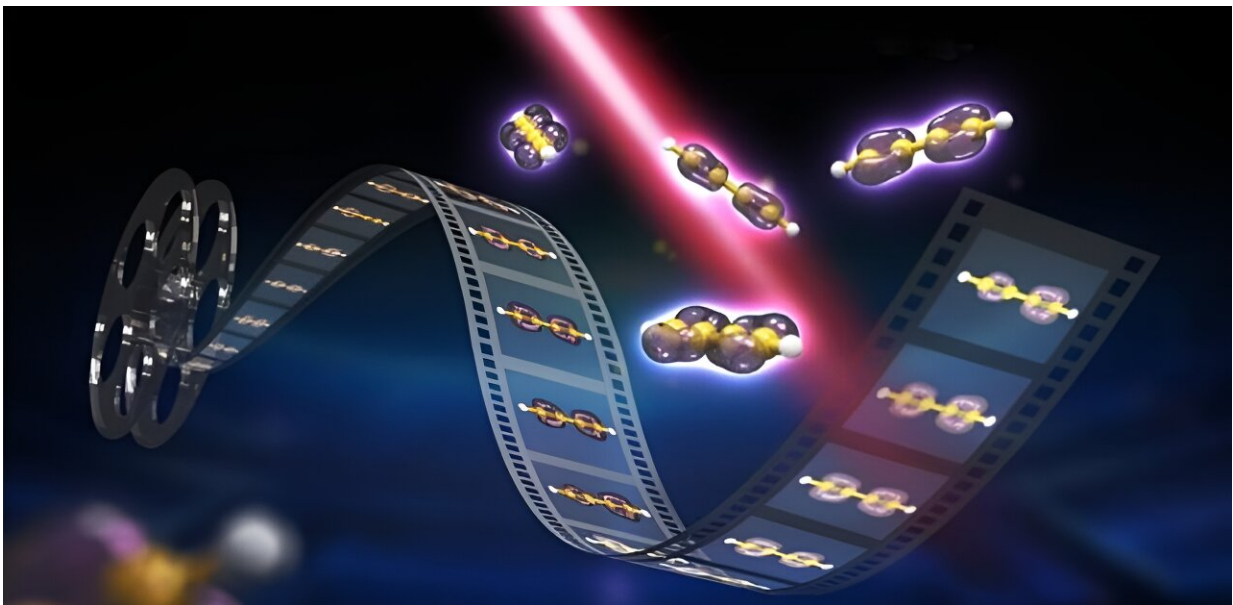


New experimental research measures the speed of molecular charge migration for the first time

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Charge migration in a linear carbon-chain molecule, captured by high harmonic spectroscopy (artistic rendering). Credit: Lixin He and Pengfei Lan.

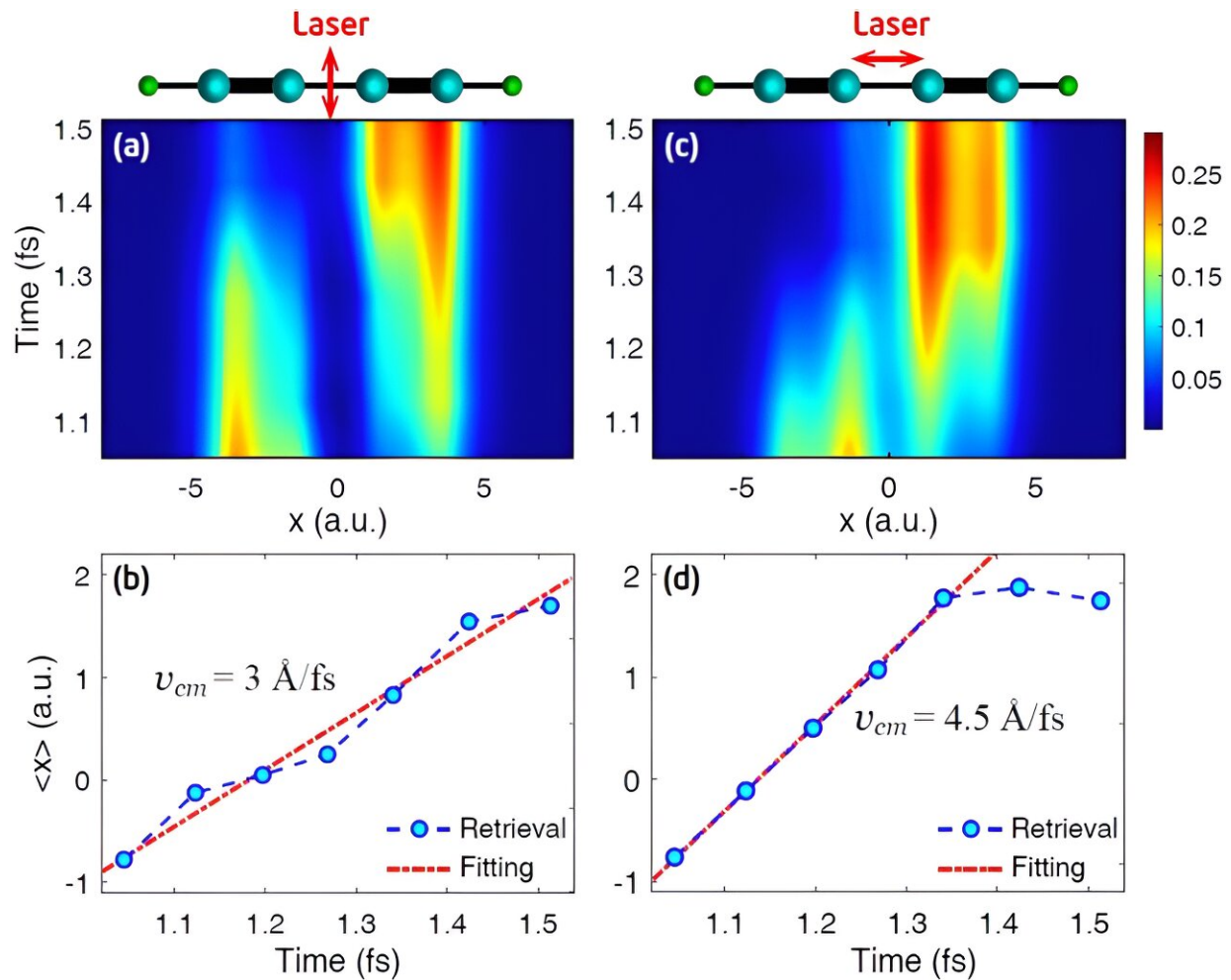
To discover how light interacts with molecules, the first step is to follow electron dynamics, which evolve at the attosecond timescale. The dynamics of this first step have been called charge migration (CM). CM plays a fundamental role in chemical reactions and biological functions associated with light–matter interaction. For years, visualizing CM at the

natural timescale of electrons has been a formidable challenge in ultrafast science due to the ultrafine spatial (angstrom) and ultrafast temporal (attosecond) resolution required.

Experimentally, the sensitive dependence of CM on [molecular orbitals](#) and orientations has made the CM dynamics complex and difficult to trace. There are still some open questions about molecular CM that remain unclear. One of the most fundamental questions: how fast does the charge migrate in molecules? Although molecular CM has been extensively studied theoretically in the last decade by using time-dependent quantum chemistry packages, a real measurement of the CM [speed](#) has remained unattainable, due to the extreme challenge.

As reported in *Advanced Photonics*, a research team from Huazhong University of Science and Technology (HUST), in cooperation with theoretical teams from Kansas State University and University of Connecticut, recently proposed a high harmonic spectroscopy (HHS) method for measuring the CM speed in a carbon-chain molecule, butadiyne (C_4H_2).

The principle of HHS is based on the three-step model of high-order harmonic generation (HHG): ionization, acceleration, and recombination. Strong field ionization first creates a hole wave packet in the ion, which evolves in the laser field and is probed by the returning electron wave packet at the recombination moment, with the hole dynamics recorded in the generated harmonic spectra. The researchers used a two-color HHS scheme in combination with an advanced machine learning reconstruction algorithm to reconstruct the CM in C_4H_2 at the most fundamental level for each single fixed-in-space angle of the molecule. The method achieved a temporal resolution of 50 as.



Charge migration in a C₄H₂ molecule. (a) Time-dependent hole densities along the molecular backbone reconstructed for the perpendicular alignment of the molecule with respect to the driving laser polarization direction. (b) Time-dependent center of charge position (dashed line with circles) retrieved from the hole densities in (a). Red dash-dotted line is a linear fitting to extract the CM speed. (c)-(d) Same as (a)-(b), but for the parallel alignment of the molecule. Credit: *Advanced Photonics* (2023). DOI: 10.1117/1.AP.5.5.056001

From the retrieved time-dependent hole densities, the movement of the center of charge is identified. From there, the CM speed is quantified, which is about several angstrom per femtosecond. Moreover, the

dependence of the CM speed on the alignment angles of molecule with respect to the laser polarization is also revealed. The CM under the laser control is demonstrated to be faster than the field-free one. This work for the first time offers an experimentally derived answer regarding the speed of CM in a molecule.

Corresponding author Pengfei Lan, a professor in the HUST School of Physics, says, "This work provides deep insight into CM dynamics in [molecules](#) and could strengthen our understanding of these ultrafast dynamics." Lan notes that the control of CM speed by molecular alignment also suggests a promising way to manipulate the rate of a chemical reaction—a path his team aims to explore in the near future.

More information: Lixin He et al, Attosecond probing and control of charge migration in carbon-chain molecule, *Advanced Photonics* (2023). [DOI: 10.1117/1.AP.5.5.056001](https://doi.org/10.1117/1.AP.5.5.056001)

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