Manipulation of electron dynamics calls for electromagnetic forces that can be confined to and controlled over sub-femtosecond (fs) time intervals. Tailored transients of light fields can provide these forces. We report on the generation of sub-cycle field transients spanning the infrared, visible, and ultraviolet frequency regimes with a 1.5-octave three-channel optical field synthesizer and their attosecond sampling. To demonstrate applicability, we field-ionized krypton atoms within a single wave crest and launched an electron wavepacket into a valence-shell orbit with a well-defined initial phase. Half-cycle field excitation and attosecond probing revealed fine details of atomic-scale electron motion, such as the instantaneous rate of tunneling, the initial charge distribution of a valence-shell wavepacket, the attosecond dynamic shift (instantaneous ac Stark shift) of its energy levels, and its few-fs coherent oscillations.

The generation and measurement (briefly: synthesis) of electric field transients permitted the characterization of electric circuits with sub-picosecond temporal resolution (1) and constitutes a base technology for advancing high-speed electronics and electron-based information technologies. Electronic processes on the atomic scale typically evolve on a few-femtosecond to sub-femtosecond time scale. Time-domain access to these dynamics requires the extension of electric field control to optical frequencies.

As a first step to this end, measurement (2,3) and control (4–9) of the phase of field oscillations relative to their envelope (carrier-envelope phase, CEP) yielded reproducible few-cycle light waveforms (10). Attosecond metrology (11) was further advanced by the reproducible generation and measurement of isolated attosecond pulses (12–14). Control (15) and real-time observation (16–21) of electronic processes would greatly benefit from sub-femtosecond sculpting and confinement of strong light fields. Here we demonstrate this capability along with some of its consequences.

Tailoring light fields on the electronic time scale requires the coherent superposition and manipulation of frequencies over more than an octave in the visible and flanking spectral ranges. So far, this demand could only be met by the technique of molecular modulation (22–27). This approach recently allowed the sub-cycle shaping of optical fields via the superposition of quasi-monochromatic waves in the infrared-visible range (28). Although these periodic waveforms are highly relevant to advancing modern electronics, time-domain access to electronic phenomena calls for the temporal confinement of the sculpted waveform to a single cycle or just a few oscillation cycles. We refer to such super-octave optical waveforms as light transients. Recent experiments have paved the way towards the synthesis of light transients (29–33), but they have not yet achieved the goal of sub-cycle field shaping and measurement.

Here, we report on the shaping, confinement, and attosecond sampling of the fields of intense light transients within their carrier wave cycle (~2.4 fs) over the frequency band of 0.3 to 0.9 PHz. A variety of on-demand waveforms with controlled sub-cycle field evolution, yielding sub-femtosecond rise times or sub-cycle confinement of instantaneous intensity, demonstrate the power of PHz field synthesis. As an application of enhanced atomic-scale electron control, we field-ionize atoms within a single wave crest and trigger valence electron motion on a sub-femtosecond scale. By providing a sub-femtosecond optical field trigger and a robust attosecond probe, sub-cycle light transients establish sub-femtosecond pump-probe spectroscopy.

Synthesized Light Transients

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1.5-octave optical field synthesizer. We produced coherent supercontinua by propagating ~0.8 mJ, ~25-fs pulses carried at a wavelength of $\lambda_0 \sim 780$ nm in a hollow-core fiber filled with neon gas. Spectral broadening was enhanced with respect to previous experiments (34) by raising the gas pressure to ~3.5 bar, resulting in a nearly uniform (to within 20 dB) energy distribution over a bandwidth of > 0.6 PHz (330 to 1100 nm), see Fig. 1B. Manipulation of individual spectral components requires their spatial separation and subsequent recombination. The conventional approach, based on prisms and liquid crystal modulators (31), is hardly scalable for super-octave spanning operation; therefore we instead implemented chirped multilayer mirror technology, proposed in (34), which offers scalability to several octaves in the visible and nearby spectral regions. Our prototypical three-channel device (Fig. 1A) subdivides the aforementioned ~0.6-PHz spectral range into three bands of nearly equal width—ChNIR: 700 to 1100 nm, ChVIS: 500 to 700 nm and ChVIS-UV: 350 to 500 nm—with the help of dichroic beamsplitters DBS_VIS-NIR and DBS_UV-VIS [see supporting online material (SOM) fig. S1]. Dispersive chirped mirrors CM_VIS_UV, CM_VIS and CM_NIR compensate for the chirp carried by the pulse as well as that introduced by the thin fused silica wedge pairs incorporated in each channel and the beamsplitters near the edges of the spectral bands. As a result, the pulses in the individual channels are compressed to their bandwidth-limited durations (Fig. 1C).

The chirp, the CEP and the delay of the pulses formed in ChNIR, ChVIS and ChVIS-UV can be precisely controlled by wedges and nanometer-precision delay stages, respectively. The adjustment of the beam size in each channel—via an iris—allows control of the pulse’s energy (see SOM, section 1). These control knobs offer both sub-cycle shaping of the generated fields and compression of the fields close to their bandwidth limit. Owing to the high efficiency of the chirped multilayer optics used (fig. S2), the device transmits some ~83% of the incident continuum beam (dotted line in Fig. 1B), resulting in a pulse energy of ~0.3 mJ/pulse at the exit of the apparatus (ChNIR ~250 µJ, ChVIS ~35 µJ and ChVIS-UV ~15 µJ). The setup is assembled on a monolithic aluminum base plate with active thermal and interferometric path-length stabilization (fig. S3).

By analogy with femtosecond electro-optic sampling of THz transients (1), we can use attosecond streaking (12,35) for sampling the electric field of PHZ transients. To this end, the PHZ transients exiting our three-channel synthesizer were gently focused (to a peak intensity of $\sim 10^{12}$ W/cm$^2$) into a neon gas jet, where they generated broadband extreme ultraviolet (XUV) radiation (14) emitted in a near-diffraction-limited beam collinear with the driving radiation (see SOM, section 2). Bandpass filtering (width of ~13 eV centered at ~85 eV) near the cut-off energy (~90 eV), implemented with multilayer optics and thin metal foils, isolated a single attosecond pulse (12,14). Both pulses were then focused into a second neon gas jet placed near the entrance of a time-of-flight electron spectrometer (TOF) for measuring the XUV-induced, laser-field-streaked photoelectron spectra versus delay (i.e. an attosecond streaking spectrogram) (35).

The left panels of Fig. 2 display attosecond streaking spectrograms of sub-cycle waveforms synthesized from near-bandwidth-limited fields exiting ChNIR, ChVIS and ChVIS-UV. The spectrograms are composed of a series of laser-field-streaked XUV photoelectron spectra recorded as a function of delay between the XUV pulse and the sub-cycle field. A delay step of 0.2 fs was used, which safely allows sampling up to the highest frequency components (~0.9 PHz) in the waveform. Remarkably, each spectrogram reveals an isolated sub-200-as XUV pulse. This is a direct consequence of the substantial sub-cycle variation of field amplitude (see right panels in Fig. 2), which provides an efficient temporal gate—via ionization confinement and/or energy filtering—for isolating a single attosecond burst in the XUV radiation emitted by the ionizing atoms. The feasibility of generating a robust isolated attosecond probe for a wide range of waveforms is required not only for the sampling of the transients but is most important also for the interrogation of processes triggered and/or controlled by the optical field transients.

Any of the retrieved electric field waveforms (central panels in Fig. 2) permits full characterization of the properties of the apparatus and subsequently on-demand synthesis of prescribed fields. To this end, we used the waveform in Fig. 2A as a reference and retrieve the values of the control parameters of the system such as the field amplitudes, phase delays and the CEPs of the three channels by numerical band-pass filtering of the measured output waveform within the spectral ranges defined by ChNIR, ChVIS and ChVIS-UV. The field transients shown in Fig. 2B and 2C are then synthesized by delaying ChVIS-UV in steps of $\pi/4$ (~200 attoseconds) with respect to the reference waveform. Fig. 2D displays a more complex, non-sinusoidal transient, which is generated by delaying ChNIR such that the fields from the three channels cancel each other at the center of the waveform. The transient shown in Fig. 2E is generated via delay of ChNIR by $\sim \pi$ with respect to the waveform shown in Fig. 2D, resulting in a single intense field crest pointing in the opposite direction to the peak field in Fig. 2A. The full red and the dashed black lines in the central panels depict, respectively, waveforms measured or calculated from the constituent ChNIR, ChVIS and ChVIS-UV fields retrieved from the reference waveform (Fig. 2A), with control parameters changed by known amounts with respect to those of the reference waveform. The agreement between prediction and measurement demonstrates controlled sub-femtosecond shaping, complete.
characterization, and reproducibility (i.e. synthesis) of petahertz field transients.

The instantaneous intensity shown in the right panels of Fig. 2 reveals substantial variations of the strength of consecutive wave crests upon these transformations. The reference waveform (Fig. 2A) exhibits half cycles with relative intensities of (0.65, 1, 0.32) underpinning the sub-cycle character of the transient. Delaying ChVIS-UV gradually transforms the field into the highly asymmetric transient of Fig. 2C with the temporal extension left almost unchanged. This transient carries its most intense field crest right at its leading edge, followed by half cycles of decreasing intensity (1, 0.88, 0.57, 0.17), resulting in a sub-femtosecond rise time of its instantaneous intensity. Fig. 2D, on the other hand, reveals a transient with its two most intense field crests separated by ~4.5 fs and a half-cycle virtually annihilated in between. Fig. 2F shows the transient with the largest degree of temporal energy confinement, with field crest intensities of (0.39, 1, 0.64) and with ~35% of its energy carried in a single wave crest.

**Field ionization and its real-time sampling.** We used transients with a central field crest ~1.7 times more intense than the adjacent half cycles (Fig. 3B) to ionize krypton atoms enclosed in a quasi-static gas cell (length l=0.74 mm) at a density of ≈5.6-1016 cm−3. The gas cell was positioned at the laser focus, replacing the neon gas jet previously used for recording the streaking spectrograms shown in Fig. 3A (experimental setup shown in fig. S4). This procedure reveals the absolute timing of any process initiated or impacted by the laser field acting on the XUV initiated, time dependent ionic, polarization response. Moreover, our simulations (based on an adiabatic tunnel-ionization calculation) suggest that under the conditions of our experiments—the emerging absorption lines coincide with the population dynamics of the relevant ionic states, permitting retrieval of their transient evolution from the peak absorbances with good (~10%) accuracy (see fig. S9). Fig. 4A shows the sub-femtosecond evolution of the effective transient population in the ground state manifold 4p_j=3/2 (black dots), defined as

\[ \rho_{j=3/2}^{\text{eff}}(t) = \rho_j^{(3/2)}(t) + a \rho_j^{(1/2)}(t) \],

where a = 2/3 reflects the higher transition cross-section for the (m_j = ±1/2) transitions between 4p_j=1/2 and 3d_j=1 states; the 4p_j=1/2 manifold population \( \rho_j^{(1/2)}(t) \) is represented by black diamonds. These populations are retrieved from the 4p_j=1/2 \rightarrow 3d_j=1 and 4p_j=3/2 \rightarrow 3d_j=1 absorbances versus pump-probe delay, respectively. Because attosecond streaking is performed in the same apparatus, this evolution can be directly timed and contrasted with the evolution of the ionizing field (\( |E_i(t)|^2 \) shown by the dashed line).

The buildup of the retrieved ionic populations exhibits steps that are in synchrony with the field crests of the transient. This becomes even more evident from the population rates obtained by taking the time-derivative of the ionic populations in Fig. 4A. The ionization rate, estimated as

\[ \frac{d}{dt} \rho_{j=3/2}^{\text{eff}}(t) \] and shown in Fig. 4B (dots and red line),

exhibits three main features at the crests of the ionizing field. The main ionization burst is responsible for approximately 80% of the ion population and has a full width at half maximum of <0.7 fs, indicating a sub-femtosecond confinement of field ionization. It is this confinement to a
single field crest that allows, quantitative evaluation of the time-dependent rate of optical field ionization and state-selective population dynamics. For the populations depicted, we evaluate a peak production rate for \( \rho_{j=3/2, 3/2}^{\text{eff}}(t) \) of
\[
\Gamma_{\text{peak}} = (0.12 \pm 0.01) \text{ fs}^{-1}
\]
and for \( \rho_{j=1/2, 1/2}^{1/2}(t) \) of
\[
\Gamma_{\text{peak}} = (0.059 \pm 0.009) \text{ fs}^{-1},
\]
in excellent agreement with results obtained by numerically integrating the Schrödinger equation of a single-active electron model in three dimensions (see SOM, section 6 for details) yielding
\[
\Gamma_{\text{peak}} \left( \rho_{3/2, 3/2}^{\text{eff}} \right) = 0.13 \text{ fs}^{-1} \quad \text{and} \quad \Gamma_{\text{peak}} \left( \rho_{1/2, 1/2}^{1/2} \right) = 0.059 \text{ fs}^{-1},
\]
which also well reproduces details of the temporal evolution of the ionic populations (green and purple lines in Fig. 4A, and green line of 4B).

**Observation of the instantaneous optical Stark shift.** The shift \( \Delta E \) of quantum energy levels of atoms, molecules, or solids induced by an optical field \( E(t) \)—the ac Stark shift (40)—plays a central role in fundamental dynamical processes. So far, only cycle-averaged Stark effects have been accessible to experiments (41). If the laser frequency \( \omega_L \) is much smaller than atomic resonance frequencies, the Stark shift of a non-degenerate atomic level is expected to instantly follow variations of the laser field: \( \Delta E(t) = -1/2 \mu E^2(t) \), where \( \mu = \mu(\omega_L) \) is the atomic polarizability (42). When an XUV pulse creates a coherent superposition of two states with a difference in their respective polarizabilities \( \Delta \mu(\omega_L) \), the induced polarization oscillations experience a phase shift, which is approximately given by (eq. 1):
\[
\Delta \varphi_{\text{dipole}}(t) \approx -\frac{\Delta \mu}{2 \hbar} \int_{t_0}^{t} E^2_L(t') dt'
\]
where \( t_0 \) denotes the moment of arrival of the attosecond XUV pulse (with \( \tau_{\text{XUV}} << 2\pi / \omega_L \)). The instantaneous Stark shift detunes the energy at which the atom most efficiently absorbs photons from the XUV probe pulse, in our case by the \( 3d \rightarrow 4p \) transition in Kr\(^+\) ions, and modulates the frequency of the respective coherent dipole emission. Because the decay of the emission lasts several field cycles \( (\tau = \tau_{3d} \approx 7.5 \text{ fs}) \), the Stark effect does not merely shift but also reshapes the transient absorption lines shown in Fig. 3D(i-ii). Thanks to the nearly instantaneous triggering of the polarization oscillations and their subsequent rapid decay, signatures of the instantaneous ac Stark shift come to light in our transient absorption spectra (Fig. 4C), in agreement with our simulations (figs. S8 and S9).

**Creation of a valence wavepacket with a well-defined quantum phase.** Attosecond probing of few-cycle-driven field ionization of Kr atoms has revealed the emergence of a valence electron wavepacket in the \( 4p \) sub-shell of the Kr\(^+\) ensemble, as a consequence of liberation of electrons from the \( 4p_{j=3/2} \) as well as the \( 4p_{j=1/2} \) manifolds, separated in energy by spin-orbit coupling in the krypton atoms (19). By repeating this attosecond absorption spectroscopic experiment with our sub-cycle transient shown in Fig. 3B, and supplementing it with attosecond streaking, we can now launch a valence wavepacket within a sub-femtosecond interval and with sub-femtosecond absolute timing accuracy.

From the recorded absorption spectrogram (Fig. 5A) we retrieve (SOM text, section 4) the fractional populations of the six ionic states, in the form of the diagonal matrix elements \( \rho_{\text{ionic}}^{(n_j)} \) of the reduced density matrix—\( \rho_{3/2, 3/2}^{(3/2)} + \rho_{3/2, 2/2}^{(-3/2)} = 0.315 \pm 0.024 \), \( \rho_{3/2, 2/2}^{(3/2)} + \rho_{3/2, 2/2}^{(-3/2)} = 0.400 \pm 0.024 \), \( \rho_{1/2, 1/2}^{(1/2)} + \rho_{1/2, 1/2}^{(-1/2)} = 0.285 \pm 0.004 \)—and a degree of coherence of \( g = 0.85 \pm 0.06 \), which exceeds that measured in the previous experiment (19) by ~1.4 times, and is unparalleled for long-lived (> 1 fs) coherences in the valence shell. Our simulations (based on a state-of-the-art 3D time-dependent configuration-interaction singles approach (43) that includes correlation dynamics between the field-generated hole and the photoelectron and has been extended to include spin-orbit interaction (44)) well predict the measured coherence as well as fractional populations in the \( 4p_{j=3/2} \) and \( 4p_{j=1/2} \) manifolds. For more details see SOM text, section 7.

The nearly perfect coherence is, once again, a direct consequence of the sub-fs width of the ionization gate. This confinement, along with sub-fs absolute timing information from streaking, has far reaching consequences. The former allows launching of the wavepacket with a well-defined initial phase, whereas the latter permits reliable determination of this initial quantum phase. Figs. 5B and 5C show the retrieved phase \( \phi(t) \) (Eq. 2 in (19)) of the quantum superposition along with representative snapshots of the generated ensemble-averaged hole density distributions, as evaluated from our data in Fig. 5A. Linear extrapolation of \( \phi(t) \) to “time zero”, the birth of the hole at the peak of the ionizing field transient (blue line in Fig. 5B), yields \( \phi(t_0) = (0.99 \pm 0.04) \pi \), in very good agreement with the prediction of our configuration-interaction based simulations: \( \phi(t_0) = 1.06 \pi \). This initial quantum phase implies an elongated initial hole-density distribution aligned with the ionizing field vector, commensurate with our intuitive expectation.
Outlook. Sub-cycle engineering of optical field transients opens new prospects for steering the atomic-scale motion of electrons (15) with the electric force of light and for driving complex valence-shell dynamics in molecules (45). As a simple manifestation of enhanced control over valence shell dynamics they allow sub-femtosecond temporal confinement of ionization and precise associated triggering of a wealth of subsequent electronic phenomena. They also provide an isolated attosecond photon probe for interrogating the unfolding electronic and—in molecules—nuclear motions by attosecond absorption and/or photoelectron spectroscopy, as well as an isolated electron probe for tracing these dynamics via electron diffraction (46) or high-harmonic interferometry (47). This constitutes a substantial extension of the repertoire of attosecond science, which was previously restricted to either triggering or probing electronic processes within a sub-femtosecond time window. The feasibility of sub-femtosecond pump-probe interrogation of strong-field phenomena, opens exciting research prospects. Real-time femtosecond pump-probe interrogation of strong-field fields, or into strong-field-induced electron correlations in atoms, molecules, or solids (48,49) are but a few examples.

References and Notes
36. nl−1 indicates that relative to the ground state configuration a hole (or electron vacancy) is created in the nl sub-shell, where j denotes the total angular momentum and mj its projection on the z axis, the latter being aligned with the laser polarization.
47. O. Smirnova et al., Nature 460, 972 (2009).

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Fig. 1. Apparatus for infrared-visible-ultraviolet field synthesis. (A) Schematic representation of a prototypical three-channel light field synthesizer. (B) Spectrum of the coherent radiation at the exit of the hollow-core fiber (dashed line). Spectra exiting the individual channels (not to scale) are shown in red for Ch\textsubscript{NIR} (700–1100 nm), yellow for Ch\textsubscript{VIS} (500–700 nm), and blue for Ch\textsubscript{VIS-UV} (350–500 nm). (C) Temporal intensity (solid lines) and phase profiles (dashed curves) of the respective pulses. The thin black lines depict the intensity profiles of the corresponding bandwidth-limited pulses, with durations of \( \tau_{Ch(NIR)} = 6.8 \) fs, \( \tau_{Ch(VIS)} = 5 \) fs, and \( \tau_{Ch(VIS-UV)} = 4.5 \) fs. Insets show photos of the respective beam profiles taken at the exit of the apparatus.

Fig. 2. Synthesis of petahertz light field transients. (A)-(F) Attosecond streaking spectrograms composed of photoelectron spectra normalized to their integral (left panels) and the respective retrieved electric fields (central panels) and instantaneous intensity (right panels). Relative intensities for the most intense field crests—normalized to the maximum—are given in brackets. From (A) to (C), the delay of Ch\textsubscript{VIS-UV} is varied in steps of 200 fs (\(- \pi / 4 \)). Dashed lines in (B) and (C) show the field transients calculated from the reference waveform of (A). In (D) relative delays and CEPs of the individual channels are adjusted so as to create twin transients with a field minimum in between them. In (E), Ch\textsubscript{NIR} is delayed by 1.45 fs (\(- \pi \)), resulting in a high-frequency leading transient followed by a low-frequency tail. Dashed line in (E) shows the field transients calculated from the reference waveform of (D). Transients in (A), (B), (C), (E), and (F) carry less than one cycle within the FWHM of their temporal intensity profile. For (F) \( \tau_{FWHM} \sim 2.1 \) fs, incorporating only \( \sim 0.88 \) field cycles at the carrier wavelength of \( \lambda_c \sim 710 \) nm.

Fig. 3. Ionization with a sub-cycle light field transient. (A) Spectrogram and (B) retrieved electric field of the transient used for field ionization of krypton atoms. (C) Spectral density recorded with attosecond pulses averaged over delays between -20 and -5.7 fs (preceding the pump), and \( I(h\omega, \tau) \) is the spectral density recorded at a pump-probe delay \( \tau \). The delay is varied in steps of 100 as in the range of (-3 fs, 3 fs) and 300 as in the range of (-6 fs, 3 fs) and (3 fs, 6 fs). (D) Absorption spectra for different delays. The dots and error bars represent the mean value and standard error evaluated from 12 spectra recorded at each delay step.

Fig. 4. Attosecond ionization and Stark effect dynamics in Kr\textsuperscript{+}. (A) Population dynamics in the ground-state \( 4p_{j=3/2}^{-1} \) manifold (dots), \( \rho_{3/2,3/2}^{eff}(t) \), and in the excited-state \( 4p_{j=1/2}^{-1} \) manifold (diamonds), \( \rho_{1/2,1/2}^{eff}(t) \), retrieved from the absorption spectrogram of Fig. 3C and contrasted with the instantaneous intensity (dashed line) as well as with the prediction of numerical simulations convolved with the XUV probe pulse duration (green and purple lines). (B) Ionization rate \( d\rho_{3/2,3/2}^{eff}(t)/dt \) evaluated from the data in (A) (dots and red line) in comparison with the theoretical prediction (green line). (C) Shift of the central energy of the \( 4p_{5/2}^{-1} \rightarrow 3d_{5/2}^{-1} \) transition evaluated by fitting a Lorentzian profile to the attosecond transient absorption spectra shown in Fig. 3C. Red curves in (A) to (C) and magenta curve in (A) are guides to the eye obtained by three adjacent point FFT (Fast Fourier Transform)-smoothing.

Fig. 5. Initial quantum phase and density distribution of a valence electron wavepacket. (A) Attosecond XUV transient absorption spectrogram of krypton atoms field-ionized by a sub-cycle field transient shown by the blue line in (B). Linear extrapolation of the retrieved quantum phase \( \phi(t) \) (shown by the red line in (B)) to time zero as determined by attosecond streaking, allows access to the initial quantum phase \( \phi(t) = (0.99 \pm 0.04) \pi \) of the valence electron wavepacket. (C) Ensemble-averaged initial hole density distribution in the valence shell at the instant of ionization and its subsequent evolution, as evaluated from (A).