Non-dynamical multi-photon phase shifts in attosecond science

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Ab initio simulations of a range of interferometric experiments are used to identify a strong dependence on non-dynamical multi-photon phase shifts in above-threshold ionization. A simple rule of thumb for non-dynamical phase shifts is derived to explain both the conservation of photoelectron yield and its absolute CEP-dependence. As an example, it is found that interferometric above-threshold ionization experiments are shifted by $\pi/4$ relative to RABBIT experiments. Thus, our work helps to resolve the issues of separability of quantum dynamics in attosecond and free-electron laser sciences.

Attosecond science has opened up the possibility to measure dynamical time-delays in photoionization and to probe chemical reactions in the time domain. As with any interferometric experiment, it is essential to understand the phase evolution along each arm of the interferometer in order to interpret the final outcome of experiments. In attosecond science such arms take many different manifestations. While traditional spectroscopy allows for measurements of phase differences between separate sources, e.g. high-order harmonic generation (HHG) from atoms [1] or molecules [2,3], the technique is limited to probe transitions to the same final energy. In contrast, novel types of non-linear spectroscopies make use of probe laser beams to change the dynamics in the interferometric arms, thus, making it possible to interfere different energy components of the system [4]. Prime examples of such non-linear arms include perturbed quasi-classical trajectories in HHG in atoms [5–8], molecules [9] and solids [10]; perturbed photoelectrons in laser-assisted photoionization (LAP) from atoms [11–15] and molecules [16]; resonant two-photon ionization by bound [17] or autoionizing states [18–20]; and perturbed photoelectrons in above-threshold ionization (ATI) [21]. Non-linear interferometric arms have recently also found applications in free-electron laser (FEL) experiments [22]. This plethora of interferometric arms makes it possible to inherently study non-linear processes, such as HHG and ATI, but they also open the question of separability between unperturbed and perturbed dynamics.

In this Letter we address of separability of quantum dynamics in attosecond experiments by performing a systematic study using the time-dependent configuration interaction singles [23,24] (TDCIS) method. Photoelectron distributions are obtained with the time-dependent surface flux [25] (t-SURFF) method and compared with simulations based on the Keldysh-Faisal-Reiss [26,27] (KFR) theory for strong-field ionization. Specifically, we consider three recent attosecond experiments, which utilize different non-linear interferometric arms, and compare them with the traditional RABBIT experiment [11]. First, we consider a LAP experiment by Laurent et al. [15], where even and odd harmonics from HHG are coupled by a laser photon from the probe beam. Second, we consider a LAP experiment by Maroju et al. [22], where attosecond pulse structures of FEL beams were coupled to twin sidebands by a mix of one and two exchanged probe photons. Lastly, we consider the ATI experiment by Zipp et al. [21], where an ATI process was probed by generation of sidebands created by exchange of probe photons. Interestingly, we find all three recent experiments exhibit interference patterns that depend on non-dynamical phase shifts and on the directionality of the photoelectron, phenomena that are not observed in the RABBIT experiment [11]. Using KFR theory, we derive a simple rule of thumb capable of explaining the general phase and amplitude effects in all above mentioned experiments on an absolute CEP-scale. In this work atomic units are used: $\hbar = e = a_0 = a_e = 4\pi\epsilon_0 = 1$.

The one-photon ionization process of an atom by an XUV photon, $\Omega > I_p$, is described by the reaction: $A + \gamma_\Omega \rightarrow A^+ + e^-$. The associated quantum phase is [29]

$$\arg[e^{(1)}_{k_1;\ell_1 m}]_{\ell_1 m} = \arg[Y_{\ell_1 m}(k_1)] - (1 + \ell_1) \frac{\pi}{2} + \eta_{\ell_1}(k_1),$$

(1)

where $\eta_{\ell_1}(k_1)$ is the scattering phase of the final angular momentum channel with linear momentum, $k_1$, and orbital angular momentum, $\ell_1$, and where $|a\rangle$ is the initial atomic state with orbital angular momentum, $\ell_0$, and binding energy, $I_p$. The magnetic quantum number, $m$, is conserved for linearly polarized fields, $E_\Omega(t) = E_\Omega \sin(\Omega t)\hat{\mathbf{z}}$. If the photoelectron further interacts with a laser field, $E_\omega(t) = E_\omega \sin(\omega t - \varphi)\hat{\mathbf{z}}$, with $\omega \ll I_p$ it undergoes a laser-assisted photoionization process: $A + \gamma_\Omega \pm \gamma_\omega \rightarrow A^+ + e^-$. The associated quantum phase is approximately [14]

$$\arg[e^{(2)}_{k_{\omega};\ell_2 \ell_1 m}]_{\ell_2 \ell_1 m} \approx \arg[Y_{\ell_2 m}(k_2)] - \frac{\pi}{2} + \phi_{CC}^{(\pm)} + \varphi$$

$$- (1 + \ell_1) \frac{\pi}{2} + \eta_{\ell_1}(k_1),$$

(2)

for the angular momentum sequence: $\ell_0 \rightarrow \ell_1 \rightarrow \ell_2$. The dynamical phase induced due to exchange of the $\omega$ photon, $\phi_{CC}^{(\pm)} \approx -\phi_{CC}^{(-)}$, is called the continuum–continuum (CC) phase and it arises due to the long-range Coulomb potential. The CC phase has been the subject of recent attention as it must be compensated...
for to separate the one-photon dynamical Wigner delay \[59\]. At higher kinetic energies the CC phases are universal and tend to zero \[14\]. In addition to the dynamical phase shift, there is further a non-dynamical phase factor, \(-\pi/2 + \arg[Y_{\ell z,m}(k_2)/Y_{\ell z,m}(k_1)]\), where the latter term is due to the fact that exchange of a laser photon changes the parity of the photoelectron. The relative sign of up \((\theta = 0\) degrees) and down \((\theta = 180\) degrees\) photoelectrons then follows from the parity transformation of the spherical harmonics, \(P Y_{\ell m}(k) = (-1)^\ell Y_{\ell m}(k)\). In general, the remaining phase factor, \(-\pi/2\), is larger than the dynamical phase shifts and is important to understand the CEP-dependence in LAP experiments with mixing parity processes from the perspective of perturbation theory \[13,31\].

With the use on Eqs. \([12]\), Laurent et al. proposed that there exists an additional non-dynamical phase shift, \(\delta = \pi/2\), between odd and even harmonics from HHG, based on an experimental checkerboard pattern over CEP between the up and down photoelectrons \[13\]. This phase shift has caused controversy because it opposes the idea that one attopulse per cycle is generated from HHG with an \(\omega/2\omega\) driving field \[22\]. In fact, Eq. \(2\) was derived using asymptotic wavefunctions for hydrogenic systems and several effects beyond this approximation has been observed in more recent experiments \[33-36\]. For this reason, we present in Fig. 1 (b) the result of LAP simulations with a comb of odd, \((2n+1)\omega\), and even, \(2n\omega\), high-order harmonics coupled by a laser field, \(\omega\). We find that the relative phase of \(\delta = \pi/2\) leads to the checkerboard structure, while the \(\delta = 0\) case exhibits no such structure (not shown). Thus, we have confirmed the result of Laurent et al. \[13\], and open the quest for understanding the general role of non-dynamical phases in attosecond experiments.

The simulation presented in the left part of Fig. 1 (b) is performed using KFR theory with the initial state \(|a(t)\rangle = |a\rangle \exp[i\mathbf{p}t]\) taken to be a scaled hydrogenic 1s orbital with a binding energy equal that of the Hartree-Fock (HF) 2p orbital in neon, \(I_p = -e_{2p}^\text{HF} = 23.142\) eV. The complex amplitude for photoionization in velocity gauge using KFR is given by

\[
c_{k_0} \approx \frac{1}{\hbar} \int \mathrm{d}t \langle \chi_k^{(V)}(t) \mid A(t) \cdot \mathbf{p} + \frac{A^2(t)}{2} \mid a(t) \rangle,
\]

where the linearly polarized electric field is \(E(t) = -dA/dt\) and \(\mathbf{p}\) is the momentum operator. The continuum is described by time-dependent Volkov states, \(\langle \chi_k^{(V)}(t) \rangle\). The simulation presented in the right part of Fig. 1 (b) is performed using the TDCIS method in velocity gauge with the active space restricted to excitations from the 2p orbital with \(m = -1, 0, 1\), which implies that \(I_p = -e_{2p}^\text{HF}\). Throughout, we consider \(\omega = 1.55\) eV and a pulse duration of 9.9 fs.

The good agreement between the two simulations in Fig. 1 (b) shows that neither the initial angular-momentum, nor electron correlation effects are required to capture the checkerboard CEP-dependence. In Fig. 1 (c), agreement between KFR and TDCIS for angle and CEP-resolved photoelectrons is observed, where the TDCIS result shows a slightly broader distribution in angle.

In order to study systematically non-dynamical multi-photon phase shifts, we now consider the general LAP process: \(A + q\gamma \rightarrow A^q + e^-\), where \(q\) laser photons are exchanged. The KFR transition amplitude for photoionization, expressed in Eq. \(3\), can be used to express LAP

\[
c_k \approx -\frac{1}{2} \sum_{n=-\infty}^{\infty} (-i)^n \exp\{in\varphi\}J_n \left(\frac{k \cdot \Lambda}{\omega}\right)
\times (\langle k | \mathbf{p}_z | a \rangle \int \mathrm{d}t \Lambda_{\Omega}(t) \exp[i(\varepsilon_k + I_p - \Omega - n\omega)t])
\]

to a given final plane wave state, \(|k\rangle\) \[37\]. Here the laser photon interaction number, \(n = 2q\), describes the interaction amplitude with the laser field in terms of Bessel functions, \(J_q(2\xi)\), from a Jacobi-Auger expansion. Bessel functions are odd and even functions depending on the integer \(q\), and they can be approximated as \(J_q(2\xi) \approx \frac{2}{\xi} p^q\) for weak interactions. Hence, the argument of the Bessel functions is important for the parity of the photoelectron, but it is suppressed here for brevity. The field envelopes of the XUV- and IR-fields, \(\Lambda_{\Omega/\omega}\), are defined to be positive along the z-axis. In Eq. \(4\) we have assumed that the laser envelope is constant and that the XUV-field can be treated with the rotating wave approximation. The LAP process is linear in the XUV field, which makes it easy to interpret interference features as energy-shifted replicas of the elementary photoionization process, \(f_{k_0}\), for each number of laser interactions, \(n\). We note that the processes of absorption \((n > 0)\) and emission \((n < 0)\) of laser photons have the same non-dynamical phase factor because \((-i)^{-q}J_{-q} = (-i)^qJ_q\). The non-dynamical phase factor accumulates for each laser interaction, thus extending the lowest-order results, in Eqs. \(1-2\), to any order, \(q\), in interaction with the laser field. Hence, the transition amplitude for the \(n\)th LAP process can be written as

\[
c_{k_0}^{(n)} \approx (-i)^n J_{|n|} \exp\{in\varphi\}f_{k_0}(n)
\]

referred to as the “rule of thumb” for ATI.

In order to understand the checkerboard pattern, shown in Fig. 1 (b), we now apply Eq. \(5\) for laser-photon processes up to second order, \(0 < q \leq 2\), illustrated in Fig. 1 (a). We consider a plateau of equally strong harmonics, with a phase shift of \(\delta\) between even and odd harmonics, which means that \(f_{k_0}\) (odd) = 1 and \(f_{k_0}\) (even) = \(\exp\{i\delta\}\). The transition amplitude for the laser-assisted harmonics, corresponding to the diagrams in Fig. 1 (a), are constructed using the rule of thumb as
This leads to photoelectron peaks that modulate with the CEP as

\[
\begin{cases}
|c_k(\text{odd})|^2 &= J_0^2 + 4J_1 \sin(\delta) \cos(\varphi) \\
|c_k(\text{even})|^2 &= J_0^2 - 4J_1 \sin(\delta) \cos(\varphi),
\end{cases}
\]

for odd (+) and even (−) laser-assisted harmonics respectively. While the CEP-dependent term vanishes for \( \delta = 0 \), it is maximal for \( \delta = (\frac{1}{2} + N)\pi \), leading to the checkerboard pattern. In contrast, the rule of thumb does not exhibit a modulation at twice the CEP-rate, denoted as the “RABBIT” term by Laurent et al. \[13\]. The cancellation of this higher-order term is explained by the inclusion of transitions with two laser photons, shown in the outermost diagrams in Fig. 1(a). Thus, the rule of thumb explains the robustness of the checkerboard pattern with substantial increase in laser intensity observed experimentally \[13\]. It also shows that the total probability of even and odd photoelectron peak pairs is conserved in the plateau, due to the opposite signs of the CEP-dependent terms in Eq. (7).

In Fig. 2(b) we show the result of an actual RABBIT simulation for neon using KFR (left part) and TDCIS (right part), for both sideband (SB) and high-order harmonic (HH) photoelectron peaks, with respect to both CEP-dependence and relative peak intensity. In Fig. 2(c) we show the sideband resolved in both CEP, \( \varphi \), and emission angle, \( \theta \), with good agreement between the two simulations. In Fig. 2(a), we show the lowest-order photon processes, that contribute to the sideband peak with \( q = 1 \) (left diagram) and to the harmonic peak with \( q = 0 \) and 2 (right diagram). While the sideband modulation over CEP is the main measurable in RABBIT experiments \[11\], recent work has also included the modulation of harmonics to improve the experimental statistics of the measurements without any formal justification \[30\].

Given a sequence of synchronized odd harmonics, \( f_{k_a}(\text{odd}) = 1 \) and \( f_{k_a}(\text{even}) = 0 \), the rule of thumb for ATI gives

\[
\begin{align*}
|c_{k_a}(\text{SB})|^2 &= -iJ_1 \exp[-i(\varphi - \delta)] - iJ_3 \exp[i\varphi] \\
|c_{k_a}(\text{HH})|^2 &= -J_2 \exp[-i2\varphi] + J_0 - J_2 \exp[2i\varphi].
\end{align*}
\]

While the two terms in the sideband amplitude carry the same non-dynamical phase due to the same order \( q = 1 \), the odd harmonic peaks comprise of different orders \( q = 0 \) and 2, which leads to different non-dynamical phases. Using Eq. (6), the corresponding probabilities are

\[
\begin{align*}
|c_{k_a}(\text{odd})|^2 &= J_0 \cos^2(\varphi) \\
|c_{k_a}(\text{even})|^2 &= J_0[1 + J_2(4 - 8 \cos^2(\varphi))].
\end{align*}
\]

Hence, the fact that SB and HH peaks in RABBIT behave in opposite ways with CEP is a phenomenon directly related to non-dynamical multi-photon phase shifts. Eq. (9) also shows that the probability of SB+HH pairs are conserved for perturbative laser fields. Similar analysis can be used to understand the general CEP-dependence of sidebands in higher-order RABBIT schemes, proposed by Harth et al. \[38\], where the exchange of \( q \) photons is used to reach the sideband in each interferometric arm. The rule of thumb then gives that the central sidebands modulate as \( 4J_q^2 \sin^2(q\varphi) \) over CEP, which shows that sidebands in any RABBIT-like experiment are insensitive to the non-dynamical phase shifts. However, non-dynamical phase shifts are essential to describe the depletion of the harmonics and, hence, the probability conservation of the overall process.

In Fig. 3(b), we show the result of simulations of the FEL experiment by Maroju et al. \[22\], where...
two FEL beams with a photon energy difference of $\Delta \Omega_{\text{FEL}} = \Omega_{\text{FEL}}^\text{high} - \Omega_{\text{FEL}}^\text{low} = 3\omega$ are used to photoionize neon atoms with an assisting laser with photon energy $\omega$. This results in the formation of twin sidebands that are created by a combination of one and two laser photons, as illustrated in Fig. 3 (a). The “low” (“high”) twin is generated by absorption of one (two) laser photon and emission of two (one) laser photons. In contrast to the above studied experiments, the present result shows a non-trivial CEP-transformation. Further, the CEP-dependence of the twin sidebands are reversed under a parity transformation, as shown in Fig. 3 (c) for photoelectrons in the up and down directions, respectively. A natural question arises as to what physical mechanism determines the different directionalities of the photoelectrons in the twin peaks? In order to understand the CEP-dependence, we apply the rule of thumb for ATI process, with $f_{ka}(\Omega^\text{FEL}_{\text{KFR}}) = \exp[i\delta]$ and $f_{ka}(\Omega^\text{FEL}_{\text{TDCIS}}) = 1$, which leads to

\[
\begin{align*}
|c_{ka}(\text{high})|^2 &= J_1^2 - J_1 J_2 [2 - 4 \sin^2 \left(\frac{\pi}{4} + \frac{3 \varphi + \delta}{2}\right)] \\
|c_{ka}(\text{low})|^2 &= J_1^2 - J_1 J_2 [2 - 4 \cos^2 \left(\frac{\pi}{4} + \frac{3 \varphi + \delta}{2}\right)],
\end{align*}
\]  

(11)

showing that the two sidebands have equal strength for $\varphi = \delta = 0$, but evolve in different ways with CEP. Further, the relative sign of the two terms in Eq. (10) is flipped under a parity transformation due to the properties of the Bessel functions. In this sense the physics at play in Maroju’s experiment is a higher-order version of the orbital parity mixing experiment by Laurent et al. [13]. Indeed, in Fig. 3 (c), it is shown that it is possible to perform a smooth parity transformation of the twin peaks by tuning the CEP of the laser field by $\Delta \varphi = 60$ degrees.

Finally, we present in Fig. 4 (b) ATI simulations for the experiment by Zipp et al. using KFR theory. Here, ATI peaks are generated by a strong $2\omega$ laser field. Intermediate sidebands (SB) are created by the exchange of $\omega$ photons from a weaker laser field, as illustrated in the left diagram of Fig. 4 (a). The ATI peaks are further perturbed by the $\omega$ laser field as illustrated in the right diagram of Fig. 4 (a). The data presented in Fig. 4 (b) is normalized to each ATI and SB peak individually. In reality the probability of ATI decreases with increasing order, see the insert in Fig. 4 (b). While there is great a level of similarity...
between Zipp’s experiment in Fig. 4 (b) and the RABBIT experiment in Fig. 2 (b), there is a marked difference in the CEP-dependence on the absolute scale by close to $\pi/4$. To understand this numerical result, we turn to the rule of thumb. The initial wave packet is here created by the non-linear ATI process, and no longer the LAP process. Accordingly, each interaction yields an additional $-i$ to the transition amplitude. Therefore, we model the higher ATI peak to have a non-dynamical phase shift of $-\pi/2$ compared to the lower adjacent ATI peak. The resulting probabilities are

$$
\begin{align*}
|c_{\text{SB}}(\text{SB})|^2 &= 4J_z^2 \cos^2 \left( \frac{\pi}{4} + \varphi \right), \\
|c_{\text{ATI}}(\text{ATI})|^2 &= J_0 \{ 1 + J_2 [4 - 8 \cos^2 \left( \frac{\pi}{4} + \varphi \right)] \},
\end{align*}
$$

which is identical to the RABBIT result in Eq. (9) apart from the CEP-translation by $-\pi/4$. In Fig. 4 (c,d) we show a sideband and an ATI peak resolved over CEP and emission angle. These drastically different photoelectron distribution, as compared to RABBIT in Fig. 2 (c), are explained by the alternating sign of the $2\omega$-ATI peaks: $f^{(k)}(\text{ATI}) = (-i)^k J_k$, where $k$ is the number of $2\omega$ photons absorbed.

In this Letter we have studied theoretically the separability of quantum dynamics in a range of recent attosecond experiments that rely on weak probe laser beams. We have found that RABBIT sidebands are exceptional, as they are not affected by non-dynamical multi-photon phase shifts. In contrast, experiments with an unbalanced number of photons may suffer from substantial CEP-shifts. We have identified such effects at play in both LAP and HHG, by ab initio simulation of the “checkerboard” structure reported by Laurent et al. [13], and by simulation of the FEL experiment with twin sidebands, reported by Maroju et al. [22]. Further, we have shown that the total photoelectron probability is conserved over CEP in these LAP experiments, which is not understood by naive usage of perturbation theory [13]. Finally, we have shown that $2\omega/\omega$ ATI experiments, akin to the work of Zipp et al. [21], have a large CEP-shift of $-\pi/4$, relative to synchronized RABBIT experiments, which has neither been identified nor explained previously. This opens up a call for investigations of CEP effects on an absolute scale, relative to known reference experiments. All numerical results have been confirmed by a rule of thumb, derived from KFR theory, which shows that many attosecond experiments can be analysed in a simple universal framework.

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