments, passes well through the corresponding experimental data. For lower intensities, it deviates from the experimental result, but reproduces the trend of the experimental angle which increases rapidly for lower intensities.

Next, we turn to the prediction of Eq. (1) which corresponds to the nonadiabatic intensity scaling in experiments. One can see that both the exact and analytical curves of Eq. (1) agree well with the nonadiabatic experimental data for $I \geq 1 \times 10^{14}\text{W/cm}^2$. The remaining difference between theory and experiment for cases of low intensities can be due to that the Coulomb effect plays a more important role in the momentum $p_f$ for these cases, and this role is underestimated in our theory.

In Fig. 4, we further compare the predictions of Eq. (3) for the lag $\tau$ with experimental and 3D-TDSE data of $H$ abstracted from [14]. For $I \leq 2 \times 10^{14}\text{W/cm}^2$, the predictions of Eq. (3) agree with 3D-TDSE simulations. For higher laser intensities, the predictions of Eq. (3) are somewhat higher than experimental and 3D-TDSE results, with a difference around 10 attoseconds. With using the exact solution of SPE to evaluate the exit position $r(t_0)$ in Eq. (2), or using the expression of $x(t_0) = x(t_0) + \Delta x$, which considers the displacement difference $\Delta x \approx v_x'(t_0)/(2E_0) \approx Z'/\langle 6I_p \rangle$, to replace $x(t_0)$ in Eq. (3), the calculated results become somewhat smaller and nearer to the experimental and 3D-TDSE results. The difference between our results and experiments at high laser intensities can be due to that the ionization of $H$ with $I_p = 0.5 \text{ a.u.}$ is also strong for high intensities. Our TDSE simulations show that Eq. (3) works better for cases of intermediate ionization probabilities.

We have also compared our theory predictions to more experimental and TDSE data with different targets such as $H$ [29], $\text{Ar}$ [14] and $\text{He}$ [17] and diverse laser parameters (see Figs. S1-S5 in [23]). In all cases, our response-time model manifested with Eqs. (1)-(3) well reproduces the main characteristics of relevant results. We therefore believe that it holds the essence of the physics behind the phenomena studied.

Discussions.-Our theory is based on the existence of a measurable response process in laser-atom interaction. This agreement between our theory, TDSE and experiments in a wide parameter region strongly supports the ansatz. Conversely, this agreement is also a direct exhibition of the ability of attoclock at ultrahigh time-resolution measurement, and gives a clear physical definition of the time measured in attoclock. Because the response process occurs after the tunneling electron exits the barrier, our results do not contradict with the argument that tunneling is instantaneous [12, 17, 28, 30]. As the response process also occurs before the tunneling electron is ionized, our results also stand for the argument that tunneling ionization needs finite times [18, 19, 31].

The response process discussed here can be described semiclassically, and the response time can be clearly defined with the classical expression of Eq. (2). This is different from the Wigner time delay, the definition of which depends on the phase of the wave function $\phi$.

Ionization is the first step of many strong-field processes. The existence of the response process in tunneling ionization will inevitably affect the following dynamics of the electron after tunneling, such as high-order harmonic generation (HHG) and high-order above-threshold ionization (ATI). This also provides a potential chance for probing the response process with HHG.

As the response process occurs near to the core of the target, one can anticipate that the interior structure of the target has an important effect on the process. One of such cases is the influence of the molecular structure and property on the process, raising the issue of molecular response time in tunneling ionization. Conversely, the internal information of the target is encoded in the response time and finally mapped to the PMD. This provides an opportunity for people to access this information from experimental observables.

Methodologically, Eq. (2) is applicable for the MPR. For a general SFA electron trajectory $(p_f, t_f)$ with the amplitude $c(p_f, t_f)$ [26], we have $\tau \approx \sqrt{\langle |v(r(t_0))|^2 / \langle p_f / E'(t_0) \rangle}$ with $E'(t_0) = \sqrt{(E_0 \sin \omega t_0)^2 + (E_1 \cos \omega t_0)^2}$. Then using the mapping relation $v(\infty) = v(t_0) - A(t_i - t_i + \gamma)$, one can directly obtain the Coulomb-modified PMD with the drift momentum $p_f = v(\infty)$ and the amplitude $c(p_f, t_i) \approx c(p(t_0))$, without the need of solving Newton equation. This approach, which can be called as TRCM (see Fig. 1), provides a simple and effective tool to study strong-field phenomena of a real system (see methods and Figs. S8-S9 in [27] for more details).

Conclusion.-In summary, we have addressed the subtle issue of electronic response to an intense-field ionization event. Semiclassical theory has been developed to study the response process, and a compact expression has been obtained to describe the response time. The response process occurs just between tunneling and ionization. It has a time scale of $\tau \sim 100$ as and has a space scale of $\Delta x \sim 0.3 \text{ a.u.}$. These scales are so small that this process is ignored before the present study. During the response process, the electron, which just exits a barrier through tunneling, acquires some necessary energy from the external field to match the Coulomb tail and recover its natural state distorted due to tunneling. Such energy encodes both space and time information of the response process and can be measured in experiments. The response process, serving as the boundary of quantum and classical, provides a narrow time-space window for “observing” the end of tunneling and “monitoring” the beginning of classical motion. Further studies on electronic ultrafast response to an event, associated with different targets (including gas, solid and liquid) and processes (including photoionization, light absorption and radiation), promise to open up new prospects for attosecond measurement.

We thank Y. F. He for discussions. This work was
supported by the National Natural Science Foundation of China (Grant Nos. 12174239, 11904072).

Appendix: Numerical method

The Hamiltonian of the He atom studied here has the form of \( H(t) = H_0 + \mathbf{E}(t) \cdot \mathbf{r} \) (in atomic units of \( h = e = m_e = 1 \)). Here \( H_0 = \mathbf{p}^2/2 + V(r) \) is the field-free Hamiltonian and \( V(r) = -Z/\sqrt{x^2 + y^2 + \xi} \) is the Coulomb potential in 2D cases. With the effective charge \( Z = 1.45 \) a.u. and the soft-core parameter \( \xi = 0.5 \) a.u., the ionization potential of the ground state of He reproduced here is \( I_p = 0.9 \) a.u.. The term \( \mathbf{E}(t) = f(t)[\mathbf{e}_x E_x(t) + \mathbf{e}_y E_y(t)] \) with \( E_x(t) = E_0 \sin(\omega t) \) and \( E_y(t) = E_1 \cos(\omega t) \) is the external electric field. Here, the term \( \mathbf{e}_x \) (\( \mathbf{e}_y \)) is the unit vector along the \( x \) \( (y) \) axis (i.e., the major (minor) axis of the polarization ellipse). The term \( \epsilon \) is the laser ellipticity, \( \omega \) is the laser frequency, and \( f(t) \) is the envelope function. \( E_0 = E_L/\sqrt{1+\epsilon^2} \), \( E_1 = \epsilon E_L/\sqrt{1+\epsilon^2} \), and \( E_L \) is the maximal laser amplitude related to the peak intensity \( I \) of the laser pulse. The value of \( \epsilon \) used here is \( \epsilon = 0.87 \). We use trapezoidally shaped laser pulses with a total duration of 15 optical cycles and linear ramps of three optical cycles. The TDSE of \( i\Psi(t) = H(t)\Psi(t) \) is solved numerically using the spectral method \([24]\). We work with a grid size of \( L_x \times L_y = 409.6 \times 409.6 \) a.u.. The space steps used are \( \Delta x = \Delta y = 0.4 \) a.u., and the time step is \( \Delta t = 0.05 \) a.u..

To avoid the reflection of the electron wave packet from the boundary and obtain the momentum space function, the coordinate space is split into the inner and the outer regions with \( \Psi(t) = \Psi_{in}(t) + \Psi_{out}(t) \), by multiplication using a coordinate function \( F(x,y) = F(x) \) for \( r_x < r_f \) and \( F(x,y) = 1 \) for \( r_x \geq r_f \). Here, \( r_x = \sqrt{x^2 + y^2/\epsilon^2} \), \( L_r = L_x \) and \( r_f = 150 \) a.u. is the absorbing boundary. In the inner region, the wave function \( \Psi_{in}(t) \) is propagated with the complete Hamiltonian \( H(t) \). In the outer region, the time evolution of the wave function \( \Psi_{out}(t) \) is carried out in momentum space with the Hamiltonian of the free electron in the laser field. The mask function is applied at each time interval of 0.5 a.u. and the obtained new fractions of the outer wave function are added to the momentum-space wave function \( \Psi_{out}(t) \) from which we obtain the PMD. Then we find the local maxima of the PMD and the offset angle \( \theta \) is obtained with a Gaussian fit of the angle distribution of local maxima.

Appendix: Analytical method

Analytically, we begin our discussions with a MSFA model introduced in \([21]\). First, according to the electron-trajectory theory based on the SFA, the mapping between the ionization time \( t_0 \) and the photoelectron momentum \( \mathbf{p} \) can be obtained with solving the following saddle-point equation \([3, 26]\):

\[
|\mathbf{p} + \mathbf{A}(t_s)|^2/2 = -I_p. \tag{A.1}
\]

Here, \( \mathbf{A}(t) \) is the vector potential of the electric field \( \mathbf{E}(t) \). The solution \( t_s \) of the above equation is complex which can be written as \( t_s = t_0 + i\tau \). The real part \( t_0 \) can be understood as the ionization time. The corresponding momentum-time pair \( (\mathbf{p}, t_0) \) have been termed as electron trajectory \([22]\). For \( I_p = 0 \), one can return to the classical SM mapping condition \( \mathbf{p} = -\mathbf{A}(t_0) \).

1. Coulomb-modified momentum-time mapping

To consider the Coulomb effect, one can further solve the following Newton equation \([3, 11]\)

\[
\ddot{\mathbf{r}}(t) = -\mathbf{E}(t) - \nabla_r V(r) \tag{A.2}
\]

for each SFA electron trajectory, with initial conditions \([11]\) of the exit velocity \( \mathbf{v}(t_0) = \dot{\mathbf{r}}(t_0) = \mathbf{p} + \mathbf{A}(t_0) \) and the exit position \( \mathbf{r}(t_0) = R \mathcal{I}_{t_0}^\infty \left[ \mathbf{p} + \mathbf{A}(t') \right] dt' \). This velocity condition reflects the quantum effect related to \( I_p \) on the momentum \( \mathbf{p} \). It also indicates the SFA mapping relation between time and momentum

\[
\mathbf{p} = \mathbf{v}(t_0) - \mathbf{A}(t_0). \tag{A.3}
\]

The integration of Eq. (A.2) from \( t = t_0 \) to \( t \rightarrow \infty \) gives the following expression \( \mathbf{v}(\infty) + \mathbf{A}(t_0) = \mathbf{v}(t_0) - \int_{t_0}^\infty \nabla_r V(r(t')) dt' \). Here, the term \( \mathbf{v}(\infty) \equiv \mathbf{p}_f = \ddot{\mathbf{r}}(t \rightarrow \infty) \) is the final Coulomb-modified drift momentum in the MSFA. As discussed in \([21]\), due to the existence of the Coulomb potential, the electron which exits the barrier at the time \( t_0 \) can not ionize immediately. Instead, a small period of time \( \tau \) is needed for the electron to acquire some additional kinetic energy from the laser field. Then it ionizes at \( t_i = t_0 + \tau \). According to Eq. (A.2), at the time \( t_i \), we also have

\[
\mathbf{v}(\infty) + \mathbf{A}(t_i) = \mathbf{v}(t_i) - \int_{t_i}^\infty \nabla_r V(r(t')) dt'. \tag{A.4}
\]

Here, the term \( \mathbf{v}(t_i) = \mathbf{v}(t_0) + \mathbf{A}(t_i) - \mathbf{A}(t_0) - \int_{t_0}^{t_i} \nabla_r V(r(t')) dt' \) is the instantaneous velocity at \( t_i = t_0 + \tau \). The concept of ionization implies the condition that the Coulomb potential plays no role or plays a small role in the motion of the electron after it is ionized. The following mapping relation guarantees such a condition

\[
\mathbf{v}(\infty) + \mathbf{A}(t_i) = \mathbf{v}(t_0); \tag{A.5}
\]

$$\mathbf{v}(t_i) - \int_{t_0}^{t_i} \nabla_r V(r(t')) dt' = \mathbf{v}(t_0).$$
of $\tau$ relative to the time $t_0$ of the general SFA prediction. The second expression $v(t_i) = v(t_0) + \int_{t_0}^{t_i} \nabla_x V(r(t')) dt'$ indicates that due to the work of the electric field in the period of $\tau$, the electron has a nonzero velocity at the time $t_i$. This velocity is able to compensate the impulse of the Coulomb force from $t_i$ to $t \to \infty$ so that the effect of the Coulomb potential on $p_f$ can be neglected after $t_i$. We therefore need to consider the Coulomb effect on $p_f$ only in the time region of $t_0$ to $t_i$. It should be stressed that we are doing discussions for the cases of elliptical laser fields with high ellipticity, for which a strong rescattering event does not appear.

2. Coulomb-modified angle-time mapping

In experiments, especially for the attoclock experiment, people are usually interested in the brightest part of PMD related to the most probable route (MPR). This route corresponds to the electron trajectory $(p, t_0)$ with the time $t_0$ around the peak of $E_x(t)$. Some properties of MPR obtained from MSFA-based analyses are as follows. Firstly, the initial velocity $v_z(t_0)$ for MPR is zero, i.e., $v_z(t_0) = 0$, and that of $v_y(t_0)$ has a nonzero value arising from the nonadiabatic effect [28]. When $\gamma_L \approx 1$, the value of $|v_y(t_0)|$ is comparable to $|A_y(t_0)| = E_1/\omega$, but for $\gamma_L \ll 1$, $|v_y(t_0)|/|A_y(t_0)| \ll 1$. Here, $\gamma_L = w\sqrt{2\gamma p}/E_L$ is the Keldysh parameter [27]. Secondly, the impulse of the Coulomb force is mainly along the direction of the main axis $E_x$ of the polarization ellipse. This implies that $\int_{t_0}^{t_\infty} \nabla_x V(r(t')) dt' \neq 0$ and $\int_{t_0}^{t_\infty} \nabla_y V(r(t')) dt' \approx 0$. By the second expression of Eq. (A.5), we have $v_x(t_i) \approx v_y(t_0)$. We therefore need to focus mainly on $v_x(t_i)$ in the following discussions. Thirdly, the lag $\tau$ is small with a scale of about 100 attoseconds [21]. This implies that the displacement difference $\Delta x = x(t_i) - x(t_0)$ is also small, we therefore have $|E_x(t_i)| \approx |E_x(t_0)| = E_0$, $|v_x(t_i)| \approx |A_x(t_i)| > |A_x(t_0)| = 0$ and $V(r(t_i)) \approx V(r(t_0))$.

The above analyses for MPR show that at the ionization time $t_i = t_0 + \tau$, the electron still has the potential energy $V(r(t_0))$, which is finally offset by the work of the electric field in the time region of $t_i$ to $t \to \infty$, as revealed by the second expression in Eq. (A.5). From the viewpoint of quantum mechanics, this implies that at the ionization time $t_i$, the electron is still located at a bound state with the average potential energy $V(r(t_0))$. But for the laser-driven time-dependent system, such a bound state can be considered an ionized state, since the potential energy of the bound state can be fully compensated by the work of the laser field when $t \to \infty$. Below, this property will be used to deduce the analytically expression of $\tau$.

For MPR, considering that $v_z(t_0) = 0$, with the Coulomb-modified mapping relation $v(\infty) = v(t_0) - A_x(t_i)$ of Eq. (A.5), we can define the offset angle related to MPR with (Eq. (1) in the main text)

$$\tan \theta = v_x(\infty)/v_y(\infty) = A_x(t_i)/(A_y(t_i) - v_y(t_0)).$$

The above expression also indicates the Coulomb-modified mapping relation between the offset angle $\theta$ and the ionization time $t_i = t_0 + \tau$. It stands as a deduction of Eq. (A.5). Through the expression, one can deduce the lag $\tau$ with the offset angle obtained in experiments or TDSE simulations. When $\gamma_L \ll 1$, the absolute value of $v_y(t_0)$ is also far smaller than that of $A_y(t_0)$, the Coulomb-modified mapping relation of Eq. (A.5) can be approximated as $p_f = v(\infty) \approx -A(t_i)$. To do so, we in fact introduce the lag $\tau$ into the SM mapping relation $p = -A(t_0)$. Then we have

$$\tan \theta \approx A_x(t_i)/A_y(t_i).$$

This expression can be understood as the adiabatic version of Eq. (1) in the main text.

3. Response time and its expression

To further understand the physics behind Eq. (A.5), next, we explore the analytical expression of the lag $\tau$ for MPR. Specifically, we find the relation between the lag and the laser and atomic parameters and obtain the expression of $\tau$ which is independent of the final observation $p_f = v(\infty)$. To do so, first, we evaluate the value of $v_x(t_i)$. For a hydrogen-like atom, according to the virial theorem, the average kinetic energy $\langle E_k \rangle$ of the electron at a bound state is a half of the negative average potential energy $V(r)$.

For real three-dimensional (3D) cases, that is $\langle E_k \rangle = (p_x^2/2 + p_y^2/2 + p_z^2/2) = 3(p_z^2)/2 = -(V(r))/2$. For 2D cases, it is $p_y^2 = -\langle V(r) \rangle/2$. As a classical correspondence to this quantum property, we assume $|v_x(t_i)|^2 = |V(r(t_i))|/n_f \approx |V(r(t_0))|/n_f$. Here, $n_f = 2, 3$ is the freedom degree of the system studied. For MPR, the Coulomb force $\nabla_x V(r)$ at the exit position $r(t_0)$ is far smaller than the electric-field force $|E_x(t_0)| = E_0$. Therefore, in the small time interval of $t_0$ to $t_i = t_0 + \tau$, the main impulse on electron comes from the force of the electric field. The electric-field impulse from $t_0$ to $t_i$ can be written as $E_0 \tau \approx |v_x(t_i)| \approx \sqrt{|V(r(t_0))|/n_f}$. Then we obtain (Eq. (2) in the main text)

$$\tau \approx \sqrt{|V(r(t_0))|/n_f/E_0}.$$

With neglecting the field $E_y(t)$ in solving Eq. (A.1), the exit position $r(t_0)$ can be approximated as $x(t_0) \approx (E_0/\omega^2)^{1/(\sqrt{\gamma^2 + 1} - 1)}$ and $y(t_0) \approx 0$ with $\gamma = w\sqrt{2\gamma p}/E_0$. In the single-active electron approximation, the potential $V(r)$ for a hydrogen-like atom has the form of $V(r) = -Z^*/r$. Then we obtain (Eq. (3) in the main text)

$$\tau \approx \sqrt{Z^*\omega^2/[n_f E_0^2(\sqrt{\gamma^2 + 1} - 1)].$$

Here, $Z^*$ is the effective charge. For real 3D cases such as in experiments, the value of $Z^*$ can be evaluated with $Z^* = \sqrt{2\gamma p}$. For TDSE simulations, the value of $Z^*$ can
be chosen as that of $Z$ in the expression of $V(r)$ used in calculations. Equation (3) shows that the lag $\tau$ decreases with the increase of the laser amplitude $E_0$ and the laser wavelength $\lambda$ (the decrease of the laser frequency $\omega$) on the whole. It also shows that in TDS simulation, the value of $\tau$ is larger in 2D cases than 3D ones. For $\gamma \ll 1$, we also have $x(t_0) \approx (I_p/E_0)[1-\gamma^2/4]$. In Eq. (3), $\gamma$ can be further approximated as $\tau \approx \sqrt{(V(r(t_0)))^2 + V(t_0)}$. The derivation of Eq. (3) indeed reveals the physical meaning of the lag $\tau$, namely, it reflects the response time of the electronic wave function to the strong-field ionization event. Specifically, when the electron tunnels out of the barrier at the time $t_0$, it has minus potential energy $-V(r(t_0))$ but is absent about kinetic energy. The lag is needed to make the electron acquire some specific kinetic energy $[x_z(t_0)]^2/2 \approx |V(r(t_0))|/2n_f$ such that it matches a certain high-energy bound state of the field-free Hamilton $\text{H}_0 = p^2/2 + V(r)$. For the time-dependent system driven by the strong laser field, such a bound state behaves similarly to an ionized state.

It should be noted that Eq. (3) is applicable only for the long-range Coulomb potential. For a short-range potential, $V(r(t_0)) \to 0$ and therefore $\tau \to 0$. In addition, the appearance of the lag $\tau$ is due to the nonvanishing Coulomb tail $V(r)$ at the tunnel exit $r(t_0)$. However, as discussed in Eq. (A.5), the impulse of the Coulomb force basically plays no role in the final momentum $p_f = v(t_0) - A(t_0 + \tau)$ while the impulse of the electric field in the small time interval of $\tau$ does so. Considering that at the tunnel exit, the electric-field force is generally one order of magnitude larger than the Coulomb one, one can expect that the direct influence of the Coulomb potential on the electronic motion can be neglected just as treated in SFA. It is the response process, where the state of the tunneling-out electron rapidly

**4. TRCM model**

Equation (2) is obtained for MPR. For general SFA electron trajectories ($p,t_0$), similarly, we have

$$\tau \approx \sqrt{|V(r(t_0))|/n_f/E'(t_0)}. \quad (A.7)$$

Here, $E'(t_0) = |E(t_0)|$ is the amplitude of the laser electric field $E(t_0)$ at the time $t_0$. For the present elliptically polarized case explored here, that is $E'(t_0) = \sqrt{(E_0 \sin \omega t_0)^2 + (E_1 \cos \omega t_0)^2}$. Once the lag $\tau$ is obtained, using the first expression of Eq. (A.5), we can obtain the Coulomb-modified drift momentum $p_f = v(\infty) = v(t_0) - A(t_0)$ with $t_i = t_0 + \tau$ and $v(t_0) = p + A(t_0)$. Assuming that the amplitude $c(p_f,t_i)$ for the Coulomb-modified electron trajectory $(p_f,t_i)$ is equivalent to the corresponding amplitude $c(p,t_0) \sim e^b$ for the SFA trajectory $(p,t_0)$ \cite{29}, we can obtain the Coulomb-modified PMD directly from the SFA without the need of solving Eq. (A.2). Here, $b$ is the imaginary part of the quasiclassical action $S(p,t_j) = \int_{t_j}^{t_f} (\mathbf{p} + A'(r'))^2/2 + I_p \mathbf{d}r'$ with $t_s = t_0 + it$. As the above approach naturally arises from the simple picture of tunneling, response and classic motion of the strong-laser-driven electron of a real atom, depicted in Fig. 1 in the main text, we would like to call it TRCM. This TRCM approach provides a new and simple tool to study strong-field phenomena of real atoms. It bypasses the integration of Eq. (A.2) including the complex interplay of the electric-field force and the Coulomb force. This integration calls for a high accuracy of numerical calculations and is time consuming in 3D cases.
mental and 3D-TDSE data for laser intensities lower than prediction of Eq. (1) with considering the initial velocity
parison for the offset angle. One can observe that the main text. In Fig. S1, we further show the com-
the offset angle. Blue square dots: experimental results in [14]. Orange dotted and gray lines: predictions of Eq. (1) with 
...deviates from the theory prediction. In [29], it has been shown that this remarkable decrease is associated with 
the large depletion of the ground-state population indicating the strong ionization of the system in these cases.

From Figs. S1 and S2, one can also observe that the theory predictions of Eq. (1) with \( \tau \) and \( v_0(t_0) \) evaluated using the exact solution of SPE (Eq. (1) Analy.) are similar at intermediate and high laser intensities and differ somewhat from each other for low intensities. As most of at-
toclock experiments are performed at intermediate laser intensities, in practice, one can calculate the response 
time (the lag \( \tau \)) and the velocity \( v_0(t_0) \) with the full analytical manner, and bypass the numerical solution of saddle-point equation.

2. Cases of He

In Fig. S3, we present comparisons of He for the offset angle with experimental results in [14] over a wider range of laser intensity. It can be seen that for laser intensities lower than \( I = 1.4 \times 10^{15}\text{W/cm}^2 \), the theoretical curves basically fall within the error range of the experimental data. For higher laser intensities, there is a difference of about 2 degrees between theory and experimental results.

Figure S3: Comparisons with experimental results of He for the offset angle. Blue square dots: experimental results in [14]. Orange dotted and gray lines: predictions of Eq. (1) with \( \tau \) and \( v_0(t_0) \) evaluated using the exact solution of SPE (orange dotted) or using full analytical expressions (gray). The laser wavelength used is \( \lambda = 740\text{ nm} \) and \( \epsilon = 0.78 \).

Figure S4: Comparisons with experimental results of Ar for the offset angle. Blue square dots: experimental results in [14]. Orange dotted and gray lines: predictions of Eq. (1) with \( \tau \) and \( v_0(t_0) \) evaluated using the exact solution of SPE (orange dotted) or using full analytical expressions (gray). The laser wavelength used is \( \lambda = 740\text{ nm} \) and \( \epsilon = 0.78 \).

Appendix: Extended comparisons

In the main text, we have compared the predictions of Eqs. (1)-(3) with experimental and TDSE results of He and H atoms. Here, comparisons of our theory with more acquirable experimental and TDSE data associated with different targets and laser parameters are presented.

1. Cases of H

In [16], the offset angle \( \theta \) of experimental and 3D-TDSE results and the deduced time delay \( \tau \) are plotted in a figure with the mapping relation \( \theta \approx \omega \tau \). In our approach, the time lag \( \tau \) is predicted by Eq. (2) or Eq. (3) and the offset angle \( \theta \) is deduced from the lag \( \tau \) with the mapping relation of Eq. (1) which is somewhat different from the relation \( \theta \approx \omega \tau \). Relevant comparisons for the time delay (lag) have been shown in Fig. 4 in the main text. In Fig. S1, we further show the comparison for the offset angle. One can observe that the prediction of Eq. (1) with considering the initial velocity \( v_0(t_0) \) (nonadiabatic treatment) agrees with the experimental and 3D-TDSE data for laser intensities lower than \( I = 2 \times 10^{14}\text{W/cm}^2 \), similar to the case of the time delay in Fig. 4 in the main text. There is a difference of about two degrees for higher intensities. The potential reason may be that the ionization is strong for H at these laser intensities. It is worth noting that for \( I > 2 \times 10^{14}\text{W/cm}^2 \), different from cases of low laser intensity, the experimental and TDSE offset angles decrease slowly with the increase of laser intensity. This trend agrees with our theory prediction.

Comparisons with 3D-TDSE results in [29] calculated over a wider parameter range of laser intensity give further supports to our theory, as shown in Fig. S2.

It can be seen that our theory predictions agree well with the TDSE results from \( I = 0.4 \times 10^{14}\text{W/cm}^2 \) to \( I = 2.5 \times 10^{14}\text{W/cm}^2 \). For higher laser intensities, the TDSE curve shows a remarkable trend of decrease and deviates from the theory prediction. In [29], it has been shown that this remarkable decrease is associated with the large depletion of the ground-state population indicating the strong ionization of the system in these cases.
experimental data, suggesting that our approach can also

be surprising that our theory predictions also agree with the
ing dynamics of the molecule. In this situation, it is not
does not influence remarkably on strong-field tunnel-

angles data of Ar and H

S5 are also similar to those in Fig. S4. Both the measured
Ar. One can observe that the experimental results in Fig.
1.4 eV which is similar to

2 degrees.

3. Cases of Ar

Comparisons for Ar are presented in Fig. S4. The ex-
perimental offset angles taken from Ref. [14] show a re-
markable increase as decreasing the laser intensity. This
phenomenon is well reproduced by our theory. For higher
laser intensities, the theory curves are still slightly higher
than the experimental one, with the difference of 1 degree
to 2 degrees.

4. Cases of H₂

Besides atoms, we have also performed comparisons
with acquirable experimental data for H₂ [17]. Relevant
results are presented in Fig. S5. For the small molecule
H₂ with the internuclear distance \( R = 1.4 \) a.u., it has the
ionization potential of \( I_p = 15.6 \) eV which is similar to
Ar. One can observe that the experimental results in Fig.
S5 are also similar to those in Fig. S4. Both the measured
angle data of Ar and H₂ decrease from about 16 degrees
to about 8 degrees as increasing the laser intensity in a
similar parameter range. These results suggest that for
the H₂ molecule with a small \( R \), the molecular prop-
ties do not influence remarkably on strong-field tun-
eling dynamics of the molecule. In this situation, it is not
surprising that our theory predictions also agree with the
experimental data, suggesting that our approach can also
be applied to molecules with a small \( R \).

Appendix: Effects of wavelengths and ionization
potential

Present experiments mainly focus on the effect of laser
intensity on the tunneling dynamics. In Fig. 2 in the

main text, we have shown the effects of laser wavelength
and ionization potential on the offset angle \( \theta \) and the time
lag \( \tau \). Here, we give more discussions on these issues.

In Fig. S6, we show the theoretical predictions of \( \theta \) and
\( \tau \) for different atoms in 3D cases, calculated with the full
analytical approaches both for \( \tau \) and \( v_y(t_0) \) introduced
above. Firstly, as we fix the laser wavelength and increase
the laser intensity (the first column), or with the contrary
manipulation (the second column), the calculated offset
angles and the time lags of the targets both decrease,
but the lag decreases slowly with the increase of the wave-
length. In particular, in all of cases, for fixed laser param-
eters, the calculated angles and lags are larger for atoms
with smaller \( I_p \). This phenomenon can be understood
with considering the limit case of \( \tau \approx (1.5 \sqrt{2I_pE_0})^{-0.5} \)
at \( \gamma^2 \to 0 \). This expression shows that the lag \( \tau \) de-
creases with increasing \( I_p \). For the elliptical laser field
with a high ellipticity, the approximation \( \theta \sim \omega \tau \) also
holds. Therefore, as changing \( I_p \), the offset angle \( \theta \) be-
haves similarly to the lag \( \tau \). As a case, in each panel of
Fig. S6, we show the corresponding limit result for He. In comparison with full analytical results, the limit
results are remarkably lower for cases of lower laser in-
tensities and shorter laser wavelengths corresponding to
larger values of the parameter \( \gamma = \omega \sqrt{2I_p/E_0} \), but ap-
proach the full analytical ones for cases of small \( \gamma \). As in
the limit case, the value of $\tau \approx (1.5 \sqrt{2 T_0 E_0})^{-0.5}$ does not depend on $\omega$, the lag $\tau$ generally has a weak dependence on laser wavelength for smaller values of the parameter $\gamma$. Because of $\theta \sim \omega \tau$, the offset angle $\theta$ decreases as the laser wavelength increases.

It should be stressed that in experiments, the $I_p$-dependent phenomena discussed above can change, since the ionization probability of a system depends strongly on its ionization potential.

More insights into roles of laser wavelength and ionization potential in the offset angle are obtained when we compare the drift momentum $(p_x, p_y)$ of MPR between TDSE and theory predictions. Relevant results are shown in Fig. S7 and the laser parameters used are as in Fig. 2 in the main text. Firstly, the theory predictions by our model agree well with TDSE ones both for $p_x$ and $p_y$. Secondly, when fixing the laser intensity and increasing the laser wavelength (the left column), the value of $p_x$ almost does not change and the value of $p_y$ increases. Therefore, it is the wavelength dependence of $p_y$ that mainly contributes to the wavelength dependence of the offset angle here. Thirdly, for the present parameter region, both the values of $p_x$ and $p_y$ are insensitive to the small change of the ionization potential. As a result, the offset angle is also insensitive to $I_p$. The results shed light on $\lambda$ and $I_p$-dependent phenomena in Fig. 2 in the main text. Note, the TDSE results presented here are obtained with simply finding the peak of PMD and therefore show somewhat small fluctuations.

Appendix: Applicability of TRCM

As discussed in the method part, the TRCM is able to consider the effect of Coulomb potential on the electronic dynamics, without the need of solving Newton equation including both the electric-field force and the Coulomb force. An introductory comparison between TDSE and TRCM has been shown in Fig. 1 in the main text. Here, we give more discussions on TRCM.

In Fig. S8, we show the comparisons between predictions of TDSE and TRCM at two different laser intensities in elliptical laser fields. One can observe that for both intensity cases, the TRCM well reproduces the main characteristics of PMD of TDSE, with showing a ring-like distribution and a remarkable offset angle $\theta$ (indicated by the white arrows). This angle predicted by TRCM is very close to the TDSE one, as shown by the numbers. The radius (size) of PMD of TRCM is also similar to the TDSE one.

In Fig. S9, we also show the results of PMD for He in an orthogonal ($\omega + 2\omega$) two-color (OTC) laser field, obtained with different methods. The result of TDSE in Fig. S9(a) shows a butterfly-like structure with a striking up-down asymmetry. This structure is absent in SFA
By comparison, the MSFA result in Fig. S9(d), where a absorbing procedure is used to eliminate the contributions of the rescattering electrons with distances \( r \) to the core smaller than 4 a.u., is similar to the TDSE one, with showing small amplitudes around the origin. We therefore expect that the recapturing process, which occurs when the rescattering electron moves to a distance near to the core, plays a dominating role in the distribution of TDSE around the origin. This process is not considered in TRCM. The TRCM directly maps the SFA electron trajectory \( (\mathbf{p}, t_0) \) into the Coulomb-modified one \( (\mathbf{p}_f, t_1) \) through Eq. (A.5) and Eq. (A.7) with \( E'(t_0) = \sqrt{(E_0^2 \sin \omega t_0)^2 + (E_1 \sin(2\omega t_0 + \phi))^2} \). Here, \( E_0 \) is the amplitude of the fundamental field, \( E_1 = \varsigma E_0 \) is that of the second-harmonic field, and \( \phi \) is the phase difference between these two colors.

The agreement between predictions of TDSE and TRCM for PMD shown above gives direct evidence to the applicability of Eq. (A.7) and Eq. (A.5). Equation (A.7) stands for the time scale of the response process associated with a certain electron trajectory \( (\mathbf{p}, t_0) \), and Eq. (A.5) shows that the complex Coulomb effect can be attributed to a simple temporal correction (quantified by the response time \( \tau = t_1 - t_0 \)) to the electron trajectory.

It should be stressed that the TRCM approach introduced here is applicable for cases where the rescattering effect is not strong, such as the case of the elliptical laser field with a relatively large ellipticity. For cases where the rescattering effect is strong (i.e., the Coulomb potential has an important role in the dynamics of the rescattering electron when it approaches this nucleus), such as the case of the orthogonal \( (\omega + 2\omega) \) two-color laser field discussed above, the TRCM can still give a good description of the main characteristic of the PMD. In this case, the difference between TDSE and TRCM mainly comes from the effects which are neglected in TRCM, such as the recapturing (recombination) and the back rescattering. With further including these effects into TRCM, we expect that the TRCM can also be used to explain relevant phenomena.

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**Figure S9:** Extended comparisons for cases of orthogonal two-color laser fields. Here, we show PMDs of He obtained with 2D-TDSE (a), SFA (b), TRCM (c) and MSFA (d) in an orthogonal \((\omega + 2\omega)\) two-color laser field. The laser field has the form of \( E(t) = f(t)[\vec{e}_x E_x(t) + \vec{e}_y E_y(t)] \) with \( E_x(t) = E_0 \sin(\omega t) \) and \( E_y(t) = \varsigma E_0 \sin(2\omega t + \phi) \). The laser parameters used are \( I = 5 \times 10^{14} \text{W/cm}^2 \) \((E_0 \approx 0.12 \text{ a.u.})\), \( \lambda = 1000 \text{ nm} \) \((\omega \approx 0.0456 \text{ a.u.})\), \( \varsigma = 0.5 \) and \( \phi_0 = \pi/2 \). The log_{10} scale is used here.

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