

Control of attosecond photoemission time delays by quantum state interference

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(Received 26 January 2026; accepted 4 September 2026; published 9 October 2026)

Scattering theory has long established that wave packets can experience time delays and advances, while developments in attosecond interferometry have made such temporal shifts experimentally accessible. They are encoded in the spectral phase of the emitted electron wave packet and are commonly accessed through interferometric techniques. Here, we report an intensity-driven reversal of the angular phase of a threshold photoelectron sideband in helium. By varying the intensity of an infrared dressing field, we control the direction of the phase change at large emission angles, which is associated with interference between partial waves near angular nodes. Solving the time-dependent Schrödinger equation within the single-active-electron approximation confirms the existence of an angular phase inversion. Our results reveal a mechanism for manipulating the angular phase structure of photoelectron wave packets over a large range via laser-induced interference.

DOI: 10.1103/yjyp-kx19

I. INTRODUCTION

Interference is a universal phenomenon that arises from the superposition of two or more waves, creating spatial or temporal patterns depending on their relative phases. In quantum mechanics, interference is even more fundamental as it underlies the wave-particle duality that defines the quantum realm. In photoionization, electrons can reach the same final continuum state via distinct quantum pathways, each accumulating a phase determined by the ejected electron's interaction with the nucleus and the other electrons (often described by a potential), and the external electric field [1,2]. The interference among these pathways shapes both the spatial and temporal characteristics of the emitted electron wave packet (EWP). The result can then be observed as modulations in the photoelectron spectrum or angular distribution. The interference also reflects the underlying temporal dynamics of the electrons as they transition from bound states to the continuum [2–6]. The Wigner time delay, as introduced in scattering theory, describes how the interaction with a potential modifies the wave packet's motion relative to free propagation. This

delay (τ_d) is obtained from the spectral derivative of the phase shift $\phi(E)$ accumulated during the interaction: $\tau_d = \hbar \frac{\partial \phi(E)}{\partial E}$ [7]. For interfering partial waves, the concept was extended to an angular-dependent delay [8].

Techniques such as RABBITT (Reconstruction of Attosecond Beating By Interference of Two-photon Transitions), attosecond streaking, and the attoclock have been used to probe attosecond time delays in photoionization. RABBITT employs an attosecond pulse train and an infrared (IR) field to measure phase differences between quantum pathways contributing to the EWP [9,10]. Streaking employs an isolated attosecond extreme ultraviolet (XUV) pulse and an IR field to map ionization times onto electron momenta [3,11], while the attoclock technique uses elliptically polarized fields to infer ionization dynamics from angular distributions [12]. These techniques have since become essential for studying naturally occurring time delays in photoionization, from atoms to complex molecules and clusters. Additionally, the angular dependence of the photoelectron wave packet and its associated emission delay directly reflects the relative weights and phases of interfering partial waves, offering insights into the underlying photoionization dynamics that are not accessible through energy-resolved measurements alone [13–15]. Several factors can modify these partial-wave contributions. Fano's propensity rule predicts different angular-momentum compositions in the absorption and emission pathways [8], while resonances and antiresonances can strongly alter both their amplitudes and phases [16,17].

Here, we report how the angular profile of the RABBITT phase $\phi_R(\theta)$ [see Eq. (1)] can be actively controlled on a relatively large scale by varying the intensity of an external laser field. This phase, multiplied by $\hbar$ and divided by the corresponding energy difference between the interfering pathways, is commonly called the “angle-dependent

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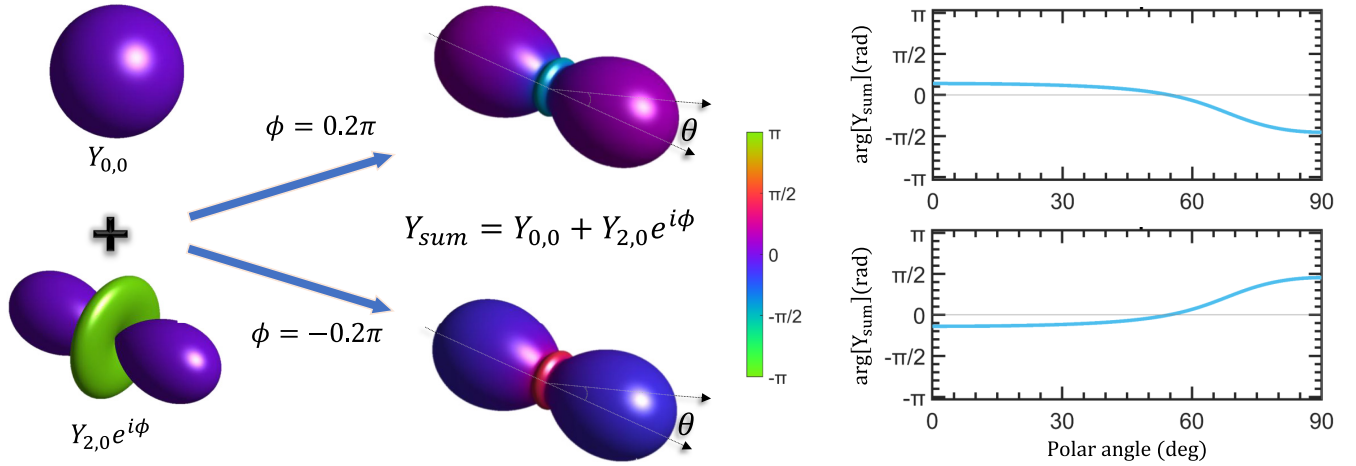


FIG. 1. Angular phase control by interference of two wave functions. Visualization of the magnitude and phase in the coherent superposition of s and d waves with relative phases $\phi = 0.2\pi$ and $\phi = -0.2\pi$. In the three-dimensional surface plots, the radius represents the magnitude, while the color encodes the phase as a function of the polar (emission) angle θ . The line plots show the phase of the combined spherical waves as a function of θ .

photoemission delay” [8]. It should, however, not be confused with a travel time of the photoelectrons to reach a detector.

We find that the angular phase of the threshold photoelectron sideband exhibits a rapid but continuous variation near a partial-wave node. As the infrared intensity is tuned across a specific value, the direction of this angular phase evolution reverses, causing the relative phase (set to 0 at small angles) to approach either positive or negative π at large emission angles, depending on the intensity. The key idea is illustrated in Fig. 1, employing the superposition of s - and d -wave angular-momentum states as an example. It is well known that such superpositions substantially modify the resulting spatial structure of the states. However, the phase of the resulting superposition state is also strongly dependent on the emission angle. In fact, as shown in the figure, a small change in the relative phase between the two waves from lagging to leading can reverse the direction of the rapid angular phase variation at angles beyond the angular node.

II. EXPERIMENTAL DETAILS

A detailed description of the experimental setup and measurement procedures is provided in Ref. [19]. We employed a RABBITT-like interferometric scheme using collinearly polarized XUV and IR pulses [18,19]. A commercial fiber-based laser produced 50 fs (full width at half maximum of intensity) ultrashort infrared pulses centered around 1030 nm (angular frequency ω) with a pulse energy of 1.2 mJ. The pulse was divided into two arms of a Mach-Zehnder-type interferometer by a holey mirror, which reflected approximately 85% of the beam into the pump arm and transmitted the rest into the probe arm. Within the pump arm, a barium borate crystal was employed to produce the second harmonic (515 nm) of the laser pulse with an efficiency of 25%–30%. The fundamental IR beam was subsequently filtered via a dichroic mirror, with the second harmonic focused onto an argon gas jet to produce an XUV pulse train through high-order harmonic generation (HHG). The driving 515 nm beam was first spatially filtered by the holey mirror. The XUV beam

and the remaining copropagating 515 nm light were then directed onto a 150-nm-thick aluminum filter, which transmitted the relevant XUV harmonics while blocking the residual 515 nm light. Meanwhile, the IR beam within the probe arm was directed through a retroreflector mounted on a piezoelectric translation stage, before it was spatially and temporally recombined with the XUV beam. Both the XUV and IR beams were focused inside a reaction microscope (ReMi) [20] onto a supersonic helium gas jet. The IR and XUV fields were linearly polarized parallel to each other and along the spectrometer axis of the ReMi. The ReMi allowed us to measure the electron momentum and hence the emission angle. The IR intensity was adjusted by utilizing an iris in the probe arm. RABBITT measurements were performed at three IR peak intensities: $I_1 \approx 5 \times 10^{11}$ W/cm², $I_2 \approx 7 \times 10^{11}$ W/cm², and $I_3 \approx 12 \times 10^{11}$ W/cm². The XUV-IR temporal delay was sampled at regular intervals of $T_0/60$, where $T_0 = 3.44$ fs is the period of the IR pulse. The delay was scanned over a range equivalent to one and a half times the optical cycle of the IR pulse. The XUV-IR beamline was actively stabilized [18,21] to achieve a stability of approximately 30 as over a data acquisition period of more than 10 h. The resulting spectral comb in the XUV beam consisted of odd harmonics of the 515 nm driving field, with adjacent harmonics separated by four times the fundamental frequency of the 1030 nm IR field. From the photoelectron spectra of helium, we observed harmonics up to H_{17} . The bandwidth of each harmonic was estimated to be around 180 meV.

III. DESCRIPTION OF THE PROCESS

Figure 2(a) shows the helium energy-level structure and the transition pathways. The two key frequencies in our experiment are the 9th and 11th harmonics, labeled as H_9 and H_{11} , respectively. The photon energy of H_9 (21.67 eV) alone is insufficient to ionize helium with its ionization potential of $I_p = 24.6$ eV. However, absorption of the harmonic H_{11} (26.49 eV) provides sufficient energy to ionize the atom, thus leading to the creation of a photoelectron in the

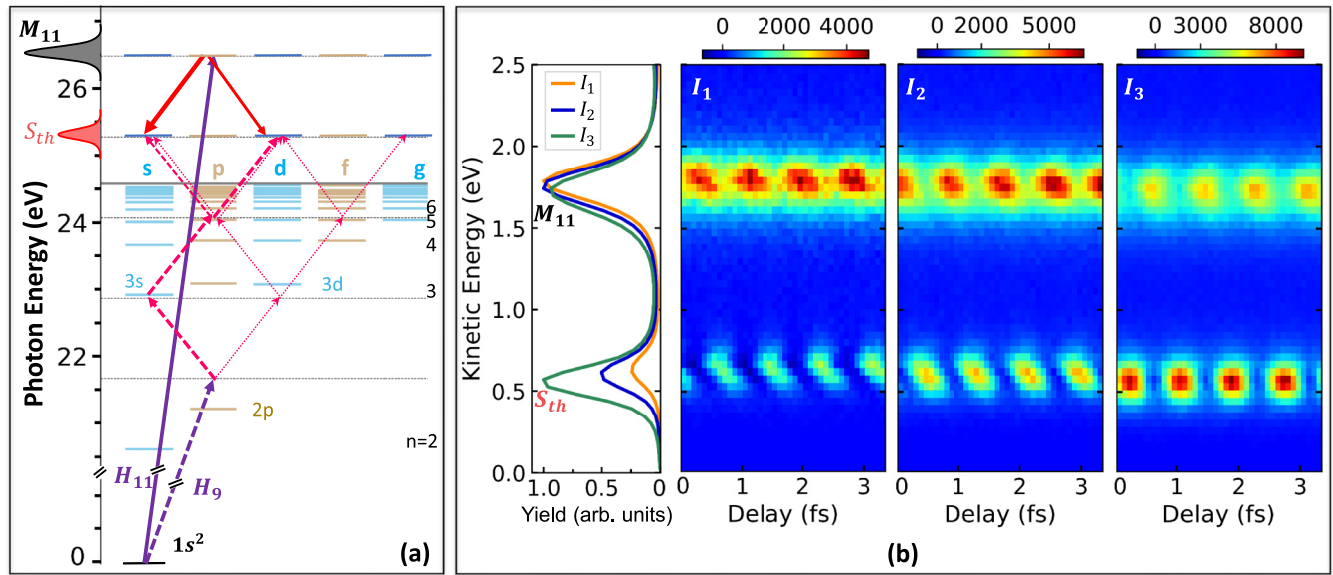


FIG. 2. Transition scheme and delay-dependent photoelectron spectra. (a) Pathways participating in the formation of the threshold sideband (S_{th}). Solid (dashed) lines represent the emission (absorption) pathways. The horizontal black dashed lines mark the energy positions corresponding to an integer number of IR photons. (b) Measured delay- and angle-integrated photoelectron spectra and RABBITT traces at three IR intensities: $I_1 \approx 5 \times 10^{11} \text{ W/cm}^2$ (orange), $I_2 \approx 7 \times 10^{11} \text{ W/cm}^2$ (blue), and $I_3 \approx 12 \times 10^{11} \text{ W/cm}^2$ (green).

continuum with p -wave angular-momentum character. This process forms the main peak labeled as M_{11} in the photoelectron spectrum shown in Fig. 2(a). The lowest sideband S_{th} (“threshold sideband”) in the photoelectron spectrum arises from two distinct quantum pathways that involve combinations of both XUV and IR photons to produce photoelectrons with the same final energy.

These pathways are classified into emission and absorption channels. The emission pathway primarily involves a two-photon process, wherein an electron absorbs the energy of an XUV photon from H_{11} and subsequently loses the energy corresponding to one IR photon to the infrared field, reaching the final photoelectron energy at S_{th} . This process generates partial waves with s and d angular momenta, as illustrated by the solid lines in Fig. 2(a). In contrast, the absorption pathway is more complex, involving the absorption of one H_9 XUV photon and three IR photons to get to S_{th} . Although H_9 on its own is far from any resonant bound state, the energy of H_9 plus one IR photon effectively coincides with the energy of the $(1s3s)^1S$ bound state at 22.92 eV. This intermediate resonance enhances the further absorption of a second IR photon, driving the valence electron from the $3s$ to a superposition of the $5p$ and $6p$ orbitals, which are near-resonant with $H_9 + 2$ IR photons. Finally, a third IR photon ionizes the atom by producing an EWP consisting of s and d partial waves. This four-photon process is illustrated by the thick dashed lines in Fig. 2(a). Additional absorption pathways are also possible. These involve near-resonant transitions with the real $(1s3d)^1D$ state or involve a virtual d orbital instead of the $3s$ orbital. From the d orbital, the active electron can absorb additional IR photons, transitioning to higher-lying bound orbitals such as p and f before reaching the continuum. These processes generate an EWP with s , d , and g partial waves in the continuum, as illustrated by the thin dotted lines in Fig. 2(a).

The photoelectron momentum distribution in the sideband is thus a coherent sum of all angular-momentum components, each with a distinct amplitude and phase created by a specific quantum pathway. By adjusting the temporal delay τ between the XUV and IR pulses, a phase shift of $4\omega\tau$ is introduced between the absorption and emission pathways [22,23]. This delay-dependent phase shift modulates the S_{th} signal, exhibiting constructive or destructive interference between the two pathways.

IV. RESULTS AND DISCUSSION

We measured the photoelectron angular distribution as a function of pulse delay for three IR laser intensities: $I_1 \approx 5 \times 10^{11} \text{ W/cm}^2$, $I_2 \approx 7 \times 10^{11} \text{ W/cm}^2$, and $I_3 \approx 12 \times 10^{11} \text{ W/cm}^2$, while keeping the XUV field constant across all measurements. The left panel of Fig. 2(b) exhibits the measured delay-averaged photoelectron spectra for the three IR intensities, integrated over all emission angles, with each spectrum normalized to its maximum value. The corresponding RABBITT traces are shown as color plots in Fig. 2(b) after removing the delay-independent background signal.

For each intensity, the photoelectrons emitted at different angles relative to the linear polarization direction of the fields were collected over angular windows of 10° , incremented by steps of 5° . The signal in each angular window was fitted according to the following expression to extract the interference phase $\phi_R(\theta)$ as a function of the emission angle:

$$S_{th}(\tau, \theta) = A(\theta)[1 + B(\theta) \cos(4\omega\tau - \phi_R(\theta))]. \quad (1)$$

Here, $A(\theta)$ is the magnitude, while $B(\theta)$ represents a relative modulation amplitude depth.

Figure 3(a) displays the measured interference phases as a function of emission angle for the three IR intensities. Since the absolute phase cannot be determined experimentally, the

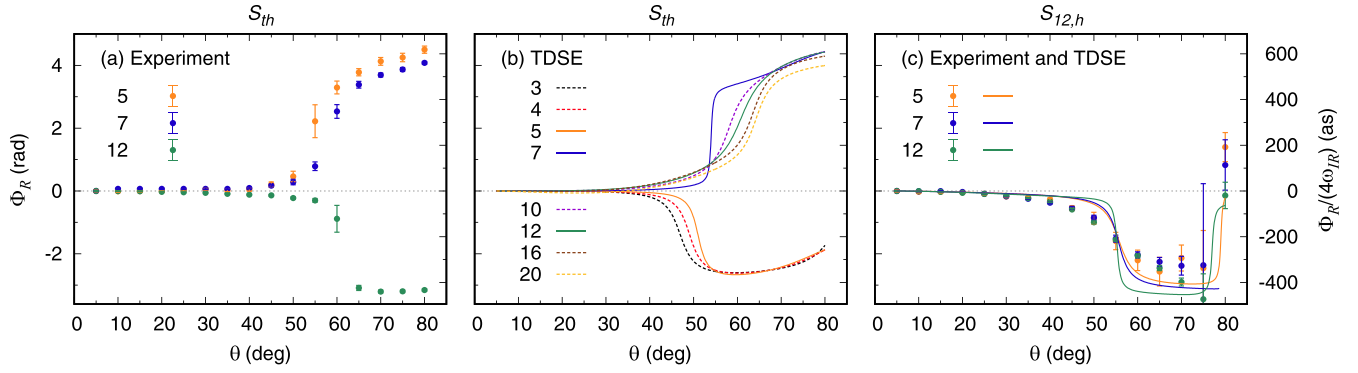


FIG. 3. Angle-dependent phase ϕ_R (left scale) and corresponding delay $\phi_R/(4\omega)$ (right scale) retrieved from the oscillation in S_{th} . Panels (a) and (b) display the measurements and results of the TDSE simulation, respectively, for the threshold sideband, while panel (c) shows results for the highest sideband of the $S_{12,h}$ group. The legend gives the IR intensities in multiples of 10^{11} W/cm^2 . To show the gradual change of the phase and the flip in the angular interval of 40° – 70° , theoretical predictions for additional intensities are shown by dashed lines in panel (b).

phase at the first measured angle (5°) was set to zero for all cases.

For the lower intensities, I_1 and I_2 , the extracted phase initially stays constant and then gradually increases as the angle approaches 55° before undergoing a steady but rapid increase of approximately π . In contrast, at the highest intensity, I_3 , the phase moves in the opposite direction, initially showing a slight decrease before undergoing a sharp π transition around 55° . This abrupt phase transition suggests that a significant d -wave contribution is present among the interfering paths for S_{th} , as d waves exhibit a node (a zero in their angular distribution) near 55° .

In multiphoton single ionization, near-resonant states can act as transient stepping stones, enhancing or suppressing certain transitions depending on the energy detuning. The phase and amplitude of the resulting photoelectron wave packet are therefore highly sensitive to the exact detuning, which in turn can be dynamically modified by the IR-induced Stark effect [16,17,24–28]. The angular dependence of the RABBITT phase reflects the coherent superposition of the outgoing partial waves, mainly s , d , and possibly also g waves, generated in both the absorption and emission pathways. Only for the highest intensity studied here, the g wave (only in the absorption channel) produces a small contribution (see Fig. 4 for an example). The absorption pathway is especially sensitive to the IR intensity, as it couples multiple intermediate states. As shown in Fig. 2(a), several of these states are near-resonant with the multiphoton energies involved,

and their energies shift due to the Stark effect when the IR intensity is varied. This changes the detuning dynamically and modifies the composition of the absorption pathways as well as the phase relationship between the angular-momentum channels contributing to the photoelectron wave packet. These shifts alter the final photoelectron phase in specific angular-momentum channels obtained along the different paths and thereby changes the phase and thus the delay extracted from the sideband oscillations.

In our experiment, we observe that the RABBITT phase increases with emission angle at the two lower IR intensities, while it decreases at the highest applied intensity. This reversal in the angular variation of the phase originates from a change in the relative phase between the s - and d -wave components of the absorption pathway (cf. Fig. 1), here driven by intensity-dependent detuning of the coupled resonances. The observed flip in the angular dependence provides evidence for a resonance-mediated control mechanism over the phase structure of the photoelectron wave packet.

To provide a theoretical perspective, the results of a single-active-electron (SAE) solution of the time-dependent Schrödinger equation (TDSE) for the interference phase as a function of emission angle for a range of IR intensities are shown in Fig. 3(b). We used a new version of our SAE code in which the field-free potential depends on the angular momentum of the active electron. This ℓ -dependent version of the SAE yields, in particular, much improved energy levels and electric dipole matrix elements compared with those obtained using other potentials previously used in the literature. More details can be found in Ref. [29]. The calculation qualitatively reproduces the observed behavior, showing a nearly constant phase up to about 40° followed by a slow increase or decrease, culminating in a similar sharp change by approximately $\pm\pi$ near 55° , exactly as in the experiment. While the phases above $\approx 70^\circ$ could be reconstructed by adding or subtracting 2π and hence might suggest an ambiguity without physical meaning, it is important to note that this is *not possible* in the 40° – 70° range. There is a clear trend regarding the direction (up or down), in which the phase, and hence the delay, will go. The data shown in Figs. 3(b) and 3(c) were obtained with a step size of 0.1° and a window of 0.3° . However, the analysis of the theoretical predictions was also performed for several

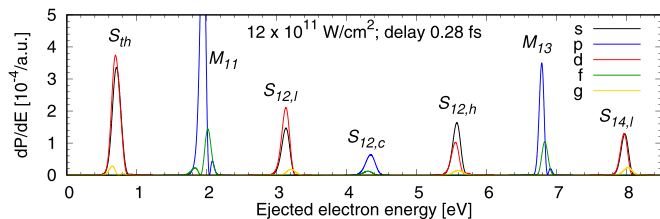


FIG. 4. Predicted partial-wave contributions to the various peaks in the ejected-electron spectrum for a peak intensity of $12 \times 10^{11} \text{ W/cm}^2$ and a delay of 0.28 fs. As expected, the sidebands are dominated by contributions from s and d waves, while the main peaks contain p and f waves.

angular steps and windows, with hardly any change in the quantitative and certainly not in the qualitative results. Therefore, we are confident regarding the way the points need to be connected when passing the d -wave node near 55° . This smooth evolution through the angular node can be understood from the interference between the contributing partial waves [8]. If these contributions were exactly in or out of phase, the complex interference amplitude would pass through zero at the node, producing an abrupt π phase jump. A finite relative phase smooths this jump as the relative weights of the contributions vary with emission angle. Our results show that the direction of this transition can be controlled via the intensity of the driving field.

Nevertheless, an important discrepancy emerges when comparing theory and experiment: In the experiment, the RABBITT phase increases as a function of emission angle at lower IR intensities, while it decreases at higher intensities. The theoretical model predicts the opposite trend, where the phase near the angular node decreases with increasing angle at low intensities and increases at higher intensities. We checked for a consistent sign convention and currently do not have an explanation for this discrepancy. We confirm numerical convergence by employing a partial-wave decomposition of the wave function with $\ell_{\max} = 10$, together with a nonuniform radial grid with spacing $dr \leq 0.1$ a.u., and a second-order propagation scheme as described in Ref. [30], using a time step $dt = 0.001$ a.u. Also, as shown in earlier work [19,31] and again demonstrated in Fig. 3(c), the new code reproduces the angular dependence of the RABBITT phase very well for the higher-energy sidebands, e.g., for the corresponding $S_{12,h}$ of the next group.

Most likely, the predictions for S_{th} are particularly sensitive to the details of the model. Ideally, one should carry out all-electron calculations and investigate the convergence with the number of states in a close-coupling expansion. Initial tests using the R -matrix with time-dependence approach [32], however, showed that the computational demand is prohibitive for our currently available resources.

V. CONCLUSION AND OUTLOOK

In conclusion, we demonstrated the possible manipulation of interferometric phases (delays) measured in photoemission setups by strong laser coupling of atomic states. When studying a threshold sideband in an angle-resolved RABBITT setup, we observed a rapid angular phase variation near a partial-wave node, whose direction reversed with increasing infrared intensity. A theoretical calculation within the improved SAE approximation qualitatively supported our experimental findings.

While the mechanism was demonstrated here for the specific application of phase control in laser-atom interactions, we emphasize that the presented interference mechanism is general and might also be applied to more complex quantum systems, including molecules, clusters, nanostructures, or solid-state quantum dots. Enabling significant changes in the angle-dependent phase of emitted electron wave packets represents a powerful tool in attosecond science and ultrafast spectroscopy. In the future, we envision applications, for example, in attosecond electron microscopy [33] and pulse-shaped electron sources for the exploration and steering of transient topological phases of matter and time-dependent quantum materials.

ACKNOWLEDGMENTS

The experimental part of this joint study was supported by the DFG-QUTIF program under Project Ref. No. 396 HA 8399/2-1 and IMPRS-QD, while the theoretical work was funded by NSERC (A.T.B.) and the NSF (A.T.B., J.C.d.V., and K.B.) under Grants No. PHY-2110023 and No. PHY-2408484.

DATA AVAILABILITY

There are no publicly available research data or software supporting this manuscript. Requests for further information or data should be sent to the authors.

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