Strong Field Ionization in $\omega$-2$\omega$ Laser Pulses: Phase-of-the-Phase Spectroscopy

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The presence of a weak second-harmonic field in an intense-laser ionization experiment affects the momentum-resolved electron yield, depending on the relative phase between the $\omega$ and the $2\omega$ component. The proposed ‘phase-of-the-phase spectroscopy’ quantifies for each final electron momentum a relative-phase contrast (RPC) and a ‘phase of the phase’ (PP), describing how much and with which phase lag, respectively, the yield changes as function of relative phase. Experimental results for RPC and PP spectra for rare gas atoms and CO$_2$ are presented. The PP spectra demonstrate a rather universal structure with pronounced features that allow to distinguish photoelectrons that rescatter during the first or the second return to their parent ion. Details in the PP and RPC spectra are target sensitive and thus may be used to extract structural (or even dynamical) information with high accuracy.

Momentum-resolved photoelectron spectra from strong-field ionization of atoms and molecules contain a wealth of information about the ionizing laser field, the target, and ultrafast processes that may occur in it (see, e.g., Refs. [1, 2] for recent overviews). If one is able to disentangle this information one may use photoelectron spectra to image the entire ionization dynamics and the structural information convoluted into it. Thanks to the appealing possibility to analyze most of strong-field ionization dynamics in terms of semi-classical electron trajectories it has been demonstrated that many of the complex and, at first sight, puzzling features could finally be explained in simple and intuitive terms. Examples are various low-energy structures [3–14], intra and inter-cycle interferences [15–17], “holographic side-lobes” [18, 19], molecular strong-field ionization [20, 21], or “interference carpets” [22].

Refined strong-field experiments make use of additional experimental “knobs,” such as a pump-probe delay, a carrier-envelope phase (CEP) in the case of few-cycle pulses (see, e.g., Refs. [23] and [24, 25] for reviews), or a relative phase between two laser fields of different frequency [26–28]. The resulting changes in the photoelectron spectra may then help to unequivocally indentify certain ionization scenarios. In this Letter, we present experimental results obtained with a co-linear, two-color laser set-up in which the strong $\omega$-component drives the actual ionization dynamics, and the sole purpose of the weak $2\omega$-component is to introduce a relative-phase dependence.

By Fourier-transforming the momentum-dependent ionization yield with respect to the relative phase, phase-of-the-phase (PP) and relative-phase-contrast (RPC) spectra are calculated, analyzed in terms of “simple man’s theory” (SMT), and compared to the corresponding results from the numerical solution of the time-dependent Schrödinger equation (TDSE). The main findings are: (i) the overall structure of the PP spectra is largely target-independent and displays features that can be assigned to certain electron trajectories via comparison with SMT; (ii) target-dependent features are clearly visible in the RPC and PP spectra, thus making PP spectroscopy an attractive approach for determining, e.g., scattering cross sections with higher accuracy than from “ordinary” photoelectron spectra.

The basic structure of photoelectron spectra is qualitatively well understood. For linear laser polarization, the so-called “direct electrons” extend up to energies $p^2_z/2 = A^2_0/2 = 2U_p$, where $p_z$ is the photoelectron momentum along the polarization direction. $A_0$ is the vector potential amplitude, and $U_p = A^2_0/4$ is the ponderomotive energy (atomic units are used unless otherwise stated). If electrons are driven back to their parent ion by the laser field, energies up to $10U_p$ may occur upon rescattering. A wealth of information is encoded in this high-energy above-threshold ionization (HATI) part of the photoelectron spectra about both (i) the driving laser field and (ii) the structure of the target (via its scattering cross section [29, 30]). Moreover, HATI is more robustly accessible to theoretical modeling than the low-energy part of photoelectron spectra, which is plagued by the necessity to take Coulomb-corrections into account [31–36].

Bichromatic $\omega$-2$\omega$-pulses with parallel polarizations of the field components and adjustable relative phase $\varphi$ were generated with a set-up similar to the one in Ref. [37]. Briefly, a Ti:sapphire laser system provides 100-fs pulses at 794 nm. The second harmonic is generated in a 100-µm thick BBO I crystal. The $\omega$-2$\omega$ intensity ratio is controlled by detuning the phase matching conditions through a tilt of the crystal. Birefringent calcite crystals compensate for the time lag between the $\omega$ and $2\omega$ pulse. The relative phase lag is controlled by two glass wedges mounted on piezo-driven motors. The laser pulses are focused into the extraction region of a homebuilt high-energy velocity-map-imaging (VMI) spectrometer [38] by a concave silver mirror having a focal length of 300 mm. Photoemission from Xe was used to optimize the pulse overlap. The MCP back plate of the detector system is gated using a fast electronic switch to suppress spurious signals.

Our two-color field is described by the vector potential

$$A(t) = A_0 e_z [\sin \omega t + \xi \sin (2\omega t + \varphi)]$$

in dipole approximation. The use of long pulses ensures that
envelope effects are unimportant. Only the ionization dynamics due to the \( \omega \) component is probed as long as the \( 2\omega \) field is weak (\( \xi \ll 1 \)).

Figure 1 introduces schematically the two quantities—RPC \( P \) and PP \( \Phi \)—used to describe how the photoelectron yield \( Y(\varphi) \) changes as function of relative phase. For each final momentum \( p_z, p_x \), the fundamental \( (N = 1) \) oscillation component of the yield \( Y_1(\varphi) = P \cos(\varphi + \Phi) \) is extracted via Fourier analysis. The fundamental component is found to dominate over higher-harmonic contributions \( Y_N, N > 1 \) for most final momenta. A similar analysis has been performed previously with respect to the CEP for SiO\(_2\) nanospheres [39].

Experimentally determined RPC and PP spectra for Ar are presented in Fig. 2. Figure 2a shows that the direct electrons vary most with \( \varphi \) (black and dark gray), while the rescattered electrons vary with about one order of magnitude less contrast (lighter gray areas within the black rescattering circle). The direct electrons with \( p_z > 0 \) in the PP spectrum in Fig. 2b behave predominantly \( \sin \varphi \) to \( -\cos \varphi \)-like (blue to black area labelled ’1’), the ones with \( p_z < 0 \) behave \( \cos \varphi \) to \( -\sin \varphi \)-like (green to red, ’2’). Most of the rescattered electrons behave phase-wise similar to the direct electrons in the opposite direction (’3’ red to black, and ’4’ blue to green). However, in the semi-circle-shaped momentum regions ’5’ and ’6’ beyond the respective \( 2U_p \)-cutoffs, the electrons continue to follow phase-wise the direct electrons in the same direction, i.e., ’2’ and ’1’, respectively. This effect will be discussed in more detail below. Note that the overall absolute phase is not determined by the experiment but by comparison with theory (see below).

In Fig. 3 we show RPC and PP spectra for Kr, Xe, and randomly aligned CO\(_2\). Figures 3a,b,d,f show that the overall structure of the PP spectra is universal: they resemble two overlapping clubs (indicated in Fig. 3), one colored in red to green (regions ’2’, ’5’, and ’4’ in Fig. 2b), the other one blue to black (’1’, ’6’, and ’3’). The blunt parts of the clubs (regions ’3’ and ’4’) represent rescattered electrons. The tip regions ’5’ and ’6’ are investigated below. While the overall structure is similar for all species, a target dependence is most obvious in the RPC spectra Figs. 3a,c,e and 2a, and also reflected in detailed features of the PP spectra. This allows PP spectroscopy to be employed for imaging and as a sensitive test of theoretical models. In what follows, we aim at reproducing the results of Fig. 2 by SMT and the numerical solution of the single-active electron TDSE, using an effective Ar potential.

SMT (see, e.g., [22, 41, 42]) should be able to reveal the origins of the common, overall features observed in the PP spectra. Given a vector potential \( (\mathbf{A}) \) the electron momentum before rescattering reads \( p(\tau)/A_0 = \mathbf{p}(\tau) = \mathbf{a}(\tau) - a(\tau_1) \). The dimensionless time \( \tau = \omega t \), momentum \( \mathbf{p} = p/A_0 \), and vector potential \( \mathbf{a} = \mathbf{A}/A_0 \) are introduced to highlight the universal scaling of SMT, and \( \tau_1 = \omega t_1 \) with \( t_1 \) the ionization time. Assuming isotropic scattering and considering the planar motion in, e.g., the \( xz \)-plane, the momentum after rescattering and once the pulse is off reads
\[
\frac{\mathbf{p}_z}{\mathbf{p}_x} = \begin{pmatrix} -a(\tau_2) \\ 0 \end{pmatrix} + [a(\tau_2) - a(\tau_1)] \frac{\cos \theta}{\sin \theta}.
\] (2)
Here, \( \tau_2 = \omega t_2 > \tau_1 \) with \( t_2 \) the rescattering time, and \( \theta \) the scattering angle. The maximum momentum \( \mathbf{p}_{\text{max}} = \sqrt{5} \) corresponds to the well-known cut-off energy \( 10U_p \), which
occurs if ionization happens at $\tau_1/2\pi = 0.543$ (or at 0.043 in opposite direction) and the $\theta = \pi$-backscattering at $\tau_2/2\pi = 1.22$ (0.72). The $10U_p$ rescattering rings, i.e., rings in the momentum plane centered at $-a(\tau_2)e_z$ with radius $|a(\tau_2) - a(\tau_1)|$, are shown in Fig. 4b and indicated in all experimental and TDSE spectra. If electrons do not scatter during the first but the second return they end up in the opposite $z$-direction and generate second-return rescattering rings. In particular, if ionization and rescattering occur at $\tau_1/2\pi = 0.519$ (0.019) and $\tau_2/2\pi = 1.74$ (1.24), respectively, a local maximum cutoff energy of $7U_p$ is reached, also indicated in Fig. 4b, with the respective trajectory shown in Fig. 4b.

The fundamental structure of the PP spectra is easy to reproduce within SMT: (i) by looping over $\tau_1$ and $\tau_2 > \tau_1$ all electron trajectories that actually rescatter are considered, and (ii) weighted by their probability $W = W_iW_r$ where $W_i$ is an ionization probability and $W_r$ a rescattering probability.

An instantaneous tunneling rate through a triangular barrier $W_i(\tau_1) \sim \exp[-2/3|e(\tau_1)|]$ (with $e(\tau) = -e, a(\tau)$ the dimensionless electric field) has been taken. We are not interested in absolute numbers here, as our SMT modeling should be robust and depend only on $a$ and $\tau$. In a first, crude approximation $W_i(\tau_2 - \tau_1) = (\tau_2 - \tau_1)^{-a}$ with a spreading exponent $s > 0$ has been chosen. Such a rescattering probability accounts for wave-packet spreading (see, e.g., [25, 41]) but neglects any momentum and angular dependence of the scattering cross section. If several trajectories end up in the same final momentum bin only the most probable has been considered, thus neglecting interference effects.

The direct electrons have been weighted by $W_r^{i(direct)}(\tau_1) = W_i(\tau_1)\exp(-\beta\tau_1^2/e(\tau_1))$ with $\beta = 15$ (to produce a reasonable lateral spread of the momentum distribution). The result of such a simple modeling for $s = 3/2$ is shown in Fig. 4a. The spreading exponent has been chosen smaller than $s = 3$ for free, 3D Gaussian wave-packet spreading in order to mimic Coulomb focusing [44]. The structure of the two overlapping clubs—one red, the other blue—is clearly visible. Although the $2\omega$-component was weak ($\xi = 0.05$) an extension of the $10U_p$-cutoff is clearly seen as a small strip of opposite color around the blunt, high-momentum parts of the clubs.

Obvious disagreements between Fig. 4 and all experimental results are (i) the oversimplified PP behavior consisting of essentially only two PP s (red or blue), and (ii) the too short low-momentum tip of the club ending at $|\vec{p}_z| = 1$, corresponding to the $2U_p$ cut-off for direct electrons. In all experimental and TDSE results the club tips extend beyond $2U_p$ (see regions ’5’ and ’6’ in Fig. 2b). On the other hand, in all experimental RPC plots the $2U_p$ cut-offs match well with a pronounced drop in contrast. Hence the question arises why the electrons in a well-defined region beyond $2U_p$ still follow phase-wise the direct electrons before the oppositely behaving rescattered electrons take over.

In the SMT modeling of Fig. 4, the trajectories that rescatter at their first return dominate because of the $(\tau_2 - \tau_1)^{-s}$-penalty for later returns. This is the reason why there is essentially only a sin-like or a $-\sin$-like behavior for all final momenta (i.e., blue or red). In a refined modeling, a rescattering probability $\sim W_i(\tau_2 - \tau_1)\sigma(q, \theta)$ with a differential scattering cross section (in first Born approximation) of the form

$$\sigma(q, \theta) = \frac{(2Z)^2}{|\vec{p}^2 + 4q^2\sin^2(\theta/2)|^2}$$

for a screened potential $V(r) = -Z\exp(-\mu r)/r$ has been employed. The corresponding PP spectrum in Fig. 4a is in much better agreement with the experimental results. In fact, given the simple modeling, the agreement is striking. Going from Fig. 4a to d, the PP in regions 3 and 4 changes from sin (blue) to $-\cos$-like (black) for $\vec{p}_z < 0$, and from $-\sin$ (red) to cos-like (green) for $\vec{p}_z > 0$. The reason for this change lies in the introduction of an additional functional dependence on the relative phase via the cross section $\sigma(q(\varphi), \theta)$.

As in the SMT modeling all spectral features can be understood in terms of electron trajectories, we were able to identify the origin of the ring-like extensions ’5’ and ’6’ beyond $2U_p$. Figure 4a suggests that these features are due to electrons that rescatter during the second return because the corresponding rescattering rings are centered around the $2U_p$ cutoffs. A cross section of the form (3) favors forward scattering (i.e., small $\theta$), and the more so the larger the instantaneous scattering momentum $q = |a(\tau_2) - a(\tau_1)|$ is. As a consequence, the trajectories which scatter with lower-momentum later during the second return (incident from the opposite direction) may have a higher probability for large $\theta$ than the trajectories that rescatter during the first return, despite the wave-packet-spreadings.
penalty for late returns. It is thus the competition between the probability factors governing wave packet spreading and the momentum-dependent large-angle scattering which is responsible for the club tips beyond $2U_p$.

Figure 5 shows RPC and PP spectra for Ar from a TDSE simulation [40] in single-active electron approximation. Abel-projected photoelectron momentum spectra were calculated for different $\phi$ and subsequently treated like the experimental spectra. The correct ionization potential of Ar was imposed by choosing the $3p_0$-orbital in the effective potential $V(r) = -[1 + 17 \exp(-2.11375r)]/r$ as the initial state. Comparison with Fig. 2 yields satisfactory agreement with respect to the overall club structure in the PP and the qualitative features in the RPC spectra. We note that the dynamic range of the experimental detection is two orders of magnitude lower than the one shown for the TDSE. This might be the reason why the marked features close to the second-return cut-off in the TDSE PP spectrum (labelled 'A' and 'B') are absent in the experiment. The remaining differences could be due to the idealized pulses and the neglect of focal averaging in the simulation, slightly different laser parameters in experiment and simulation, or the possibly inadequate (but computationally unavoidable) assumption of a single active electron in the TDSE calculations. In any case, such differences show the sensitivity of PP spectroscopy and its power to discriminate between various effects.

In summary, we have introduced PP spectroscopy and demonstrated its usefulness by unambiguously identifying the role of second-return electrons for certain final electron momenta. This information could only be extracted because of the additional relative-phase knob at hand. The presence of the $2\omega$-field component tags each emission time according to how ionization probability changes as function of relative phase. This change is subsequently mapped through the actual electron dynamics to the final photoelectron momentum. The overall structure of the PP spectra for various rare gas atoms and CO$_2$ is universal and simple to analyze in terms of SMT, provided a reasonable rescattering cross section is employed and Coulomb focusing is taken into account. However, there are features that differ from target to target in both the RPC and the PP, paving the way to employ them for imaging electronic structure or dynamics, and as tests for theoretical predictions or cross sections. We also anticipate that PP will be able to discriminate among coherent, delayed, or thermal electron emission, e.g., in non-sequential ionization of multi-electron systems. For applying the method to a given target (atom, molecule, cluster, or nanostructure), we imagine the following course of action in practice: a certain spectral feature in an “ordinary” photoelectron spectrum is identified and hypothetically attributed to a certain phenomenon (such as rescattering or focusing of certain trajectories, polarization of the target, or internal dynamics). A PP experiment will then reveal if the PP and RPC signature of this feature is in accordance with the predictions from the conjectured theory.

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FIG. 4: (color online). (a) Final SMT photoelectron momenta for $\xi = 0$, absolute value of electric field at ionization time $|e(\tau_i)|$ color-coded. Rings representing local maxima in final momentum $|\mathbf{p}_z|$ due to rescattering during first, second and third return are labelled. For better visibility, final momenta in respective opposite directions (originating from electrons emitted half a laser cycle earlier) are plotted light gray. (b) Normalized excursion $\overline{\tau}_r$ of electron rescattering at first (solid) and second return (dotted), respectively, leading to final momenta indicated ‘x’ in (a). SMT PP spectra for $\xi = 0.05$, taking into account (c) ionization and wave packet spreading ($s = 3/2$) and (d) additionally a scattering cross section [5] (with $Z = 18$ and $\mu = 1/2$). Same color coding in (c) and (d) as in previous figures. White numbers in (d) analogously to Fig. 2b.

FIG. 5: (color online). RPC (a) and PP (b) spectra for Ar from TDSE $(10^{14}$ W/cm$^2$, $\xi = 0.05$). Durations of $\sin^2$-shaped pulses for 800 and 400 nm were 60 and 50 fs, respectively. Additionally to $2U_p$, cut-offs and $10U_p$-rings, second-return cut-off rings are included, which form boundaries for features labelled 'A' and 'B'. Slight left-right asymmetries visible in (a) are remnants of CEP-dependence.
