compute the ICS and DCS from the $S$-matrix, with or without resonance contributions. The results for the final $(5/2f)$ state of NO, where the effects are most pronounced, are shown in Fig. 3. The peaks in the ICS corresponding to both resonances II and III vanish when we only included the background contribution (Fig. 3, top). The effect of the resonances on the DCS at energies close to these resonances is illustrated in Fig. 3, bottom. The background contributions (Fig. 3, dashed lines) show the usual pattern of diffraction oscillations, which are most pronounced for small scattering angles and decrease in amplitude for larger angles. The effect of the resonance contributions is substantial; they lead to additional strong scattering near the forward and backward directions. Also shown in Fig. 3 to additional strong scattering near the forward amplitude for larger angles. The effect of the

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4. R. T. Skodje

The ultrafast motion of electrons and holes after light-matter interaction is fundamental to a broad range of chemical and biophysical processes. We advanced high-harmonic spectroscopy to resolve spatially and temporally the migration of an electron hole immediately after ionization of iodoacetylene while simultaneously demonstrating extensive control over the process. A multidimensional approach, based on the measurement and accurate theoretical description of both even and odd harmonic orders, enabled us to reconstruct both quantum amplitudes and phases of the electronic states with a resolution of ~100 attoseconds. We separately reconstructed quasi-field-free and laser-controlled charge migration as a function of the spatial orientation of the molecule and determined the shape of the hole created by ionization. Our technique opens the prospect of laser control over electronic primary processes.

Measurement and laser control of attosecond charge migration in ionized iodoacetylene

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The ultrafast motion of electrons and holes after light-matter interaction is fundamental to a broad range of chemical and biophysical processes. We advanced high-harmonic spectroscopy to resolve spatially and temporally the migration of an electron hole immediately after ionization of iodoacetylene while simultaneously demonstrating extensive control over the process. A multidimensional approach, based on the measurement and accurate theoretical description of both even and odd harmonic orders, enabled us to reconstruct both quantum amplitudes and phases of the electronic states with a resolution of ~100 attoseconds. We separately reconstructed quasi-field-free and laser-controlled charge migration as a function of the spatial orientation of the molecule and determined the shape of the hole created by ionization. Our technique opens the prospect of laser control over electronic primary processes.

Ultrashort charge transfer plays a key role in chemical reactions, biological processes, and technical applications. For example, charge transfer after photoexcitation on the femtosecond time scale and the associated long-lived coherences observed in photostimulated systems (1) or photovoltaic blends (2) have been invoked to explain the high efficiencies of energy conversion in these systems. These charge transfer phenomena are driven by nuclear motion but typically involve a much faster, purely electronic response. This phenomenon was predicted theoretically and called charge migration to distinguish it from the nuclear dynamics–driven charge transfer (3–6). Charge migration arises whenever multiple electronic states are
coherently populated. Charge migration is likely to be responsible for the unexpected selectivity in photofragmentation of ionized peptides (7), leading to the concept of charge-directed chemistry (8). Moreover, it offers approaches to probing electron correlation and orbital relaxation phenomena (3, 5, 6). These prospects make the measurement of charge migration a key goal of attosecond science (9). Even more promising is the prospect of controlling charge migration. Steering electrons inside molecules is expected to yield control over reactivity, thereby providing access to regions of potential energy surfaces and consequent reaction pathways that usually remain unexplored (10).

Previously described techniques for measuring electronic dynamics include transient absorption (11), sequential double ionization (12), and photo-fragmentation spectroscopy (13). Although pioneering work on phenylalanine recently reported a resolution of better than 4.5 fs (13), such measurements of intramolecular dynamics are inherently limited in temporal resolution by the use of an infrared pulse. Electronic dynamics after strong-field ionization (SFI) have also been inferred from high-harmonic spectra. This approach has been established in seminal studies (14, 15), but a direct reconstruction of the dynamics has remained out of reach, either because no reconstruction approach was established or because experimental data were lacking. Another pioneering approach demonstrated the tomographic reconstruction of time-dependent orbitals at the expense of temporal resolution (16).

We advanced high-harmonic spectroscopy (HHS) to reconstruct, at a resolution of ~100 attoseconds (as), the full quantum dynamics of charge migration in spatially oriented polar molecules. We measured both amplitudes and phases of high-harmonic emission from oriented molecules at multiple wavelengths of the driving field. Scaling the wavelength is equivalent to tuning the excursion time of the photoelectron wave packet on the attosecond time scale (17). We show that this set of experimental data is sufficient to reconstruct the amplitudes and phases of the transiently occupied electronic states of the cation and to determine the initial shape of the hole created by SFI.

We demonstrated our technique on iodoacetylene (HCCI) because it encompasses several general characteristics of polar polyatomic molecules. These include (i) the simultaneous population of multiple electronic states of the cation by SFI, (ii) strong laser-induced coupling between these states, and (iii) a strong alignment dependence of their coupling. An energy level diagram of HCCI is shown in Fig. 1. The \( \tilde{X}^+ \)–\( \tilde{II} \) ground state and \( \tilde{\Lambda}^+ \)–\( \tilde{II} \) first excited state [with vertical ionization potentials \( I_p^\tilde{X} = 9.71 \text{ eV} \) and \( I_p^\tilde{\Lambda} = 11.94 \text{ eV} \)] are both appreciably populated by strong-field ionization [supplementary materials (SM) section 4] and are coupled by a large transition dipole moment (1.35 atomic units, which is equivalent to 3.43 Debye) lying parallel to the molecular axis. This large value originates from the charge-transfer nature of this \( \pi \rightarrow \pi^* \) transition (19).

We use control over the spatial orientation of the molecule to separately reconstruct quasi-field-free and laser-driven charge migration. For molecules aligned perpendicular to the laser polarization (Fig. 1B), the effect of the laser field on charge migration is negligible. For parallel molecules (Fig. 1C), the laser field induces substantial population transfer between the \( \tilde{X}^+ \) and \( \tilde{\Lambda}^+ \) states, which depends on the head-to-tail orientation of the molecule.

The concept of the experiment is presented in Fig. 2A. Control over the spatial orientation of the ensemble of molecules is achieved by impulsively orienting the molecules (20). The fixed-in-space ensemble of molecules is then interrogated by a high-harmonic generation (HHG) pulse one rotational period (157.0 ps) later under otherwise field-free conditions. The time resolution in our experiment arises from the subcycle nature of the HHG process: Every emitted harmonic order can be associated with a unique transit time of the electron wave packet in the continuum by experimentally selecting the short electron trajectories (21, 22).

The reconstruction of charge migration in the cation requires several observables. The ratios of emission intensity between molecules aligned parallel or perpendicular to the polarization of the probe pulse (Fig. 2B) exhibit minima located at markedly different photon energies: 23.2 eV for 800 nm (laser field period 2.67 fs) and 35.3 eV for 1300 nm (laser field period 4.33 fs). Such a strong wavelength dependence of the observed

![Fig. 1. Electronic energy levels and orientation dependence of charge migration in the iodoacetylene cation. (A) Energy level diagram. (B and C) Illustration of quasi-field-free charge migration (B) and laser-controlled charge migration (C). The energies are computed at the EOM-IP-CCSD/cc-pVTZ level of theory. Strong-field ionization by infrared photons (red arrows) prepares the electron hole. Its evolution is encoded in the high-harmonic emission (violet arrows) at the instant of recombinations. For molecules aligned perpendicular to the laser field as shown in (B), the populations of the \( \tilde{X}^+ \) and \( \tilde{\Lambda}^+ \) states are time-independent. Charge migration takes place as under field-free conditions. The laser field induces strong population transfer between the \( \tilde{X}^+ \) and \( \tilde{\Lambda}^+ \) states for molecules aligned parallel to the laser field, as depicted in (C). The evolution of the hole density is represented by \( \left| \langle \Phi^{\tilde{X}+} | \Phi^{\tilde{\Lambda}+} \rangle \right|^2 \), where \( \Phi^{\tilde{\Lambda}+} \) and \( \Phi^{\tilde{X}+} \) are the N and N – 1 electron wave functions of the neutral and ionized molecule, respectively.](image-url)
minimum cannot be contained in the photo-recombination matrix elements and indicates attosecond electronic dynamics in the cation (23, 24). Purely structural minima contained in the photocombination matrix elements do not shift in photon energy when the driving wavelength is changed (25).

Spatially resolving charge migration in the general class of polar molecules additionally requires orientation (i.e., a head-to-tail order), resulting in even-harmonic emission. This was achieved using one phase-controlled two-color (800 nm + 400 nm) laser pulse for impulsive orientation (26). Figure 2C shows the intensity ratio of the even harmonics to the averaged intensity of the two adjacent odd harmonic orders (the even-to-odd ratio) observed using a HHG driving wavelength of 800 nm.

A full characterization of electronic quantum dynamics becomes possible through the additional measurement of the phase of high-harmonic emission. We measured the phase $\phi_{\text{HHG}}$ as a function of the alignment angle of the molecules by generating high harmonics in two spatially separated sources in the gas jet (27). Here, we extended this technique to a broad range of wavelengths by working with a liquid crystal mask that introduces a voltage-tunable phase shift in one-half of the unfocused probe-beam wave front (SM section 1). Phase measurements for driving wavelengths of 800 nm and 1300 nm are shown in Fig. 2, D and E, for selected harmonic orders. The phase variation as a function of the alignment angle strongly depends on the harmonic order and driving wavelength, not just the emitted photon energy, again suggesting a pronounced charge migration on the attosecond time scale. The phase differences for molecules aligned parallel or perpendicular to the polarization of the laser field for all harmonic orders are summarized in the SM, section 1.

The reconstruction of charge migration in terms of initial (as prepared by ionization at $t'$) and final (at $t$, the instant of photoionization) populations and phases of the states of the cation is a numerical inversion problem employing a generalized theory of HHG (SM section 2) that describes all experimental intensity ratios and phase differences between molecules aligned parallel or perpendicular to the laser field. The initial and final populations and phases were retrieved in a global nonlinear least-squares optimization using a Levenberg-Marquardt algorithm with multiple starting values. Our theory includes all relevant electronic states, the continuum structure through the use of scattering-wave-matrix elements (28), nuclear motion through autocorrelation functions derived from photoelectron spectra (SM section 2), and the molecular axis distribution.

The photorecombination dipole matrix elements and the angular variation of the ionization rates were calculated theoretically. These quantities are experimentally accessible using narrow-band extreme ultraviolet sources and/or charged-particle detection, which were not available in this study. Because the emission from several channels adds coherently, our experiment is sensitive to both the amplitudes and the phases of the involved electronic states. The mapping from photon energy to transit time ($\tau = t - t'$) was performed using quantum electron trajectories obtained by the saddle-point method (29–31).

We first reconstructed charge migration for molecules aligned perpendicular to the 800 nm driving field. The reconstructed initial populations are given in Fig. 2B. These results compare well both with time-dependent density functional theory (TDDFT) (SM section 4) (32) and with weak-field asymptotic theory (WFAT) (SM section 2) (33). Combined with the experimentally retrieved initial phase $\Delta \phi = \phi_A (t = 0) - \phi_K (t = 0)$ (Fig. 3C) between the ground state $|\psi_0\rangle$ and the instant of photodetachment $|\psi_f\rangle$, the phase terms $\Delta \phi$ were calculated as the difference of the phases at the time of photodetachment $t_f$ and of the initial state $t_i$.

$\Delta \phi = \phi_A (t_f) - \phi_K (t_f) - \phi_A (t_i) + \phi_K (t_i) = \phi_A (t_f) - \phi_K (t_f)$.

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Fig. 2. HHG monitoring of charge migration. (A) Concept of the experiment: HHG (upper panel) provides a unique transit time–energy mapping as a consequence of the subcycle timing of the electron trajectories (thin lines, lower panel) in the laser field (thick lines). (B) Intensity ratio of high harmonics emitted from molecules aligned parallel or perpendicular to the laser field for 800 nm [11 (±0.2) × 10^14 W/cm²] and 1300 nm [0.9 (±0.1) × 10^14 W/cm²]. The insets show the variation of the intensities of selected harmonic orders of 800 nm. (C) Even-to-odd ratio for oriented molecules (800 nm). (D and E) Alignment dependence of the high-harmonic phase $\phi_{\text{HHG}}$ using 800-nm and 1300-nm pulses, respectively. A sum of the four lowest even-order Legendre polynomials was fitted to the experimental data. The error bars (shaded areas) in all figures correspond to the standard deviation of the measured signal fluctuations. All data were obtained by averaging the signals from at least 1500 laser shots.
and the first excited electronic state of the cation, charge migration is reconstructed as shown in Fig. 3A. The spatial representation of the electron densities further requires the computation of molecular orbitals, which must be chosen to be consistent with those used in the calculation of the photorecombination matrix elements (SM section 2). It also requires the knowledge of the difference of the vertical ionization potentials of the field-free eigenstates, which determines the oscillation period of 1.85 fs and is known from photoelectron spectroscopy (36). Strong-field ionization was found to create a one-electron hole localized at the iodine side of the molecule (Fig. 3D), compatible with the low ionization potential and high polarizability of the I atom. Subsequently, the hole delocalizes over the molecule and then localizes at the acetylene side after 930 as. Because the differential Stark shift of the $\tilde{X}^+$ and $\tilde{A}^+$ levels at the maximal applied field amounted to only 18 meV (SM, section 3, and fig. S11) or 0.8% of the energy level separation, and population transfer is absent by symmetry, the reconstructed dynamics correspond to quasi-field-free charge migration.

We found a $\sim$ difference in the relative initial phase of the $\tilde{X}^+$ and $\tilde{A}^+$ states for parallel alignment (Fig. 3C), which implies that the hole is created on the acetylene side (Fig. 3E). This means that the electron hole is created on the opposite side from where the electron tunneled because ionization via the iodine atom is dominant (fig. S5). Although this result appears counter-intuitive at first sight, the reconstructed hole is consistent with ionization on the lowest-lying multielectron eigenstate of the cation in a static field (fig. S12).

The subsequent charge migration for parallel alignment is controlled by the laser field (Fig. 4) and differs substantially from the field-free evolution (Fig. 3A). We first discuss the results obtained with 800-nm pulses. Figure 4A shows the fractional population of the $\tilde{X}^+$ ground state (solid blue line) and the relative phase $\Delta \phi(t) = \phi_{\tilde{X}^+}(t) - \phi_{\tilde{X}^+}(t)$ between the $\tilde{X}^+$ and $\tilde{A}^+$ states (dashed cyan line) for all reconstructed delays. The first striking feature is the strong depletion of the $\tilde{X}^+$ state for early transit times. The population of the $\tilde{X}^+$ state reaches a minimum at 1.02 fs before increasing again. The relative phase between the two eigenstates shows a pronounced jump around the time of maximal depopulation of the ground state. Figure 4A also shows the reconstructed hole density at selected transit times for electron tunneling via the iodine atom. For electron tunneling via the hydrogen atom (Fig. 4B), the population transfer is markedly different from tunneling via iodine (Fig. 4A). Again, the ground state is strongly depopulated at the first reconstructed delay, but its repopulation begins earlier than in Fig. 4A. One of the crucial observables for the reconstruction of side-dependent charge migration is the relative intensity of the even harmonics. Our reconstruction procedure additionally includes the sign dependence of the transition dipole moment on the molecular orientation (SM section 2). The temporal uncertainty in the reconstruction is harmonic order-dependent and amounts to $\pm 110$ as on average. The temporal accuracy is limited by the uncertainty in the peak intensity needed to connect the electron transit time to the emitted photon energy (22, 29) and the different transit times associated with the electronic states of the cation involved in the dynamics. The shaded area represents the combined error from all experimental uncertainties.

Turning to the results obtained at 1300 nm, we again find strong population transfer in the laser field, as shown by the reconstructed populations in Fig. 4C for tunneling via the iodine atom. The fractional ground-state population gradually increases until 1.53 fs and then decreases again. Our reconstruction shows a second minimum of the $\tilde{X}^+$ population at 1.71 fs and a subsequent rise for longer delays. For tunneling via the hydrogen atom (Fig. 4D), the behavior is similar, with the maximal population shifted to earlier delays. The electron hole densities reconstructed from the populations and relative phases demonstrate the strong laser control over the dynamics. This fact is further illustrated by movies S1 to S4, which show very rapid changes in the hole density associated with the reconstructed phase jumps. These results clearly show that both the site of electron tunneling and the laser wavelength offer extensive control over charge migration. They also highlight the highly non-adiabatic nature of the strongly driven electronic dynamics (fig. S10).

Our experimental reconstruction of the time-dependent populations and phases is supported by independent calculations (Fig. 4E). The population transfer was calculated by solving the time-dependent Schrödinger equation for the two
that the dynamics are accurately predicted when only the $\tilde{X}^+$ and $\tilde{A}^+$ states of the cation are included (fig. S8).

The theoretical population transfer for 800 nm and tunneling via the I side (blue line in Fig. 4E) correctly predicts the near-complete depletion and repopulation of the ionic ground state, including the associated phase jump. The theoretical depopulation of the ground state is weaker than the reconstructed one (Fig. 4A), and the phase jump is consistently less pronounced. Such jumps are most pronounced for complete de- populations and repopulations of states (see fig. S6, which also shows calculations for tunneling via the H atom). For 1300 nm, the calculations qualitatively agree with the reconstructed dynamics, but the population maximum occurs later than in the experimental results by ~300 as. Given the complexity of the nonadiabatic electronic dynamics (fig. S10), the agreement between experiment (Fig. 4, A and C) and calculations (Fig. 4E) is highly encouraging. The nonadiabatic electronic dynamics introduces a strong dependence of the population transfer on the exact parameters of the laser pulse. The remaining discrepancies may result from electron correlation between the continuum wave packet and the bound electrons beyond that taken into account in the present mean-field description of photorecombination.

Our work outlines several general aspects of the preparation and control of charge migration by intense laser pulses. First, the initial phase of the wave packet strongly depends on the orientation of the molecule with respect to the ionizing laser field (Fig. 3, D and E). This suggests molecular alignment and orientation both as necessary for a well-characterized measurement and as an attractive control parameter. Second, the laser field can be used to achieve extensive control over charge migration, especially when the relevant transition dipole moments are large and the level separations are small, resulting in strongly nonadiabatic electronic dynamics. The demonstrated laser control can be further refined by using laser pulses with subcycle-controlled waveforms (34)–for example, to steer the electron hole to a particular position within the molecule where it can trigger a desired chemical reaction.

REFERENCES AND NOTES
Evidence for primordial water in Earth’s deep mantle

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The hydrogen-isotope [deuterium/hydrogen (D/H)] ratio of Earth can be used to constrain the origin of its water. However, the most accessible reservoir, Earth’s oceans, may no longer represent the original (primordial) D/H ratio, owing to changes caused by water cycling between the surface and the interior. Thus, a reservoir completely isolated from surface processes is required to define Earth’s original D/H signature. Here we present data for Baffin Island and Icelandic lavas, which suggest that the deep mantle has a low D/H ratio (δD more negative than −218 per mil). Such strongly negative values indicate the existence of a component within Earth’s interior that inherited its D/H ratio directly from the protosolar nebula.

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stablishing Earth’s initial D/H ratio is important for understanding the origin of our planet’s water, as well as the dynamical processes that operated during planet formation in the solar system. However, evolution of this ratio occurs over time as a result of surface and mantle processes. Collisions with hydrogen-bearing planetesimals or cometary material after Earth’s accretion should have altered the D/H ratio of the planet’s surface and upper mantle (1). In addition, experimentally based chemical models suggest an increase in the atmospheric D/H value by a factor of 2 to 9 since Earth’s formation (2). Preferential loss of the lighter hydrogen isotope from the upper atmosphere causes this increase, driven by thermal atmospheric escape or plasma interactions with the atmosphere. As atmospheric D/H is linked with that of ocean water and sediments, the D/H ratio of the mantle also increases with time via subduction and convective mixing. Only areas of the deep Earth that have not participated in this mixing process are likely to preserve Earth’s initial D/H ratio.

Studies of the trace-element, radiogenic-isotope, and noble gas isotope characteristics of mid-ocean ridge basalts (MORBs) and ocean-island basalts (OIBs) reveal the existence of domains within Earth’s mantle that have experienced distinct evolutionary histories (3, 4). Although alternative theories exist (e.g., (5)), most studies suggest that high δ3He/4He ratios in some OIBs indicate the existence of relatively undegassed regions in the deep mantle compared to the upper mantle, which retain a greater proportion of their primordial He (6, 7). Helium-isotope (3He/4He) ratios more than 30 times the present-day ratio of Earth’s atmosphere (R = 1.38 × 10⁻⁶) (8) can be found in volcanic rocks from oceanic islands, including Iceland and Hawaii (9–12). Early Tertiary (60-million-year-old) lavas from Baffin Island and west Greenland, which represent volcanic rocks from the proto/early Iceland mantle plume, contain the highest recorded terrestrial 3He/4He ratios of up to 50 R (6, 7). These lavas also have Pb and Nd isotopic ratios consistent with primordial mantle ages [4.45 to 4.55 billion years (Ga)] (13), indicating the persistence of an ancient, isolated reservoir in the mantle. The degassed and primitive nature (14) of this reservoir means that it could preserve Earth’s initial D/H ratio. This study targets mineral-hosted melt inclusions in these rocks in search of this primordial signal. A range of D/H ratios are found on Earth. We compare the ratio of deuterium (²H or D) to hydrogen (¹H) relative to Vienna Standard Mean Ocean Water (VSMOW, D/H = 1.5576 × 10⁻⁶) using δD = [(D/H)sample/(D/H)VSMOW] – 1) × 1000, in units of parts per thousand [per mil (‰)]. The hydrological cycle fractionates hydrogen, creating a glacial ice [standard Greenland Ice Sheet Precipitation 6D = −190‰ (15)], ocean water (VSMOW δD = 0‰) and fresh water (δD = 0 to −300‰ (16)) reservoirs. Subduction provides a means to mix water back into the mantle, producing a variation in δD from −126 to +46‰ from slab dehydration and sediment recycling (17, 18). The MORB source appears to be better mixed, with a uniform δD of −60 ± 5‰ (19).

We measured the D/H ratios of olivine-hosted glassy melt inclusions in two depleted picrite samples (basaltic rocks with abundant Mg-rich olivine) from Padloping Island, northwest Baffin Island (20), and in three picrite samples from Iceland’s western and northern rift zones (9–11). The high forsterite (Fo) contents of these olivines (Fo₈₀ to ₉₀) suggest crystallization from primitive melts (21). We monitored possible contamination from crustal materials, or meteoric water due to weathering, by measuring the oxygen-isotope ratios of the samples (22). One Icelandic sample shows slightly raised δ¹⁸O, indicative of crustal contamination. All other samples fall within the range expected for uncontaminated mantle-derived samples.

Baffin Island melt inclusions are characterized by extremely low D/H ratios, from δD −97 to −218‰.