Revealing laser-coherent electron features using phase-of-the-phase spectroscopy

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Abstract

Phase-of-the-phase (PoP) spectroscopy is extended to two-color laser fields having a circularly counter-rotating polarization. In particular, the higher harmonics of the (two-color) phase information are analyzed in order to extract the laser-coherent part of the photoelectron spectra taken under complex target conditions. We illustrate this with a proof-of-principle simulation by considering strong-field electron emission from argon atoms within helium nanodroplets under realistic experimental conditions, i.e. a limited number of photoemission events. Multiple elastic scattering on neutral helium atoms creates a laser-incoherent background, but the higher harmonics of the PoP-signal allow to resolve the coherent contribution to the photoemission.

Keywords: strong-field ionization, photoelectron spectra, attosecond physics

1. Introduction

Photoelectron momentum distributions obtained in experiments with strong laser fields are a treasury containing valuable information related to the target, the laser field, and the ultrafast phenomena during the process of ionization. However, in complex targets scattering may destroy the phase relation between the photoelectron yield and the laser field, and these electrons may clutter the coherent electron spectral features known from atomic gas-phase targets (such as plateaus, low-energy structures etc.). The recently proposed phase-of-the-phase (PoP) technique [1] can serve as Occam’s razor with respect to incoherent processes that spoil photoelectron spectra (PES). The general principle of the PoP and similar techniques is based on the irradiation of a target with a laser field that has a periodic parameter such as the relative phase between two fields with different colors [1–9] or the carrier-envelope phase [10]. However, in contrast to techniques where the forward-backward asymmetry [11–16] or side streaking [17–22] of the PES is analyzed at selected values of the periodic parameter, the PoP suggests to perform the Fourier transform with respect to this periodic parameter and to reveal how the PES follow it, e.g. whether the PES change with the same periodicity as the relative phase (‘first harmonic’) but a certain phase lag \( \Phi_1 \) (the ‘phase of the phase’), twice per period (‘second harmonic’, with the corresponding phase lag \( \Phi_2 \)), and so on. It turns out that these Fourier components have well distinguished features at certain momenta. Specifically, the phase lag of some momentum-resolved Fourier components sharply flips along certain curves in momentum-space. Those ‘flipping curves’ are sensitive to the parameters of the system, i.e. the laser intensity and frequency, and the type of target used.

In [9], the PoP technique was developed for the case of two-color circularly polarized counter-rotating intense laser fields with frequency ratio 1:2. Here, we extend the method to arbitrary ratios. We establish the general property of the PoP-flipping curves, showing that they all have a quite simple, circular geometry and therefore are easier to analyze than, for instance, the features observed in the PES for two-color linearly polarized fields [1, 2]. One of the flipping curves was predicted in [9]. However, for fast photoelectrons, where the signals are low as a consequence of the significantly reduced ionization probability, flipping curves may not be resolved due to the small number of events on the detector. In the present paper, we propose a way to bypass this problem extending the theory so that it can be used for a broader domain of parameters. We choose the specific frequency ratio \( \omega - 2\omega \) in most of our examples, as it is widely used in both experimental and theoretical investigations [23–29]. Note,
however, that for other frequency ratios \( \omega - b\omega \), being of particular interest nowadays (see, e.g. [30–34]), the PoP technique can also be applied.

The paper is organized as follows. Section 2 is devoted to the derivation of the relevant formulas within the Strong Field Approximation (SFA) [35–37], and the general properties of PoP spectra are derived. In section 3, the PES obtained for a complex target where multiple elastic scattering of electrons after ionization are taken into account is presented. Elastic scattering gives rise to a huge contribution of the incoherent electron signal (with respect to the phase shift between the laser field components). The PoP technique is then applied in order to reveal the small coherent features in the electron yield. We conclude in section 4. Atomic units are used throughout this paper unless otherwise stated.

2. Theory

2.1. Strong field approximation

Let the vector potential of a laser field in dipole approximation be of the form

\[
A(t) = A_\alpha(a_\omega t) + A_\beta(b_\omega t + \phi). \tag{1}
\]

The PoP technique is based on the momentum-resolved PES, Fourier-transformed with respect to the phase shift \( \phi \) between the laser field components,

\[
Y(p, \phi) = Y_0(p) + |Y_1(p)| e^{i\phi + \Phi_1(p)} + |Y_2(p)| e^{i2\phi + \Phi_2(p)} + \cdots + c.c. \tag{2}
\]

The phases \( \Phi_k(p) = \arg Y_k(p) \) describe the phase lag of the change in the photoelectron yield as a function of the relative phase \( \phi \), hence the name ‘phase(s) of the phase’. In the previous papers [1, 2, 9], only \( Y_1(p) \) was considered. A representation of the series (2) is shown in figure 1 (yields are normalized to a maximum value of 1 throughout the paper). The curves \( Y_n(p) = 0 \) in momentum space are very sensitive to the parameters of the system under consideration. Let us derive simple, analytical properties of \( Y_n(p) \) using the SFA [35–37]. Within the SFA, we can write the photoionization rate as

\[
Y = \left| \int_0^T P(p, t) e^{iS(t)} dt \right|^2, \tag{3}
\]

where the prefactor \( P(p, t) \) (see the explicit form, e.g. in [38]) is slowly varying on the timescale of a laser period \( T \), the fast oscillating function \( S(t) \) in the exponent

\[
S(t) = \int_0^t \left[ \frac{(p + A(t))^2}{2} + I_p \right] dt', \tag{4}
\]

is the classical action of the photoelectron with final momentum \( p \), \( A(t) \) is the vector potential determining the laser electric field \( E(t) = -\partial_t A(t) \), and \( I_p \) is the ionization potential.

We calculate the yield (3) with exponential accuracy using the saddle-point method (see, e.g. [38–40] and references therein) and obtain

\[
Y(p, \phi) \sim \left| \sum s e^{iS(t_s)} \right|^2, \tag{5}
\]
where the complex times $t_i$ are solutions to the saddle-point equation

$$\frac{(p + A(t_i))^2}{2} + I_p = 0. \tag{6}$$

In the present paper, we aim at the case of two-color, circularly polarized, counter-rotating fields described by a vector potential of the form

$$A_x(t) = A_0 [\cos(a\omega t + \xi \cos(h\omega t + \phi))]$$

$$A_y(t) = A_0 [\sin(a\omega t) - \xi \sin(h\omega t + \phi)], \tag{7}$$

and $A_z = 0$, with the second component being weak ($\xi \ll 1$), and $a$, $b$ coprime, and $b > a$. For the sake of simplicity, we assume an infinitely long pulse with a constant envelope $A_0 = E_0 / a\omega$, where $E_0$ is the amplitude of the main (i.e. strong) component of the laser field.

Introducing the Keldysh parameter $\gamma = \sqrt{2I_p / A_0}$ and a dimensionless momentum $q = p / A_0$, one can rewrite (6) as

$$0 = 1 + \gamma^2 + q^2 + \xi^2 + 2q \cos(a\omega t + \alpha)$$

$$+ 2\xi (q \cos(h\omega t + \alpha + \phi) + \cos((a + b)\omega t + \phi)]. \tag{8}$$

Here, $\alpha$ is the angle of photoelectron emission in the $xy$ plane, i.e.

$$q = (e_x \cos \alpha + e_y \sin \alpha). \tag{9}$$

Most electrons will be emitted within the $xy$ plane as long as non-dipole effects are negligible [37, 41]. Hence we do not consider final photoelectron momenta out of the $xy$ plane in this work.

In the monochromatic case ($\xi = 0$), the emission is isotropic with respect to the angle $\alpha$ [41] (see also [42]). Therefore, it is convenient to introduce a new variable

$$a\tau = a\omega t - \alpha. \tag{10}$$

In the two-color case, it is useful to introduce additionally

$$a\theta = (a + b)\alpha + a\phi. \tag{11}$$

One can then rewrite (8) as

$$0 = 1 + \gamma^2 + q^2 + \xi^2 + 2q \cos(a\tau)$$

$$+ 2\xi (q \cos(b\tau + \phi) + \cos((a + b)\tau + \theta)], \tag{12}$$

and the action (4) as

$$S(\tau) = \frac{A_0^2}{2a\omega} \left[ (1 + \gamma^2 + q^2 + \xi^2) a\tau + 2q \sin(a\tau)$$

$$+ 2\xi (q \frac{a}{b} \sin(b\tau + \phi)$$

$$+ \frac{a}{a + b} \sin((a + b)\tau + \theta) \right] \right|_{\tau = 0} \tag{13}.$$

The yield only depends on $\alpha$ and $\phi$ in the combination (11). The coefficient in front of $\alpha$ in (11) is related to the $(a + b)$-fold symmetry of the electric field and, hence, of the vector potential and the yield. As a consequence, the yield has the property

$$Y(q, \alpha, \phi) = Y(q, \alpha + \frac{a}{a + b} \phi, 0)$$

$$= Y(q, 0, \phi + \frac{a + b}{a} \alpha). \tag{14}$$

### 2.2. General properties of the PoP

Now let us pass over to the main quantity of interest, namely, the Fourier components of the yield

$$Y_0(q, \alpha) = \frac{1}{2\pi} \int_0^{2\pi} Y(q, \alpha, \phi) e^{-i\phi} d\phi. \tag{15}$$

Property (14) suggests to factor out the $\alpha$-dependence of each $Y_0$,

$$Y_0(q, \alpha) = Y_0(q, 0) e^{ia\phi}. \tag{16}.$$

This form allows to conclude that, due to the periodicity in $\alpha$, only harmonics which are multiples of $a$ are non-zero. In figure 2, two 5-fold-symmetric configurations are shown,
Figure 3. Left panel: total yield $Y$ and absolute values of several of the Fourier components $Y_n$ for $n = 0-3$, taken along the $q_x = p_x/A_0 > 0$ direction. All parameters are the same as in figure 1. Spiky minima correspond to momenta where the phase flipping occurs and are indicated by squares on the right panel. Right panel: Radial momenta of the phase flipplings for the first (black line) and the second (gray lines) harmonics of the Fourier expansion for argon in a two-color $\omega-2\omega$ laser field with the laser main component’s wavelength $\lambda_1 = 800\,\text{nm}$ plotted versus the intensity of that laser field. The gray area represents a domain with a yield above 0.1% of its maximum, and the dashed line indicates the position of this maximum.

where in the $a:b = 1:4$ case the first non-vanishing phase-dependent term is $Y_1(p)$ while in the $a:b = 2:3$ case it is $Y_2(p)$.

Another general property of the $Y_n$ is revealed if we consider the action and the saddle-point equation. One may easily verify that if $t_0(p, \alpha = 0, \phi)$ is a solution to (12) for a given $\phi$, then $t_0(\alpha = 0, -\phi) = 2\pi a - t_0^*(p, \alpha = 0, \phi)$ is a solution for $-\phi$. Next, inserting these solutions into the action (13) and evaluating the imaginary part of it, one obtains the same value in both cases, i.e. $\Im S(\phi, t_0(\alpha = 0, \phi)) = \Im S(-\phi, t_0(\alpha = 0, -\phi))$. Thus, the yield is symmetric in $\phi$ for $\alpha = 0$. This allows to rewrite (15) as

$$Y_n(q, \alpha) = \frac{1}{\pi} \int_0^{2\pi} Y(q, 0, \phi) \cos(n\phi) d\phi \, e^{i\alpha \phi}.$$

Finally, let us pass to the arguments $\Phi_n = \arg Y_n$ of these Fourier components (i.e. to phases-of-the-phase) that can now be written as

$$\Phi_n(q, \alpha) = n \frac{a + b}{a} \alpha + \pi \delta_n(q).$$

Here, $\delta_n(q)$ is related to the sign of the purely real $Y_n(q, 0)$ and can only have values equal to 0 for $Y_n(q, 0) > 0$ or 1 for $Y_n(q, 0) < 0$ (without loss of generality). Expression (18) highlights an essential advantage of the PoP technique related to circular, counter-rotating fields: the only type of momentum-dependent features that appear in the PoP spectra are circles centered around $q = 0$ at which the phase has a sharp flip of $\pi$. This makes the PoP flipping curves one dimension simpler compared to the case of linear+linear two-color fields and allows to represent their behavior for a broad variety of parameters within a single plot (see the example shown in figure 3), thus making PoP a possibly handy laser-calibration technique.

2.3. PoP flipping of the higher fourier components

Electron emission in circularly polarized fields at intensities above $10^{13}\,\text{Wcm}^{-2}$ and $I_p/\omega \gg 1$ has the highest probability for radial momentum $q = 1$, i.e. $p = A_0 = E_0/\omega$ [41]. In [9], the position of the flipping of the PoP of the first Fourier component $\Phi_1$ for $\omega-2\omega$ laser fields (i.e. $a = 1$, $b = 2$ in the current notation) has been analytically described. It appeared that the circle of the $\Phi_1$ flipping is in general located in the momentum space region far beyond the peak of the ionization probability where the yield might be too low in actual measurements. Here we show that in such cases the PoP technique using higher harmonics of the Fourier expansion should be more practicable from the experimental point of view. In particular, for the laser parameters considered above, $\Phi_2$ flips close to the maximum yield (see example in figure 3) and, thus, should be easier to observe than the flipping of $\Phi_1$.

3. PoP-based exclusion of incoherent scattering

In order to demonstrate another practical application of the PoP technique, we introduce a model describing the PES generated by ionization of atoms in a complex environment. In particular, we consider helium nanodroplets [43]. They are widely used as finite spectroscopic matrices for embedded particles, called dopants [44–48]. The ultralow temperature of the droplets, the weak electronic interaction between dopant and helium, and the optical transparency up to about $20\,\text{eV}$ allows for almost undisturbed high resolution spectroscopy in the microwave, IR and UV spectral range of targets varying in complexity from simple atoms [49] to large biomolecules [49–51]. In contrast, photoelectrons emitted by the dopant might scatter on the surrounding helium atoms before leaving the droplet. In that way, an incoherent electron yield is added to the PES since the phase information is almost lost in the scattering. This incoherent contribution might mask the interesting, laser-coherent features in the ordinary PES while PoP spectra still show them, as will be demonstrated in the following.

In our computer experiment, we use argon as the dopant. The laser parameters considered are as follows: two counter-rotating, circularly polarized components with wavelengths 800 and 400 nm ($a = 1$, $b = 2$), intensities are $I_1 = 2 \times 10^{14}\,\text{Wcm}^{-2}$ and $I_2 = 2 \times 10^{12}\,\text{Wcm}^{-2}$ ($\xi = 0.05$). A sketch of the experimental setup to produce single-atom-doped helium droplets is shown in figure 4. We consider droplets of size $D \approx 60\,\text{Å}$, that is $N \approx 3 \times 10^4$ atoms. At 20 bar and for a 5 $\mu$m diameter gas exit, nozzle temperatures of about 16 K have to be established [52]. The partial pressure of argon gas in the pick-up region is adjusted to conditions such that only a single Ar atom is present in each droplet on average. DFT calculations show that rare gas impurities reside near the center of the droplet [53, 54].

The photoelectrons have a significant probability of multiple scattering on the enclosing neutral helium atoms before leaving the droplet and flying towards the detector. As the mean distance between the He is larger than their size, we treat each scattering within a single-atom approach. Due to
the significant difference between the ionization potentials of argon (15.8 eV) and helium (24.6 eV), we neglect the ionization probability of the latter and consider photoelectrons produced from argon only. For the sake of simplicity, we reduce our problem to the two-dimensional case, taking into account the polarization plane only, since most of the photoelectrons produced from an atom in a circularly polarized laser field have their momenta within the polarization plane. As a result, most of the coherent signal is in the momentum distribution in this plane. As typical kinetic energies of photoelectrons produced from argon with the chosen laser intensity are too low to ionize or excite helium, we only take elastic scattering into account. Moreover, most electrons have an energy close to the average \( \langle p^2 \rangle / 2 = 15.4 \text{ eV} \) so that we neglect the dependence of the cross section on the energy of the scattered electrons. The total cross section \( \sigma \) and the angular distribution \( d\sigma / d\Omega \) at \( p^2 / 2 = 15 \text{ eV} \) are taken from [55]. For \( N_\phi = 20 \) phases \( \phi \in [0, 2\pi] \) (i.e. 10 sampling points per period of \( Y_2 \)) we simulate a finite number \( N_e \) of ionization events with the initial momentum distribution taken from the SFA. Then we simulate their scattering with the probability of traveling a distance \( s \) before a new scattering event as

\[
w(s) = 1 - e^{-nR},
\]

where the concentration of helium atoms \( n \) is assumed to be constant all over the droplet. Thus, \( s \) is chosen randomly as \( s = -(\sigma R)^{-1} \log R \) where \( R \) is a random number between 0 and 1. If \( s \) is smaller than the distance to the boundary of the droplet, the scattering occurs. The angle of scattering \( \eta \) is then chosen randomly in a range from \(-\pi\) to \(\pi\) according to the angular distribution \( d\sigma / d\Omega \). The total number of scattering events for each initial electron is not restricted.

An example of a PES obtained with the SFA is shown in figure 5(a). If an atom is trapped inside a helium droplet the PES is significantly distorted because of electron-He scattering (see figure 5(b)). The three-fold symmetric laser-coherent part of the total signal is now deeply suppressed by an almost circular PES of multiply scattered electrons. The percentage of electrons as a function of the number of scattering events in figure 6 shows that only about 6% of the laser-coherent signal survives. After having simulated a set of PES for different relative phases \( \phi \), we apply the PoP technique and obtain the phase flipping curves (see figure 7). Due to the low ionization probability of photoelectrons

\[
\Phi_1 \text{ predicted by the SFA. (b) } \Phi_1 \text{ obtained with a finite number of } N_e = 2 \times 10^7 \text{ photoelectrons for each phase } \phi. \text{ (c) } \Phi_2 \text{ predicted by the SFA. (d) } \Phi_2 \text{ obtained with a finite number of } N_e = 2 \times 10^7 \text{ photoelectrons for each phase } \phi. \text{ (e) } \Phi_3 \text{ same as (d) but with elastic scattering on neutral helium atoms in the droplet taken into account. In each case } N_\phi = 20 \text{ phases } \phi \in [0, 2\pi] \text{ have been simulated.}
\]
with high momenta, the flipping curve for $\Phi_2$ (which is at $p = 1.76$ a.u.) is only visible if a huge number of ionization events is considered ($N_e \geq 2 \times 10^8$ for a grid with $dp = 0.02$ a.u.) even without scattering taken into account. However, as previously noted, instead of increasing the number of particles (i.e. the measurement time in the experiment) one may simply consider $N_e$, the phase of the second harmonic in the Fourier expansion. As was predicted in figure 3, it requires a significantly smaller $N_e$ for the phase-flipping to remain visible, even when the spectra are distorted by scattering, as shown in figure 7(e).

4. Conclusions

To wind up, we showed that a slight extension of the PoP technique applied to a two-color, circularly polarized and counter-rotating intense laser field can significantly improve its practicability: considering higher terms in the Fourier series of the photoelectron yield, PoP spectra with flipping curves that are located in regions of high photoelectron yield can be obtained. Being based on the observation of sharp and intensity-sensitive features, PoP spectroscopy could be an instrument for accurately tuning and benchmarking the laser intensity.

Secondly, we performed a simulation of photoelectrons originating from the ionization of a single argon atom inside a helium droplet by a $\omega$–$2\omega$ laser field. The electrons may scatter on their way to the detector so that laser-coherent features survive in the second-harmonic PoP spectrum calculated in such a manner.

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