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Attosecond probing and control of charge migration in carbon-chain molecule

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In quantum mechanics, when an electron is quickly ripped off from a molecule, a superposition of new eigenstates of the cation creates an electron wave packet that governs the charge flow inside which has been called charge migration (CM). Experimentally, to extract such dynamics at its natural (attoseconds) timescale is quite difficult. Here, we report the first such experiment in a linear carbon-chain molecule, butadiyne (C$_4$H$_2$), via high harmonic spectroscopy (HHS). By employing advanced theoretical and computational tools, we showed that the wave packet and the CM of a single molecule is reconstructed from the harmonic spectra for each fixed-in-space angle of the molecule. For this one-dimensional molecule, we calculate the center of charge $\langle x \rangle (t)$ to obtain $v_{cm}$, to quantify the migration speed and how it depends on the orientation angle. The findings also uncover how the electron dynamics at the first few-tens to hundreds attoseconds depends on molecular structure. The method can be extended to other molecules where HHS technique can be employed.

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I. INTRODUCTION

Ultrafast electron dynamics are of crucial importance for understanding and steering complex chemical and biological reactions. When subjected to an intense laser field, a pure electron-driven dynamics can be initiated by strong-field ionization of the molecule. Theoretically, this dynamics manifests itself as a migration of the initially created hole density through the molecular backbone and was dubbed charge migration (CM) since 1999 [1]. In the last decade, CM has been extensively studied theoretically using time-dependent quantum chemistry packages [2–12]. Topics of interest include the dependence of CM on molecular species and structures [9–12], as well as the role of nuclear dynamics and decoherence in CM [13–15]. In these numerical studies, calculations were made to follow the time evolution of the density of the wave packet under the field-free condition. However, these results cannot be tested against experiments directly. In a real experiment, to know the field-free wave packet (or CM), a probe pulse is needed, which will inevitably affect the evolution of the wave packet. Thus, to retrieve the field-free CM from the measurement is a monumental task and rarely possible.

Since it’s a fascinating goal of ultrafast science to follow electron and/or nuclear dynamics, in spite of the difficulty, it is still desirable to perform pump-probe experiments to extract some partial information. For CM in molecules, it occurs at few-ten attoseconds timescale. Then one would like to have pump and probe pulses shorter than a few attoseconds. Such pulses, however, would have bandwidth of a few-ten electron volts and the created wave packet will be very complicated for studying valence electrons in a molecule. Additionally, the currently available attosecond pulses are still not intense enough for such applications. To date, most electron dynamics studies have used attosecond pump and intense IR probe pulses, or vice versa [16–20]. Among the experiments that studied electron dynamics or CM, most of them are unable to obtain electron wave packet or the dynamics of CM for individual molecules directly.

From strong field physics, it has long been suggested that one can use high-harmonic spectroscopy (HHS) to study attosecond electron dynamics using intense infrared lasers (see the reviews [21, 22]). The principle of HHS is based on the three-step process of high-order harmonic generation (HHG) [23, 24], where an electron is first ionized near the peak of the laser field, the released electron then drifts in the laser field and some portion will be driven
back to recombine with the ion and emits harmonic radiation. At the time of recombination, the information of the electron wave packet of the cation is encoded in both the amplitude and phase of each generated harmonic. The intrinsic time-to-frequency mapping underlying HHG allows a temporal measurement of the electron dynamics with a resolution of tens to a hundred attoseconds [25–30]. Up to now, HHS has been well used for ultrafast molecular detection with both attosecond temporal [25–30] and ångström spatial resolutions [31–33].

While the HHS principle is easy to understand, to implement it for extracting accurate CM for each single molecule is not so straightforward since the experimentally measured harmonic spectra are due to the coherent summation of the individual radiation weighted by the angular distribution of the molecules. In the first experiment of using HHS to extract the CM by Kraus et al. [25], the measured angularly integrated harmonic spectra were directly taken as the single-molecule results. In doing so, large errors are introduced into the reconstructed dynamics. In our recent paper [34], we developed a machine learning (LM) algorithm to retrieve the single-molecule harmonics from the angularly integrated harmonic spectra, opening an avenue for accurate measurement of CM on the single-molecule level.

In spite of that, the sensitive dependence of CM on molecular orbitals and orientations still makes the CM dynamics complex and difficult to trace. There are still some open questions about CM that remain unclear, for example, how fast the charge migrates in molecules? Very recently, by creating a localized wave packet, the hole has been predicted to migrate along the molecular backbone in particle-like manner at a speed of a few Å/fs [10]. However, in most reactions involving valence electrons, delocalized wave packets are created. In addition, the electron wave packet cannot be measured directly. They have to be reconstructed from other experimental data. In this work, we focus on a linear carbon chain molecule, butadiyne (C₄H₂), where the movement of the hole is expected to be along the carbon backbone [9]. By using a ML-based two-color scheme, we successfully retrieved the hole wave packet of the cation in C₄H₂ from the harmonic spectra. From the center of charge at each time the CM speed was measured for the first time. Our approach allows us to extract how migration speed depends on the alignment angle of the molecule whether charge density is localized [10–12] or delocalized.
FIG. 1: Probing CM in C$_4$H$_2$ with HHS. (a) Schematic layout of the multichannel HHG in C$_4$H$_2$ molecule that involves the ground $\tilde{X}^2\Pi_g$ ($\tilde{X}$) and first excited state $\tilde{A}^2\Pi_u$ ($\tilde{A}$) of the molecular ion. In C$_4$H$_2$, there are four channels labeled as XX, AA, XA, and AX, respectively, contributing to HHG. Here the first and second letters label the ionic state after ionization and before recombination, respectively. (b)-(c) Experimentally retrieved population amplitude ($|P_X|$) of the $\tilde{X}$ state (b) and the relative phase ($\Delta \phi_{XA}$) between the wavefunctions of $\tilde{X}$ and $\tilde{A}$ states (c) for the parallel (0°, green squares) and perpendicular (90°, red circles) alignment of the C$_4$H$_2$ molecule. The solid lines show the TDDFT results for comparison. Error bars in (b)-(c) represent the standard deviations of the reconstructions, which are estimated from the experimental errors of the HHG signals with the bootstrap method.

II. RESULTS

Figure 1 sketches the multichannel mechanism of HHG from the C$_4$H$_2$ molecule. Exposed to an intense external laser field, the ground $\tilde{X}^2\Pi_g$ ($\tilde{X}$) and the first excited $\tilde{A}^2\Pi_u$ ($\tilde{A}$) states of the C$_4$H$_2^+$ ion, which correspond to the removal of an electron from the highest- and second highest-occupied molecular orbitals HOMO and HOMO-1, respectively, can be simultaneously populated by the strong-field ionization due to their close vertical ionization potentials ($\Delta E=2.4$ eV). The coherent superposition of multiple electronic states of
the molecular ion creates a many-electron wave packet, which evolves in time, leading to the time-dependent variation of the charge density, i.e., the CM in the molecular ion. In HHG process, these ionic states act as the intermediate that connects the same initial and final state of the system. The presence of different ionic states between the ionization and recombination provides different channels for the harmonic radiation [35–37]. For \( \text{C}_4\text{H}_2 \), it includes two diagonal channels XX, AA, and two off-diagonal channels XA, AX, as shown in Fig. 1(a). The off-diagonal channels reflect the laser-induced coupling between the two ionic states during the HHG process. Eventual harmonic radiation is a coherent superposition of each channel [35–37], i.e.,

\[
D(\omega, \theta) = \sum_{ij=X,A} C_{ij}(\theta) d_{ij}(\omega, \theta),
\]

where \( D(\omega, \theta) \) is total dipole moment for HHG, \( d_{ij}(\omega, \theta) \) is the dipole related to each channel, which in our reconstruction is calculated by the quantitative rescattering (QRS) theory [38–41]. \( C_{ij}(\theta) \) is a complex mixing coefficient related to the laser-induced transitions between the two ionic states during the HHG process. Eq. (1) suggests that the multi-electron dynamics is naturally recorded in the harmonic spectrum and can be deciphered by disentangling the multichannel contributions from the total dipole moment.

In our experiment, the CM dynamics in \( \text{C}_4\text{H}_2^+ \) was reconstructed by using a two-color driving scheme in combination with the ML algorithm in [34]. Briefly, the ML algorithm was used to disentangle the coherent angular average of high harmonics originated from the imperfect molecular alignment in experiment, since this angular average could compromise the measurement from the real single-molecule response [42–44]. This is especially important for a low degree of molecular alignment in our experiment which has \( \langle \cos^2 \theta \rangle \approx 0.5 \) (see Supplementary Material). A two-color driving scheme was used to generate odd and even harmonics to replenish the data set required for the decomposition of the multichannel contributions to each harmonic by varying the two-color relative phase (see Methods and Supplementary Material for more details). By applying the ML-based reconstruction procedure to the two-color measurements, we are able to obtain the complex mixing coefficients of the multiple orbitals of the molecule, for each fixed-in-space angle, at the instant of recombination that each harmonic order occurs. To obtain a one-to-one mapping between the instant time and harmonic order, we ensure that phase matching in the experiment is achieved such that only short-trajectory electron contributes to each harmonic.
FIG. 2: Reconstruction of CM in C₄H₂⁺ for perpendicular alignment. (a) Snapshots of the reconstructed hole densities for the alignment angle of 90°. (b) Time-dependent hole densities along the molecular backbone obtained by integration over the y direction. For clarity, the molecular backbone has been plotted on the top of panel (b). (c) Time-dependent COC position ⟨x⟩(t) (dashed line with circles) retrieved from the hole densities in (b). Here the dash-dotted line is a linear fitting of ⟨x⟩(t) to evaluate the CM speed, and the green squares represent the SD of the x-coordinate. (d) Flux of charge density crossing the x=0 plane. Negative value means CM from −x side to +x side.

Figure 1(b) shows the time-dependent population amplitudes of the ground state ̃X (|PX⟩) of C₄H₂⁺ ion extracted from the experimental data of H11 to H17 (including both even and odd orders) for the molecules aligned at 0° (squares) and at 90° (circles) to the polarization of the driving field, respectively. Figure 1(c) displays the corresponding relative phases between the population coefficients of the ̃X and ̃A states (Δφₓₐ). In our reconstruction,
FIG. 3: Reconstruction of CM in C$_4$H$_2^+$ for parallel alignment. (a)-(d) are the same as Fig. 2(a)-(d), but for the case of parallel alignment of the C$_4$H$_2$ molecule.

The uncertainties of the retrieved parameters [error bars in Figure 1(b)-(c)] were estimated from the experimental errors of the HHG signals with the bootstrap method [45]. For comparison, we have simulated the above parameters based on the time-dependent density functional theory (TDDFT) with the OCTOPUS package [46–48]. As shown in Figs. 1(b)-(c), the simulation results agree well with the experimental reconstructions.

With the retrieved parameters in Figs. 1(b)-(c), we can reconstruct the hole wave packet of the C$_4$H$_2^+$ ion. We first examine the results for the 90° alignment of the molecules. In this case, by symmetry the two states are not coupled by the laser, such that populations of the $\tilde{X}$ and $\tilde{A}$ states do not change with time, see the red line in Fig. 1(b). In this situation, the reconstructed charge density does change with time, as can be seen in Fig. 2(a), which displays the reconstructed hole densities in C$_4$H$_2^+$ ion at the recombination times of H12.
to H17. These figures also show substantial hole migration along the molecular backbone (x-axis). Moreover, the hole densities are always symmetric about the $y = 0$ plane because of the symmetries of the HOMO and HOMO-1 orbitals of C$_4$H$_2$. Note that these results are similar to the field-free CM defined by Cederbaum et al. [1].

To provide simpler information from the charge density distribution, we extracted the reduced hole density $\rho_x(t)$ by integrating the hole density over the $y$ direction. As shown in Fig. 2(b), the hole created by ionization is initially localized at the left C$_1$≡C$_2$ triple bond, then spreads over the molecule, and finally distributed around the right C$_3$≡C$_4$ triple bond after about 500 as. To quantify the CM dynamics, we further define the expectation value of the abscissa $x$ (carbon chain) as the center of charge (COC) position. As shown in Fig. 2(c), the extracted COC position (blue circles) increases almost linearly with time, indicating a unidirectional migration of the hole from the $-x$ side to the $+x$ side. By a linear fitting of $\langle x \rangle(t)$ [the dash-dotted line in Fig. 2(c)], we can then evaluate the CM speed from the slope, which is about 2.94 Å/fs. This result is close to the theoretical predictions in halogenated hydrocarbon chains [10]. We have also calculated the standard deviation (SD) of the abscissa $x$ to quantify the degree of localization of the hole during the evolution. As shown in Fig. 2(c), the SD of $x$ (green squares) increases in the first 200 as and turns to decrease in the last 300 as, which reflects a delocalization-to-localization behavior of the created hole wave packet. The decrease is due to the wave packet is beginning to bounce back from the right C≡C bond. At longer time, the hole has been predicted to travel back and forth between the C≡C triple bonds on both sides [9, 10]. Such a periodic behavior cannot be fully visualized from the reconstructions in Figs. 2(b)-(c) due to the limited temporal window in our measurement. With the retrieved CM speed, we can estimate the CM-mode period or frequency that is critical for describing the periodic dynamics, by $T_{cm}=2\pi/\omega_{cm}=2d_{cm}/v_{cm}$, where $d_{cm}=4.88$ a.u. is the distance between the centers of the two C≡C triple bonds in C$_4$H$_2$ which is approximately the CM distance in C$_4$H$_2^+$. For the quasi-field-free CM, the CM-mode period and frequency are estimated to be 1.76 fs and 2.35 eV, which are close to the results 1.72 fs and 2.4 eV defined by the energy difference between the two ionic states of C$_4$H$_2^+$. Rigorously, single-hole dynamics is to be obtained with quantum mechanics. From the retrieved complex wave packet, real values of probability density $\rho_e$ and current density $\mathbf{J}$ can be obtained and they have been found to satisfy the equation of continuity, $\frac{\partial \rho_e}{\partial t} + \nabla \cdot \mathbf{J} = 0$. 
FIG. 4: TDDFT simulations of the CM dynamics in $\text{C}_4\text{H}_2^+$. (a) TDDFT calculations of the reduced hole density $\rho_x(t)$ in the experimental temporal range for the $90^\circ$ alignment of the $\text{C}_4\text{H}_2$ molecule. (b) Same as (a), but for the parallel alignment (the alignment angle of $0^\circ$) of the $\text{C}_4\text{H}_2$ molecule.

In Fig. 2(d), we show the time-dependent total flux crossing the $x = 0$ plane calculated from the retrieved complex hole wave packet. One can see that the total flux is always negative over the 500 as duration, indicating continuous migration of the charge from $-x$ side to the $+x$ side. A near-zero flux near 1.5 fs also reflects that the hole has moved to the right terminal C≡C bond. These results are consistent with the analysis in Fig. 2(c). On the other hand, the flux changes rapidly with time, indicating that the constant speed in Fig. 2(c) is valid only for the speed of COC, which in turn would imply that $v_{cm}$ would be the same also for a localized wave packet if other conditions stay the same. From the other viewpoint, this would imply that $v_{cm}$ itself, or the hole density alone, would not provide adequate information on electron dynamics except from the retrieved wave packet. Like electron and hole in solids, $v_{cm}$ could provide a first order interpretation on carrier dynamics even though the carriers are not localized. This is how the speed of CM is to be understood. It is a measure of how the average position moves.

Proceeding further, next we consider in Fig. 3 the reconstructed results for the parallel
alignment of the molecule. In this case, the ground \( \tilde{X} \) and first excited \( \tilde{A} \) states of \( \text{C}_4\text{H}_2^+ \) are strongly coupled by the laser field due to the large transition dipole moment between these two states along this direction. The resulting CM is controlled by the laser field. Comparing Fig. 2 with Fig. 3, we can see that laser coupling would drive the hole density to +x side faster [Fig. 3(a)], as well as for the reduced hole density [Fig. 3(b)]. The COC position also rises faster from which the CM speed has increased to 4.5 Å/fs, even though the SD remains roughly the same, except the feature changes at earlier time [Fig. 3(c)]. Note that, here the CM speed is obtained by fitting the COC position \( \langle x \rangle (t) \) before 1.34 fs, where \( \langle x \rangle (t) \) increases almost linearly. Afterwards, the hole reaches and localizes at right terminal \( \text{C} \equiv \text{C} \) bond. In Fig. 3(d) the flux also shows faster changes with time, indicating that laser coupling has substantially changed the hole density distribution.

To evaluate the reconstructed hole dynamics, we have further performed TDDFT simulations. Figure 4(a) shows the time-dependent hole density \( \rho_x(t) \) simulated in the same temporal range as in the experiment (i.e., 1.04–1.51 fs) for 90° alignment of the molecule. One can see that the TDDFT results in Fig. 4(a) are in good agreement with the experimental reconstructions presented in Fig. 2(b). The laser-controlled CM in the parallel aligned molecules has also been simulated. As shown in Fig. 4(b), under the influence of the laser field, the simulated CM dynamics is far different from the field-free result in Fig. 4(a). By
comparing Fig. 3(b) with Fig. 4(b), the experimental reconstructions in this case also agree quite well with the corresponding TDDFT simulations.

Using our retrieval method, we are able to obtain CM for all alignment angles at the same time. The CMs for other alignment angles of the fixed-in-space molecules have also been reconstructed (see Supplementary Material). The CM speeds retrieved as a function of the alignment angles are shown in Fig. 5 (squares). For comparison, the TDDFT results are also presented (solid line). The CM is demonstrated to depend sensitively on the alignment angles. Moreover, the speed of the laser-controlled CM (nonperpendicular alignments) is faster than the field-free result at $90^\circ$. This is similar to the well-known fact that the field-free two-level oscillation is slower than the Rabi oscillation when the two levels are driven by an intense laser pulse. This alignment dependence, on the one hand, proves that one should not obtain the single-molecule dynamics directly from the experiment without angular deconvolution. On the other hand, it also provides a potential way to control the CM speed and even ultimately to manipulate the rate of a chemical reaction.

## III. DISCUSSION

In summary, CM in a carbon-chain molecule $C_4H_2$ was measured by using a ML-based two-color HHS method. The CM dynamics is reconstructed at the most fundamental level for each single fixed-in-space molecule. By analyzing the time-dependent hole densities, the speed of CM was extracted for the first time for each alignment angle of the molecule. The retrieval results demonstrate that laser-controlled CM is much faster than the field-free one. These results are consistent with the TDDFT simulations. Our result provides a simple and intuitive way to understand and quantify the complex CM in molecules.

Looking ahead, the method presented here may be extended to driving lasers with much longer wavelengths, in which the whole periodic hole migration may be directly observed from the measurement. On the other hand, oriented halogen-functionalized carbon-chain molecules are also ideal candidates for the CM studies in experiment, where the influence of the hole localization on the CM speed may be examined. Moreover, the halogen functionalization can provide other degree of freedom to control the CM.
IV. MATERIALS AND METHODS

1. Experimental method

We conducted our experiment using a commercial Ti:sapphire laser system (Astrella-USP-1K, Coherent, Inc.), which provides 35-fs, 800-nm laser pulses at a repetition rate of 1 kHz. The laser output was divided into two pulses: an alignment pulse and a probe pulse. The alignment pulse had moderate intensity and was utilized to induce nonadiabatic alignment of molecules along its polarization. The probe pulse was intense and was used to interact with the aligned molecules to generate high-order harmonics. The alignment and probe pulses had parallel polarizations and were focused onto a supersonic gas jet by a spherical mirror ($f=250$ mm). A motorized delay line was installed in the alignment pulse arm to adjust the time delay between the two pulses. The gas jet was positioned 2 mm after the laser focus to ensure good phase matching for short-trajectory harmonics, and the backing pressure was kept at 0.2 bar. The high harmonics produced were detected by a homemade flat-field soft X-ray spectrometer [49]. More details about the experiment can be seen in Supplementary Material.

In this work, we first conducted HHG experiment employing a single-color 800-nm driving laser. We measured harmonic signals at different time delays, which enabled us to determine the distribution of molecular alignment using the technique outlined in [50]. To further extract the CM dynamics, it necessitates multiple experimental observables to decompose the multichannel contributions in HHG. Whereas, in a single-color laser field, only one set of $D(\omega, \theta)$ can be obtained for each harmonic order, which is inadequate for the decomposition. To overcome this limitation, we employed a two-color driving scheme, where the laser field is synthesized by an intense 800-nm fundamental field and a parallel second-harmonic (SH) field. Note that the SH field is weak in our experiment, which barely alters the electron dynamics of the molecular ion, but could affect the harmonic radiation of each channel (see Supplementary Material). In the two-color experiment, we measured time-dependent harmonic signals at various relative phases of the two-color laser field, providing a two-dimensional dataset for decomposing the multiple orbital contributions in the HHG process.

2. Reconstruction method
In our experiment, HHG from the partially aligned molecular ensemble can be expressed as a coherent superposition of the individual radiation weighted by the angular distribution of the molecules \([42–44]\), i.e.,

\[
S(\omega, \tau) = \left| \int_0^\pi D(\omega, \theta)\rho(\theta, \tau)\sin\theta d\theta \right|^2,
\]

(2)

where \(\theta\) is the alignment angle of the molecule, \(\tau\) is the time delay between the alignment and driving pulses, \(D(\omega, \theta)\) is single-molecule harmonic dipole moment, and \(\rho(\theta, \tau)\) is the angular distribution of molecules at delay \(\tau\). Since the coherent average over molecular alignment distribution could severely deviate the measurement from the real single molecule response, especially for a low degree of molecular alignment, it is critical to decode the fixed-in-space single-molecule dipole moment \(D(\omega, \theta)\) from the measured harmonic signals. In this work, the complex deconvolution was resolved with recently developed ML algorithm \([34]\), of which the accuracy and robustness have been verified in both theory and experiment.

By implementing the ML algorithm onto the two-color measurement, we obtained the single-molecule dipole moment \(D(\omega, \theta)\) for each relative phase \(\varphi\) of the two-color field. Since the SH field used in our two-color experiment is weak enough and hardly alters the electron dynamics in the molecular ion, that means the complex mixing coefficients \(C_{ij}(\theta)\) can be assumed to be independent of the relative phase \(\varphi\), we thus can get

\[
D(\omega, \theta; \varphi) = \sum_{i,j=X,A} C_{ij}(\theta)d_{ij}(\omega, \theta; \varphi).
\]

(3)

The complex mixing coefficients \(C_{ij}(\theta)\) then can be obtained by solving Eq. (3) with the genetic algorithm. By repeating the above procedure for different harmonic orders, we can ultimately construct the hole dynamics in the ionized molecular ion according to time-frequency mapping underlying HHG.

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Author contributions
P.F.L. and P.X.L. conceived this research. L.X.H. and Y.Q.H. performed the experiments and the simulations. S.Q.S. developed the reconstruction algorithm. X.S.Z, E.G., and A.T.L helped with the TDDFT and QRS calculations. C.D.L., L.X.H., and P.F.L. analyzed the data and wrote the manuscript.

Data availability
All the data that support the findings of this study are available from the corresponding author upon reasonable request.

Conflict of interest
The authors declare no conflicts of interest.

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