# Light control of intramolecular nuclear dynamics by vortex electron localization

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In strong-field ionization of molecules, intense light pulses are thought to have a negligible direct influence on atomic nuclei. Molecular dissociation is thus expected to be determined by the geometrical configuration of the molecular ion at the ionization instant. Contrary to this picture, we observe a counterintuitive electron-proton angular correlation and the formation of proton vortices following strong-field ionization of  $H_2$  molecules by bicircularly polarized two-color laser fields. We explain this phenomenon by the pathway interference and localization of the residual  $H_2^+$  electron in different angular-momentum states formed in the tail of the driving laser pulse. We validate this interpretation by combining a quantum-mechanical numerical simulation of the field-driven coupled electronic-nuclear dynamics and a semiclassical-trajectory model for the phase accumulation of the laser-driven electron localization which can be used for controlling molecular-bond breaking with circularly polarized laser fields.

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### I. INTRODUCTION

Control of molecular bond breaking with light fields is a long-term goal in modern molecular science [1-3]. Although laser pulses with tailored waveforms can be used to control the photoelectron emission direction from molecules [4,5], the direct interaction between light fields and molecular nuclei is rather limited for femtosecond pulses, mainly due to the heavy nuclear mass and the limited force imposed by the light fields. Prototypical strong-field dissociative ionization of hydrogen molecules,  $H_2 + n\omega \rightarrow e^- + H_2^+ \rightarrow e^- + H +$ H<sup>+</sup>(a proton), has attracted much attention over the past three decades [6-19]. There, the dominant photoelectron emission approximately follows the laser vector-potential direction at the instant when the electric field is maximal, while the protons are primarily emitted along the molecular axis. It has been shown that phase-stable few-cycle [11-13] or two-color [14,20,21] laser pulses can control on which side along the molecular axis the proton is emitted, resulting from the coherent superposition of gerade and ungerade  $H_2^+$  electronic states (for recent reviews, see Refs. [22,23]). The control of proton emission in two dimensions in the laboratory frame has been demonstrated convincingly in Refs. [24,25]. The synthesized electric field prefers to ionize the molecules aligned along its maximum electric-field direction and the direction is sensitive to the two-color phase. As a result, the proton emission direction appears to be steered by the two-color field. However, the time-resolved two-dimensional nucleus motion with coincidence photoelectron motion has not been thoroughly examined. The electrons and the nucleus are strongly coupled in the dissociative process.

In this article, we find that the proton emission in strongfield dissociative ionization of H2 molecules does not have to be along the molecular axis. Using bicircularly polarized twocolor laser fields, we observe a spiral distribution of the proton emission asymmetry in the polarization plane and a counterintuitive electron-proton angular correlation. These surprising observations can be explained by the two-dimensional localization and interference of coupled electron-nuclear wave packets with nonzero magnetic quantum numbers that form so-called vortex states or vortices. The interference of these vortices can map the relative time delay of the nuclear motion between different pathways to the slope of the spiral fringes with respect to energy. Most of our experimental findings are reproduced by both a quantum simulation including both electronic and nuclear degrees of freedom and a semiclassical-trajectory model for the electron-nuclear wavepacket propagation.

### **II. EXPERIMENTAL RESULTS**

Experimentally, the fundamental-field laser pulses were delivered by a multipass Ti:sapphire amplifier with a central wavelength of 800 nm, a repetition rate of 3 kHz, and a pulse

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FIG. 1. Measured electron and proton coincident momentum distributions in strong-field dissociative ionization of H<sub>2</sub>. Panels (a)–(c) show the phase-averaged photoelectron and proton coincident momentum distributions from two-color corotating laser fields where the ellipticity of the strong fundamental field is 0.6, 0.8, and 1.0, respectively. Red curves on top of these panels indicate the polarization of the 800-nm electric-field pulse and the molecule aligned along the major axis of the polarization ellipse is highlighted, which will be preferentially ionized. The ellipticity of the weak second harmonic field is fixed at 1 (circular polarization). (d), (e) Photoelectron (g), (h) and proton coincident momentum distributions at (d), (g)  $\phi_{RP} = 0$  and (e), (h)  $\phi_{RP} = \pi$ . Both fields are circularly polarized. Insets on the top of panels (d) and (e) sketch the synthesized electric fields. Arrows mark instants when the electric-field maximum is reached. Panels (f) and (i) show the difference of photoelectron and proton momentum distributions between these two phases.

duration of 25 fs (full width at half maximum in intensity). The 400-nm central wavelength second-harmonic pulses were produced by frequency doubling using a 200-µm-thick type-I  $\beta$ -barium borate (BBO) crystal (29.2° cut). The two light beams were synchronized in a Mach-Zehnder interferometer with 5-as precision for their relative phase  $\phi_{\rm RP}$  [26]. Their polarizations and intensities were controlled independently by a combination of wire-grid polarizer and half- and quarter-wave retardation plates. The peak intensities of the fundamental and second-harmonic fields were calibrated to be  $1.6 \times 10^{14}$ and  $1.0 \times 10^{13}$  W/cm<sup>2</sup>, respectively. The polarization of the strong fundamental field was adjusted from elliptical to circular and the major axis was fixed along the x axis. The weak second-harmonic field was adjusted with corotating circular polarization in the same polarization plane (x-z plane). Both light beams were focused by a silver-coated concave mirror with f = 75 mm focal length onto a supersonic gas jet of randomly oriented H<sub>2</sub> molecules (backing pressure,  $\sim 1$  bar), which was delivered into the high-vacuum interaction chamber ( $<10^{-10}$  mbar) along the x direction by a 30-µm nozzle. We used a cold target recoil ion momentum spectroscopy reaction microscope setup [27,28] to detect electrons and protons in coincidence. Here only the events with one electron in coincidence with one proton are presented. Static electric (3.2 V/cm) and magnetic (5.4 G) fields were applied along the z axis. The particle momenta were integrated along the light propagation direction (y axis). The y axis is also the quantization axis of the vortex states.

Figures 1(a)-1(c) show our measured coincident photoelectron and proton momentum distributions in the polarization plane at fundamental-field ellipticities of 0.6, 0.8, and 1.0, respectively. The data are averaged over the relative phase  $\phi_{RP}$  between the two colors. In the elliptically polarized laser fields, molecules aligned along the major axis (i.e., the *x* axis) of the polarization ellipse will be preferentially ionized, causing the photoelectron to be emitted along the direction of the corresponding vector potential (i.e., along the *z* axis) [29]. During the subsequent dissociation, rotation of the molecular ion can be neglected, such that protons are emitted along the maximum-electric-field direction (i.e., the *x* axis). The photoelectron momentum distributions in Figs. 1(a) and 1(b) show a dipolar structure that is tilted by the so-called attoclock offset angle [30–34]. The proton momentum distribution displays several discrete peaks corresponding to dissociation pathways accessed by absorption of different numbers of photons. Our experimental results in Figs. 1(a) and 1(b) demonstrate that the dominant electron and proton emission directions are approximately perpendicular to each other (with an attoclock offset angle). This validates that elliptically polarized fields barely spin molecular nuclei during dissociation.

For circular polarization there is no preferred ionization direction and phase-averaged momentum distributions for both photoelectrons and protons exhibit an isotropic, donutlike shape as shown in Fig. 1(c). Nevertheless, electron-proton angular correlation still can manifest itself at a specific relative phase  $\phi_{\rm RP}$  that breaks the symmetry of the synthesized light field. Figures 1(d), 1(e), 1(g), and 1(h) show measured coincident photoelectron and proton momentum distributions at  $\phi_{\rm RP} = 0$  and  $\phi_{\rm RP} = \pi$ . The measured photoelectron distributions are single-lobed and governed by the shape of the synthesized vector potential, which is perpendicular to the synthesized electric field. When changing  $\phi_{\rm RP}$ , the synthesized vector potential will rotate in the polarization plane and thus streak the photoelectron-emission angle. To highlight the angular anisotropy, we also display their difference as "asymmetry" graphs in Figs. 1(f) and 1(i).

Interestingly, low-energy protons (corresponding to the  $1\omega$  dissociation channel) are predominantly emitted in the direction nearly opposite to the electrons. This indicates that the general physical picture, where a proton is emitted along the



FIG. 2. (a), (b) Measured energy distribution of the coincident (a) photoelectrons and (b) protons as a function of the relative phase between the two-color fields. The electron and the proton emission angles are both confined in the same angular range,  $\phi_e = \phi_i = 0 \pm 10^\circ$  in the laboratory frame.

electric field direction and a photoelectron follows its vectorpotential [as observed in Figs. 1(a) and 1(b)], is no longer valid. The synthesized electric field still prefers to ionize molecules oriented along the *x* axis, but the nuclei acquire an additional rotation of 90° from the light field, which cannot be explained within classical pictures. Moreover, we observe a spiral pattern for the high-energy protons above the 1- $\omega$ channel in Fig. 1(i), suggesting that the relative emission angle between the electron and proton changes as a function of the proton energy.

When scanning the relative phase between the two colors, the shape of the synthesized field does not change, but the synthesized field overall rotates in space. In Fig. 2, we show the measured coincident electron and proton energy spectra as a function of the two-color relative phase. The electron and the proton emission angles are both confined to the same angular range. A slight tilt of the vertical stripes in the photoelectron distribution show that the photoelectron emission angle has a weak energy dependence. For protons with energies of less than 0.4 eV, i.e., for the 1 $\omega$  dissociation channel, the emission angle is out of phase with the electron, indicating back-toback emission. For protons with energy above 0.4 eV, the oscillation of the center energy position of the proton band corresponds to the spiral pattern shown in Fig. 1(i).

Spiral patterns are usually formed by the interference of electron wave packets in vortex states with different magnetic quantum numbers [35-39]. They have been observed in photoelectron spectra resulting from multiphoton ionization of atoms by counter-rotating circularly polarized fields [38,39] and predicted for ions emitted in molecular photodissociation [40,41]. Here, we observe the spiral pattern for protons resulting from the dissociative ionization of H<sub>2</sub><sup>+</sup> in bicircularly polarized two-color fields. Figures 3(a) and 3(c) show the  $\phi_{\rm RP}$ -averaged proton energy spectrum and the schematic diagram of the involved pathways, respectively. We observe a series of peaks in the proton energy spectrum, corresponding to  $1\omega$ ,  $2\omega$ , and  $3\omega$  dissociation channels and chargeresonance enhanced ionization (CREI) (see Refs. [10,14] for details of channel identification). In Fig. 3(b), we illustrate the  $\phi_{\rm RP}$ -resolved proton asymmetry degree which is defined by  $[Y(\phi_i = 0^\circ, E_i) - Y(\phi_i = 180^\circ, E_i)]/[Y(\phi_i =$  $0^{\circ}, E_i) + Y(\phi_i = 180^{\circ}, E_i)$ , where  $Y(\phi_i, E_i)$  is the proton yield with the energy  $E_i$  and the emission angle  $\phi_i$ . By scanning  $\phi_{\rm RP}$ , the proton emission angle is streaked. Furthermore,



FIG. 3. Experimental control of proton emission asymmetry in laboratory and molecular frames in bicircularly polarized two-color fields. (a) Proton energy spectrum averaged over the relative twocolor phase  $\phi_{RP}$ . (b) Lab-frame proton emission asymmetry as a function of the proton energy and  $\phi_{RP}$ , where the proton emission angle is confined in two angular ranges,  $0 \pm 15^{\circ}$  and  $180 \pm 15^{\circ}$ , in the laboratory frame. (c) Schematic diagram of the relevant dissociation pathways in the fundamental-wave (FW) only and the secondharmonic (SH) involved cases. The gray curves are the ground the first excited ionic states of H<sub>2</sub><sup>+</sup>. (d) Molecular-frame proton emission asymmetry, averaged over  $\phi_{RP}$ , as a function of proton energy and molecular-frame electron emission angle. The length of the arrow corresponds to the photon energy and its color is used to assign its pathway.

we observe that the asymmetry distribution has a  $\pi$  phase shift between the rising and falling edges of any two adjacent channels. This phenomenon tends to validate the photonphase-representation theory [42]. Within this approach, any two-color quantum phenomena can be explained by the interference of the pathways involving different numbers of photons in regions where these pathways overlap in energy. Since the adjacent pathways have alternating opposite parities, their interference gives rise to asymmetry with a changing sign.

The asymmetric proton distribution can also be revealed in the molecular frame using the photoelectron emission angle  $\phi_e$  to tag the ionization instant by angular streaking [43,44]. Correspondingly, the ionization instant (or phase) in the molecular frame is defined by the electron and proton emission angle difference,  $\phi_{mol} = \phi_e - \phi_i$ . Figure 3(d) shows our measured proton asymmetry distribution in the molecular frame as a function of  $\phi_{mol}$  and  $E_i$ ,  $[Y(\phi_{mol}, E_i) - Y(\phi_{mol} + \pi, E_i)]/[Y(\phi_{mol}, E_i) + Y(\phi_{mol} + \pi, E_i)]$ , where the two-color phase  $\phi_{RP}$  is averaged. The similarity of Figs. 3(b) and 3(d) confirms that the proton emission asymmetry can be controlled by  $\phi_{RP}$  in the laboratory frame and visualized by  $\phi_{mol}$  in the molecular frame, supporting the conclusion of a previous study [44] with few-cycle pulses. The interference contrast in the asymmetry distribution for the 1 $\omega$  channel ( $E_i \sim 0.2 \text{ eV}$ ) in Fig. 3(d) peaks around  $\phi_{mol} = 180^\circ$ . This is consistent with the results in Figs. 1(d)–1(i), showing that low-energy protons are predominantly emitted in the direction opposite to the coincident electrons. The slanted fringes in the polar coordinate representation correspond to the spiral pattern in the momentum distribution shown in the Cartesian coordinates [Fig. 1(i)].

The ionization step is affected by the preferential ionization effect, which gives us the experimental "knob" to control it by changing the shape of the electric field via the two-color phase. This has been well studied in previous publications [24,25]. In the second step, i.e., dissociation, the pathway interference imposes new structures or modulations on top of the result from the ionization step. In this article, our idea is to show the effect of the second step can even dramatically change the result from the first step, leading to a counterintuitive phenomenon.

The preferential ionization induced by a noncircular vector shape can explain previous findings that the proton emission direction is sensitive to the two-color relative phase. However, it does not account for the vortexlike feature in the relative emission angle between the proton and the proton. To explain this, pathway interference with different vortex states must be considered.

### **III. THEORETICAL MODELS**

## A. Classical and quantum-mechanical interpretations of molecular-frame emission angles

The molecular-frame emission angle,  $\phi_{mol} = \phi_e - \phi_i$ , which is directly related to the molecular-frame ionization instant, is defined as the angular difference between the direction of electron and proton emission. It plays an important role in the proton emission asymmetry. Here we interpret it both classically, based on angular streaking, and quantummechanically, based on vortex interference.

From the classical perspective, the electron emission angle is defined by the birth time of the nuclear wave packet in H<sub>2</sub><sup>+</sup>, i.e.,  $t_0 = (\phi_e + \pi/2 + 2k\pi)/\omega$ , where  $k \in \mathbb{Z}$  and  $\pi/2$  comes from the angle between the electric field and the vector potential. Here the constant attoclock offset angle is ignored. With the assumption that the molecular axis does not rotate during the dissociation, for a given molecular orientation  $\phi_i$  the laser electric field projection along this molecular axis has the form of  $E \sim \cos(\omega t - \phi_i) = \cos[\omega(t - t_0) + \phi_{mol} + \pi/2]$ . Therefore, the wave packet will acquire the phase factor  $e^{i(\phi_{mol}+\pi/2)}$ per photon absorption from the light field. Considering the superposition between the one-photon and net two-photon channel, the final nuclear wave function is given by  $\Psi(E_i) \sim e^{2i(\phi_{mol}+\pi/2)+i\phi_g}|g\rangle + e^{i(\phi_{mol}+\pi/2)+i\phi_u}|u\rangle$ , where  $\phi_g$  and  $\phi_u$  are the accumulated phases for H<sub>2</sub><sup>+</sup> nuclear wave packets along the gerade and ungerade final states, respectively. Their interference fringes are governed by  $\cos(\phi_{mol} + \pi/2 + \phi_g - \phi_u)$ , with a periodicity of  $\phi_{mol}$ . In this article, we use the classical words like "streaking" or "rotation" to describe the lightcontrol nuclear motions. This is because the phase of the vortex states,  $\phi_{mol} = \phi_e - \phi_i$ , is proportional to the electron emission angle  $\phi_e$ , which is streaked by the laser field.

From the quantum perspective, the dissociative ionization must comply with the conservation of energy and angular momentum between the photoelectron and the residual ion, i.e.,

$$E_e + E_i = E_{H_2} + n\omega,$$
  
$$m_e + m_i = n.$$
 (1)

where  $E_{H_2}$  is the ground-state energy of the H<sub>2</sub> molecule, *n* is the total number of absorbed photons, and  $m_{e/i}$  is the angular momentum quantum number of electrons/ions. Because only the channels with the same electron energy and the same nuclear energy can interfere with each other, n is same for the two interfering channels. Comparing the one-photon channel and net two-photon channel, in the dissociation process the numbers of photons absorbed by the nuclei differs by +1; therefore, the photon number absorbed by the photo electron in the photoionization process must differ by -1accordingly. The final electron-nuclear wave packet (which must be considered as a whole) can be described by  $\Psi(E_i) \sim$  $e^{im_e\phi_e+im_i\phi_i+\phi_g}|g\rangle + e^{i(m_e-1)\phi_e+i(m_i+1)\phi_i+\phi_u}|u\rangle$ . Thus, the corresponding interference fringes are governed by  $\cos(\phi_{mol} +$  $\phi_g - \phi_u$ ). Note that here  $\phi_g$  and  $\phi_u$  are the phases of the electron-nuclei-coupled wave packet and, compared to those defined in the classical picture, include the electron phase factor of  $\pi/2$ .

#### B. Semiclassical trajectory model

The next question is how to calculate the accumulated phases  $\phi_g$  and  $\phi_u$  semiclassically. Since the shapes of potential energy curves change dramatically with respect to the unperturbed ones at the intensity used in the present experiment ( $I \sim 10^{14} \text{ W/cm}^2$ ), we first calculate the light-dressed potential energy curves. The Hamiltonian matrix in the basis of { $|1s\sigma_g\rangle$ ,  $|2p\sigma_u\rangle$ } at a given internuclear distance can be written as

$$H = \begin{pmatrix} E_g & 0\\ 0 & E_u \end{pmatrix} + \begin{pmatrix} 0 & \mu\\ \mu & 0 \end{pmatrix}$$
$$\times E_0 f(t) \cos[\omega(t - t_0) + \phi_{\text{mol}} + \pi/2], \qquad (2)$$

with the state energies  $E_g$  and  $E_u$  and the transition dipole  $\mu$  computed by solving the Schrödinger equation of fixed nuclear H<sub>2</sub><sup>+</sup> [45].  $E_0 = \sqrt{I_0/2}$  is the amplitude of the electric field along the molecular axis. Ignoring the field envelope f(t) and further introducing the Floquet basis  $|g/u, n\rangle = e^{-in[\omega(t-t_0)+\phi_{mol}+\pi/2]}|g/u\rangle$ , one arrives at the Floquet Hamiltonian

$$H_F = \begin{pmatrix} E_g & 0\\ 0 & E_u \end{pmatrix} + \begin{pmatrix} 0 & \mu\\ \mu & 0 \end{pmatrix} E_0 \frac{\hat{a} + \hat{a}^{\dagger}}{2} + \hat{N}\omega.$$
(3)

After diagonalizing  $H_F$  for every internuclear distance, we get the Floquet energy curves plotted in Fig. 4 as gray solid



FIG. 4. Floquet energy curves for an intensity of  $1.6 \times 10^{14}$  W/cm<sup>2</sup> and a wavelength of 800 nm.

lines. They are quite different from the unperturbed curves (dashed lines) especially in the region of 3 a.u.  $\leq R \leq 6$  a.u. We assign the one-photon and net two-photon pathways to the red and green curves, respectively. Notice that the one-photon channel requires one more Landau-Zener tunneling, compared to the net two-photon channel. The tunneling point is determined by the minimal gap condition.

In the Floquet picture, the time dependence of the Hamiltonian is removed, and we can compute the phase difference accumulated in those two "static" pathways, for which the energy of the wave packet remains constant. Formally, we start from the Feynman path integral for the nuclear wave function:

$$\Psi(x_f, t_f) = \int K(x_f, t_f; x_i, t_i) \Psi(x_i, t_i) dx_i$$
$$= \int \exp\left\{ i \int_{t_i}^{t_f} L[x(t), \dot{x}(t)] dt \right\}$$
$$\times \Psi(x_i, t_i) D[x(t)] dx_i.$$
(4)

The initial nuclear wave packet  $\Psi(x_i, t_i)$  is close to the vibrational ground of H<sub>2</sub>, according to the Franck-Condon approximation. *L* is the Lagrangian for all trajectories x(t) that satisfy the boundary conditions  $x(t_i) = x_i$  and  $x(t_f) = x_f$ . The propagation time  $t_f - t_i$  should be large enough to guarantee that all the physical observables are converged. In the end, one measures the distribution in momentum space  $|\Phi(p_f, t_f)|^2$ , instead of that in real space. Thus, an additional Fourier transformation is required [46],

$$\Phi(p_f, t_f) = \int e^{-ip_f x_f} \Psi(x_f, t_f) dx_f$$
  
= 
$$\int \exp\left\{-ip_f x_f + i \int_{t_i}^{t_f} L[x(t), \dot{x}(t)] dt\right\}$$
  
× 
$$\Psi(x_i, t_i) D[x(t)] dx_i dx_f.$$
 (5)

We apply a saddle-point approximation to the above integral. The first saddle-point condition on the path x(t) results in the Lagrange equation

$$\frac{\delta\phi}{\delta x} = 0 \Rightarrow \frac{d}{dt} \left( \frac{\partial L}{\partial \dot{x}} \right) - \frac{\partial L}{\partial x} = 0.$$
 (6)

This means that x(t) moves classically. Since the Lagrangian is time independent, the energy  $E = \dot{x}(\partial L/\partial \dot{x}) - L$  is conserved. Introducing the canonical momentum  $p \equiv \partial L/\partial \dot{x}$ , the phase term can be written as

$$\phi = -p_f x_f + \int_{t_i}^{t_f} (p\dot{x} - E) dt$$
  
=  $-p_f x_f + \int_{x_i}^{x_f} p \, dx - E(t_f - t_i).$  (7)

One has the following saddle point conditions for the usual variables  $x_i$  and  $x_f$ :

$$\frac{\partial \phi}{\partial x_i} = 0 \Rightarrow -p|_{t=t_i} = 0, \tag{8}$$

$$\frac{\partial \phi}{\partial x_f} = 0 \Rightarrow -p_f + \left. p \right|_{t=t_f} = 0. \tag{9}$$

We now have all the required information to determine the classical trajectory: it starts at turning point  $x_i$ , with zero momentum  $p_i = 0$ , and reaches asymptotic momentum  $p_f$  in the end. The accumulated phase is given by

$$\phi = -p_f x_f + \int_{x_i}^{x_f} p \, dx - E(t_f - t_i)$$
  
=  $\int_{x_i}^{x_m} p \, dx - p_f x_m + E(t_f - t_i).$  (10)

Here we introduce a given end point  $x_m$  which is large enough such that  $p \approx p_f$  for  $x > x_m$ . The last two terms in above phase expression are the same for the two interference channels. Note that here the energy *E* here should be understood in the Floquet picture and equals the final kinetic energy of the nuclei. Thus, the only important term is  $\int_{x_i}^{x_m} p \, dx$ , the classical action from the initial position  $x_i$  to a given final position  $x_m$ .

Two more steps lead to the accurate phase difference [3]. First, since the net two-photon channel has one less Landau-Zener tunneling, there is a so-called Stokes phase [47],

$$\varphi_{\mathcal{S}} = \frac{\pi}{4} + \delta(\ln \delta - 1) + \arg \Gamma(1 - i\delta), \qquad (11)$$

added to it. Here the adiabatic parameter  $\delta = \Delta^{3/2}/(4v\sqrt{\Delta''})$  is determined by the gap energy  $\Delta$ , the curvature  $\Delta''$ , and the nuclear velocity v at the minimal gap. Second, since the momentum of the emitted electron does not exactly follow the vector potential at the ionization time, but has an attoclock offset angle  $\gamma$ , we also have to add this constant correction to  $\phi_{\text{mol}}$ .

In summary, the phase difference is given by

$$\phi_g - \phi_u = \int_{x_i}^{x_m} p^{(g)} \, dx - \int_{x_i}^{x_m} p^{(u)} \, dx + \varphi_S. \tag{12}$$

It is interesting to note that its energy derivative,

$$\partial_E(\phi_g - \phi_u) = \int_{x_i}^{x_m} \frac{dx}{v^{(g)}} - \int_{x_i}^{x_m} \frac{dx}{v^{(u)}} + \frac{\partial\varphi_S}{\partial E}$$
$$= t^{(g)} - t^{(u)} + \frac{\partial\varphi_S}{\partial E}, \tag{13}$$

roughly equals the time difference for nuclear wave packets that move from their initial position to a given final position. The last term is relatively small compared to the previous two and can also be regarded as the transition time during the Landau-Zener tunneling.

The final wave function has the form

$$\Phi(p_f) \sim e^{i\phi_g} |g, n\rangle + e^{i\phi_u} |u, n+1\rangle$$
  
 
$$\sim |g\rangle + e^{i(\phi_u - \phi_g) - \phi_{\text{mol}} - \pi/2 - \gamma} |u\rangle, \qquad (14)$$

with maximal asymmetry at  $\phi_{mol} = \phi_u - \phi_g + \pi/2 + \gamma$ . The comparison between the model calculation and the *ab initio* simulation is shown in Fig. 6 (more discussions there). The calculation of the phase difference for other channels is similar and thus not reproduced here.

Finally, we need to point out the differences between our current model and the one used in previous studies [21,44,48]. First, in those studies the nuclear wave packet was propagated along unperturbated potential energy curves. Second, in those studies the phase  $-p_f x_f$  due to the Fourier transformation from coordinate to momentum space was not included. Third, in those studies the initial position was taken to be the equilibrium internuclear distance of H<sub>2</sub> ( $r_e = 1.4$  a.u.), instead of the classical turning point that we use here. Finally, in those studies a  $\pi$  phase ws added for each photon absorption, while the correct Stokes phase at the zero-field limit should be  $\pi/4$  following from Eq. (11).

## C. Quantitative quantum simulation

To quantitatively model the proton spiral pattern and the electron-proton angular correlation, we resort to the quantitative quantum simulation [49], where the electron part is treated using the strong-field approximation with the Coulomb-Volkov correction and the nuclear part is modeled by solving the time-dependent Schrödinger equation (TDSE) for the two lowest electronic states of  $H_2^+$  (1s $\sigma_g$  and 2 $p\sigma_u$ ). In this approach, we remove the degrees of freedom of the emitted electron in the TDSE, while the correlation between the emitted electron and the residual  $H_2^+$  is still preserved. A similar approach has successfully described the energy correlation between the electron and the ion [3,50]. In Figs. 5(a) and 5(b), we present the calculated photoelectron and proton momentum distributions at  $\phi_{\rm RP} = 0$ , respectively. In Fig. 5(c), we show the obtained proton asymmetry distribution defined by the difference between  $\phi_{\rm RP} = 0$  and  $\phi_{\rm RP} = \pi$ . The simulation confirms that the  $1\omega$  protons are emitted approximately in the direction opposite to the coincident electrons, while high-energy protons form a spirally shaped asymmetry distribution. To quantitatively compare with the experimental results shown in Fig. 3(d), in Fig. 5(d) we illustrate the calculated proton asymmetry parameter in the molecular frame, The slanted fringes are clearly visible for energies  $E_i >$ 0.5 eV corresponding to the spiral pattern.



FIG. 5. (a), (b) Calculated photoelectron (a) and proton (b) momentum distributions at the two-color relative phase of 0.0. (c) Calculated difference between the proton momentum distributions at the two-color relative phases of 0 and  $\pi$ , corresponding to the experimental data in Fig. 1(i). (d) Calculated proton asymmetry distribution in the molecular frame, averaged over the two-color relative phase. (e) Schematics of the interference of temporally delayed wave packets with different magnetic quantum numbers. (f) Formation of the spiral pattern by vortex electron localization. The remaining bound electron in H<sub>2</sub><sup>+</sup> forms a vortex distribution and then the electron localization will transfer the angular momentum to the emitting proton, creating proton vortices. The time delay of the interfering nuclear pathways is corresponding to the slope of their interference fringe with respect to energy.

Figures 5(e) and 5(f) illustrate the mechanism of light control over molecular nuclei. The asymmetry pattern is formed by the interference of the gerade and ungerade electronnuclear wave packets in H<sub>2</sub><sup>+</sup> with helical phase factors  $m(\phi_e - \phi_i) + E_i t$ , where *m* is the magnetic quantum number due to the circular polarization and *t* is the lifetime of the nuclear wave packet, which is defined as the time interval from the creation to the detection. The two pathways have different *m*. Their interference fringes are proportional to  $\cos[\Delta m(\phi_e - \phi_i) + E_i \Delta t]$ . Since the residual electron in H<sub>2</sub><sup>+</sup> absorbs spin angular momenta of photons, a spiral pattern of the bound electron (yellow curve) emerges from pathway interference. As sketched in Fig. 5(f), the formation of the proton pattern can be understood as electron localization, achieved through constructive interference, which forces the protons (green dots) to change their original direction of motion. The fringe slope with respect to energy is the lifetime difference (also referred to as "time delay") of the interfering pathways divided by the difference of magnetic quantum numbers. For the special case when the time delay  $\Delta t$  is energy independent, the interference fringes will form a standard Archimedes spiral, corresponding to the stripe shown in Fig. 5(f). Therefore, the spiral proton pattern provides an approach to probe the energy-dependent time delay of nuclear pathways in molecules spectroscopically.

## **IV. DISCUSSION**

To validate the correspondence between the slope of spiral fringes and the nuclear time delay, we did the simulations based on the semiclassical trajectory model for the propagation of a coupled electron-nuclei packet in the laser fields. For simplicity, we first only include the fundamental field, where just two dissociation channels (1 $\omega$  and net-2 $\omega$ ) interfere. Figure 6(a) illustrates the positions of the interference maxima of the spiral fringes obtained from our quantum-mechanical and semiclassical simulations. In Fig. 6(b), we compare the quantum-mechanically calculated spiral slope with the lifetime difference of the wave packets calculated by numerical propagation along the two semiclassical trajectories. The results show that the time delay of the nuclear motion in the two pathways is on the femtosecond timescale and decreases with the proton energy. The good agreement between them provides evidence for the validity of our semiclassical model and suggests a new type of nuclear dynamics chronoscopy.

The back-to-back emission of the electron and the lowenergy proton in the two-color field is a special case of vortex interference when the time delay is between two interference pathways  $\Delta t = 0$ . As sketched in the inset of Fig. 3(c), two pathways dominate the release of protons with energies below 0.3 eV, the 1 $\omega$  channel due to the fundamental field only and the net-1 $\omega$  channel triggered by both fields. In Fig. 6(c), we show the positions of interference maxima for those two pathways based on the semiclassical trajectory calculation in comparison with our quantum-mechanical simulation. Both curves are almost flat and are located near  $\phi_{mol} = 180^{\circ}$ , in agreement with the  $1\omega$  channel shown in Fig. 1(i). This indicates that there is no prominent time delay between the two dissociation paths, such that constructive interference occurs around  $\phi_{\rm mol} = 180^{\circ}$ . As illustrated by the light-dressed potential curves in Fig. 6(d), the  $1\omega$  pathway and the net- $1\omega$ pathway are symmetric in the Floquet picture and thus have almost equal lifetimes.

We note that for the experimental results in Fig. 3(d), the spiral fringes above 0.5 eV are not as smooth as in the numerical simulation [Fig. 5(d)]. We attribute this discrepancy to our TDSE solution being restricted to the two lowest electronic states of  $H_2^+$  and expect it to be resolved by more accurate solutions of the two-electron TDSE. Another possible reason for this discrepancy is the use of electron angular streaking to



FIG. 6. (a) Angular positions of the interference maxima between 1 $\omega$  and net-2 $\omega$  dissociation channels extracted from the TDSE result (blue solid line) and predicted by our semiclassical model (red dashed line). (b) Time delay between the 1 $\omega$  and net-2 $\omega$  channels. Note that the TDSE result is more noisy than the model result due to the numerical difference used for evaluating the curve slope. (c) Angular positions of the interference maxima between the 1 $\omega$  and net-1 $\omega$  channels. (d) Sketch of related energy curves. Red and orange dashed lines represent unperturbed one-photon and net one-photon paths, respectively. Gray and green solid lines represent gerade and ungerade states, respectively.

tag the ionization time. The mapping between electron angular direction and ionization time is not sharply defined and more importantly exhibits energy dependence, as demonstrated in Refs. [26,51].

### V. CONCLUSION

To conclude, for dissociative ionization of H<sub>2</sub> in bicircularly polarized two-color laser fields, we observed a surprising strongly correlated back-to-back emission of electrons and protons and spiral proton asymmetry distribution. Although it is difficult to directly control nuclei with laser light, we demonstrated an optical mechanism for controlling the intramolecular nuclear dynamics. This mechanism follows the ionization step and proceeds by the residual electron of the molecular cation absorbing circularly polarized photons from the tail of the laser field, giving rise to the pathway interference of vortex electron wave packets with helical phases. Vortex electron localization imposes a two-dimensional spiral structure on the fragment (proton) momentum distribution, allowing one to indirectly control the nuclear dynamics. We developed a semiclassical trajectory model which clarifies the phase of electron-nuclear-coupling wave packets in strong laser fields. We validate our interpretation by both a quantum-mechanical numerical simulation of the field-driven coupled electronic-nuclear dynamics and a semiclassical-trajectory model for the phase accumulation of the laser-driven electronic-nuclear wave packet.

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- P. Brumer and M. Shapiro, Laser control of molecular processes, Annu. Rev. Phys. Chem. 43, 257 (1992).
- [2] A. H. Zewail, Laser femtochemistry, Science 242, 1645 (1988).
- [3] J. Wu, M. Kunitski, M. Pitzer, F. Trinter, L. P. H. Schmidt, T. Jahnke, M. Magrakvelidze, C. B. Madsen, L. B. Madsen, U. Thumm, and R. Dörner, Electron-nuclear energy sharing in above-threshold multiphoton dissociative ionization of H<sub>2</sub>, Phys. Rev. Lett. **111**, 023002 (2013).
- [4] L. Holmegaard, J. L. Hansen, L. Kalhøj, S. Louise Kragh, H. Stapelfeldt, F. Filsinger, J. Küpper, G. Meijer, D. Dimitrovski, M. Abu-Samha *et al.*, Photoelectron angular distributions from strong-field ionization of oriented molecules, Nat. Phys. 6, 428 (2010).
- [5] M. Wollenhaupt, V. Engel, and T. Baumert, Femtosecond laser photoelectron spectroscopy on atoms and small molecules: Prototype studies in quantum control, Annu. Rev. Phys. Chem. 56, 25 (2005).
- [6] P. H. Bucksbaum, A. Zavriyev, H. G. Muller, and D. W. Schumacher, Softening of the H<sub>2</sub><sup>+</sup> molecular bond in intense laser fields, Phys. Rev. Lett. 64, 1883 (1990).
- [7] A. Giusti-Suzor, F. H. Mies, L. F. DiMauro, E. Charron, and B. Yang, Dynamics of H<sub>2</sub><sup>+</sup> in intense laser fields, J. Phys. B: At., Mol. Opt. Phys. 28, 309 (1995).
- [8] M. Thompson, M. Thomas, P. Taday, J. Posthumus, A. Langley, L. Frasinski, and K. Codling, One and two-colour studies of the dissociative ionization and Coulomb explosion of H<sub>2</sub> with intense Ti:sapphire laser pulses, J. Phys. B: At., Mol. Opt. Phys. **30**, 5755 (1997).
- [9] H. Niikura, F. Légaré, R. Hasbani, A. Bandrauk, M. Y. Ivanov, D. Villeneuve, and P. Corkum, Sub-laser-cycle electron pulses for probing molecular dynamics, Nature (London) 417, 917 (2002).
- [10] A. Rudenko, B. Feuerstein, K. Zrost, V. L. B. de Jesus, T. Ergler, C. Dimopoulou, C. D. Schröter, R. Moshammer, and J. Ullrich, Fragmentation dynamics of molecular hydrogen in strong ultrashort laser pulses, J. Phys. B: At., Mol. Opt. Phys. 38, 487 (2005).
- [11] M. Kling, C. Siedschlag, A. J. Verhoef, J. Khan, M. Schultze, T. Uphues, Y. Ni, M. Uiberacker, M. Drescher, F. Krausz *et al.*, Control of electron localization in molecular dissociation, Science **312**, 246 (2006).
- [12] M. Kremer, B. Fischer, B. Feuerstein, V. L. B. de Jesus, V. Sharma, C. Hofrichter, A. Rudenko, U. Thumm, C. D. Schröter, R. Moshammer, and J. Ullrich, Electron localization in molecular fragmentation of H<sub>2</sub> by carrier-envelope phase stabilized laser pulses, Phys. Rev. Lett. **103**, 213003 (2009).
- [13] B. Fischer, M. Kremer, T. Pfeifer, B. Feuerstein, V. Sharma, U. Thumm, C. D. Schröter, R. Moshammer, and J. Ullrich, Steering the electron in H<sub>2</sub><sup>+</sup> by nuclear wave packet dynamics, Phys. Rev. Lett. **105**, 223001 (2010).

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- [14] D. Ray, F. He, S. De, W. Cao, H. Mashiko, P. Ranitovic, K. P. Singh, I. Znakovskaya, U. Thumm, G. G. Paulus, M. F. Kling, I. Litvinyuk, and C. L. Cocke, Ion-energy dependence of asymmetric dissociation of D<sub>2</sub> by a two-color laser field, Phys. Rev. Lett. **103**, 223201 (2009).
- [15] H. Xu, J.-P. MacLean, D. Laban, W. Wallace, D. Kielpinski, R. Sang, and I. Litvinyuk, Carrier-envelope-phase-dependent dissociation of hydrogen, New J. Phys. 15, 023034 (2013).
- [16] H. Xu, T.-Y. Xu, F. He, D. Kielpinski, R. T. Sang, and I. V. Litvinyuk, Effect of nuclear mass on carrier-envelope-phasecontrolled electron localization in dissociating molecules, Phys. Rev. A 89, 041403(R) (2014).
- [17] K. P. Singh, F. He, P. Ranitovic, W. Cao, S. De, D. Ray, S. Chen, U. Thumm, A. Becker, M. M. Murnane, H. C. Kapteyn, I. V. Litvinyuk, and C. L. Cocke, Control of electron localization in deuterium molecular ions using an attosecond pulse train and a many-cycle infrared pulse, Phys. Rev. Lett. **104**, 023001 (2010).
- [18] G. Sansone, F. Kelkensberg, J. Pérez-Torres, F. Morales, M. F. Kling, W. Siu, O. Ghafur, P. Johnsson, M. Swoboda, E. Benedetti *et al.*, Electron localization following attosecond molecular photoionization, Nature (London) **465**, 763 (2010).
- [19] F. He, A. Becker, and U. Thumm, Strong-field modulated diffraction effects in the correlated electron-nuclear motion in dissociating H<sup>+</sup><sub>2</sub>, Phys. Rev. Lett. **101**, 213002 (2008).
- [20] B. Sheehy, B. Walker, and L. F. DiMauro, Phase control in the two-color photodissociation of HD<sup>+</sup>, Phys. Rev. Lett. 74, 4799 (1995).
- [21] J. Wu, A. Vredenborg, L. P. H. Schmidt, T. Jahnke, A. Czasch, and R. Dörner, Comparison of dissociative ionization of H<sub>2</sub>, N<sub>2</sub>, Ar<sub>2</sub>, and CO by elliptically polarized two-color pulses, Phys. Rev. A 87, 023406 (2013).
- [22] H. Li, X. Gong, K. Lin, R. de Vivie-Riedle, X. Tong, J. Wu, and M. F. Kling, Sub-cycle directional control of the dissociative ionization of H<sub>2</sub> in tailored femtosecond laser fields, J. Phys. B: At., Mol. Opt. Phys. **50**, 172001 (2017).
- [23] H. Ibrahim, C. Lefebvre, A. D. Bandrauk, A. Staudte, and F. Légaré, H<sub>2</sub>: the benchmark molecule for ultrafast science and technologies, J. Phys. B: At., Mol. Opt. Phys. **51**, 042002 (2018).
- [24] X. Gong, P. He, Q. Song, Q. Ji, H. Pan, J. Ding, F. He, H. Zeng, and J. Wu, Two-dimensional directional proton emission in dissociative ionization of H<sub>2</sub>, Phys. Rev. Lett. **113**, 203001 (2014).
- [25] K. Lin, X. Gong, Q. Song, Q. Ji, W. Zhang, J. Ma, P. Lu, H. Pan, J. Ding, H. Zeng *et al.*, Directional bond breaking by polarization-gated two-color ultrashort laser pulses, J. Phys. B: At., Mol. Opt. Phys. **49**, 025603 (2016).
- [26] M. Han, P. Ge, J. Wang, Z. Guo, Y. Fang, X. Ma, X. Yu, Y. Deng, H. J. Wörner, Q. Gong *et al.*, Complete characterization of

sub-Coulomb-barrier tunnelling with phase-of-phase attoclock, Nat. Photon. **15**, 765 (2021).

- [27] R. Dörner, V. Mergel, O. Jagutzki, L. Spielberger, J. Ullrich, R. Moshammer, and H. Schmidt-Böcking, Cold target recoil ion momentum spectroscopy: A 'momentum microscope' to view atomic collision dynamics, Phys. Rep. 330, 95 (2000).
- [28] J. Ullrich, R. Moshammer, A. Dorn, R. Dörner, L. Schmidt, and H. Schmidt-Böcking, Recoil-ion and electron momentum spectroscopy: reaction-microscopes, Rep. Prog. Phys. 66, 1463 (2003).
- [29] W. Quan, V. V. Serov, M. Z. Wei, M. Zhao, Y. Zhou, Y. L. Wang, X. Y. Lai, A. S. Kheifets, and X. J. Liu, Attosecond molecular angular streaking with all-ionic fragments detection, Phys. Rev. Lett. **123**, 223204 (2019).
- [30] U. S. Sainadh, H. Xu, X. Wang, A. Atia-Tul-Noor, W. C. Wallace, N. Douguet, A. Bray, I. Ivanov, K. Bartschat, A. Kheifets *et al.*, Attosecond angular streaking and tunnelling time in atomic hydrogen, Nature (London) **568**, 75 (2019).
- [31] N. Camus, E. Yakaboylu, L. Fechner, M. Klaiber, M. Laux, Y. Mi, K. Z. Hatsagortsyan, T. Pfeifer, C. H. Keitel, and R. Moshammer, Experimental evidence for quantum tunneling time, Phys. Rev. Lett. **119**, 023201 (2017).
- [32] A. S. Kheifets, The attoclock and the tunneling time debate, J. Phys. B: At., Mol. Opt. Phys. 53, 072001 (2020).
- [33] M. Klaiber, K. Z. Hatsagortsyan, and C. H. Keitel, Tunneling dynamics in multiphoton ionization and attoclock calibration, Phys. Rev. Lett. **114**, 083001 (2015).
- [34] M. Han, P. Ge, Y. Fang, X. Yu, Z. Guo, X. Ma, Y. Deng, Q. Gong, and Y. Liu, Unifying tunneling pictures of strong-field ionization with an improved attoclock, Phys. Rev. Lett. 123, 073201 (2019).
- [35] J. M. Ngoko Djiokap, S. X. Hu, L. B. Madsen, N. L. Manakov, A. V. Meremianin, and A. F. Starace, Electron vortices in photoionization by circularly polarized attosecond pulses, Phys. Rev. Lett. 115, 113004 (2015).
- [36] J. M. Ngoko Djiokap, A. V. Meremianin, N. L. Manakov, S. X. Hu, L. B. Madsen, and A. F. Starace, Multistart spiral electron vortices in ionization by circularly polarized UV pulses, Phys. Rev. A 94, 013408 (2016).
- [37] J. M. Ngoko Djiokap, A. V. Meremianin, N. L. Manakov, L. B. Madsen, S. X. Hu, and A. F. Starace, Dynamical electron vortices in attosecond double photoionization of H<sub>2</sub>, Phys. Rev. A 98, 063407 (2018).
- [38] D. Pengel, S. Kerbstadt, L. Englert, T. Bayer, and M. Wollenhaupt, Control of three-dimensional electron vortices from femtosecond multiphoton ionization, Phys. Rev. A 96, 043426 (2017).

- [39] D. Pengel, S. Kerbstadt, D. Johannmeyer, L. Englert, T. Bayer, and M. Wollenhaupt, Electron vortices in femtosecond multiphoton ionization, Phys. Rev. Lett. 118, 053003 (2017).
- [40] Z. Chen, P.-L. He, and F. He, Spiral nuclear momentum distribution for the dissociation of H<sub>2</sub><sup>+</sup> in a circularly polarized laser pulse, Phys. Rev. A **101**, 033406 (2020).
- [41] Z. Chen and F. He, Interference of nuclear wave packets carrying different angular momenta in the dissociation of  $H_2^+$  in strong circularly polarized laser pulses, Phys. Rev. A **102**, 033107 (2020).
- [42] V. Roudnev and B. D. Esry, General theory of carrier-envelope phase effects, Phys. Rev. Lett. 99, 220406 (2007).
- [43] J. Wu, M. Magrakvelidze, L. P. H. Schmidt, M. Kunitski, T. Pfeifer, M. Schöffler, M. Pitzer, M. Richter, S. Voss, H. Sann *et al.*, Understanding the role of phase in chemical bond breaking with coincidence angular streaking, Nat. Commun. 4, 2177 (2013).
- [44] S. Kangaparambil, V. Hanus, M. Dorner-Kirchner, P. He, S. Larimian, G. Paulus, A. Baltuška, X. Xie, K. Yamanouchi, F. He *et al.*, Generalized phase sensitivity of directional bond breaking in the laser-molecule interaction, Phys. Rev. Lett. **125**, 023202 (2020).
- [45] H. Liang, X.-R. Xiao, Q. Gong, and L.-Y. Peng, Accurate computation of above threshold ionization spectra for stretched H<sub>2</sub><sup>+</sup> in strong laser fields, J. Phys. B: At., Mol. Opt. Phys. 50, 174002 (2017).
- [46] N. I. Shvetsov-Shilovski, M. Lein, L. B. Madsen, E. Räsänen, C. Lemell, J. Burgdörfer, D. G. Arbó, and K. Tőkési, Semiclassical two-step model for strong-field ionization, Phys. Rev. A 94, 013415 (2016).
- [47] S. N. Shevchenko, S. Ashhab, and F. Nori, Landau–Zener– Stückelberg interferometry, Phys. Rep. 492, 1 (2010).
- [48] S. Pan, Z. Zhang, C. Hu, P. Lu, X. Gong, R. Gong, W. Zhang, L. Zhou, C. Lu, M. Shi *et al.*, Wave-packet surface propagation for light-induced molecular dynamics, Phys. Rev. Lett. 132, 033201 (2024).
- [49] H. Liang and L.-Y. Peng, Quantitative theory for electronnuclear energy sharing in molecular ionization, Phys. Rev. A 101, 053404 (2020).
- [50] C. He, H. Liang, M.-M. Liu, L.-Y. Peng, and Y. Liu, Laser-wavelength and intensity dependence of electron-nuclear energy sharing in dissociative ionization of H<sub>2</sub>, Phys. Rev. A 101, 053403 (2020).
- [51] D. Trabert, N. Anders, S. Brennecke, M. S. Schöffler, T. Jahnke, L. P. H. Schmidt, M. Kunitski, M. Lein, R. Dörner, and S. Eckart, Nonadiabatic strong field ionization of atomic hydrogen, Phys. Rev. Lett. **127**, 273201 (2021).