Diffraction and microscopy with attosecond electron pulse trains

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Attosecond spectroscopy¹⁻⁷ can resolve electronic processes directly in time, but a movie-like space-time recording is impeded by the too long wavelength (~100 times larger than atomic distances) or the source-sample entanglement in re-collision techniques⁸⁻¹¹. Here we advance attosecond metrology to picometer wavelength and sub-atomic resolution by using free-space electrons instead of higher-harmonic photons¹⁻⁷ or re-colliding wavepackets⁸⁻¹¹. A beam of 70-keV electrons at 4.5 pm de Broglie wavelength is modulated by the electric field of laser cycles into a sequence of electron pulses with sub-optical-cycle duration. Time-resolved diffraction from crystalline silicon reveals a <10-as delay of Bragg emission and demonstrates the possibility of analytic attosecond-Angstrom diffraction. Real-space, low-magnification electron microscopy visualizes with sub-light-cycle resolution how an optical wave propagates in space and time. This unification of attosecond science with electron microscopy and diffraction will enable space-time imaging of light-driven processes in the entire range of sample morphologies that electron microscopy can access.

Our concepts for (a) generating attosecond electron pulses, (b) direct streaking-oscilloscope characterization, (c) atomic diffraction and (d) proof-of-principle attosecond electron microscopy of electromagnetic fields are depicted in Figure 1. A femtosecond laser¹² triggers electron emission from a photocathode (yellow) and produces electron pulses of ~1 ps duration at a

central energy of $E_{el} = 70 \text{ keV}$ ¹³. The de Broglie wavelength is $\lambda_{el} \approx 4.5 \text{ pm}$ and space charge effects are avoided by using less than one electron per pulse at the sample. A first laser beam (“modulation”) is used to compress the electron pulse into a sub-cycle pulse train. For this purpose, we let the laser beam and the electron beam intersect at a 50-nm-thick silicon nitride membrane which lets the electrons pass through. The membrane is also transparent to the laser beam (1030 nm wavelength), but the refractive index of ~ 2 in combination with thin-layer interferences generates a phase shift between the incoming and outgoing electromagnetic waves. Therefore the periodic electromagnetic acceleration and deceleration of the propagating electrons in the optical field cycles before and after the membrane do not cancel out after passage through the laser focus, as it would happen in free space¹³. In the end there remains a time-dependent overall momentum kick that is proportional to the change of vector potential at the membrane and therefore dependent on the optical phase. In contrast to metal foils^{13,14} or nanostructures¹⁵, a dielectric membrane has negligible linear or nonlinear optical absorption and can therefore sustain an extreme level of power and fields, enabling strong/effective compression and metrology.

In the experiment, we let the electron beam ($0.48 \times$ speed of light) and the laser beam (p-polarized, $\sim 15 \text{ mW}$, peak field $\sim 5 \times 10^7 \text{ V/m}$) hit the membrane under 35° and 60° from the surface normal, respectively. In this geometry (close to Brewster’s angle) there is predominantly a periodic acceleration and deceleration of the electrons in dependence on the arrival time at the membrane with respect to the laser cycles. Calculations predict¹⁶⁻¹⁸ that such a periodic energy modulation¹⁸ can reshape the incoming electron packet after some free-space propagation into a train of attosecond pulses (see Fig. 1). A homogeneous compression of the entire electron beam (diameter $135 \text{ }\mu\text{m}$) is achieved by setting the optical focus larger (diameter $\sim 300 \text{ }\mu\text{m}$) and the pulses longer in time ($\sim 1.7 \text{ ps}$). For temporal pulse characterization, we apply at 3.7 mm distance a second dielectric membrane (60 nm of silicon) and a second laser beam (“excitation” in Fig. 1). The optical peak field at the silicon membrane is several times higher ($\sim 2 \times 10^8 \text{ V/m}$) and the foil’s refractive index is greater (~ 3.5). Therefore the previously negligible sideways forces are now substantial and create a time-dependent beam deflection, i.e. a cathode ray oscilloscope at optical frequencies (PHz). Further modulations in time are now irrelevant. Because the sub-cycle

electron pulse train generated at the first membrane is temporally synchronized to the characterization laser's optical cycles¹⁶, each attosecond electron pulse in the pulse train sees the same kind of deflection dynamics, and a time-integrated measurement characterizes the average pulse duration and shape¹.

Figure 2a shows the electron beam shape when all lasers are off. Figure 2b depicts the beam when the characterization oscilloscope ("excitation" in Fig. 1) is activated. We observe an elongated spot with two maxima at the ends (Fig. 2b), which can be identified as the turning points of the periodic deflection at the optical frequency. We infer a streaking speed of ~ 0.2 mrad/fs, which is ~ 25 times faster than previously achieved with THz radiation¹³. When the compression laser is also activated and adjusted to proper field strength, the streak of Fig. 2b turns into a localized spot that moves up and down with a sinusoidal dependence on the optical phase delay between the two laser beams (see Fig. 2c). This real-space streaking result directly demonstrates in probably the clearest possible way the presence of electron pulses with sub-optical-cycle localization in time, for which there has been rather indirect evidence so far¹⁷⁻¹⁹.

The duration of the laser-compressed electron pulses can be inferred from the streaked spot-width divided by the streaking speed¹³, but numerically fitting the entire deflectogram to a model is more accurate. We assume a Gaussian electron pulse shape of width τ_{el} on top of a time-independent background originating from uncompressed electrons from the non-converging optical cycles¹⁶. The deflection is modelled by a sinusoidal oscillation with free amplitude and phase at the fixed laser-cycle period of 3.44 fs. Optimization of these four parameters results in the blue lines in Fig. 2d, in noteworthy agreement with the experiment (dots). Figure 2e shows the electron pulse shape in time and reveals a pulse duration (above the background) of 0.35 fs root-mean-square or 0.82 fs full-width-at-half-maximum. These values are upper limits, because potential short-time interferometric instabilities in the experiment cannot entirely be excluded. Figure 2f shows the electron pulse duration in dependence on the compression laser's peak field strength, showing a characteristic shortening (compression) and subsequently an increase (over-compression) of the pulse width^{13,20}. The best-compressed electron pulses in the train are about 35 times shorter than previously reported for atomic-resolution diffraction²⁰.

Beam quality is not substantially altered by the compression. Figures 2g reports magnetic waist scans as a measurement of emittance, i.e. product of beam size and divergence. The beam's transverse and longitudinal coherence lengths, which define an electron's ability to interfere with itself when diffracted from a complex material, are directly related²¹. In the y direction (perpendicular to the laser polarization) there is no measurable change of emittance ($\epsilon_{ps}^y \approx \epsilon_{as}^y \approx 0.94$ nm) and in x direction we observe a slight increase from $\epsilon_{ps}^x \approx 0.85$ nm to $\epsilon_{as}^x \approx 1.37$ nm. This difference originates from tiny sideways deflections at the compressor stage in combination with a non-perfectly collimated electron beam there, an effect that can be avoided at appropriate incidence angles (see Fig. 2h). With $\epsilon_{as}^x \approx 1.37$ nm, the predicted transverse coherence for a beam with radius $\sigma_x \approx 100$ μm is $\lambda_{el}\sigma_x/(2\pi\epsilon_{as}^x) \approx 50$ nm and therefore larger than a complex molecule or unit cell. Figure 3a shows the inner part of a diffraction pattern of 60-nm-thick single-crystalline silicon in $[-1/\sqrt{2}, \sqrt{2}, 1/\sqrt{2}]$ direction, obtained with the attosecond electron pulses. Tens of Bragg spots are discernible and demonstrate the atomic resolution. Figure 3b shows the rocking curve (angle-dependent intensity) of the $1\bar{1}3$ spot, revealing a width that is limited by sample thickness (~ 70 nm in beam direction) and almost identical traces for uncompressed and compressed electron pulses. The asymmetry is a sign of substantial multiple-scattering interferences, but equally strong in both cases. The attosecond pulses do not obtain any substantial energy spread ΔE . The time-energy uncertainty principle and the measured Bragg spot width changes by the compression restrict ΔE to 4.5–60 eV and $\Delta E/E_{el}$ to 10^{-4} – 10^{-5} . This monochromaticity is ~ 100 times better than in high-harmonic pulses or re-colliding electrons and might be useful for attosecond electron energy loss spectroscopy (EELS) of, for example, inner-shell dynamics in atoms or some high-energy plasmonic processes. Figure 3c shows that the overall intensity of the Bragg spots, i.e. the structure factor representing the scattering potential within the unit cell, does not change beyond the experimental shot-noise limit when activating the compression. Diffractive imaging of laser-driven electronic motion in molecules or condensed matter requires percent-level sensitivities to intensity changes²²⁻²⁵, a signal-to-noise ratio that our attosecond electron pulses can provide (see Fig. 3c), even although our electron source was operated far from optimum conditions (see methods). These results demonstrate that laser-compression of an electron beam into attosecond pulses does not substantially modify any

spatial beam parameter; only the effective current is approximately halved by the presence of a constant background (see Fig. 2e) and somewhat further reduced by scattering and absorption at the compressor membrane. In summary, the experimental tools are now available for analytic attosecond-Angstrom diffraction with fundamental space-time resolution.

We quickly report two proof-of-principle applications. Many photoelectric processes in atoms, molecules, solids or liquids occur with subtle delays on attosecond time scales^{7,26-30} which are related to tunnelling or propagation of quasi-free electrons in the potential or band structure of the material, modified by electron-electron interactions and screening. Here we investigate whether Bragg diffraction, i.e. the conversion of crystal momentum to electron momentum, takes time or not. The silicon membrane was oriented for diffraction and simultaneously laser-excited. In this geometry, the electromagnetic streaking dynamics of the attosecond electron wavepackets in the laser field before and after the foil superimposes with the Bragg condition inside the foil, causing diffraction and deflection in synchrony. All Bragg spots show a pronounced time-dependent streaking when scanning the laser phase delay (see Fig. 3d). A centre-of-mass analysis^{26,30} reveals any potential delays between a Bragg spot and the direct beam (000-spot) with extreme precision (see Fig. 3e). Figure 3f summarizes the results: Electron-crystal scattering at tens of keV occurs with no delay on attosecond time scales (<40 as for every single Bragg spot, <10 as root-mean-square for all eight spots). Multiple scattering, plasmonic losses, inner-shell processes, electron-electron exchange interactions or electromagnetic fields exceeding 10^8 V/m or 0.3 T do not affect the attosecond-level timing with which Bragg electrons are emitted from a crystal.

In a second application, we invoke real-space electron microscopy and report a proof-of-principle demonstration at low magnification. While the motion of bound electrons occurs naturally on atomic dimensions, where diffraction is an applicable approach for imaging, collective carrier dynamics and transport often involve nanometre dimensions and above, for example in metamaterials or biological/technological light-energy conversions. We illuminated a $130 \times 150\text{-}\mu\text{m}$ big silicon membrane with the electron pulse train and adjusted the excitation laser slightly off-angle with the compression laser, in order to generate a travelling optical excitation wave on the sample. Figure 4a shows the membrane's static image and Figure 4b reveals the

time-dependent intensity changes observed with the attosecond pulses; see also supplementary video 1. Fitting a temporal phase to each location reveals the delay map depicted in Fig. 4c, showing a continuous and substantial phase advance from left to right (x direction). Figure 4d illustrates the contrast mechanism: Each point of the sample deflects the electron beam with a phase delay and amplitude that is determined by the optical waveform at that location. Deflection angles $\alpha(x, t)$ originating from the tilted incidence of the excitation wave can be convergent or divergent in x , leading to local intensity increases and decreases, which follow in space and time the travelling-wave propagation (see Fig. 4d). A condition for such combined spatial/angular imaging³¹ in the experiment is an out-of-focus magnification (see Fig. 4e); otherwise any ray originating from a certain sample location would end up at the same detector position regardless of deflection. Integrating over $y = \pm 20 \text{ } \mu\text{m}$ reveals the space-time results of Fig. 4f in comparison to a simulation based on ray optics and $\alpha(x, t)$ from a travelling wave. While in the middle region the intensity is highest at times where $d\alpha/dx$ is maximum (converging rays, see Fig. 4d), the highest or lowest intensities are observed at the membrane edges ($\pm 64 \text{ } \mu\text{m}$) at times where α is highest or lowest, respectively. Indeed, the phase shift of the edge features is close to $\pi/2$ (see Fig. 4f, right panel). Figure 4g shows the y -integrated phases along x , revealing the travelling-wave parameter ($12 \text{ as}/\mu\text{m}$), the phase shift at the edges and a ~ 10 -as accuracy of this proof-of-principle microscopy experiment. State-of-the-art instrumentation¹⁸ and appropriate waveform electron microscopy³¹ with our attosecond pulses will reveal nanoscale and potentially smaller electric and magnetic near-fields with spatial, temporal and vectorial resolution, hence the entire nanophotonic information.

Electron pulses at tens or hundreds of keV can in principle have multi-keV bandwidths and therefore zeptosecond duration. In our experiment, the degree of compression is limited by the available peak field strength at the compression membrane, by the distance to the sample, by the nonlinear shape of the optical cycles and by the uncorrelated energy spread of the incoming electron packet ($\sim 0.5 \text{ eV}$). If these parameters are improved, even shorter electron pulses could be obtained, which might be useful to study electron-electron interactions on sub-attosecond time scales. While pulse trains may offer space-time access to cycle-reversible processes (e.g. linear/nonlinear optics/plasmonics at atoms/molecules/nanostructures, Bloch oscillations,

tunnelling in laser fields, cycle-induced macroscopic currents) or such with <few-fs time constants (e.g. Auger decays, multi-electron ionization, damping in plasmonics), certain experiments on cycle-irreversible/slower dynamics or processes that cannot be multi-cycle excited due to damage (e.g. dielectric breakdown, molecular dissociations) will require isolated electron pulses and not a pulse train. The reported concept offers several ways to do so. First, the incoming electron packet can be pre-compressed by terahertz radiation¹³ or other means to few-femtosecond duration, i.e. short enough to inject it into a single cycle of the compression laser. Alternatively, optical single-cycle pulses can be used to deflect an isolated electron pulse out of the pulse train. It is also possible to filter out via single-cycle sideways deflection an attosecond electron pulse directly out of longer pulses or even out of a continuous beam. With compact multi-electron sources and/or sub-MHz laser repetition rates (see methods), about 10^6 - 10^8 e/s will impinge on the sample, enough for pump-probe experiments. Any future advances in source technology will directly apply.

Modern electron microscopy and diffraction have a huge application range and almost no sample-type or morphology restrictions. We therefore think that attosecond electron diffraction and microscopy may become a versatile alternative to conventional attosecond spectroscopy for visualizing fundamental light-matter interactions in space and time.

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Contributions

YM and PB conceived the experiment, YM measured the data, YM and PB evaluated the data and YM and PB wrote the manuscript.

Competing financial interests

The authors declare no competing financial interests.

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Figure 1 | Concept and experiment. A photo-emitted picosecond electron pulse (blue) with a sub-atomic de Broglie wavelength is temporally modulated at a dielectric foil (green) by the optical cycles of a femtosecond laser (red) into a train of attosecond electron pulses (blue). Electron diffraction from a cycle-excited sample (green) can produce atomic resolution in space and time. Alternatively, real-space electron microscopy can reveal optical near-field vectors and phase delays with sub-cycle time resolution.

Figure 2 | Attosecond electron pulses. **a**, Basic electron beam profile. **b**, Streaking deflection of long electron pulses by optical field cycles. **c**, Streaking deflection of laser-compressed electron pulses in dependence of delay. The oscillatory shape demonstrates the presence of sub-optical-cycle electron pulses. The streaking widths at the zero-crossings indicate the pulse duration. **d**, Comparison of experiment (dots) and numerical simulation (blue lines). **e**, Electron pulse shape, evaluated directly from the raw data (dots) or via the simulation fit (blue). The electron pulse duration (above background) is 820 as (full width, see black arrow) or 350 as (root-mean-square). **f**, Electron pulse duration in dependence on the compression laser's peak field strength E_{peak} . **g**, Beam quality measurements (emittance) for picosecond pulses (black) in comparison to the attosecond pulse train (blue). **h**, Numerical simulation (finite difference time domain, Lumerical Inc.) of the compression efficiency ($\parallel$) and residual sideways deflection ($\perp$) in dependence on the laser-electron angle ($\alpha_{laser-e}$) for a fixed electron-foil incidence of 35° and a peak field strength of 10^8 V/m. An angle of $\sim 40^\circ$ provides efficient compression without sideways deflection.

Figure 3 | Atomic diffraction with attosecond electron pulses. **a**, Diffraction pattern of single-crystalline silicon, taken with the attosecond electron pulse train. The not labelled Bragg spots are forbidden but visible via multiple scattering. **b**, Rocking curve of the $1\bar{1}3$ spot for picosecond pulses in comparison to the attosecond electron pulse train. Inset, crystal structure of silicon. **c**, Absolute intensity of all Bragg spots, measured with the attosecond pulses and normalized to the picosecond case. Error bars denote the shot noise of the measurement (~ 140 s integration). **d**, Streaking deflectograms of three example Bragg spots; all of them behave similarly. **e**, Centre-of-mass of the deflectogram of the $1\bar{1}3$ spot vs. the direct beam for delay analysis. Inset, illustration of delays in Bragg spot emission. **f**, Measured attosecond-level delay of different Bragg spot

emissions with respect to the direct beam. The filled and open circles are the results of centre-of-mass analysis and deflectogram fit, respectively. Error bars represent standard deviation. The average delay is (4.6 ± 5.4) as.

Figure 4 | Attosecond electron microscopy of electromagnetic waveform propagation. **a**, Shadow image of a 60-nm thick silicon membrane (grey) with SiN/SiO overcoat (black) at low magnification. The excitation field's polarization direction $E(t)$ is indicated by the arrow. **b**, Image changes in dependence on time delay. The arrows indicate the polarization direction of the excitation fields. **c**, Phase map of the measured intensity oscillations for every position on the membrane. **d**, Illustration of how a travelling-wave excitation (red) produces converging and diverging electron trajectories (blue) of the attosecond pulse train. **e**, Illustration of the imaging geometry with an image plane slightly off the detector in order to observe positions and angles at the same time (differential phase contrast). **f**, Position-dependent intensity changes vs. time in comparison to a trajectory simulation. **g**, Phase and delay across the sample (dots) in comparison to the simulation (blue). The travelling wave advances in space-time by 12 as/ μm .

Supplementary Video 1 | Attosecond electron microscopy of a traveling wave. The left panel shows the raw microscopic image of the silicon window in time. The right panel shows the change of the images with respect to the excitation delay. The scale bars represent 100 μm .

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Figure S1 | Details of the experimental setup and field-driven electron motion in Silicon.

a, Setup and beam paths. Red: laser beam (1030 nm wavelength), green: second-harmonic beam (515 nm wavelength), blue: electron beam, yellow: gold photocathode, BS: beam splitter, BBO: β -barium-borate crystal, HWP: half-wave plate, L: lens, ML: magnetic lens, CCD: optical beam profiling camera. **b**, Delay-dependence of the contrast of the electron deflectogram when scanning the two laser pulses. Red circles: experimental result. Black curve: fitting curve. **c**, Numerical simulation of a possible electric-field-induced change of the charge density in Si, as seen by the electron beam and averaged along the propagation direction. The small circles represent the position of Si nuclei. **d**, Angle-dependence of streaking deflection (left panel) and temporal compression (right panel). White lines denote impossible configurations, dotted long lines indicate the experimental conditions and the short dotted lines show a compressor setting without deflection. For more information, see methods section.