

# Coulomb-corrected reconstruction of ionization and recombination times in high-order harmonic generation with orthogonally polarized two-color fields

Mohammad Monfared<sup>\*</sup> and Manfred Lein<sup>†</sup>

*Institute of Theoretical Physics, Leibniz University Hannover, Appelstrasse 2, 30167 Hannover, Germany*



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Accurate knowledge about electron dynamics in strong-field phenomena is crucial for understanding ultrafast processes at the attosecond scale. In this work, we improve the time-retrieval method for extracting electron ionization and return times from experimental observables in high-order harmonic generation. We address an existing approximation for the condition to maximize the harmonic intensity, and we introduce an accurate alternative equation. Furthermore, we introduce a numerical approach to incorporate Coulomb corrections into the retrieval procedure by calculating the Coulomb-induced shift of the electron velocity via integration of the Coulomb force, and the Coulomb action via integration of the Coulomb potential, both along the electron trajectories. Our refined method significantly improves the precision and reliability of the retrieved electron dynamics, establishing a robust foundation for future attosecond-scale investigations.

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## I. INTRODUCTION

The field of ultrafast science has seen remarkable strides in probing, understanding, and controlling ultrafast quantum phenomena, with advances in laser-based techniques enabling time measurements on the attosecond scale. Among these techniques, high-order harmonic generation (HHG) has emerged as a key tool for the production of coherent extreme ultraviolet (XUV) radiation and for the study of ultrafast quantum dynamics such as electron and nuclear dynamics in gases, liquids, and solids [1–3]. HHG has been widely used for various purposes, including the generation of isolated attosecond pulses [4,5], ultrafast spectroscopy [6], the study of tunneling dynamics [7], the characterization of molecular structure [8,9] and molecular orbital tomography [10–12], the tracking and control of charge migration [13,14], and the investigation of electron and lattice dynamics [15–18] in solids.

HHG is commonly described as a three-step process [19,20], where an electron is first freed by a strong laser field, then accelerated by the field, and finally recombined with the parent ion, emitting a high-energy photon. Measurement of ionization and return times in HHG is essential to understand the underlying electron trajectories and their role in the emission of high-order harmonics. Accurate knowledge of electron trajectories provides deeper insight into attosecond phenomena and enhances the capabilities of HHG-based ultrafast spectroscopy.

In 2012, the ionization and return times in the HHG process were experimentally measured using an orthogonally polarized two-color (OTC) laser field [21]. For that purpose, a set of two equations was used as time-retrieval equations along with two experimental observables as inputs to reconstruct the ionization and return times. Within this method, a weak second-harmonic field, polarized perpendicular to the driving field, was introduced to laterally perturb the electron motion after ionization. By adjusting the phase delay between the two fields, the electron trajectory corresponding to each harmonic order can be controlled to reach two distinct optimal phases: the optimal phase  $\phi_h$  that maximizes the harmonic intensity, and the optimal phase  $\phi_a$  that maximizes the return angle at the moment of recombination. These two optimal phases act as key input observables for the time-retrieval equations, enabling the reconstruction of both ionization and recombination times. In experiment,  $\phi_h$  is obtained by scanning the two-color delay  $\phi$  and locating the maximum of the harmonic-yield modulation, typically constructed from the *sum of adjacent even and odd harmonics*, while  $\phi_a$  is obtained from the same delay scan by locating the maximum of the *ratio between adjacent even and odd harmonics*, which provides a measure of the return angle in the OTC geometry. In the pioneering experimental and theoretical studies of Refs. [21,22], the influence of the Coulomb force on the liberated electron was neglected in the time-retrieval equations. However, Coulomb effects are predicted to play a significant role in HHG [23,24]. In particular, the analytical R-matrix (ARM) model predicts that the Coulomb interaction can shift the ionization and return times to earlier times by approximately 33–37 and 5–19 attoseconds (as), respectively, for a helium atom driven by an 800 nm field [23]. Despite this, theoretical studies demonstrate that ionization times can be extracted with good accuracy using higher-frequency probe fields ( $\geq 4\omega$ , where  $\omega$  is the fundamental field frequency), even when Coulomb effects are neglected in the retrieval formalism [25].

<sup>\*</sup>Contact author: [m.monfared@yahoo.com](mailto:m.monfared@yahoo.com)

<sup>†</sup>Contact author: [lein@itp.uni-hannover.de](mailto:lein@itp.uni-hannover.de)

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The results of the high-frequency probing are in good agreement with the ARM theory. In fact, at high probe frequencies, the Coulomb terms in the time-retrieval equations become negligible, and the Coulomb influence is instead reflected in the optimal phases. Conversely, for lower probe field frequencies, such as  $2\omega$  with an 800 nm fundamental field, neglecting the Coulomb effect introduces an error of approximately 35 as in the retrieved ionization time [25,26]. Recently, Yue *et al.* attempted to incorporate the Coulomb effect into the time-retrieval equations by considering a Coulomb-induced instantaneous kick in the electron velocity perpendicular to the fundamental field [26]. While the results from such a time retrieval showed a shift of the ionization times toward earlier times (8–20 as), they still deviated from the ARM predictions by 13–27 as, highlighting that treating the Coulomb effect as a Coulomb kick is not sufficiently accurate.

The extraction schemes for the ionization and return times have two main applications. First, they facilitate the measurement of these times when applied to experimental data. Second, when applied to exact numerical solutions of the time-dependent Schrödinger equation (TDSE), they allow us to check whether the analytical models are accurate. In experiments, the measured HHG signal generally includes macroscopic propagation effects; however, these can be minimized and the retrieval can be related to the single-atom response by selecting the emission dominated by the short trajectory. This can be achieved using standard phase-matching conditions together with on-axis (low-divergence) spatial selection [27,28] and a thin interaction region (e.g., a short gas jet) [29,30], so that propagation mainly acts as a trajectory filter and does not significantly distort the phase-dependent observables used for the retrieval.

In this study, we refine the time-retrieval equations used in the OTC method by a more accurate treatment of the Coulomb force. Unlike Ref. [26], which treated the velocity change induced by the Coulomb force as an initial kick under the assumption of constant laser electric field, our approach incorporates the Coulomb force by numerical integration over electron trajectories. The optimal phases, which serve as inputs to the time-retrieval equations, are obtained using the TDSE. Our results demonstrate reasonable agreement between the retrieved ionization times and the ARM predictions in both the low- and high-frequency regimes. We note that, for the same driving laser parameters as those used in the present work, classical trajectory calculations based on Newton's equation were previously shown to yield times very close to the ARM predictions [25,26]. Therefore, we use ARM as the primary reference for benchmarking the retrieved times in this work. Additionally, we clarify that one of the equations used in the time-retrieval method of prior studies, specifically the one related to maximizing the total harmonic intensity, is an approximation that can lead to errors of up to 11 as in the retrieval of the ionization time for certain harmonic orders. We introduce an accurate equation to replace this approximation and obtain retrieved times in better agreement with the ARM model. These refinements improve the accuracy and robustness of the time-retrieval method, establishing a more reliable foundation for future investigations.

The article is organized as follows. Section II describes the numerical method for solving the TDSE, which is used

to obtain the optimal phases as input observables, and the theoretical framework of the time-retrieval method applied to extract the ionization and recombination times. The results of our analysis are presented and discussed in Sec. III, followed by a summary of the key findings and conclusions in Sec. IV.

## II. THEORETICAL MODEL

### A. Time-dependent Schrödinger equation

The TDSE in the length gauge and dipole approximation is given by (atomic units are used unless otherwise stated)

$$i\frac{\partial}{\partial t}\psi(\mathbf{r}, t) = \left[-\frac{1}{2}\nabla^2 + V(\mathbf{r}) + \mathbf{r} \cdot \mathbf{E}(t)\right]\psi(\mathbf{r}, t). \quad (1)$$

Here,  $\psi(\mathbf{r}, t)$  denotes the time-dependent electron wave function at position  $\mathbf{r}$ . A two-dimensional soft-core potential,  $V(\mathbf{r}) = -1/\sqrt{\mathbf{r}^2 + a}$ , with a softening parameter  $a = 0.0684$  is employed to model the helium (He) atom, which has an ionization potential of  $I_p = 24.6$  eV. The electric field of the OTC pulse,  $\mathbf{E} = -\partial_t \mathbf{A}$ , is described by the vector potential as follows:

$$\mathbf{A}(t) = -\frac{E_0}{\omega}f(t)\left(\sin(\omega t)\hat{\mathbf{e}}_x + \frac{\epsilon}{n}\sin(n\omega t + \phi)\hat{\mathbf{e}}_y\right), \quad (2)$$

where  $\hat{\mathbf{e}}_x$  and  $\hat{\mathbf{e}}_y$  are unit vectors. The parameters  $E_0$  and  $\omega$  denote the peak field amplitude and frequency of the fundamental field. The pulse consists of three optical cycles of period  $T = 2\pi/\omega$ . The probe field is characterized by the frequency  $n\omega$  and the field amplitude  $\epsilon E_0$ . The relative phase  $\phi$  (two-color delay) controls the delay between the fundamental and probe fields. The envelope function  $f(t)$  includes a one-cycle ramp-on, a one-cycle flat top, and a one-cycle ramp-off, with the ramps shaped by a  $\cos^6(\omega t/4)$  function. The flat-top segment begins at  $t = 0$ . The choice of a flat-top envelope aligns our simulations with the constant-field assumption underlying the theoretical models and the retrieval method developed in the following sections. This is also consistent with the long-pulse regime of typical experiments, where the dominant HHG emission originates from the central cycles for which the envelope is nearly constant. This expectation is explicitly supported by the results below, which show that the deviations in the extracted emission times and optimal phases between  $\sin^2$  and flat-top pulses decrease systematically with increasing pulse duration and become negligible in the long-pulse limit. All simulations in this study are performed for an 800 nm fundamental laser pulse with an intensity of  $4 \times 10^{14}$  W/cm<sup>2</sup> and probe fields with various  $n$  and  $\epsilon = 0.005$ . The simulation box is defined on a Cartesian grid with dimensions  $L_x \times L_y = 300 \times 160$  a.u., using a uniform grid spacing of 0.1 a.u. The wave function is propagated in time using a time-reversal symmetric propagator [31] with a time step of 0.02 a.u. To prevent artificial electron reflections at the boundaries, a mask function is applied. The mask function is set to 1 for  $|x| \leq x_0$  and  $|y| \leq y_0$  and gradually decreases to zero at the boundaries following a  $\cos^2$ -shaped function. The absorber parameters are set to  $x_0 = 1.1E_0/\omega^2 = 36.2$  a.u. and  $y_0 = (7/18)L_y = 62.2$  a.u. The value of  $x_0$  is chosen such that the mask function effectively absorbs long-trajectory electrons. Note that absorbing the long trajectories is not crucial for the present method because the short trajectories are selected

by analyzing the time-frequency distribution; see below. The TDSE simulations are carried out using the Octopus code [32].

Given the time-dependent wave function, the dipole acceleration can be calculated using the Ehrenfest theorem [33],

$$\mathbf{a}(t) = \langle \psi(\mathbf{r}, t) | \partial_{\mathbf{r}} V(\mathbf{r}) + \mathbf{E}(t) | \psi(\mathbf{r}, t) \rangle. \quad (3)$$

Since harmonic emission consists of contributions from several trajectories, the harmonic spectrum is not an ideal observable for retrieving emission times. Instead, we employ the Gabor transform to extract the emission data. The total Gabor time-frequency distribution is defined as

$$I_G(\Omega, t) = \left| \int dt' \mathbf{a}(t') e^{-(t'-t)^2/(2\sigma^2) + i\Omega t'} \right|^2, \quad (4)$$

where  $\Omega$  is the harmonic frequency and  $\sigma = 1/(3\omega)$ . The emission time  $t_e$  is determined as the local maximum of the total Gabor intensity  $I_G$  for each harmonic frequency  $\Omega$ . For this purpose, only trajectories launched in the first half-cycle of the flat-top region of the pulse and only short trajectories are considered. Consequently, the emission intensity at the harmonic  $\Omega$  is obtained as  $I_e(\Omega) = I_G(\Omega, t_e)$ , and the amplitude ratio is determined by  $R_e(\Omega) = \sqrt{I_{G_x}(\Omega, t_e)/I_{G_y}(\Omega, t_e)}$ . Here  $I_{G_x}$  and  $I_{G_y}$  correspond to the  $x$ - and  $y$ -components of the dipole acceleration, respectively. Finally, by varying the two-color delay  $\phi$ , one can determine the optimal phase  $\phi_h$  that maximizes the harmonic intensity and  $\phi_a$  that maximizes the amplitude ratio as follows:

$$\partial_{\phi} I_e \Big|_{\phi=\phi_h} = 0, \quad \partial_{\phi} R_e \Big|_{\phi=\phi_a} = 0. \quad (5)$$

These optimal phases serve as two observables that can be used as inputs for the time-retrieval equations.

### B. Strong-field approximation and quantum-orbit model

The time-retrieval equations are formulated based on the strong-field approximation (SFA) [20] and the quantum-orbit (QO) model [34]. These models do not include the Coulomb interaction. Therefore, they can serve as a reference for verifying the validity of results obtained from the time-retrieval equations in the limit where the Coulomb correction is disabled in the retrieval method, as well as for assessing the effect of the Coulomb force on the retrieved times. In the following, we briefly review the main equations of these models as employed in this study. In the SFA evaluated within the QO model, the intensity of the emitted harmonic with frequency  $\Omega$  at the return time  $t_r$  is given by

$$I_s = |e^{-i(S - \Omega t_r)}|^2. \quad (6)$$

Within the SFA framework, the action  $S$  corresponds to the Volkov phase, defined as

$$S_V = \int_{t_i}^{t_r} \left[ \frac{1}{2} v_x^2(t) + \frac{1}{2} v_y^2(t) + I_p \right] dt. \quad (7)$$

Here we assumed that only one trajectory is relevant.

The complex ionization and recombination times based on the QO model ( $t_i^{\text{QO}}$ ,  $t_r^{\text{QO}}$ ) are determined by the saddle-point equations:

$$\mathbf{v}^2(t_i)/2 = -I_p, \quad \mathbf{v}^2(t_r)/2 = \Omega - I_p. \quad (8)$$

Here  $\mathbf{v}(t)$  represents the electron velocity

$$\mathbf{v}(t) = \mathbf{A}(t) - \frac{1}{t_r - t_i} \int_{t_i}^{t_r} \mathbf{A}(t') dt'. \quad (9)$$

The laser-induced trajectory  $\mathbf{r}_L(t) = (x_L(t), y_L(t)) = \int_{t_i}^t \mathbf{v}(t') dt'$  for the laser field of Eq. (2) is written as

$$x_L(t) = \frac{E_0}{\omega^2} \left[ \cos(\omega t) - \cos(\omega t_i) - \frac{(t - t_i)}{(t_r - t_i)} (\cos(\omega t_r) - \cos(\omega t_i)) \right], \quad (10)$$

$$y_L(t) = \frac{\epsilon E_0}{(n\omega)^2} \left[ \cos(n\omega t + \phi) - \cos(\phi_i) - \frac{(t - t_i)}{(t_r - t_i)} (\cos(\phi_r) - \cos(\phi_i)) \right], \quad (11)$$

where  $\phi_i = n\omega t_i + \phi$  and  $\phi_r = n\omega t_r + \phi$ .

The ionization and return times obtained from the QO model (a reference without Coulomb effects) can be compared with the results of the time-retrieval method to reveal the Coulomb-induced time shift.

### C. The analytical R-matrix (ARM) model

Since we will refer to the ARM model [23] as an approach that accounts for the Coulomb interaction while predicting ionization and return times, we briefly summarize its key equations here. In this model, the total action is expressed as  $S = S_V + S_C$ , where  $S_C$  denotes the Coulomb-induced correction to the action, defined as

$$S_C = \int_{t_k}^{t_f} U_C(\mathbf{r}_L(t)) dt. \quad (12)$$

Here,  $\mathbf{r}_L(t)$  represents the laser-induced Coulomb-free trajectory defined by Eqs. (10) and (11). The integration interval is defined with the lower limit  $t_k = t_i - i/(2I_p)$ , while the upper limit  $t_f$  is, following [23], obtained by enforcing the return condition

$$\sqrt{\mathbf{r}_L(t_f)^2} = \frac{\exp[2(0.5772 - \xi)]}{2v_r}, \quad (13)$$

where

$$\xi = \sum_{p=1}^{\infty} \left[ \frac{1}{p} - v_r \arctan\left(\frac{1}{v_r p}\right) \right], \quad (14)$$

and the return velocity is given by  $v_r = \sqrt{2(\Omega - I_p)}$ .

Assuming that the Coulomb-induced action  $S_C$  is small compared to  $S_V$ , one can approximately solve the Coulomb-corrected saddle-point equations. This yields the first-order corrections to the ionization and return times [23]:

$$\Delta t_i^{\text{ARM}} = -\frac{\partial S_C}{\partial I_p} - \frac{\partial S_C}{\partial \Omega}, \quad \Delta t_r^{\text{ARM}} = -\frac{\partial S_C}{\partial \Omega}, \quad (15)$$

where the partial derivatives act on those parts of  $S_C$  that depend on  $I_p$  and  $\Omega$  through the times  $t_i$  and  $t_r$ . Therefore, ARM times are obtained as

$$t_i^{\text{ARM}} = t_i^{\text{QO}} + \Delta t_i^{\text{ARM}}, \quad t_r^{\text{ARM}} = t_r^{\text{QO}} + \Delta t_r^{\text{ARM}}. \quad (16)$$

#### D. Time-retrieval equations

The time-retrieval equations proposed and used in prior studies [21,22,26] read

$$\text{Re } v_{y0} \Big|_{\phi=\phi_h} = 0, \quad \partial_\phi R \Big|_{\phi=\phi_a} = 0. \quad (17)$$

Here,  $v_{y0}$  represents the initial lateral velocity of the electron at the ionization moment  $t_i$ .  $R$  is the amplitude ratio defined as  $R = |v_y(t_r)/v_x(t_r)|$ , where  $v_x(t_r)$  and  $v_y(t_r)$  denote the return velocities along the  $x$ -axis and  $y$ -axis, respectively, evaluated at the return time  $t_r$ . For  $v_{y0}$  and  $R$ , analytical expressions are used that depend on the two-color delay  $\phi$  and the times  $t_i$  and  $t_r$ . Thus, from the knowledge of the measurable quantities  $\phi_h$  and  $\phi_a$  and using Eq. (17), the times  $t_i$  and  $t_r$  can be obtained. The initial lateral velocity and the amplitude ratio, including Coulomb corrections, can be expressed as follows:

$$v_{y0} = -\frac{\epsilon E_0}{n\omega} \left[ \sin(\phi_i) + \frac{\cos(\phi_r) - \cos(\phi_i)}{n\omega(t_r - t_i)} \right] - \frac{\Delta y^C(t_i, t_r)}{t_r - t_i}, \quad (18)$$

$$R = \frac{1}{\sqrt{2(\Omega - I_p)}} \left| -\frac{\epsilon E_0}{n\omega} \left[ \sin(\phi_r) + \frac{\cos(\phi_r) - \cos(\phi_i)}{n\omega(t_r - t_i)} \right] + \Delta v_y^C(t_i, t_r) - \frac{\Delta y^C(t_i, t_r)}{(t_r - t_i)} \right|. \quad (19)$$

Here,  $\Delta v_y^C$  and  $\Delta y^C$  represent the velocity change and displacement of the electron along the  $y$ -axis, respectively, induced by the Coulomb force during the excursion time (from  $\text{Re } t_i$  to  $\text{Re } t_r$ ). Since  $t_i$  and  $t_r$  in the above equations are generally complex values, additional assumptions are required for their imaginary parts. For the imaginary part of the ionization time, a suitable choice is to adopt the Keldysh tunneling time defined as  $\text{Im } t_i = \tau_K = \sqrt{2I_p}/|E_x(\text{Re } t_i)|$  [25]. The simplest assumption for the imaginary part of the return time is  $\text{Im } t_r = 0$ . Although this assumption may be effective to some extent [26], our analysis of the new implementation of Coulomb corrections in the time-retrieval equations (presented in the subsequent section) indicates that it leaves room for improvement. A more accurate alternative, employed throughout this work, is to adopt the value of  $\text{Im } t_r$  from the ARM times Eq. (16).

If the Coulomb force effect is approximated as a short kick induced at  $t = \text{Re } t_i$ , as used in Refs. [26,35,36], then the Coulomb terms in Eqs. (18) and (19) result in

$$\Delta v_y^C(t_i, t_r) = u_0^C = \frac{\epsilon \pi E_0 \cos(n\omega \text{Re } t_i + \phi)}{(2I_p)^{3/2}}, \quad (20)$$

$$\Delta y^C(t_i, t_r) = (t_r - \text{Re } t_i) u_0^C. \quad (21)$$

By substituting Eqs. (20) and (21) into Eqs. (18) and (19), we obtain the same retrieval equations used in Ref. [26] [Eqs. (11) and (14) in [26]]. As demonstrated in [26], this approximation results in a shift of the ionization time to earlier times at lower probe frequencies; however, the retrieved times remain significantly different from the prediction of the ARM model.

To achieve a more accurate Coulomb correction for the time-retrieval equations, we propose an alternative approach. The Coulomb-induced velocity change can be calculated via

Coulomb force integration as follows:

$$\Delta v_y^C(t_i, t) = \int_{t_1}^t F_y^C(t') dt', \quad (22)$$

where  $t_1$  represents the starting time of electron excursion, and the  $y$ -component of the Coulomb force is given by

$$F_y^C(t) = -\frac{\partial V(\mathbf{r})}{\partial y} = -\frac{y_L(t)}{(x_L^2(t) + y_L^2(t))^{3/2}}. \quad (23)$$

To obtain the time-dependent Coulomb force, we require the electron trajectories. Here, we use the Coulomb-free laser-induced trajectories as an approximation [Eqs. (10) and (11)]. By evaluating Eq. (22) at the end of the electron excursion,  $t = t_2$ , we obtain the Coulomb-induced change in velocity accumulated over the entire excursion as

$$\Delta v_y^C(t_i, t_r) = \int_{t_1}^{t_2} F_y^C(t') dt'. \quad (24)$$

Finally, the electron displacement along the  $y$ -axis for the entire excursion interval  $[t_1, t_2]$  is obtained as

$$\Delta y^C(t_i, t_r) = \int_{t_1}^{t_2} \Delta v_y^C(t_i, t') dt'. \quad (25)$$

Two possible choices for the integral limits  $t_1$  and  $t_2$  in the above equations are  $[\text{Re } t_i, \text{Re } t_r]$  and  $[t_k, t_f]$ , which are discussed in Sec. II F.

#### E. Modified time-retrieval equations

The first relation in the time-retrieval equations [Eq. (17)] is derived by maximizing the tunneling rate with respect to the two-color delay  $\phi$  [21]. In this approach, it is assumed that the dependence of the tunneling rate on  $\phi$  occurs through the real-valued initial transverse velocity as  $\Gamma \propto \exp[-(\text{Re } v_{y0})^2 \tau_K/2]$ . In fact, this equation arises from a tunneling picture. To derive a more accurate equation for this purpose, one can begin by maximizing the harmonic intensity  $\partial_\phi I_s = 0$  based on the SFA. Using Eq. (6), this maximization simplifies to  $\partial_\phi [\text{Im } S] = 0$ . Here, we use a small scaling factor  $\epsilon$  to ensure that  $t_r$  remains independent of  $\phi$ . Therefore, we propose the following modified time-retrieval equations for the OTC scheme:

$$\partial_\phi [\text{Im } S] \Big|_{\phi=\phi_h} = 0, \quad \partial_\phi R \Big|_{\phi=\phi_a} = 0, \quad (26)$$

where  $R$  is the same quantity as defined in Eq. (19). In the Coulomb-free approximation, we have  $S = S_V$ . To incorporate the Coulomb effect, one can use  $S = S_V + S_C$ , where the Coulomb correction  $S_C$  is given by the same expression as used in the ARM model, Eq. (12).

#### F. Overview of time-retrieval methods and assumptions adopted in this study

In this study, we consider three different time-retrieval methods for extracting ionization and return times, based on the equations introduced in the previous subsections. We provide further details on the assumptions underlying these methods, along with the abbreviations used, in Table I. For the TRC-integration method, the integration limits  $t_1 = \text{Re } t_i$  and  $t_2 = \text{Re } t_r$  are chosen, as they yield more accurate retrieved



TABLE I. Overview of time-retrieval methods, assumptions, and equations adopted in this study.

| Method                                                                  | Abbreviation     | Equations                   | Assumptions                                                                                     |
|-------------------------------------------------------------------------|------------------|-----------------------------|-------------------------------------------------------------------------------------------------|
| Time-retrieval with Coulomb corrections as an initial kick [26]         | TRC-kick         | (17)–(21)                   | $\text{Im } t_i = \tau_K$ , $\text{Im } t_r = 0$                                                |
| Time-retrieval with Coulomb corrections via Coulomb force integration   | TRC-integration  | (17)–(19), (22)–(25)        | $\text{Im } t_i = \text{Im } t_i^{\text{ARM}}$ , $\text{Im } t_r = \text{Im } t_r^{\text{ARM}}$ |
| Modified time-retrieval with Coulomb corrections via the Coulomb action | MTRC-integration | (26), (12), (19), (22)–(25) | $\text{Im } t_i = \text{Im } t_i^{\text{ARM}}$ , $\text{Im } t_r = \text{Im } t_r^{\text{ARM}}$ |

times than the complex integration limits. In the MTRC-integration method, the limits  $t_1 = t_k$  and  $t_2 = t_f$  are selected to maintain consistency with the integration limits used in the Coulomb action  $S_C$  [Eq. (12)]. It should be noted that  $\text{Re } t_i$  and  $\text{Re } t_r$  in the integral bounds (appearing in  $t_1$ ,  $t_2$ ,  $t_k$ , and  $t_f$ ) are treated as unknown variables in the time-retrieval process, rather than being fixed known values (such as those from the QO or ARM models). In this study, all retrieved times are calculated by inserting the optimal phases extracted from the TDSE into the time-retrieval equations. The abbreviations in Table I are used consistently throughout the results presented in this work.

### III. RESULTS

#### A. Optimal phases and robustness of the retrieval observables

We start by presenting the optimal phases extracted from the TDSE, which will be used as input parameters for the time-retrieval equations. Figure 1 shows color-density maps of the normalized total Gabor intensity and the amplitude ratio for the  $\omega-2\omega$  and  $\omega-4\omega$  configurations. The white curves mark the optimal phases,  $\phi_h$  and  $\phi_a$ , indicating the locations of the maxima of  $I_G$  and  $R$ , respectively. With these optimal phases at hand, we can retrieve the times using the time-retrieval method.

Before proceeding to the presentation of the retrieved times and the analysis of the retrieval process, we explicitly assess the influence of the pulse envelope on the observables entering the retrieval procedure. To this end, we perform additional TDSE simulations using  $\sin^2$  pulses with total durations of 4, 6, and 16 optical cycles and compare the results with those obtained for the flat-top pulse. Figure 2(a) shows one cycle from the middle of the pulses, comparing the normalized fundamental driving fields for the flat-top and  $\sin^2$  envelopes, together with the orthogonally polarized probe field at frequency  $2\omega$ . We then focus on the short-trajectory burst emitted in the half cycle located near the center of each pulse ( $t = 0.5$ – $1$  optical cycle), where the field variation is smallest. Panels (b)–(d) present the absolute deviations of the extracted emission time and of the optimal phases, defined as  $\Delta t_e = |t_e^{(\sin^2)} - t_e^{(\text{flat})}|$ ,  $\Delta \phi_h = |\phi_h^{(\sin^2)} - \phi_h^{(\text{flat})}|$ , and  $\Delta \phi_a = |\phi_a^{(\sin^2)} - \phi_a^{(\text{flat})}|$ , respectively. In all cases, the deviations decrease systematically as the pulse duration increases, approaching zero for the longest pulse. This demonstrates that envelope-induced effects become negligible in the long-pulse regime, so that the

flat-top pulse provides a faithful approximation to the nearly constant central cycles of experimentally relevant long pulses.

Macroscopic propagation is expected to play an important role in establishing the connection to experiment by acting as a trajectory filter, in particular through the suppression of the long-trajectory contribution in the present OTC configuration. To investigate this point, we coherently average the dipole acceleration over a set of TDSE simulations with different driving laser intensities according to  $\mathbf{a}_{\text{av}}(t) = (1/N_I) \sum_I \mathbf{a}(I, t)$ , where  $N_I$  is the number of intensity values included in the average. Figure 3 compares the Gabor time-frequency profiles of the total harmonic emission  $I_G$  for a single-intensity case at  $0.40 \text{ PW/cm}^2$  and for several averaged cases obtained from progressively broader

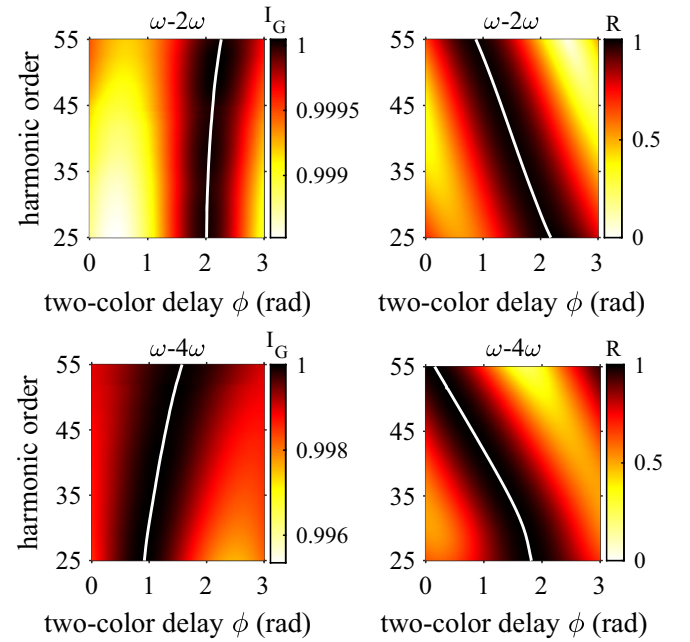


FIG. 1. Color-density maps of the normalized total Gabor intensity (left column) and the amplitude ratio (right column) obtained from TDSE simulations, plotted vs harmonic order and two-color delay. Results for the  $\omega-2\omega$  field are on the top row, and for the  $\omega-4\omega$  field on the bottom row. White solid curves indicate the optimal phases that maximize each observable. The calculations are performed for an OTC field with a fundamental wavelength of 800 nm and a peak intensity of  $4 \times 10^{14} \text{ W/cm}^2$ . The probe field has the scaling factor  $\epsilon = 0.005$ .

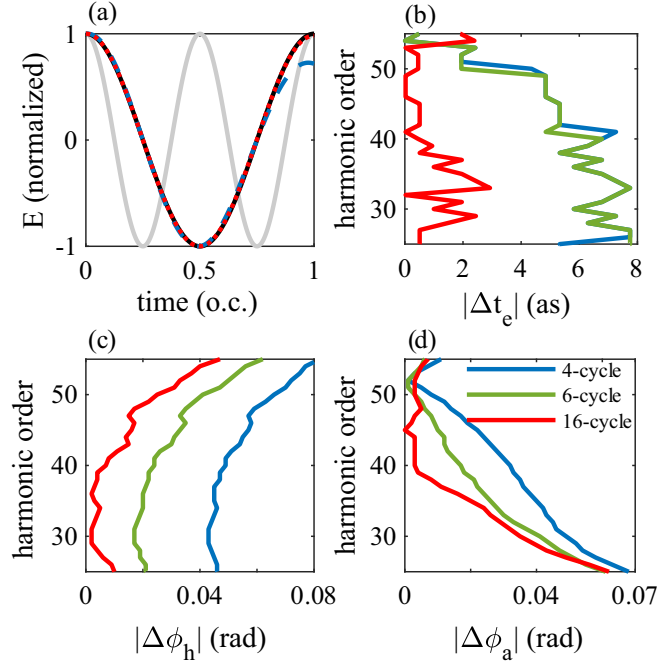


FIG. 2. Analysis of pulse-envelope effects on the HHG observables. (a) Normalized laser fields used in the comparison: the fundamental component of the flat-top pulse (black solid),  $\sin^2$  pulses with total durations of 4 optical cycles (blue dashed) or 16 optical cycles (red dotted), and the orthogonally polarized probe field at frequency  $2\omega$  (gray solid). (b) Absolute deviation in the emission time extracted from the short-trajectory burst in the central half cycle for 4, 6, and 16 optical cycles (blue, green, and red curves, respectively). (c) Absolute deviation in the optimal phase  $\phi_h$  obtained from the normalized total Gabor intensity for the  $\omega - 2\omega$  field. (d) Absolute deviation in the optimal phase  $\phi_a$  obtained from the amplitude ratio for the  $\omega - 2\omega$  field.

intensity intervals. One finds that the long-trajectory branch is systematically suppressed as the averaging range is increased, while the short-trajectory branch remains clearly visible and becomes more pronounced. This behavior is consistent with the well-known macroscopic trajectory filtering in HHG, where the short trajectory is preferentially phase-matched and enhanced on axis, whereas the long trajectory is more strongly suppressed because of its less favorable phase-matching conditions and larger divergence [27,28]. In the present calculations, the emission times extracted from the short-trajectory branch remain close to those of the single-intensity case, as shown in Fig. 3(e). These results, therefore, support the assumption that, for the present OTC scheme, macroscopic averaging over intensity acts as an additional trajectory filter, suppressing the long-trajectory contribution while leaving the short-trajectory observables entering the retrieval essentially unchanged.

### B. Retrieved times with Coulomb corrections as an initial kick

Using the optimal phases obtained in the previous section as input, we apply the TRC-kick variant of the time-retrieval method, in which Coulomb corrections are included by modeling the Coulomb interaction as an initial kick to the

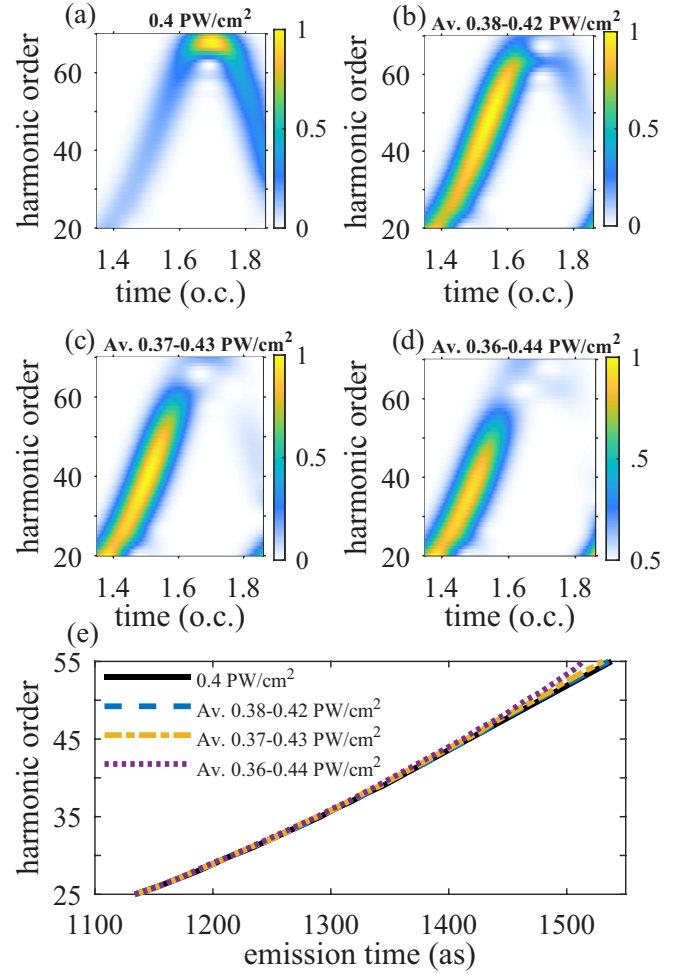


FIG. 3. Intensity-averaging of the HHG time-frequency profile in the OTC field with the two-color delay  $\phi = 0$ . (a) Gabor time-frequency profile of the total harmonic emission  $I_G$  for the single-intensity case at  $0.40 \text{ PW/cm}^2$ . The Gabor profiles of the coherently averaged dipole acceleration obtained by averaging over driving laser intensities in the ranges (b)  $0.38\text{--}0.42 \text{ PW/cm}^2$ , (c)  $0.37\text{--}0.43 \text{ PW/cm}^2$ , and (d)  $0.36\text{--}0.44 \text{ PW/cm}^2$ . (e) Emission times extracted from the short-trajectory branch for the cases shown in (a)–(d).

electron velocity. We compare the retrieved times with those predicted by the QO and ARM models, which exclude and include the Coulomb effect, respectively; see Fig. 4(a). The method and assumptions employed in TRC-kick are exactly the same as those used in [26]. For an OTC scheme with an  $\omega - 2\omega$  configuration (light blue dashed curves), the retrieved ionization time obtained by TRC-kick shows a shift toward earlier times (about 10 attoseconds at intermediate harmonics) compared to the QO model. Although this shift toward earlier times is expected, it still exhibits considerable deviation from the ARM results. The retrieved return time exhibits a similar shift toward earlier times, showing good agreement for the intermediate harmonics but slightly underestimating the shift for lower harmonics and overestimating it for higher harmonics compared to the ARM result. For the  $\omega - 4\omega$  configuration in Fig. 4(a), the retrieved ionization times are in better agreement with the prediction of the ARM model, with a

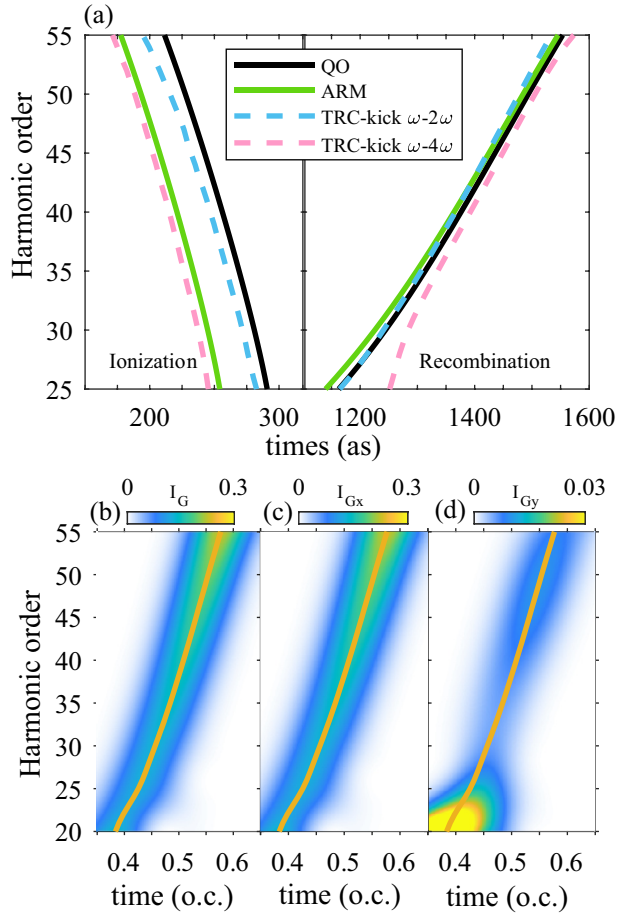


FIG. 4. (a) Retrieved ionization and return times ( $\text{Re}t_i$ ,  $\text{Re}t_r$ ) from TDSE results for an OTC scheme with  $\omega - 2\omega$  and  $\omega - 4\omega$  fields. The black solid lines indicate the real parts of the times calculated using the Coulomb-free QO model, while the green solid lines represent the times from the ARM model. Retrieved times obtained from the original time-retrieval method with Coulomb corrections modeled as an initial kick (TRC-kick) are depicted by blue ( $\omega - 2\omega$ ) and red ( $\omega - 4\omega$ ) dashed curves. The temporal regions corresponding to ionization and recombination are highlighted with light-blue and light-yellow shaded areas, respectively. (b)–(d) The Gabor time-frequency profile of the total harmonic emission  $I_G$ , the  $x$ -component  $I_{G_x}$ , and the  $y$ -component  $I_{G_y}$  for the probe frequency  $4\omega$ . The yellow solid curves represents the emission times extracted from the maxima of the total Gabor intensity in (b) for each harmonic order. Calculations are performed for an OTC field with a fundamental wavelength of 800 nm and a peak intensity of  $4 \times 10^{14}$  W/cm<sup>2</sup>. The probe field has a scaling factor  $\epsilon = 0.005$ . For panels (b)–(d), the two-color delay is set to  $\phi = 0$ .

deviation of 5–9 attoseconds. Indeed, for an OTC laser scheme with a high-frequency probe field, the Coulomb interaction significantly manifests in the optimal phases [25], while the Coulomb-related terms in the time-retrieval equations become negligible compared to the laser-driven terms. Consequently, the ionization time can be retrieved with reasonable accuracy for higher probe frequencies even without explicitly including Coulomb corrections in the time-retrieval equations [25]. The right panel of Fig. 4(a) reveals that the retrieved return times for the intermediate and high harmonics match closely

with the QO model, with a shift of approximately 12–30 attoseconds to later times, while the ARM model predicts earlier times. The return times for low harmonics significantly deviate from both QO and ARM models. This discrepancy may arise from uncertainties in accurately determining the optimal phases  $\phi_a$  in the H25–H30 region, due to the diplike structure observed in the  $y$ -component of the Gabor transform ( $I_{G_y}$ ), as illustrated in Fig. 4(d). This dip could result from the merging of two branches visible in the Gabor transform  $I_{G_y}$ . While resolving the influence of intensity minima on optimal phases is beyond the scope of this study, our approach remains applicable to regions of the plateau that are unaffected by these minima. The yellow solid curve in Fig. 4 represents the emission times extracted from the maxima of the total Gabor intensity  $I_G = I_{G_x} + I_{G_y}$  [Fig. 4(b)] for each harmonic order.

### C. Retrieved times with Coulomb corrections via Coulomb force/potential integration

Here, we present our results for the time-retrieval methods with Coulomb corrections (TRC-integration and MTRC-integration), in which the Coulomb effects are incorporated through integration of the Coulomb force or potential along the electron trajectories. Figure 5(a) compares the retrieved ionization times obtained from different methods. For the  $\omega - 2\omega$  case, times retrieved by the TRC-integration (dark blue dashed curve) and MTRC-integration (purple dotted curve) methods show a slight improvement compared to the TRC-kick method (light blue solid curve). Both methods exhibit a larger shift toward earlier times, approximately 17 as relative to the QO times. On the other hand, for the  $\omega - 4\omega$  configuration, the ionization-time retrieval improves only with the MTRC-integration method, yielding perfect agreement with the ARM model. Figure 5(b) shows the return times obtained using different methods. The retrieved return times from TRC-integration and MTRC-integration methods exhibit almost the same trend as observed with the Coulomb kick approach (TRC-kick). In Fig. 5(b), we also show a comparison with the TDSE emission times (yellow curve). There is a discrepancy with the ARM/retrieved times, which may arise from the absence of transition-dipole effects in the latter methods.

Finally, we investigate the accuracy of the retrieved times obtained using the proposed methods in this work across various probe field frequencies. Figures 6(a) and 6(b) show the retrieved ionization time as a function of probe field frequency for harmonic orders H45 and H55. As can be seen, the retrieved ionization times exhibit better agreement with the ARM prediction at higher probe frequencies, particularly for the MTRC-integration method. At higher probe frequencies, the retrieval becomes increasingly dominated by the laser-driven terms, so the relative impact of the Coulomb-induced corrections decreases and the retrieved times approach the ARM prediction more closely, consistent with the trend reported in [25]. For lower probe frequency, the deviation from the ARM model is substantial, reaching a maximum near the probe frequency  $2\omega$ . For probe frequencies below  $2\omega$ , the deviation gradually decreases. The maximum of the errors near  $2\omega$  can be attributed to the limited accuracy of Coulomb action

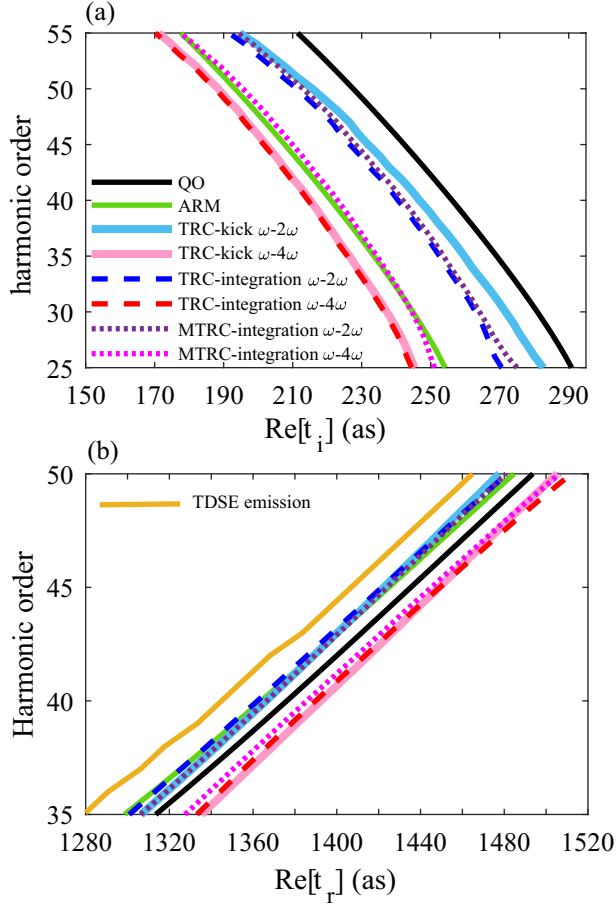


FIG. 5. Ionization (a) and return times (b) obtained using different methods. The black solid lines indicate the real parts of the times calculated using the Coulomb-free QO model, and the green solid lines represent the times from the ARM model. The TDSE emission times (yellow solid curve) are extracted from the maxima of the total Gabor time-frequency profile.

or Coulomb force integrations around this frequency. Figure 6(c) shows the errors in the optimal phases obtained from the SFA and ARM models, with  $\Delta\phi_{\text{SFA}} = \phi_{\text{TDSE}} - \phi_{\text{SFA}}$  and  $\Delta\phi_{\text{ARM}} = \phi_{\text{TDSE}} - \phi_{\text{ARM}}$ . Here,  $\phi_{\text{TDSE}}$ ,  $\phi_{\text{SFA}}$ , and  $\phi_{\text{ARM}}$  represent the optimal phases for maximizing the total harmonic intensity, obtained from TDSE, SFA, and ARM, respectively. The ARM optimal phases are obtained by evaluating the action using ARM times computed for the OTC field. The ARM phase error is expected to be smaller than the SFA phase error due to the inclusion of Coulomb corrections in the ARM model. Indeed, the results reveal a reduction of the phase error at both high and low probe frequencies compared to the SFA. However, an increased error is observed near  $2\omega$ . This counterintuitive finding may be related to the fact that the ARM model considers only first-order Coulomb corrections by construction.

#### IV. CONCLUSIONS

In this work, we have introduced two refinements to the time-retrieval equations for reconstructing ionization and return times from observables in an OTC scheme, applicable

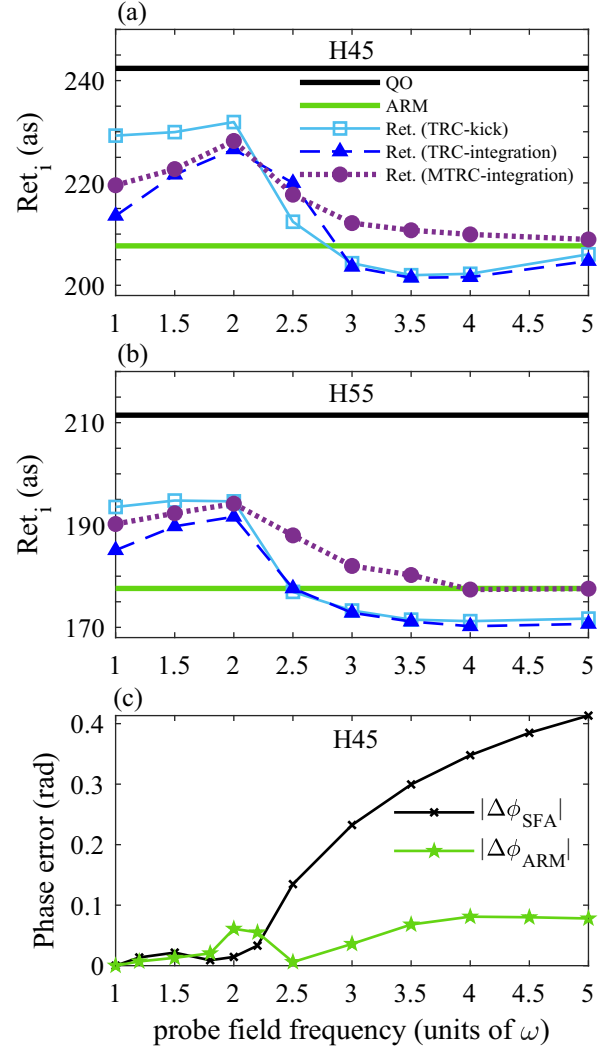


FIG. 6. Ionization times as a function of the probe frequency for harmonic orders (a) H45 and (b) H55. The black and green solid lines indicate the QO and ARM reference times, respectively. (c) The errors in optimal phases obtained from the SFA and ARM models vs the probe frequency. The errors are defined with respect to the TDSE optimal phases, which correspond to the maxima of the total harmonic intensity.

to both experiments and simulations. First, we developed an improved approach to incorporate Coulomb corrections into the time-retrieval method. Rather than modeling the Coulomb effect as an instantaneous initial momentum kick [26], we account for Coulomb effects by integrating the Coulomb force along the electron trajectories. This approach captures Coulomb-induced time shifts in the ionization time with greater accuracy and yields improved agreement with the ARM predictions. Second, we propose a revised equation for maximizing the harmonic intensity as a function of the two-color delay. The results of this refinement demonstrate a notably reduced deviation between the retrieved times and the ARM predictions at higher probe frequencies. However, even with both refinements, the retrieval method captures only about half of the time shift predicted by the ARM model when using lower probe frequencies, especially near the



experimentally convenient frequency  $2\omega$ . Overall, these refinements offer a more precise framework for attosecond-scale measurements and provide deeper insights into strong-field phenomena.

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### DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

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