Robust transverse structures in rescattered photoelectron wavepackets and their observable consequences

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Abstract

Initial-state symmetry has been underappreciated in strong-field spectroscopies, where laser fields dominate the dynamics. We demonstrate numerically that the transverse photoelectron phase structure, arising from the initial-state symmetry, is robust in strong-field rescattering, and has pronounced effects on strong-field photoelectron spectra. Interpretation of rescattering experiments need to take these symmetry effects into account. In turn, robust transverse photoelectron phase structures may enable attosecond sub-Ångström super-resolution imaging with structured electron beams.
Symmetries, exact and approximate, as well as symmetry breaking underpin our understanding of perturbative spectroscopies [1 2], where field-free symmetries of initial and final states strongly constrain the range of possible outcomes. In contrast, in intense infrared laser fields, the overall dynamics is dominated by the field itself. For example, strong infrared laser pulses can remove an electron from an atom, a molecule or in a solid (ionization), drive it away, then turn it around (propagation), and finally bring it back into the parent ion (recollision). This is the widely-used three-step model [3 4], capturing the essence of the rapidly emerging field of attosecond (asec) strong-field spectroscopies [5 7]. In the recollision step the returning electronic wavepacket (REWP) may for example recombine to the parent-ion, giving rise to high harmonics generation [5–10], or scatter elastically, giving rise to strong-field photoelectron holography [11–13] and laser induced electron diffraction (LIED) [5 6 10 13–23].

The symmetry of the laser field, and the dynamical symmetry it imposes on the continuum wavepacket, determine many qualitative features of these processes by means of selection rules [24]. In strong-field spectroscopies, such as LIED, this concept is usually taken even further: it is commonly assumed that the laser field entirely dominates the recollision step and the transverse phase structure of the ionized electron is “washed” out in the propagation step [10]. Technically, this assumption is implicitly enforced by the stationary-phase treatment of the strong-field approximation (SFA) [8 9].

Recently, an experimental/theoretical study in our institute [25] has shown that the rescattering probability in trans-butadiene is specific to the ionization channel and the molecular orientation accessed by strong-field ionization, rather than a property of the driving field alone. One possible interpretation of these results is that the REWP retains, even in the presence of the strong driving field, its transverse phase structure imprinted by the initial state. This structure should significantly influence the recollision process itself and alter the shape and the interpretation of strong-field photoelectron spectra. In turn, transverse phase structure of the REWP may enable atomic-scale engineering of structured electron beams. This is the electron-beam analogue to structuring the illumination in super-resolution light microscopy [26], giving rise to sub-Ångström attosecond super-resolution spectroscopy.

Unfortunately, due to limited statistics the work [25] was not able to obtain the angle- and ionization-channel-resolved photoelectron spectra leaving alternative interpretations possible. The goal of this Letter is to numerically explore the consequences of the initial-state
symmetry for the holographic and LIED regions of strong-field photoelectron spectra. Holographic patterns are known to be sensitive to the phase-structure of the REWP \cite{12}, which may arise from the asymmetry in the binding-potential \cite{12} or derive from the initial-state symmetry \cite{13}. On the other hand in LIED it is commonly assumed that the REWP has an asymptotically-flat wavefront. We are particularly interested in determining whether these effects are robust to the misalignment between the field-free symmetry elements and the laser-field polarization.

In molecular systems, the initial-state symmetry effects are intertwined with contributions due to the asymmetry of the potential, orientation, and nuclear motion. Combined with the cost of solving the molecular time-dependent Schrödinger equation (TDSE), this complexity makes a conclusive analysis challenging. Instead, we consider the simplest-possible example of the initial-state symmetry: a one-electron atom (He$^+$ or “Argon” \cite{27}), initially in an antisymmetric $p$ state. We solve the three-dimensional TDSE in the velocity gauge and dipole approximation \cite{28}. The grid contains 2200 radial points with spacings of $\Delta r = 0.2 \; a_0$, and angular momentum channels up to $L \leq 40$, $|M| \leq 40$. The time-step is $\Delta t = 0.03 \; \text{asec}$ and a transmission-free absorbing potential \cite{29} is applied at $r = 407.6 \; a_0$. The photoelectron spectra are calculated using surface-flux integration \cite{30, 31} continued to infinite time \cite{27}. Sine-squared envelopes are used for the vector potential $A(t)$, with carrier-envelope phase of $\pi/2$ (corresponding to the time-odd electric field $E(t) = -\partial A(t)/\partial t$).

Figure 1(a) shows strong-field photoelectron spectra for the He$^+$ ion. The 5 cycle, 800 nm electric field with peak intensity $I_{\text{max}} = 10^{14} \; \text{W/cm}^2$ is linearly-polarized along the $z$ direction. At first, we explore two initial states $|\Psi_0\rangle$: symmetric ($|\Psi_0\rangle = |2p_z\rangle$, left column) and anti-symmetric ($|\Psi_0\rangle = |2p_x\rangle$, right column) with respect to the $\sigma_{yz}$-reflection. The vertical axis ($k_{||}$) coincides with the laser polarization direction and the horizontal axis ($k_\perp$) with the “averaged” perpendicular direction. The averaging is performed in spherical coordinates over the azimuthal angle $\phi$, separately in the “left” and “right” hemispheres. Hence, the “left” side of the plot ($k_\perp < 0$) averages over $90^\circ < \phi < 270^\circ$; the “right” side ($k_\perp > 0$) over $\phi < 90^\circ$ and $\phi > 270^\circ$. Finally, the polar angle $\theta$ is defined with respect to the laser polarization direction ($k_{||}$-axis).

For the symmetric $2p_z$ case (left panel), we observe typical features of strong-field photoelectron spectra such as left–right symmetry with respect to perpendicular momenta $k_\perp$, holographic fingers in the lower-energy region below $2U_p$ ($U_p$ is the ponderomotive energy)
and recollision-circles for back-scattered electrons (small deflection angles $\theta_r$ in our notation) \cite{10}. The recollision circles corresponding to photoelectron energies around $10U_p$ ($6U_p$), and the definition of the deflection angle $\theta_r$ are indicated by the yellow (black) lines. The $10U_p$ recollision circles are due to electrons that return with maximal kinetic energy to the ion core, are scattered elastically in the backward-direction ($|\theta_r| < 90^\circ$), and gain an additional drift-momentum equal to the vector potential at the moment of recollision $A(t_r)$ \cite{10}. The maximal energy gain from the field corresponds to $\theta_r = 0^\circ$. This definition of the deflection angle is chosen to simplify the discussion of orientational-averaging below. It differs from the conventional electron-diffraction definition used e.g. in Ref. \cite{10} by $180^\circ$.

For the anti-symmetric $2p_x$ case (right panel), i.e. polarization of the laser field along a symmetry plane, the photoelectron spectra change dramatically, the most prominent feature being zero signal for $k_\perp = 0$ ($\theta_r = 0^\circ$).

For further analysis, Fig. 1(b) shows the corresponding angle-resolved photoelectron yields along the $10U_p$ circle (left) and the $6U_p$ circle (right). The yield along the $10U_p$ circle largely follows the Rutherford scattering cross-section for the symmetric case ($|\Psi_0\rangle = |2p_z\rangle$, green line). This is the basis for the retrieval of structural information in LIED \cite{10}. For the anti-symmetric case ($|\Psi_0\rangle = |2p_x\rangle$, blue line), on the other hand, the yield for back-scattering ($\theta_r = 0^\circ$) is zero and the angular dependence of the yield is changed qualitatively. Along the $6U_p$ circle (right) the same characteristic difference between the symmetric (green) and anti-symmetric (blue) case is observed.

These results are consistent with the SFA prediction for the rescattered-photoelectron amplitude \cite{32}:

$$a(k_f) = - \int dt_0 dt_c dp e^{-iS_V(t_0,t_c,p)} \times R \times I$$

where the photoionization and elastic scattering matrix elements are:

$$I = \langle p + A_z(t_0)|V_L(t_0)|\Psi_0 \rangle$$

$$R = \langle k_f + A_z(t_c)|V_c|p + A_z(t_c) \rangle.$$ 

Integration in Eq. (1) is over the ionization and recollision-times $t_0$ and $t_c$, respectively, and the canonical momentum $p$. The quantity $S_V(t_0,t_c,p)$ is the length gauge Volkov phase \cite{9} and $A_z(\tau)$ the $z$-polarized vector potential. Finally, $V_c$ is the Coulomb potential, $V_L(t_0) = zE_z(t_0)$ describes the interaction of the $z$-polarized laser field $E_z(t_0)$ and the atom and $|\Psi_0\rangle$ is the initial state.
Integrating Eq. (1) using the lowest-order stationary-phase approximation (SPA) yields [32]:

$$a(k_f) \propto e^{-iS_V(t_{0s},t_{cs},\mathbf{p}_s)} \times R_s \times I_s,$$

with the stationary matrix elements

$$R_s = \langle k_f + A_z(t_{cs})|V_c|p_{sz} + A_z(t_{cs})\rangle$$

$$I_s = \langle p_{sz} + A_z(t_{0s})|V_L(t_{0s})|\Psi_0\rangle$$

where \(t_{0s},t_{cs}\) and \(\mathbf{p}_s\) are the stationary points of the Volkov phase [9, 32]. For linearly-polarized laser fields, the stationary perpendicular canonical momentum is \(p_{s\perp} = 0\).

For the symmetric \(2p_z\) case, \(R_s\) corresponds to the standard Rutherford scattering cross-section [10] and the recollision can be represented by a single trajectory with zero transverse momentum (Fig. 1(a), upper-left panel). However, for the anti-symmetric initial state (\(|\Psi_0\rangle = |2p_x\rangle\)) the stationary hydrogenic photoionization matrix element:

$$I_s \propto \frac{(p_{sz} + A_z(t_{0s}))p_{sx}}{(4(p_{sz} + A(t_{0s}))^2 + 4)^{\frac{1}{2}}}$$

is zero by symmetry \((p_{sx} = 0)\). The lowest-order SPA rescattered amplitude in this orientation vanishes [33], and higher orders must be considered in evaluating Eq. (1).

To this end, we expand the matrix elements \(I\) and \(R\) in Eq. (1) in the transverse component of the canonical momentum \(\mathbf{p}\) around the stationary point \(\mathbf{p}_s\). After stationary-phase integration, we obtain, in the lowest non-zero order [25]:

$$a(k_f) \propto e^{-iS_V(t_{0s},t_{cs},\mathbf{p}_s)} \left(\frac{1}{i(t_{cs} - t_{0s})}\right) \times \tilde{R}_s \times \tilde{I}_s$$

$$\tilde{I}_s = \left.\frac{\partial}{\partial p_x} \langle \mathbf{p} + A_z(t_0)|V_L(t_0)|2p_x\rangle\right|_{\mathbf{p}=p_{sz}}$$

$$\tilde{R}_s = \left.\frac{\partial}{\partial p_x} \langle k_f + A_z(t_c)|V_c|\mathbf{p} + A_z(t_c)\rangle\right|_{\mathbf{p}=p_{sz}}$$

For a hydrogenic initial state, the matrix elements become:

$$\tilde{I}_s \propto \frac{(p_{sz} + A_z(t_{0s}))}{(4(p_{sz} + A(t_{0s}))^2 + 4)^{\frac{1}{2}}}$$

$$\tilde{R}_s \propto \frac{k_{fx}}{(\|k_f - p_{sz}\|^2)^{\frac{1}{2}}}.$$

This result can be intuitively understood as follows: For laser polarization along a symmetry plane, the REWP can no longer be described by an asymptotically-flat wavefront.
with well-defined return direction along the laser polarization. Instead, we can introduce an (arbitrary) small transverse momentum $\Delta p_{sx}$, and evaluate the matrix elements $\tilde{I}_s$ and $\tilde{R}_s$ by finite-differences. For an atomic $2p_x$ initial state, the finite-difference expression then factorizes into a sum of two contributions with $\pi$ phase difference. These trajectories need to be added coherently (Fig. 1(a), right panel). Hence, the high-energy (LIED) region of the strong-field photoelectron spectrum shows pronounced signatures of the phase structure of the REWP. In particular, for zero final perpendicular momentum ($k_{fx} = 0$), the rescattered-electron photoelectron signal equals zero (Fig. 1(a), right panel). Thus, the phase structure of the initial state is transferred to the REWP.

In order to explore the robustness of our results with respect to misalignment of the laser-polarization direction and the symmetry plane of the initial state, Figure 2(a) shows the photoelectron spectrum for a misalignment angle of $\alpha = 10^\circ$. The same $z$-polarized pulse as in Fig. 1 is used. The signatures of the phase structure of the REWP in the photoelectron spectra are quite different in the lower-energy (holographic) and the higher-energy (LIED) region: In the holographic region, the photoelectron signal is suppressed close to $k_{\perp} = 0$, clearly reflecting the symmetry plane of the initial state [13]. This behaviour is qualitatively similar to exact laser polarization along a symmetry plane ($\alpha = 0^\circ$) (Fig. 1(a), right panel). As expected, a “left”–“right” asymmetry of the photoelectron signal is also present for $\alpha = 10^\circ$, which breaks the overall reflection symmetry.

In the LIED region effects of the initial-state symmetry are more subtle for $\alpha = 10^\circ$. Here, the non-symmetric initial state gives rise to a transverse phase gradient of the REWP. This phase gradient results in a left–right asymmetry, and the corresponding displacement of the minimum along the recollision circles (Figure 2(b)). For the 10U$_p$ circle (left panel), the white line traces the photoelectron minimum for each $\alpha$. For $\alpha = 10^\circ$ the minimum lies around $\theta_r = -13^\circ$. This displacement deviates considerably from the Rutherford scattering cross section with minimum at $\theta_r = 0^\circ$ (Fig. 1(b), left panel). This behaviour corresponds to counter-rotation of the photoelectron signal with the initial state which prevails well beyond $\alpha = 20^\circ$ (results are shown up to $\alpha = 20^\circ$). The counter-rotation is even more pronounced along the 6U$_p$ recollision circle (Fig. 2(b), right panel). This left–right asymmetry can be understood as the interference of the signals from equations (2) and (3), sketched in Fig. 1(a).
With $|\Psi_0\rangle = \sin(\alpha) |2p_z\rangle + \cos(\alpha) |2p_x\rangle$, we get:

$$a(k_f) \propto e^{-iS_V(t_{0_s}, t_{cs}, p_s)} \times \left( \sin(\alpha) R_s \times I_s + \cos(\alpha) \left( \frac{1}{i(t_{cs} - t_{0s})} \right) \tilde{R}_s \times \tilde{I}_s \right).$$

(In the holographic region, an additional interference with the direct-electron “reference” wave will also be present.) Hence, also for imperfect alignment the photoelectron phase structure clearly influences the recollision process both in the holographic and LIED region.

Finally, Figure 3(a) shows the photoelectron spectrum for an “argon” atom interacting with the same $z$-polarized laser field used in Figs. 1 and 2 for the He$^+$ ion. Results for two different initial states are shown: For laser polarization along a symmetry plane, i.e. $|\Psi_0\rangle = |3p_x\rangle$ (left panel), similar results as for the He$^+$ ion are observed (Fig. 1(a), right panel). In particular, zero signal for $k_\perp = 0$ due to the symmetry plane of the initial state, transferred to the phase structure of the REWP, is observed both in the holographic and the LIED region. For $\alpha = 10^\circ$ (right panel), on the other hand, significant differences between the argon atom and the He$^+$ ion occur: Focussing at first on the holographic region, the photoelectron spectrum co-rotates with the initial state for the argon atom, whereas it is strongly suppressed around $k_\perp = 0$ for the He$^+$ ion (Fig. 2(a)). Hence, for imperfect alignment the specific phase structure of the REWP and its signature in strong-field photoelectron signals depends sensitively on the system.

In the LIED region of the argon photoelectron spectrum the phase structure of the REWP also results in a co-rotation of the signal for $\alpha \neq 0^\circ$, and a corresponding shift of the minima along the recollision circles (Figure 3(b)). For the $10U_p$ circle (left panel), the white line traces again the photoelectron minima for each $\alpha$. For $\alpha = 10^\circ$, the minima along $\theta_r$ are shifted by about $4^\circ$ with respect to the symmetric case ($\alpha = 90^\circ$). As for the He$^+$ ions, the left–right asymmetry is more pronounced along the $6U_p$ recollision circle (Fig. 3(b), right panel). However, we observe a co-rotation of the photoelectron signal with the initial state, whereas the signal counter-rotates with the initial state for the He$^+$-ion (Fig. 2(b)), illustrating again the sensitivity of the phase structure of the REWP on the specific quantum system.

In summary, we demonstrate that the phase structure of the REWP due to the initial-state symmetry strongly affects strong-field rescattering, opening the possibility for a complete characterization of the rescattered photoelectron. The signatures of the photoelectron’s phase structure, especially of its transverse phase gradient, are robust in the strong-field
spectra to field misalignment with respect to the symmetry plane and depend sensitively on the quantum system (Figs. 2(b) and 3(b)). These effects need to be accounted for in the interpretation of strong-field recollision experiments both in the lower-energy (holographic) and the higher-energy (LIED) region. The numerical results are supported by an analytical model based on an extension of the standard stationary-phase approximation of the rescattered-electron SFA [8, 9, 25]. While our study focussed on atomic systems, our conclusions will be as important for complex molecular systems. Accordingly, the phase-structure of the REWP, strong-field ionized along a symmetry plane, will carry a $\pi$ phase shift, leading to an interchange of maxima and minima in the LIED region of the strong-field photoelectron spectrum. While strong-field ionization along a nodal plane may be suppressed in small molecules [34], this expectation no longer applies for a broad range of typical organic molecules [25, 35].

In turn, robust transverse phase structures of the REWP are the prerequisite for attosecond sub-Ångström super-resolution imaging with structured electron beams. For example, symmetry preservation of ring-currents of the initial state [36, 37] in the rescattering process gives rise to atomic-scale electronic vortex (doughnut) beams, in analogy to light vortices used in super-resolution light microscopy [26]. Accordingly, structured attosecond electron beams may specifically probe certain regions within a molecule and thereby enhance the resolution. Furthermore, the transverse phase gradient of the returning electron beam will also emphasize and enhance the interferences between scattering centers close to the symmetry element.

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FIG. 1. (a) Angle-resolved strong-field photoelectron spectra for the He$^+$ ion for initial states that are symmetric ($|2p_z\rangle$, left column) and anti-symmetric ($|2p_x\rangle$, right column) with respect to the laser polarization direction. The strong (10$^{14}$ W/cm$^2$) 5 cycle, 800 nm driving field is linearly polarized along the $z$ axis. The 10$U_p$ (6$U_p$) recollision circles are indicated by the yellow (black) lines with the definition of the deflection angle $\theta_r$. The spectra are shown on a logarithmic scale.

(b) Angle-resolved photoelectron yields along the recollision circles indicated in (a) for $|2p_z\rangle$ (green) and $|2p_x\rangle$ (blue). The left-hand side shows yields along the 10$U_p$ circle and the right-hand side yields along the 6$U_p$ circle. Both curves are normalized to the same area.
FIG. 2. (a) Strong-field photoelectron spectrum for a “misaligned” He$^+$ ion. The initial state $|\Psi_0\rangle$ is a $2p_x$ orbital rotated by $\alpha = 10^\circ$ around the $y$ axis. The same linearly $z$-polarized laser field from Fig. 1 is used. The spectrum is shown on a logarithmic scale. (b) Angle-resolved photoelectron yield along the same recollision circles used in Fig. 1 as function of the misalignment angle $\alpha$. For the $10U_p$ circle (left), the white line traces the minimum for each $\alpha$. Both the recollision circles and the misalignment angle $\alpha$ are sketched in Fig. 2(a) and the yields are normalized to the same area for each $\alpha$. 
FIG. 3. (a) Strong-field photoelectron spectra for the model argon atom (see text) for two different initial states: $|\Psi_0\rangle = |3p_x\rangle$ (left) and a $3p_x$ orbital rotated by $\alpha = 10^\circ$ (right). The same linearly $z$-polarized laser field from Figs. 1 and 2 is used. The spectra are shown on logarithmic scale. (b) Angle-resolved photoelectron yield along the same recollision circles used in Figs. 2 and 3 as function of the misalignment angle $\alpha$. Both the recollision circles and the misalignment angle $\alpha$ are sketched in Fig. 3(a).