Dynamical electron vortices in attosecond double photoionization of H$_2$

Jean Marcel Ngoko Djiookap  
*University of Nebraska-Lincoln*, jngokodjiokap2@unl.edu

A. V. Meremianin  
*Voronezh State University*, meremianin@phys.vsu.ru

N. L. Manakov  
*Voronezh State University*, manakov@phys.vsu.ru

L. B. Madsen  
*Aarhus University*, bojer@phys.au.dk

S. X. Hu  
*University of Rochester*, shu@lle.rochester.edu

See next page for additional authors

Follow this and additional works at: [http://digitalcommons.unl.edu/physicsstarace](http://digitalcommons.unl.edu/physicsstarace)

Part of the [Atomic, Molecular and Optical Physics Commons](http://digitalcommons.unl.edu/physicsstarace), [Elementary Particles and Fields and String Theory Commons](http://digitalcommons.unl.edu/physicsstarace), and the [Plasma and Beam Physics Commons](http://digitalcommons.unl.edu/physicsstarace)

[http://digitalcommons.unl.edu/physicsstarace/232](http://digitalcommons.unl.edu/physicsstarace/232)

This Article is brought to you for free and open access by the Research Papers in Physics and Astronomy at DigitalCommons@University of Nebraska - Lincoln. It has been accepted for inclusion in Anthony F. Starace Publications by an authorized administrator of DigitalCommons@University of Nebraska - Lincoln.
Dynamical electron vortices in attosecond double photoionization of H₂

J. M. Ngoko Djiokap,1 A. V. Meremianin,2 N. L. Manakov,2 L. B. Madsen,3 S. X. Hu,4 and Anthony F. Starace1

1Department of Physics and Astronomy, University of Nebraska, Lincoln, Nebraska 68588-0299, USA
2Department of Physics, Voronezh State University, Voronezh 394006, Russia
3Department of Physics and Astronomy, Aarhus University, DK-8000 Aarhus C, Denmark
4Laboratory for Laser Energetics, University of Rochester, Rochester, New York 14623-1299, USA

(Received 13 June 2018; published 7 December 2018)

We study electron momentum vortices in single-photon double ionization of H₂ by time-delayed, counter-rotating, elliptically polarized attosecond pulses propagating along \( \hat{k} \) either parallel or perpendicular to the molecular axis \( \mathbf{R} \). For \( \hat{k} \parallel \mathbf{R} \), dynamical vortices occur similar to those found for He. For \( \hat{k} \perp \mathbf{R} \), we find dynamical vortex structures originating from an ellipticity-dependent interplay of \( ^1\Sigma^+_u \) and \( ^1\Pi^+_u \) continuum amplitudes. We propose a complete experiment to determine the magnitudes and relative phase of these amplitudes by varying pulse ellipticities and time delays.

DOI: 10.1103/PhysRevA.98.063407

I. INTRODUCTION

An unusual kind of Ramsey interference [1] was recently predicted to result in matter-wave vortex structures in the electron momentum distributions resulting from single ionization of He by a pair of time-delayed, counter-rotating, circularly polarized attosecond pulses [2]. The vortex structures and electron angular distributions were shown to be exquisitely sensitive to the pulse polarizations and their time delay. These sensitivities indicate applications to both laser pulse diagnostics and control of electron motions. Electron momentum vortices for multiphoton single ionization of He were also predicted [3]. These predictions were confirmed experimentally for three-photon [4] and four-photon [5] single ionization of potassium atoms using time-delayed, circularly polarized femtosecond pulses. The predictions of electron vortices in single ionization of He [2,3] also stimulated a number of theoretical studies of the occurrence of electron vortices in other systems and processes, including in ionization of single-electron molecular ions [6,7], in single-photon [8] and multiphoton [9] double ionization of He, in pair production processes [10], and in H-atom strong-field ionization processes [11–13].

Although Refs. [2] and [3] treated two-electron correlations in single photoionization processes, perturbation theory (PT) analyses showed that vortices produced by single-color fields originate from kinematic factors that are independent of the photoionization dynamics. Similarly, for double photoionization (DPI) of He (in which electron correlation is essential) by time-delayed, oppositely polarized attosecond pulses it was shown that such dynamical vortices occur for a particular class of electron detection geometries in which one can extract a dynamically independent kinematic factor from the sixfold differential probability (SDP) [8]. The occurrence of dynamical vortices (in which dynamical amplitudes depend on the azimuthal angles of the photoelectrons) was not observed in single-color DPI of He [8]. However, the nonspherical symmetry of the H₂ molecule offers the possibility of uncovering features that do not occur for spherically symmetric atomic targets, such as the occurrence of heretofore unexpected dynamical vortices.

In this paper we study occurrence of vortices in electron momentum distributions produced in single-photon (\( \hbar \omega = 75 \text{ eV} \)) double ionization of fixed-in-space H₂ in its \(^1\Sigma^+_g\) ground state by time-delayed, counter-rotating, elliptically polarized (chiral) attosecond pulses for two cases: \( \hat{k} \parallel \mathbf{R} \) and \( \hat{k} \perp \mathbf{R} \), where \( \hat{k} \) is the laser propagation direction and \( \mathbf{R} \) is the internuclear axis. Our results are based on ab initio numerical solutions of the six-dimensional time-dependent Schrödinger equation (TDSE) and analytic PT analyses. We report five main findings: (i) For \( \hat{k} \parallel \mathbf{R} \), only \(^1\Pi^+_u\) continuum states are excited and dynamical vortices occur under the same conditions as in DPI of He [8]. (ii) For \( \hat{k} \perp \mathbf{R} \), both \(^1\Sigma^+_u\) and \(^1\Pi^+_u\) continuum states are excited by each pulse and unexpected dynamical vortices are found to originate from Ramsey interference of the two resulting electron wave packets. (iii) The occurrence of the dynamical vortices can be controlled by the pulse ellipticities. (iv) For fixed excess energy \( E \), the time delay \( \tau \) between pulses allows control of the \( A_\Sigma \) and \( A_\Pi \) amplitude contributions to the two-electron angular distributions. (v) Combining these features, we propose a complete experiment in which both the magnitudes and the relative phase of the \( A_\Sigma \) and \( A_\Pi \) amplitudes can be determined by varying the pulse ellipticities and time delays.

This paper is organized as follows. In Sec. II we provide a brief description of our numerical methods. In Sec. III we present our analytical and numerical results for DPI of H₂ by time-delayed, counter-rotating, elliptically polarized attosecond pulses propagating along \( \hat{k} \) either parallel or perpendicular to the molecular axis \( \mathbf{R} \). For each of these two cases, we present results for the momentum distributions of the two ionized electrons. We then focus on the \( \hat{k} \perp \mathbf{R} \) case, presenting the two-electron angular distributions and proposing a complete experiment for determining the magnitudes and relative phases of the two dynamical amplitudes involved in this DPI process. In Sec. IV we summarize our key results...
and present our conclusions. Finally, in Appendix A we give results of various tests we have carried out to confirm the convergence of our numerical results, and in Appendix B we present analytical PT derivations of the key equations used to interpret our numerical results. Atomic units (a.u.) are used throughout this paper unless specified otherwise.

II. BRIEF DESCRIPTION OF NUMERICAL METHODS

In this section we begin by providing a brief overview of ab initio computational methods that have been used to study DPI of the H₂ molecule. We then briefly describe the methods we have used in this work.

Ab initio numerical calculations of the triply differential cross sections (TDCS) for DPI of H₂ have been carried out using two different representations of the two-electron wave functions. One group of calculations used a single-center spherical coordinate representation [14–17], while a second group of calculations used a two-center prolate spheroidal coordinate representation [18–20]. The first calculations for the TDSE were done in spherical coordinates using either the time-independent exterior complex scaling (ECS) method [16,17] or the time-dependent close-coupling (TDCC) method [14]. Taking into account the experimental uncertainties in the scattering angles, the agreement between the theoretical calculations and experimental results was good [14,16,17]. Subsequently, three calculations were carried out in prolate spheroidal coordinates, using the ECS method [18] and the TDSE method [19,20]. Also, another TDSE calculation was carried out in spherical coordinates using a single-center partial-wave expansion of the wave packet in terms of bipolar harmonics [15]. For a laser pulse linearly polarized either along or perpendicular to the molecular axis, the TDSE results obtained by these different methods were compared in Refs. [15,19,20]. Except for the TDSE obtained within the ECS method in prolate spheroidal coordinates [18], excellent agreement was found between the prolate spheroidal coordinate results [19,20] and those in spherical coordinates using the ECS method [17], the TDCC method with a larger box size than in [14] (see Refs. [15,19]), and a variant of the TDSE method [15].

To obtain the SDP for DPI of H₂ numerically, we have generalized our code for solving the two-electron TDSE for He interacting with chiral (i.e., elliptically-polarized) pulses [2,3,8,21] to take into account the two-center H₂ problem within the fixed-nuclei approximation (as in Refs. [14–20]). As in Refs. [14] and [15], we use a single-center, time-dependent close-coupling expansion [14,15] of the two-electron wave packet in spherical coordinates (with the origin at the center of mass of the nuclei). To discretize the radial part of the two-electron wave packet, we use a finite-element discrete variable representation (FE-DVR) [22] in which 60 finite elements equally spaced by 2 a.u. are used. An eight-point Gauss-Legendre-Lobatto basis is used within each FE, which yields a total of 421 DVR functions in each radial coordinate up to 120 a.u. [23]. To efficiently time propagate the wave packet \( \Psi(r_1, r_2; R, t) \) for chiral pulses, we employ a real-space-product algorithm [23] together with a Wigner rotation transformation [24,25]. After the end of the pair of time-delayed laser pulses with total duration \( T_f = \tau + T \) (where \( T \) is the total duration of each pulse and \( \tau \) is the time delay between the two laser pulses), we freely propagate in time the wave packet \( \Psi(r_1, r_2; R, t) \) (i.e., the solution of the TDSE) for a longer additional time \( T_p \) in order to ensure that its doubly ionized part, \( \Psi_C(r_1, r_2; R, T_f + T_p) \), is sufficiently far away from the nuclei, and also so that the two photoelectrons are well separated from each other [15,19]. At a time \( t = T_f + T_p \) (where \( T_p = 13–20 \) optical cycles), we obtain the SDP as

\[
W = \left| \langle \Psi_{\rho,\rho}^{(\infty)}(r_1, r_2) | \Psi_C(r_1, r_2; R, T_f + T_p) \rangle \right|^2.
\]

Here \( \Psi_C \) equals the two-electron wave packet (that we obtain by solving the TDSE within a radial box of 120 a.u.) from which we have removed contributions of bound and singly ionized states (as done in Refs. [15,26]). The field-free double-continuum final state \( \Psi_{p,p}^{(\infty)} \), with excess energy \( E \) is approximated by a product of two Coulomb waves with charge \( Z = 2 \) [15,19]. All our TDSE results have been verified to be converged (see Appendix A).

III. ANALYTICAL AND NUMERICAL RESULTS

For H₂ in its \( 1{\Sigma}^+_g \) ground state (with binding energy \( E_b \approx 51.4 \) eV) interacting with two pulses, each having a carrier frequency \( \omega = 75 \) eV and an intensity of \( 5 \times 10^{13} \) W/cm², first-order PT is valid. For right or left circularly polarized light with a polarization ellipse, the relative phase \( \tilde{\Delta} \equiv \tilde{\hat{k}} \cdot \tilde{\hat{p}}_1 \equiv (\tilde{\hat{R}} \cdot \tilde{\hat{p}}_1) \) being respectively the major and minor axes of the polarization ellipse. The relative phase \( \tilde{\Delta} \equiv \tilde{\hat{k}} \cdot \tilde{\hat{p}}_1 \equiv (\tilde{\hat{R}} \cdot \tilde{\hat{p}}_1) \) being respectively the major and minor axes of the polarization ellipse.

For H₂ in its \( 1{\Sigma}^+_g \) ground state (with binding energy \( E_b \approx 51.4 \) eV) interacting with two pulses, each having a carrier frequency \( \omega = 75 \) eV and an intensity of \( 5 \times 10^{13} \) W/cm², first-order PT is valid. For right or left circularly polarized light with a polarization ellipse, the relative phase \( \tilde{\Delta} \equiv \tilde{\hat{k}} \cdot \tilde{\hat{p}}_1 \equiv (\tilde{\hat{R}} \cdot \tilde{\hat{p}}_1) \) being respectively the major and minor axes of the polarization ellipse. The relative phase \( \tilde{\Delta} \equiv \tilde{\hat{k}} \cdot \tilde{\hat{p}}_1 \equiv (\tilde{\hat{R}} \cdot \tilde{\hat{p}}_1) \) being respectively the major and minor axes of the polarization ellipse.
FIG. 1. Electron detection geometries for DPI of H₂ by a pair of time-delayed oppositely elliptically-polarized pulses propagating parallel (\( \hat{k} \parallel R \), top) or perpendicular (\( \hat{k} \perp R \), bottom) to the internuclear vector \( R \), with \( \hat{k} \) the laser propagation direction. Left column: \( p_1 \parallel \hat{k} \) and \( p_2 \) in the laser polarization plane (\( \hat{\xi}, \hat{\phi} \)) with spherical angles \((\theta_1 = \pi/2, \varphi_1)\), with \( p_1 \) and \( p_2 \) denoting the momenta of the outgoing electrons. Right column: back-to-back emission of electrons in the polarization plane where \( \theta_1 = \theta_2 = \pi/2 \) and \( \varphi_2 = \varphi_1 - \pi \). In all panels, \( 0 \leq \varphi_2 \leq 2\pi \) and we fix \( \hat{k} \parallel \hat{z}, \hat{\epsilon} \parallel \hat{k}, \) and \( \hat{\xi} \parallel \hat{\phi} \). The origin of coordinates is at the center of mass of the nuclei.

Numerical TDSE results below are for counter-rotating pulses \((\eta = \eta_1 = -\eta_2)\) and two-electron detection geometries [in which \( u = (\hat{p}_1 \cdot \hat{p}_2) \) is fixed] for each of two light propagation directions (see Fig. 1): (i) \( \hat{k} \parallel R \), and (ii) \( \hat{k} \perp R \). For both cases, the amplitude (2) can be rewritten as a superposition of two kinematic vortex factors, \( \cos(\Phi/2 \pm \xi \varphi_2) \), having opposite handedness (see Appendix B):

\[
A = A^{(+)}_n \cos(\Phi/2 - \xi \varphi_2) + A^{(-)}_n \cos(\Phi/2 + \xi \varphi_2),
\]

(4) where \( p_2 \) is in the polarization plane \((\theta_2 = \pi/2)\), the dynamical amplitudes \( A^{(\pm)}_n \) depend on \( \chi \), and \( \hat{\xi} \equiv \xi/|\xi| \), with \( \xi = 2\eta/(1 + \eta^2) \) being the circular polarization degree. Note that \( \xi = +1 \) or \(-1\) corresponds to right-left or left-right circularly polarized (RLCP or LCP) pulses.

A. Kinematic vortices in the \( \hat{k} \parallel R \) geometry

In the \( \hat{k} \parallel R \) geometry [see Figs. 1(a) and 1(b)], the dynamical amplitudes in Eq. (4) for \( A \equiv A_1 \) take the form (see Appendix B),

\[
A^{(\pm)}_{n,1} = e^{i(\Phi/2 - \phi_1)} \sqrt{(1 + \ell)/(1 + \eta^2)} A_1(\chi),
\]

\[
= e^{i(\Phi/2 - \phi_1)} \sqrt{(1 + |\xi|)} A_1(\chi),
\]

(5)

where \( \ell = (1 - \eta^2)/(1 + \eta^2) \) is the linear polarization degree, \( A_1(\chi) \equiv A_{1,1}(\chi) \) for the detection geometry in Fig. 1(a), and \( A_{1}(\chi) \equiv A_{1,1}(\chi) - A_{1,1}(\chi) \) for that in Fig. 1(b). For circularly polarized pulses, \(|\eta| = 1\) so that \( A^{(\pm)}_{n,1} = 0 \). Consequently, the SDP \( \mathcal{W}_1 \equiv |A_1|^2 \) becomes

\[
\mathcal{W}_1(p_2) = 2|A_1(\chi)|^2 \cos^2(\Phi/2 - \xi \varphi_2),
\]

(7)

which has exactly the same form as in both single [2] and double [8] photoionization of the He \( 1S^0 \) ground state. The dynamical coefficient \( |A_1(\chi)|^2 \) in (7) (as for those in Refs. [2,8]) has no angular dependence, since \( n_u, u_1, \) and \( u_2 \) in \( \chi \) [see Eq. (3)] are fixed for both detection geometries. Thus, the maxima and zeros of the kinematical factor \( \cos^2(\Phi/2 - \xi \varphi_2) \) define Fermat (or Archimedean) spirals [2]. The spiral equations are \( \varphi_2^{\text{max}}(E) = \xi \Phi/2 + n\pi \) for maxima and \( \varphi_2^{\text{min}}(E) = \xi \Phi/2 + (n + 1/2)\pi \) for zeros, where \( n = 0, \pm 1, \pm 2, \ldots \) give the number of spiral arms, which is 2 since only \( n = 0 \) give independent equations. Thus, the \( p_2 \) distribution in the polarization \( x \) plane \((\theta_2 = \pi/2)\) produced by time-delayed RLCP or LCP pulses exhibits atomlike two-arm oppositely handed spiral kinematical vortices, i.e., counterclockwise for RLCP and clockwise for LCP. Our TDSE results for the \( p_2 \) distributions produced by time-delayed RLCP pulses (with parameters specified in the caption of Fig. 2) are shown in Fig. 2(a) for equal energy sharing (EES) in the detection geometry of Fig. 1(a), and in Fig. 2(b) for back-to-back (BTB) electron emission in the polarization \( x \) plane [Fig. 1(b)] with an unequal-energy-sharing (UES) partition of \( \varepsilon = E_1/E = 25\% \). Our TDSE results confirm well all vortex features predicted by PT.

B. Dynamical vortices in the \( \hat{k} \perp R \) geometry

In the \( \hat{k} \perp R \) geometry [see Figs. 1(c) and 1(d)], the dynamical amplitudes in Eq. (4) for \( A \equiv A_\perp \) take the form (see Appendix B)

\[
A_{\perp,1}(\chi) = e^{i(\Phi/2 - \phi_1)} \sqrt{(1 + \ell)/(1 + \eta^2)} A_{\perp,1}(\chi),
\]

(8)

where \( A_{\perp,1}(\chi) \equiv A_{1,2}(\chi) \), \( A_{\perp}(\chi) \equiv A_{1,2}(\chi) \), and \( A_{\perp}(\chi) \equiv A_{1,1}(\chi) \) for the geometry in Fig. 1(c), and \( A_{\perp}(\chi) \equiv A_{1,2}(\chi) - A_{1,1}(\chi) \), \( A_{\perp}(\chi) \equiv A_{1,2}(\chi) - A_{1,1}(\chi) \), with \( \chi = (p_1, p_2, \xi, 0, 0, \cos \varphi_2) \) for the geometry in Fig. 1(c), and \( A_{\perp}(\chi) \equiv A_{1,2}(\chi) - A_{1,1}(\chi) \), \( A_{\perp}(\chi) \equiv A_{1,2}(\chi) - A_{1,1}(\chi) \), with \( \chi = (p_1, p_2, R, -1, -1, \cos \varphi_2, \cos \varphi_2) \) for that in Fig. 1(d). Note that since both amplitudes, \( A_{\perp,1}(\chi) \) and \( A_{\perp}(\chi) \), in Eq. (8) depend on the azimuthal angle \( \varphi_2 \) of \( p_2 \) via...
Fig. 3(c) shows dynamical vortex structures, while there are none in Fig. 3(d) for the BTB geometry [Fig. 1(d)] and RLCP pulses, the handedness changes. For LRCP pulses, the ratio of the amplitudes $\eta$ on the ellipticity $\varepsilon$ of vortices is controlled by the amplitude ratio of the SDP in (9) reduces for circular polarization ($\ell = 0, \xi = \pm 1$) to the same form as in (7) with its kinematic vortex factor. Thus, Eq. (9) may produce vortex structures in particular cases. The analysis is best done for the general amplitude in Eq. (4) using the dynamical amplitudes $A_{\phi,1}$ and $A_{\phi,2}$.

For the geometry in Fig. 1(c), TDSE results for the two geometries in Figs. 1(c) and 1(d), our TDSE results in Figs. 3(a), 3(b) and 3(d) do show dynamical vortex structures, these are dynamical vortices rather than kinematic vortices since their occurrence depends on the interplay between the participating dynamical amplitudes, $A_{\Gamma}(\chi)$ and $A_{\Sigma}(\chi)$ amplitudes, both of which depend on the azimuthal angle $\varphi_2$ of $p_2$. (Note that for $\omega \approx 75$ eV the magnitude of $A_{\Gamma}$ can be as much as a factor of 5 larger than that of $A_{\Sigma}$.) If the SDP in (9) leads to vortex structures, these are dynamical vortices rather than kinematic vortices since their occurrence depends on the interplay between the participating dynamical amplitudes, $A_{\Gamma}(\chi)$ and $A_{\Sigma}(\chi)$ amplitudes. For the two $\hat{k} \perp \hat{R}$ geometries in Figs. 1(c) and 1(d), our TDSE results in Figs. 3(a), 3(b) and 3(d) do show dynamical vortex structures, while there are none in Fig. 3(c).

For the geometry in Fig. 1(c), TDSE results for the $p_2$ distributions by RLCP pulses delayed in time by $\tau = T \approx 331$ as are shown in Fig. 3(a) for EES and in Fig. 3(b) for an UES ($\varepsilon = 25\%$). The $p_2$ distributions exhibit vortex structures for any energy-sharing partition. These dynamical vortex patterns differ from the kinematical ones shown for $\hat{k} \parallel \hat{R}$ in Figs. 2(a) and 2(b). First, they are distorted owing to the angular dependences and the relative magnitudes of the $A_{\Sigma}$ and $A_{\Gamma}$ amplitudes. Second, they have an opposite handedness, since the $A_{\phi,1}$ term in (4) dominates, i.e., $A_{\Sigma} \approx -A_{\Pi}$ [cf. Eq. (8)]. For LRCP pulses, the handedness changes. For RLCP pulses, the ratio $\gamma$ of the energy- and angle-integrated $A_{\Sigma}$ amplitude to that of the $A_{\phi,1}$ amplitude is $\approx 2$ for both EES and UES in this geometry. We find dynamical vortex structures in this geometry occur for $0.15 \leq |\eta| \leq 1$ and are best seen for $\eta \approx 0.5$, as confirmed by our TDSE results.

For the BTB detection geometry [Fig. 1(d)] with an UES partition of $\varepsilon = 25\%$, $\gamma \approx 8$ for RLCP pulses ($|\eta| = 1$). As $A_{\Pi} \gg A_{\Sigma}$, Eq. (8) shows that $A_{\phi,1}^{\pi,1} \approx \pm A_{\Pi}$. Thus the $\chi$, spiral vortex structures are unexpected. Indeed, the SDP $W_\perp \equiv |A_{\phi,1}|^2$ takes the form (see Appendix B)

$$W_\perp(p_2) = 2(1 - \ell)|A_{\Gamma}(\chi)|^2 \sin^2 \varphi_2 \sin^2(\Phi/2)$$

$$+ 2(1 + \ell)|A_{\Sigma}(\chi)|^2 \cos^2 \varphi_2 \cos^2(\Phi/2)$$

$$+ \xi \Re \{A_{\chi,1}^2(\chi)A_{\Pi}(\chi)\} \sin(2\varphi_2) \sin \Phi,$$

which does not obviously have vortex structure. However, if $A_{\Pi}$ were equal to $A_{\Sigma}$, then the SDP in (9) reduces for circular polarization ($\ell = 0, \xi = \pm 1$) to the same form as in (7) with its kinematic vortex factor. Thus, Eq. (9) may produce vortex structures in particular cases. The analysis is best done for the general amplitude in Eq. (4) using the dynamical amplitudes $A_{\phi,1}$ and $A_{\phi,2}$.

The DPI amplitudes for the $\hat{k} \parallel \hat{R}$ and $\hat{k} \perp \hat{R}$ geometries have the same form (4), i.e., a superposition of two kinematic vortex factors $\cos(\Phi/2 \pm \xi \varphi_2)$ with opposite handedness, so that the occurrence of vortices requires one of the amplitudes in (4) to be larger than the other. Also, the $\varphi_2$ dependence of the dynamical amplitudes must be “weaker” than that of the kinematic factors $\cos(\Phi/2 \pm \xi \varphi_2)$. For $\hat{k} \parallel \hat{R}$, the $A_{\Pi}(\chi)$ amplitude in Eq. (5) has no angular dependence and the occurrence of vortices is determined by the amplitude ratio, $A_{\Pi}^\pm/A_{\Pi}^\mp = (1 - |\eta|)/(1 + |\eta|)$, which depends only on the ellipticity $\eta$. In contrast, for $\hat{k} \perp \hat{R}$ the occurrence of vortices is controlled by the amplitude ratio $A_{\phi,1}^\pm/A_{\phi,1}^\mp = (A_{\Sigma} - |\eta|A_{\Pi})/(A_{\Gamma} + |\eta|A_{\Pi})$, which depends not only on the ellipticity $\eta$, but also on the magnitudes and relative phase of the $A_{\Pi}(\chi)$ and $A_{\Sigma}(\chi)$ amplitudes, both of which depend on the azimuthal angle $\varphi_2$ of $p_2$. (Note that for $\omega \approx 75$ eV the magnitude of $A_{\Gamma}$ can be as much as a factor of 5 larger than that of $A_{\Sigma}$.) If the SDP in (9) leads to vortex structures, these are dynamical vortices rather than kinematical vortices since their occurrence depends on the interplay between the participating dynamical amplitudes, $A_{\Pi}(\chi)$ and $A_{\Sigma}(\chi)$ amplitudes. For the two $\hat{k} \perp \hat{R}$ geometries in Figs. 1(c) and 1(d), our TDSE results in Figs. 3(a), 3(b) and 3(d) do show dynamical vortex structures, while there are none in Fig. 3(c).

counter-rotating vortices in (4) overlap. 

$$A \approx A_{\Pi}(\chi)[\cos(\Phi/2 - \xi \varphi_2) - \cos(\Phi/2 + \xi \varphi_2)],$$

$$= A_{\Pi}(\chi) \sin(\xi \varphi_2) \sin(\Phi/2),$$

producing a dipolar pattern along the $y$ axis with Ramsey fringes [see Fig. 3(c)]. The same pattern is obtained (not shown) when using LRCP pulses. This absence of vortex patterns in Fig. 3(c) can also be well understood by only retaining the first term (with $\ell = 0$ for RLCP or LRCP pulses) of the SDP in (9), since the second and third terms in (9) can be dropped when $\gamma \approx 8$ (i.e., $A_{\Sigma} \ll A_{\Pi}$).

By tuning the ellipticity $\eta$ over the range $0.05 \leq |\eta| \leq 0.5$, however, spiral vortices similar to those in the $\hat{k} \parallel \hat{R}$ geometry become visible, as shown in Fig. 3(d) for $\eta = +0.2$ (or $\xi = 0.38$). To visualize the dependence of this electron phenomenon on $\eta$, we provide in the Supplemental Material [31] an animation showing the evolution with the ellipticity $\eta$ over the broader range $0.001 \leq |\eta| \leq 1$ of the dynamical vortex patterns for the BTB detection geometry [Fig. 1(d)] and the same laser parameters as in Fig. 3(c). Although $A_{\Pi} \gg A_{\Sigma}$, the vortex structures appear for $\eta$ in the range $0.05 \leq |\eta| \leq 0.5$, because $|\eta|A_{\Pi}$ becomes comparable to $A_{\Sigma}$. Consequently, depending on the relative phase of the $A_{\Sigma}$ and $A_{\Pi}$ amplitudes, one expects either $A_{\phi,1}^{\pi,1} \approx |A_{\Sigma}(\chi) - |\eta|A_{\Pi}(\chi)|$ or $A_{\phi,1}^{\pi,1} \approx |A_{\Sigma}(\chi) - |\eta|A_{\Pi}(\chi)|$ to dominate. In Fig. 3(d) for the BTB detection geometry [Fig. 1(d)] and RLCP pulses, the counterclockwise handedness of the vortices implies
In summary, vortices in DPI of $\text{H}_2$ by time-delayed counter-rotating chiral pulses are studied for two detection geometries involving fixed angular separation between electron momenta. For $\hat{k} \parallel \textbf{R}$, atomlike *kinematical* spiral vortices are predicted with a handedness given by the helicity of the first pulse. For $\hat{k} \perp \textbf{R}$, *dynamical* vortices are predicted for a wide range of ellipticities owing to the interplay of the $A_\Sigma$ and $A_\Pi$ dynamical amplitudes. The helicity of these vortices depends also upon the relative phase of the $A_\Sigma$ and $A_\Pi$ amplitudes. Finally, we have outlined a complete set of measurements allowing determination of both the magnitudes and the relative phase of these two dynamical amplitudes.

**ACKNOWLEDGMENTS**

The research of A.F.S. and J.M.N.D. is supported in part by the U.S. Department of Energy (DOE), Office of Science, Basic Energy Sciences, under Award No. DE-FG02-96ER14646; the research of S.X.H. is supported by the DOE National Nuclear Security Administration under Award No. DE-NA0001944, the University of Rochester, and the New York State Energy Research and Development Authority; the research of L.B.M. is supported by the VKR Center of Excellence, QUSCOPE; and the research of A.V.M. and N.L.M. is supported by the Russian Ministry of Science and Higher Education under Grants No. 3.1761.2017/4.6 and No. 3.7514.2017/8.9, respectively. Our computations used Stampede 2 at the Texas Advanced Computing Center (TACC) at the University of Texas at Austin under U.S. National Science Foundation Grant No. PHY-120003. This work was completed utilizing the Holland Computing Center of the University of Nebraska, which receives support from the Nebraska Research Initiative.

**APPENDIX A: CONVERGENCE TESTS FOR DPI OF $\text{H}_2$ BY TIME-DELAYED CHIRAL PULSES**

In this Appendix we describe three tests of the convergence of the angular distributions of the two ionized electrons with respect to their sensitivity to (1) our choice of the number of total electronic angular momenta $L$; (2) our choice of the projection time $T_p$ (i.e., how long we wait after the end of our pulses before calculating the SDP); and (3) our choice of the outer radial boundary $R_0$ of bound and singly ionized states (which we use to remove contributions of these states from our calculated final-state wave packet).
TABLE I. Dependence of the calculated $^1\Sigma^+$ ground-state energy (a.u.) of the H$_2$ molecule for internuclear separation $R \simeq 1.4$ a.u. on the number of total electronic angular momenta $L_i$.

<table>
<thead>
<tr>
<th>$L_i$</th>
<th>Energy (a.u.)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>-1.784972</td>
</tr>
<tr>
<td>0.2</td>
<td>-1.872914</td>
</tr>
<tr>
<td>0.2,4</td>
<td>-1.883314</td>
</tr>
<tr>
<td>0,2,4,6</td>
<td>-1.888573</td>
</tr>
</tbody>
</table>

1. Dependence of the angular distributions on the number of total electronic angular momenta $L$

In contrast to the He atom, the total electronic angular momentum $L$ is not a good quantum number for the H$_2$ molecule. While the spherically symmetric He atom ground state has $L = 0$, the nonspherical symmetry of the electron-nucleus interaction in the H$_2$ molecule Hamiltonian requires that the ground state is described using several even $L$ components. In our single-center close-coupling expansion approach, we propagate the two-electron wave packet in imaginary time to obtain the energy of the $^1\Sigma^+$ ground state of the H$_2$ molecule at the equilibrium internuclear distance $R \simeq 1.4$ a.u. The dependence of the calculated H$_2$ ground-state energy on the number of total electronic angular momenta $L_i$ is given in Table I. For each $L_i$, all combinations of individual electron orbital angular momenta $l_1, l_2 = 0 – 5$ are included. One sees that the results for two, three, and four values of $L_i$ compare well with the benchmark values $-1.888760$ a.u. [32] or $-1.888761$ a.u. [33].

Starting from the ground state obtained using either $L_i = 0$, 2 or $L_i = 0, 2, 4$ angular momenta to solve the TDSE, we employ an expansion of the two-electron wave packet including either four values ($L = 0 – 3$) or five values ($L = 0 – 5$) of $L$, respectively. Although the total electronic angular momentum $L$ is not a good quantum number, converged results are found using an expansion in only four values of $L$, $L = 0 – 3$, their azimuthal quantum numbers $|M| \leq L$, all combinations of individual electron orbital angular momenta $l_1, l_2 = 0 – 5$, and their azimuthal quantum numbers $|m_1| \leq l_1$ and $|m_2| \leq l_2$. Indeed, for our laser parameters, including six values of $L = 0 – 5$ (as in [15]) does not change our SDP results, as shown in Fig. 5 for two time delays $\tau = \tau_{12}$ [see Fig. 5(a)] and $\tau = \tau_{11}$ [see Fig. 5(b)], where $\tau_n = n\pi/\omega$. Here, the projection time is fixed at $T_p = 35$ a.u. and the outer radial boundary of bound and singly ionized states is fixed at $R_0 = 10$ a.u. The results shown are for the detection geometry in Fig. 1(c) for an UES scheme ($\epsilon = E_1/E = 25\%$) at a fixed excess energy $E = \omega - E_b$, the excess energy.

In Fig. 5 one sees that the two-electron angular distribution is sensitive to both the time delay and the number of orbital angular momenta included in the calculation. As discussed above for the angular distribution results shown in Fig. 4, for the case $\tau = \tau_{12}$ shown in Fig. 5(a) only the $^1\Sigma^+$ continuum states are excited, whereas for the case $\tau = \tau_{11}$ shown in Fig. 5(b) only the $^1\Pi_u^+$ continuum states are excited. In both cases, including only total orbital angular momenta $L = 0, 1$ leads to dipolar angular distributions, either along or perpendicular to the molecular axis. However, when one includes higher values of $L$, the angular distribution along the molecular axis develops an additional structure, whereas that perpendicular to the molecular axis does not.

2. Dependence of the angular distributions on the projection time $T_p$

For the same detection scheme as in Fig. 5 [i.e., for the detection geometry in Fig. 1(c) for an UES scheme ($\epsilon = E_1/E = 25\%$) at a fixed excess energy $E = \omega - E_b$, the excess energy].
Results are shown for seven values of \( R_0 \): 5 a.u. [dashed (red) lines], 10 a.u. [thick solid (black) lines], 15 a.u. [dash double-dotted (blue) lines], 20 a.u. [dashed (orange) lines], 25 a.u. [dashed (magenta) lines], 30 a.u. [dotted (green) lines], and 35 a.u. [thin solid (black) lines].

Angular distributions produced by two attosecond pulses delayed in time by \( \tau = \tau_{12} \) are shown in Fig. 6 for five projection times \( T_p = 20, 25, 35, 40, \) and 45 a.u. For \( R_0 = 10 \) a.u. and \( L = 0 - 3 \), it is clear that good convergence is reached for projection times \( T_p \geq 25 \) a.u. (i.e., 11–20 optical cycles after the end of pulses).

### 3. Dependence of the angular distributions on the outer radial boundary \( R_0 \) of bound and singly ionized states

To obtain the doubly ionized part, \( \Psi_c(r_1, r_2; R, T_f + T_p) \), of the solution \( \Psi(r_1, r_2; R, t) \) of the TDSE, we must remove contributions from both bound and singly ionized states. This is done by setting the wave packet \( \Psi(r_1, r_2; R, t) \) equal to zero for radial distances \( r_1 < R_0 \) and/or \( r_2 < R_0 \), where \( R_0 \) defines the outer radial boundary of bound and singly ionized states \( [15,26] \). The \( R_0 \) dependence of our TDSE results is shown in Fig. 7. For \( T_p = 35 \) a.u., we find that using a cutoff of \( R_0 = 5, 10, 15, \) or 20 a.u. leads to the same converged angular distributions. For larger values of \( R_0 \), e.g., \( R_0 = 25, 30, \) and 35 a.u., the TDSE results at \( T_p = 35 \) a.u. are unstable and decrease sharply. This behavior is expected because at that projection time \( T_p \) a significant contribution from the doubly ionized wave packet has been cut. In other words, the doubly ionized wave packet has not been given sufficient time to leave the region where either \( r_1 < R_0 \) or \( r_2 < R_0 \) [15]. All results presented in the main text have been obtained for \( R_0 = 10 \) a.u., which is the same value used in [15].

### APPENDIX B: PERTURBATION THEORY ANALYSIS

We provide in this Appendix derivations of Eq. (4) for the transition amplitude for the two cases of chiral pulses propagating along \( \hat{k} \) either parallel (\( \hat{k} \parallel \hat{R} \)) or perpendicular (\( \hat{k} \perp \hat{R} \)) to the molecular axis \( \hat{R} \). Also, we provide derivations of Eqs. (5) and (8) for the dynamical parameters of their DPI amplitudes, as well as of Eqs. (7) and (9) for their SDPs.

For convenience, we summarize first some notations applicable to all derivations. The expression for the effective polarization vector \( \mathbf{e}' \) of the pulse pair is [8]

\[
\mathbf{e}' = \mathbf{e} + e^{i\Phi} \mathbf{e}^*,
\]

where \( \Phi = \text{the Ramsey phase} \)

\[
\Phi = (E + E_b)\tau + \phi_{12},
\]

in which \( E = (p_1^2 + p_2^2)/2 \) is the energy of the electron pair in the continuum, \( E_b \) is the binding energy of the H_2 ground state, and \( \phi_{12} = \phi_1 - \phi_2 \) denotes the relative carrier-envelope phase (CEP) of the two pulses. In Eq. (B1), \( \mathbf{e} \) is the first pulse polarization vector:

\[
\mathbf{e} = (\hat{\mathbf{e}} + i\eta \hat{\mathbf{\xi}})/(1 + \eta^2)^{1/2},
\]

where \( \eta = \eta_1 = -\eta_2 \) is its ellipticity, and \( \hat{\mathbf{e}} \) and \( \hat{\mathbf{\xi}} = (\hat{\mathbf{k}} \times \hat{\mathbf{e}}) \) are the major and minor axes of the polarization ellipse, respectively. We define the \( x, y, \) and \( z \) axes of the laboratory frame respectively along \( \hat{\mathbf{e}}, \hat{\mathbf{\xi}}, \) and \( \hat{\mathbf{k}}. \) The linear and circular polarization degrees are, respectively:

\[
\ell = (1 - \eta^2)/(1 + \eta^2),
\]

\[
\xi = 2\eta/(1 + \eta^2).
\]

For later use, we note the following two relations:

\[
1 - \ell = 2/(1 + \eta^2), \quad 1 - \ell = 2\eta^2/(1 + \eta^2).
\]

In the laboratory coordinate frame the scalar product of the effective polarization vector \( \mathbf{e}' \) with an arbitrary vector \( \mathbf{p} = (p, \theta, \phi) \) can be written as

\[
(\mathbf{e}' \cdot \mathbf{p}) = e^{i\Phi/2} p \sin \theta \frac{\cos \phi \cos (\Phi/2) + \eta \sin \phi \sin (\Phi/2)}{\sqrt{1 + \eta^2}},
\]

After some simple trigonometric transformations, Eq. (B7) can be expressed in terms of the circular polarization degree \( \xi \) [see Eq. (B5) as]

\[
(\mathbf{e}' \cdot \mathbf{p}) = e^{i\Phi/2} p \sin \theta [\sqrt{1 + \xi^2} \cos (\Phi/2 - \tilde{\xi}\phi) + \sqrt{1 - \xi^2} \cos (\Phi/2 + \tilde{\xi}\phi)],
\]

where \( \tilde{\xi} = \xi/|\xi| = \text{sign}(\eta) \).

### 1. Amplitude and SDP for DPI of H_2 in the \( \hat{k} \parallel \hat{R} \) geometry

We derive here Eqs. (4), (5), and (7) of the main text for the detection geometry described by Figs. 1(a) and 1(b).

For \( \hat{k} \parallel \hat{R} \), \( (\hat{R} \cdot \mathbf{e}') = 0 \) and the perturbation theory (PT) amplitude \( A \) given by Eq. (2) in the main text reduces to

\[
A_{\parallel} = e^{-i\Phi} [A_{\parallel,1}(\chi)(\mathbf{e}' \cdot \hat{\mathbf{p}}_1) + A_{\parallel,2}(\chi)(\mathbf{e}' \cdot \hat{\mathbf{p}}_2)].
\]

Here \( A_{\parallel,1}(\chi) \) and \( A_{\parallel,2}(\chi) \) are true scalar functions of \( \chi \approx (p_1, p_2, R, u, u_1, u_2) \), where \( p_1 \) and \( p_2 \) are the magnitudes of the electron momenta \( \mathbf{p}_1 \) and \( \mathbf{p}_2 \), \( R = 1.4 \) a.u. is the internuclear separation, \( u \approx (\mathbf{p}_1 \cdot \hat{\mathbf{p}}_2) \), and \( u_{1,2} \approx (\hat{\mathbf{R}} \cdot \hat{\mathbf{p}}_{1,2}) \). For the detection geometries in which either \( \mathbf{p}_1 \parallel \hat{\mathbf{k}} \) and \( \mathbf{p}_2 \) is in the laser polarization plane [see Fig. 1(a) in the main text] or both electrons are emitted back-to-back (BTB) in the laser polarization plane [see Fig. 1(b) in the main text], the PT amplitude \( A_{\parallel} \) (B9) takes the form

\[
A_{\perp} = e^{-i\Phi} A_{\perp,1}(\chi)(\mathbf{e}' \cdot \hat{\mathbf{p}}).
\]
In Eq. (B10), for the geometry in Fig. 1(a) in the main text, \(\mathbf{p} \equiv \mathbf{p}_2\) and \(A_{n}(\chi) \equiv A_{n,2}(\chi)\), where \(\chi = (p_1, p_2, R, 0, 1, 0)\), whereas for the geometry in Fig. 1(b) in the main text, \(\mathbf{p} \equiv \mathbf{p}_2 = -\mathbf{p}_1\) and \(A_{n}(\chi) \equiv A_{n,1}(\chi) - A_{n,2}(\chi)\), where \(\chi = (p_1, p_2, R, -1, 0, 0)\). Since the two electrons have zero total spin in the \(^1\Sigma^+_g\) initial state, the coefficients \(A_{n,1}\) and \(A_{n,2}\) obey the symmetry relation,

\[
A_{n,1}(p_1, p_2, u_1, u_2) = A_{n,2}(p_2, p_1, u_1, u_2),
\]

which for brevity we have omitted \(R\) and \(u\) from the list of arguments. From the symmetry property (B11), it follows that the PT amplitude \(A_{n}(\chi)\) vanishes for BTB emission of electrons with equal energy sharing since \(A_{n,1}(p_1, p_1, 0, 0) = A_{n,2}(p_1, p_1, 0, 0)\).

Substituting the geometric factor \((\epsilon' \cdot \mathbf{p})\) in Eq. (B8) into Eq. (B10), we obtain that the PT amplitude in spherical coordinates can be represented as a superposition of two vortex amplitudes with opposite handedness:

\[
A_{n} = A_{n,1}(\chi) \cos(\Phi/2 - \xi \psi) + A_{n,2}(\chi) \cos(\Phi/2 + \xi \psi),
\]

which is Eq. (4) in the main text, where the dynamical parameters \(A_{n,1}\) and \(A_{n,2}\) are defined by

\[
A_{n,1}^{(\pm)} = e^{i(\Phi/2 - \phi)} \sqrt{(1 + \ell)/2} \sin\theta(1 \pm \eta) A_{n}.
\]

For geometries in Figs. 1(a) and 1(b), \(\theta = \pi/2\), and Eq. (B13) leads to Eq. (5) in the main text.

For circularly polarized pulses, \(|\xi| = |\eta| = 1\) and the second term in Eq. (B12) vanishes. For elliptically polarized pulses, \(|\xi| < 1\), Eq. (B12) has contributions from two kinematic factors having opposite handedness, \(\cos(\Phi/2 \pm \xi \psi)\). Taking the square modulus of Eq. (B12), the SDP, \(\mathcal{W}_R = |A_n|^2\), for two-delayed counter-rotating chiral pulses with \(\mathbf{k} \parallel \mathbf{R}\) is

\[
\mathcal{W}_R = |A_{n}(\chi)|^2 \sin^2 \theta (|1 + |\xi|)| \cos^2(\Phi/2 - \xi \psi) + (1 - |\xi|) \cos^2(\Phi/2 + \xi \psi) + 2 \eta \cos(\Phi/2 - \xi \psi) \cos(\Phi/2 + \xi \psi).
\]

One sees that for small \(\ell\), one has \(\xi \approx 1\), and the first vortex term dominates in the SDP (B14).

Alternatively, substituting the geometric factor \((\epsilon' \cdot \mathbf{p})\) in Eq. (B7) in spherical coordinates into Eq. (B10) and using Eqs. (B4) and (B5), another expression for the SDP in the polarization plane (\(\theta = \pi/2\)) can be derived:

\[
\mathcal{W}_R = 2(1 - \ell)|A_{n}(\chi)|^2 \sin^2 \theta \sin^2(\Phi/2) + 2(1 + \ell)|A_{n}(\chi)|^2 \cos^2 \theta \cos^2(\Phi/2) + \xi|A_{n}(\chi)|^2 \sin(2\phi) \sin \Phi.
\]

For time-delayed counter-rotating circularly polarized pulses, \(\ell = 0\) and \(\xi = \pm 1\). Thus, the SDP in either Eq. (B14) or in Eq. (B15) reduces to

\[
\mathcal{W}_R = 2|A_{n}(\chi)|^2 \sin^2 \theta \cos^2(\Phi/2 - \xi \psi),
\]

which is Eq. (7) in the main text.

2. Amplitude and SDP for DPI of \(^1\Sigma^+_g\) in the \(\mathbf{k} \perp \mathbf{R}\) geometry

Below we show that the amplitude in the perpendicular geometry has the same form [see Eq. (4) in the main text] as in the parallel geometry, Eq. (B12). We also derive Eq. (8) for the dynamical parameters, as well as Eq. (9) for the SDP.

For \(\mathbf{k} \perp \mathbf{R}\), the PT amplitude \((1)\) in the main text for the two detection geometries in Figs. 1(c) and 1(d) can be written, using Eq. (B1), as

\[
\mathcal{A}_\perp = e^{-i\phi}[A_{n}(\chi)(\epsilon' \cdot \mathbf{p}) + B(\chi)(\mathbf{\hat{R}} \cdot \mathbf{e})](\mathbf{\hat{R}} \cdot \mathbf{\hat{p}}),
\]

where \(B(\chi) \equiv A_{\Sigma}(\chi) - A_{n}(\chi)\). Here, for the detection geometry in Fig. 1(c) in the main text, \(\mathbf{p} \equiv \mathbf{p}_2\), \(A_{n}(\chi) \equiv A_{n,2}(\chi)\), and \(A_{\Sigma}(\chi) \equiv A_{\Sigma,2}(\chi)\), where \(\chi = (p_1, p_2, R, 0, 0, \cos \varphi)\), whereas for the detection geometry in Fig. 1(d) in the main text, \(\mathbf{p} \equiv \mathbf{p}_2 = -\mathbf{p}_1\), \(A_{n}(\chi) \equiv A_{n,1}(\chi)\), and \(A_{\Sigma}(\chi) \equiv A_{\Sigma,1}(\chi)\), where \(\chi = (p_1, p_2, R, -1, -\cos \varphi, \cos \varphi)\). As the total spin for the two electrons is zero in the initial \(^1\Sigma^+_g\) ground state, the coefficients \(A_{n,1}\) and \(A_{n,2}\) satisfy the symmetry relation (B11), and the coefficients \(A_{\Sigma,1}\) and \(A_{\Sigma,2}\) satisfy

\[
A_{\Sigma,1}(p_1, p_2, u_1, u_2) = A_{\Sigma,2}(p_2, p_1, u_1, u_2),
\]

where for brevity we have omitted \(R\) and \(u\) from the list of arguments. From the symmetry properties (B11) and (B18), it follows that the PT amplitude \(A_{\perp}\) vanishes for BTB emission of electrons with EES. (This important result holds for any laser propagation direction \(\mathbf{k}\) and molecular orientation \(\mathbf{R}\).)

For both detection geometries in Figs. 1(c) and 1(d) in the main text, \(\mathbf{k} \parallel \mathbf{R} \parallel \mathbf{R}\), and \(\mathbf{p} \equiv \mathbf{p}_2\) is detected in the polarization xy plane. The geometric factor \((\mathbf{k} \cdot \mathbf{e})\) in Eq. (B17) can be evaluated using Eq. (B7) by setting \(p = 1, \theta = \pi/2,\) and \(\varphi = 0\); one then obtains

\[
(\mathbf{k} \cdot \mathbf{e}) = e^{i(\Phi/2)/[2(1 + \eta^2)]} \cos(\Phi/2).\]

Using this equation and noting that \((\mathbf{k} \cdot \mathbf{e}) = \sin \theta \cos \varphi,\) Eq. (B17) for the amplitude becomes

\[
A_{\perp} = e^{i(\Phi/2 - \phi)}[\sin \theta/(1 + \eta^2)^{1/2}] A_{\perp},
\]

where \(A_{\perp}\) is defined by

\[
A_{\perp} = 2[A_{\Sigma} \cos \varphi \cos(\Phi/2) + \eta A_{n} \sin \varphi \cos(\Phi/2)]
\]

\[
= (A_{\Sigma} + |\eta| A_{n}) \cos(\Phi/2 - \xi \psi)
\]

\[
+ (A_{\Sigma} - |\eta| A_{n}) \cos(\Phi/2 + \xi \psi).
\]

As a result, Eq. (B20) can be written in the compact form [which is Eq. (4) in the main text]:

\[
A_{\perp} = A^{(+)}_{\perp} \cos(\Phi/2 - \xi \psi) + A^{(-)}_{\perp} \cos(\Phi/2 + \xi \psi),
\]

where the dynamical parameters \(A^{(\pm)}_{\perp}\) are defined by

\[
A^{(\pm)}_{\perp} = e^{i(\Phi/2 - \phi)} \sqrt{(1 + \ell)/2} \sin \theta[|A_{\Sigma} \pm |\eta| A_{n}|] \]

\[
\]

For \(\theta = \pi/2,\) Eq. (B23) reduces to Eq. (8) in the main text. Taking the square modulus of \(A_{\perp}\) in Eq. (B20) or in Eq. (B22) in the laser polarization xy plane (\(\theta = \pi/2\)), and using the relations (B6), one obtains

\[
\mathcal{W}_\perp = 2(1 - \ell)|A_{n}(\chi)|^2 \sin^2 \theta \sin^2(\Phi/2) + 2(1 + \ell)|A_{\Sigma}(\chi)|^2 \cos^2 \theta \cos^2(\Phi/2) + \xi \Re[A_{\Sigma}(\chi)A_{n}(\chi)] \sin(2\phi) \sin \Phi,
\]

which is Eq. (9) in the main text.


[31] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevA.98.063407 for an animation of the evolution of the dynamical vortex patterns with ellipticity η over the range 0.001 ⩽ η ⩽ 1 for the BTB UES detection geometry in Fig. 1(d) and for the same laser parameters as in Fig. 3(c). In the animation, seven values of the ellipticity η are employed in this range: η(0) ⩾ 1, η(1) ⩾ 0.5, η(2) ⩾ 0.4, η(3) ⩾ 0.3, η(4) ⩾ 0.2, η(5) ⩾ 0.05, and η(6) ⩾ 0.001. Note that dynamical vortices appear only for five values of η(j), where 1 ⩽ j ⩽ 5.
