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All-optical attoclock for imaging tunnelling wavepackets

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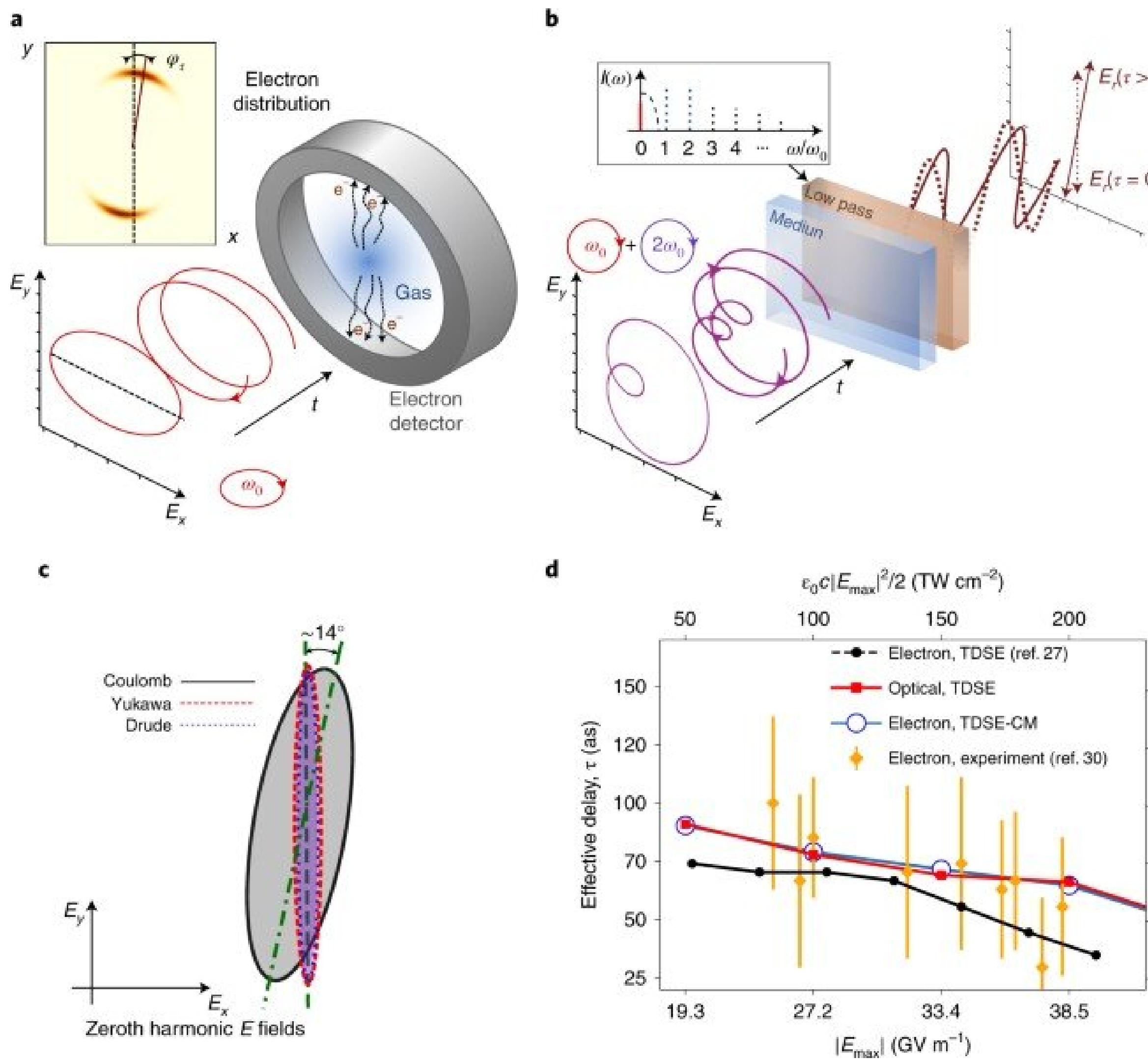
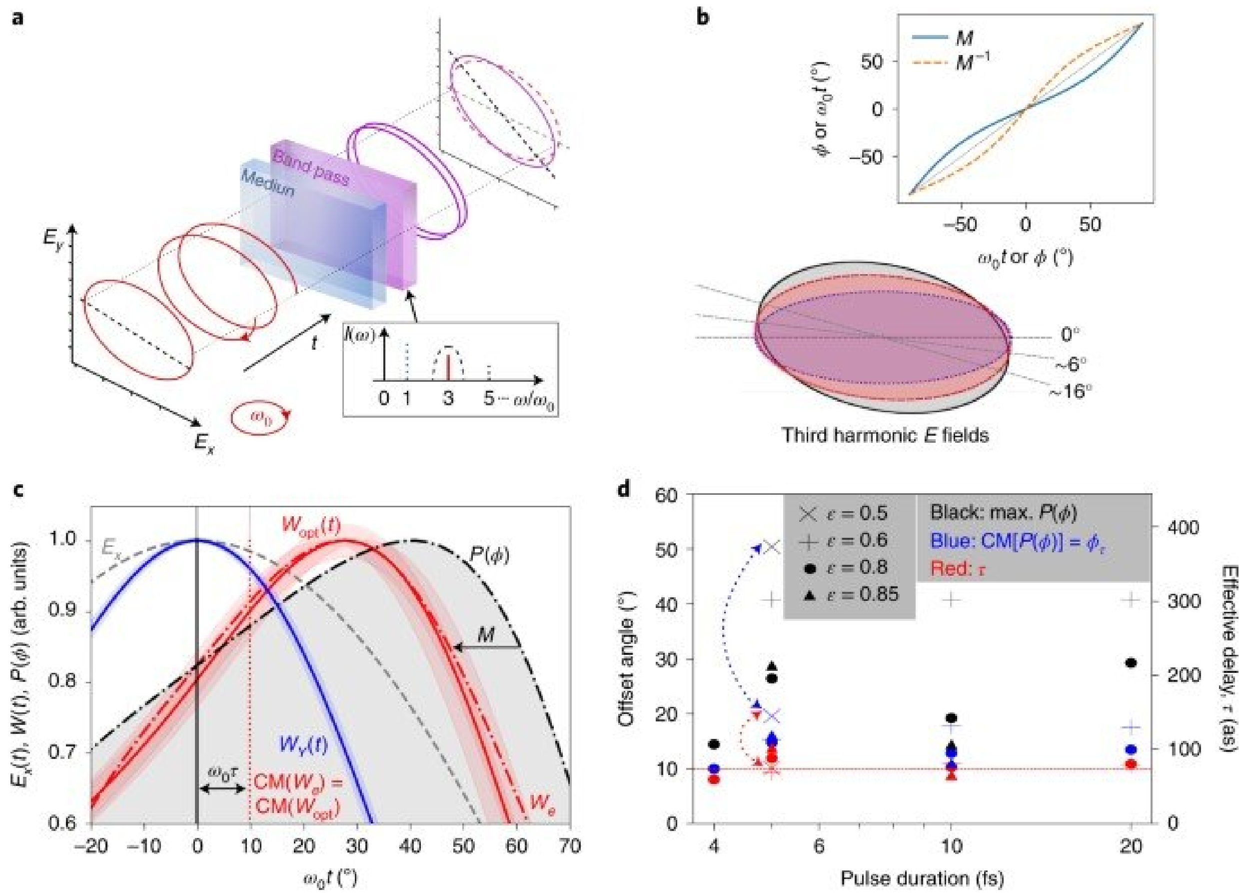


Photo-electron versus all-optical attoclock. (a) The photo-electron attoclock. The angle-resolved photo-electron spectrum generated by the driving field (red line) reveals attosecond delays and deflections of the electronic wavepacket shifting its maximum by ϕ_τ, interpreted as an effective delay $\tau = \phi_\tau/\omega_0$. b, Probing of this delay by, instead of electrons, the zeroth-order Brunel radiation in a two-colour field (magenta line). Such harmonic generated in gas or solid (blue box) can be selected by a low-pass filter (brown box). It is nearly linearly polarized (brown line) and rotated, having the same effective τ as in a. The inset shows schematically the harmonic response spectrum, including the pump (blue) and harmonic of interest (red). c, Polarization state of the zeroth harmonic for the equivalent intensity $\epsilon_0 c|E_{max}|^2/2=150$ TW cm−2, obtained from TDSE simulations with Yukawa potential, Coulomb potential and the simple-man classical Drude model. d, Effective delay τ as a function of intensity obtained from the polarization rotation of the zeroth harmonic as determined by TDSE simulations and compared with the results of the photo-electron attoclock: the centre-of-mass position of the electronic wavepacket (TDSE-CM), as well as the simulations (where the plane z = 0 instead of centre of masses was analysed) and the experiment. Credit: *Nature Physics*, https://doi.org/10.1038/s41567-022-01505-2

Physicists can study the possible time delays of light-induced tunneling of an electron from an atom after conducting measurements of time delays when cold atoms tunnel through an optically created potential barrier. In a new report now published in *Nature Physics*, Ihar Babushkin and a research team in Germany, complemented photo-electron detection in laser-induced tunneling by measuring light emitted by the tunneling electron, known as Brunel radiation. Based on combined single and two-color driving fields, they identified all-optical signatures of reshaping tunneling wave-packets as they emerged from the tunneling barrier and moved away from the core. This reshaping led to an effective time-delay and time-reversal symmetry of the ionization process, described in theory, for experimental observation. The all-optical detection method can facilitate time-resolved measurements of optical-tunneling in condensed matter systems at the attosecond time-scale.

Attosecond science

Attosecond science is a revolutionary technology, which combines optical and collision science to greatly extend the reach of each. The possibility of tunneling an electron through the potential barrier created by an oscillating electric field and the binding potential of the core is a fundamental resource in attosecond science. The phenomenon is at the heart of high harmonic generation and high harmonic spectroscopy. High harmonic generation is associated with radiative recombination based on the return of the laser-driven electron to the parent ion. But even when the electron does not return to the core, the setup emitted high harmonic radiation, referred to as the Brunel radiation or Brunel harmonics. The process is associated with bursts of current triggered by laser-induced tunneling, ubiquitous in atoms, molecules and solids. In this work, Babushkin et al. showed how Brunel harmonics generated in elliptically polarized single- and two-color laser fields provided a detailed picture of light induced tunneling of an electron. The described approach to imaging ionization dynamics distinctly differed from existing attoclock approaches based on photo-electron detection. The method allowed the introduction of a complementary, all-optical measurement protocol to establish extended measurements of tunneling dynamics in bulk solids.

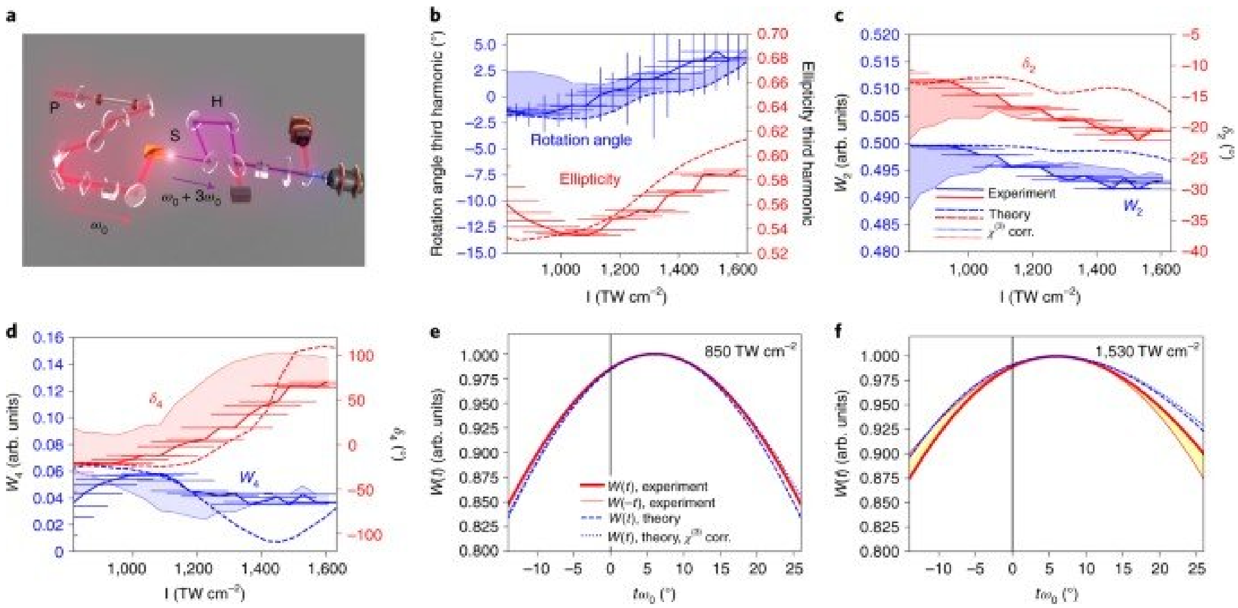


Extracting photo-ionization information from higher-order Brunel harmonics for the hydrogen atom. (a) A single-colour elliptically polarized pump (red line) produces the third Brunel harmonic (magenta line), with the polarization state (ellipticity and polarization direction) encoding the sub-cycle dynamics of the ionization process. Inset shows schematically the harmonic response spectrum, including the pump (blue) and harmonic of interest (red). (b) Polarization of the third harmonic for the single-colour driving pulse (|Emax|=36.3 GV m−1, ϵ = 0.6, I = 280 TW cm−2) calculated using TDSE with Coulomb and Yukawa potentials as well as simple-man Drude model (denotations as in Fig. 1c). Inset shows the attoclock mapping M and its inverse. (c) Optical reconstruction of the ionization dynamics (Wopt(t), solid red line) compared with the reconstruction from the photo-electron spectrum (We(t), dot-dashed red line) for the Coulomb potential. The estimation of error is given in Methods. Optical reconstruction for the Yukawa potential (WY(t), blue line) is also presented. Dot-dashed black line shows the electron wavepacket distribution P(ϕ). The attoclock delay

$\omega 0\tau$ given by the position of the center of mass (CM) of We(t) ≈ Wopt(t) is also indicated (vertical dotted red line). Dashed grey line shows the field Ex for reference. d, The positions of the maxima of the electronic spectra P(ϕ) (black markers), their CM (blue markers) and the effective delays τ=M(ϕ) reconstructed from the photo-electron spectra (red markers) for different full-width at half-maximum pulse durations and ellipticities ϵ of the driving field. Horizontal red line shows the attoclock delay extracted optically using the two-colour configuration for the corresponding peak intensity. Asymmetry of the wavepacket reveals itself from the different positions of the maximum and CM of the photo-electron spectra (traced by blue and red dotted arrows for an exemplary case of ϵ = 0.5). Credit: *Nature Physics*, https://doi.org/10.1038/s41567-022-01505-2

Physical principle and theoretical analysis

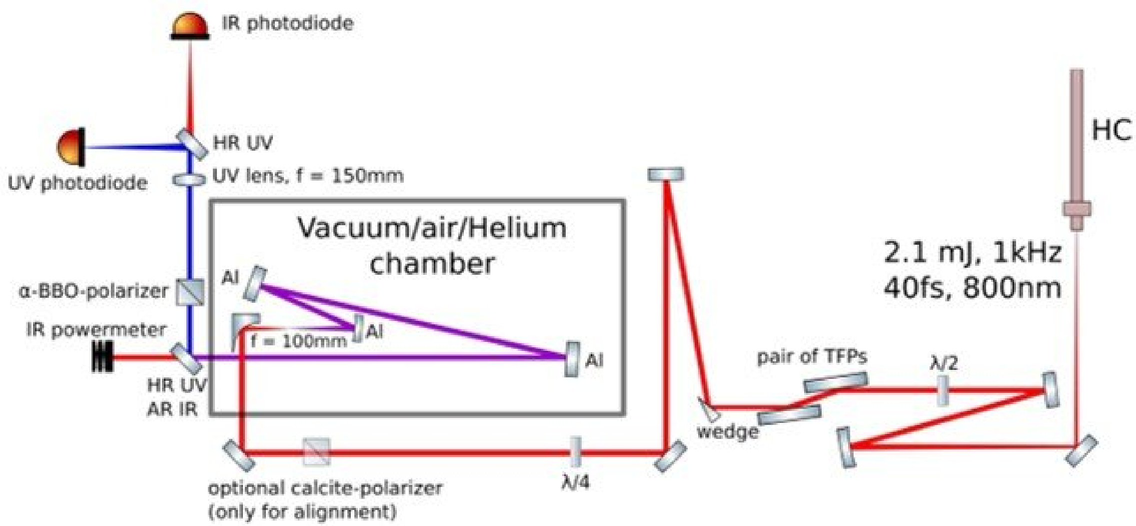
The scientists validated the central idea behind the all-optical attoclock by determining vectorial properties of the emitted light, determined by the vectorial properties of the current generated by the tunneling electron to reflect the tunneling dynamics. The team considered two field arrangements, in the first they combined an intense circularly polarized infra-red pump with its co-rotating second harmonic to generate a total electric field with a reference direction for the optical attoclock. In the second arrangement the reference direction was provided by the major axis of the single-color elliptically polarized driving field. The team began with the first arrangement where the nonlinear response contained even and odd harmonics; with a signal dominated by Brunel radiation. For instance, the team injected a classical free electron by strong-field ionization into the atomic continuum with some velocity to accelerate in the laser field and potential of the core. Babushkin et al. verified the outcomes using the ab initio time-dependent Schrödinger equation (TDSE) simulations to compute the radiated field.



Experimental reconstruction of sub-cycle ionization dynamics in Helium (He) compared with theoretical simulations. Credit: *Nature Physics*, https://doi.org/10.1038/s41567-022-01505-2

Imaging ionization dynamics and outlook

During the experiments, the team confirmed the predicted rotation of the polarization ellipse of the nonlinear response using experimental measurements with the setup. Babushkin et al. accomplished this using an 800-nm, 43-femtosecond-long, elliptically polarized pump pulse focused into a plasma spot for third harmonic generation to carefully separate and detect polarization components. The scientists compared the experimentally measured intensity-dependent parameters of the polarization ellipse with TDSE (time-dependent Schrödinger equation) simulation results to show good agreement between the experiment and simulation.



Experimental setup and investigation of errors. Experimental setup for the investigation of the polarization rotation of the third harmonic in Helium. The λ/4 plate is an achromatic plate extending from 600 to 1200 nm. The used UV polarizing beam splitter is an α-BBO Glan-Laser polarizer (ThorLabs GLB10-UV). The SiC UV photodiode is highly insensitive to other radiation frequencies than UV. The chamber was typically filled with He at 1.3 Bar pressure. All three detectors were connected to boxcar integrators triggered by the output of the regenerative amplifier. Credit: *Nature Physics*, https://doi.org/10.1038/s41567-022-01505-2

In this way, Ihar Babushkin and colleagues established a firm quantitative link between photo-electron spectra in strong-field ionization. They measured Brunel radiation generated by electrons on their way to the continuum to reveal the reshaping of electron wave packets during laser-induced tunneling. Based on Brunel harmonics imaging, the team reshaped mapping onto effective ionization delays, where Brunel harmonics in the terahertz and ultraviolet regions contained signatures of attosecond and sub-angstrom-scale electron dynamics. The researchers credited the origin of ionization asymmetry to the dynamics of the electron wave packet during and after tunneling for high intensities or saturation effects. The study provides promising capability to image tunneling and explore attosecond-scale wave packet reshaping in systems where photo-electron detection wasn't readily available. Such systems include bulk solids, where the detection of light is much simpler compared to the detection of electrons. Babushkin et al. expect the Brunel harmonics of yet higher order to allow the resolution of electron dynamics even closer to the core. The outcome will have impact beyond physics, to influence chemistry, biology and future technologies.

More information: Ihar Babushkin et al, All-optical attoclock for imaging tunnelling wavepackets, *Nature Physics* (2022). DOI: 10.1038/s41567-022-01505-2

R. E. F. Silva et al, Topological strong-field physics on sub-laser-cycle timescale, *Nature Photonics* (2019). DOI: 10.1038/s41566-019-0516-1