

ATTOSCIENCE

An attosecond stopwatch

The motion of electrons inside, around and between atoms can be captured with attosecond time resolution. A technique has now been demonstrated that can reveal electron dynamics even without attosecond light flashes.

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The microscopic dynamics of electrons have a fundamental role in excited atoms and molecules, the making and breaking of chemical bonds, as well as in light sources and electronic circuits. The observation of and ultimately control over electron dynamics on their natural timescale of attoseconds (1 as = 10^{-18} s) constitutes central topics in the emerging field of 'attoscience'. A textbook example is the oscillation of the hydrogen atom's only electron in its 1S–2P superposition state around the nucleus with a period of about 400 as. Single attosecond-duration light flashes were shown to be able to play the role of a camera with attosecond shutter speed in capturing and reconstructing the atomic-scale motion of electrons. On page 565 of this issue, Petriša Eckle and colleagues¹ demonstrate a technique for tracing electron dynamics with attosecond resolution that does not rely on attosecond light pulses.

Time-resolved spectroscopy — the extension of high-speed photography to microscopic processes — traditionally draws on two ultrashort laser pulses delayed with respect to each other. The first ('pump') pulse initiates the motion of interest whereas the second ('probe') pulse takes freeze-frame shots of the state of the system at different instants following the excitation. From these shots the motion can be reconstructed in 'slow-motion replay'. Isolated attosecond light pulses, which became available in 2001, seemed to offer a straightforward extension of this concept to tracing the attosecond-scale motion of electrons. Practical attosecond pulses, however, so far turned out to be too dim to be used for both initiating and probing microscopic dynamics. But it wasn't long before an alternative approach was at hand, when it was realized that a few-cycle

infrared laser pulse with a well-controlled electric-field waveform can take over the role of an attosecond probe pulse², for example in a technique and apparatus that were dubbed 'attosecond streaking'³ and 'attosecond transient recorder'⁴, respectively. The pulse that replaces the attosecond probe pulse has a duration of a few femtoseconds and its waveform is stabilized by drawing on the frequency comb technique, for which Theodor W. Hänsch and John L. Hall were awarded the Nobel Prize in Physics in 2005.

The attosecond transient recorder uses linearly polarized few-cycle infrared laser fields. An attosecond extreme-ultraviolet pulse (produced by the same infrared laser) liberates electrons from atoms and launches some of them along the direction of the infrared electric field. The initial velocity of these electrons is boosted or decreased by the electric field depending on the instant of the release within the oscillation cycle. The point in time at which the electron is released therefore gets mapped onto a change in final velocity (energy) of the electron. Such mapping is known as 'streaking' — the final energy distribution of the released electrons can be viewed as a streak image of the temporal shape of the electron emission within the infrared-field oscillation cycle. The measurement of near-single-cycle attosecond light pulses of 130-as duration⁵ and the real-time observation of ångström-scale electron charge transport in solids with 100-as resolution⁶ demonstrate the power of attosecond streaking in exploring electronic motion. However, the technique relies on isolated attosecond light pulses starting the motion, and these are not yet provided by commercially available laser technology.

Eckle *et al.*¹ offer a possible solution to this shortcoming. They demonstrate an implementation of attosecond streaking that does not require an attosecond 'starter' pulse. Their technique (which is based on a concept proposed by Peter Dietrich and colleagues⁷) uses few-cycle pulses with a controlled waveform, as does the attosecond transient recorder⁴, but in their case the light is not linearly but near-circularly polarized. As a result, the electrons' release

time within the infrared field cycle is connected to the final direction rather than the speed of the electrons' motion (see Fig. 1). Within one oscillation cycle of the infrared light wave, the emission angle of the electrons undergoes a full 360-degree rotation. The approach — which Eckle and co-workers refer to as 'attosecond angular streaking' — can be illustrated as a stopwatch, whose hand points in the instantaneous direction of electron emission. For the infrared wave of 725-nm wavelength used in their proof-of-principle experiments¹, a full period of this stopwatch corresponds to a span of time of approximately 2,400 as. Eckle *et al.* introduce a slight ellipticity to the infrared field polarization to trigger ionization at well-defined instants near the maxima of the field amplitude using optical-field-induced tunnelling (Fig. 1b). The ionization is then temporally imaged by angular streaking. Hence, the same field used for triggering the electron dynamics (in this case ionization) is used for probing the motion, obviating the need for an attosecond starter pulse. Eckle *et al.* report a relative timing accuracy of 24 as between the instants at which electrons are released — an impressive result that is only limited by the current signal-to-noise ratio of the measurement and might be even further improved in the future. The temporal resolution of the technique, about 200 as, is limited by the quantum mechanics and given by the angular spread of the tunnelling wavepacket in the ionization.

In the current implementation¹ of angular streaking, the waveform of the infrared laser light was not independently measured and only relative timing information between the ionization events at adjacent field peaks could be obtained. By contrast, in the attosecond transient recorder⁴, the attosecond pulse provides an absolute reference time (at which the system is excited and as a result electrons are emitted). But the independent measurement of the waveform of the few-cycle infrared streaking field should allow full exploitation of the great potential of attosecond angular streaking, for a wealth of exciting applications.

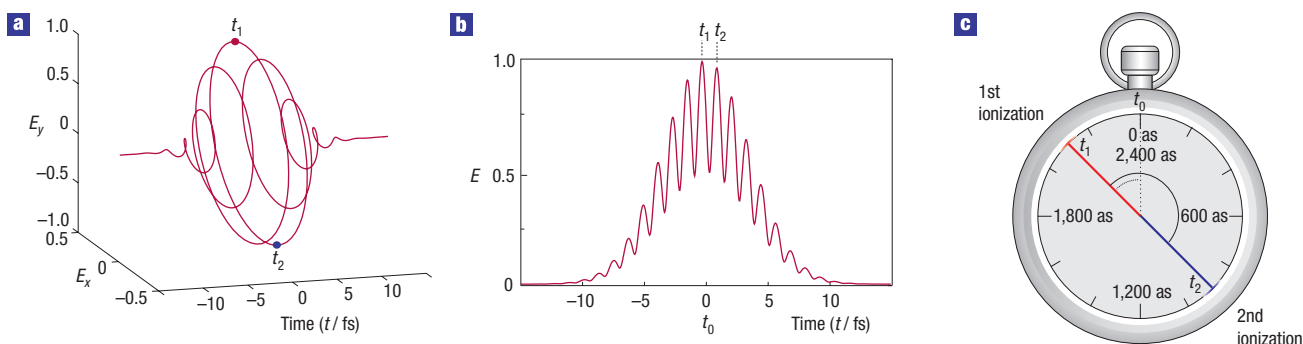


Figure 1 The attosecond stopwatch. **a**, Temporal evolution of the electrical-field vector of an elliptically polarized few-cycle laser pulse with stable waveform (shown here for an ellipticity of 0.5). **b**, Electric field $E(t) = \sqrt{E_x^2(t) + E_y^2(t)}$ of the laser pulse. **c**, Principle of attosecond angular streaking: ionization takes place at times when the field amplitude reaches a maximum. This happens two times per optical light cycle; for the case depicted in panel **b** at times t_1 and t_2 . For a known waveform of the electric field (that is, if $t_0 - t_1$ is known), the absolute time at which ionization occurs can be determined. The ionization time is reflected by the emission angle of electrons, which changes with time like the hand of a stopwatch. For 725-nm light the watch hand undergoes a full circle in $\sim 2,400$ attoseconds.

With the waveform of the infrared pulse precisely known, the launched electron wavepackets can be timed absolutely with the ‘attosecond stopwatch’ (Fig. 1). For pulses with constant carrier frequency, an independent measurement of the phase of the infrared waveform should already provide this information and seems straightforward to implement. A full retrieval of the infrared waveform, without any assumptions on the time dependence of the carrier frequency, would be achievable with the use of attosecond light pulses such as in conventional streaking measurements.

Attosecond angular streaking also holds promise for providing temporal information on multi-electron emission. In conjunction with the use of coincidence detection techniques, it might be applied to measure the correlation between electrons and used to measure the tunnelling time of electrons with high precision. In its first real-time observation, optical-field-induced tunnelling was intertwined with a preceding shake-up of electrons, and hence only an upper limit of 380 as for the overall duration of both processes could be obtained⁸. To date, the time associated with tunnelling

alone — one of the most fundamental processes in quantum mechanics — has not been measured and keeps issuing a grand challenge to attoscience. Attosecond angular streaking affords promise for rising to this challenge.

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QUANTUM DOTS

Time to get the nukes out

The ability to electrically control spin dynamics in quantum dots makes them one of the most promising platforms for solid-state quantum-information processing. Minimizing the influence of the nuclear spin environment is an important step towards realizing such promise.

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Intense theoretical efforts have produced algorithms that harness the potential power of quantum computing¹. Despite similarly vigorous experimental efforts, we have yet to identify a clear choice of the best physical platform on which a future quantum computer might be implemented.

One of the more promising solid-state candidates encodes quantum information (or qubits) in the spin of electrons confined to semiconductor quantum dots². Quantum dots are nanostructures that can be used to trap single charges. Owing to their small size, quantum dots exhibit many properties of real atoms. They have discrete electronic energy states, obey Hund’s rules, and share electrons with neighbouring dots in a manner similar to conventional molecular bonding³. But unlike real atoms, the potential that confines electrons to a quantum dot can be electrically tuned, enabling the quantum dynamics

occurring within them to be controlled by the voltage on a nearby gate. Moreover, quantum dots can be readily fabricated, and potentially monolithically integrated into larger systems, using existing semiconductor technologies. Such versatility has led to a great deal of success in preparing, manipulating and measuring the quantum spin states of semiconductor quantum dots⁴. But there’s a catch. The degree of interaction of these spins with their environment, and in particular with the nuclear spins of the quantum dots in which they are confined, is a major cause of decoherence and loss of quantum