COMMENTARY

Shining the shortest flashes of light on the secret life of electrons

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Ever since the invention of laser pulses,[1–5] one of the key directions for the development of laser technology has been the creation of ever shorter pulses of light. Over the past sixty years, improvements in technology have pushed pulse durations down from nanoseconds through picoseconds to femtoseconds, giving us access to time-resolved studies of molecular nuclear motion and chemical reactions.[6] Just in time for the 59th birthday of the standard (SI) prefixes "femto" and "atto,"[7] the 2023 Nobel Prize in Physics was awarded to the latest landmark in this effort: the generation of attosecond pulses of light,[8–10] which opens a window to the most fundamental timescale of the world around us – the timescale of electrons moving inside atoms, molecules, and condensed matter.

Attosecond pulses are the result of decades of combined advances in laser technology, raising the intensity boundary of laser pulses,[11,12] and in nonlinear optics, which expanded to use these intense laser pulses to their full potential. Attosecond pulses are born in the extreme nonlinear interaction of high-intensity laser pulses with matter – conventionally with noble gases, but increasingly with solid-state media – in a process known as high-harmonic generation (HHG).[8] HHG marks the growth of nonlinear optics from the finding of second-harmonic generation[13] to the observation of nonperturbative processes with harmonic order that can now be measured in the thousands[14] (Fig. 1).

The 2023 Nobel Prize in Physics awards the discovery of HHG by Anne L'Huillier and her team in 1988,[8] and the subsequent use of this process by Pierre Agostini and Ferenc Krausz to experimentally demonstrate pulses of light shorter than one femtosecond.[9,10]

HHG occurs when a strong and ultrafast laser drives the atoms in a gas, with enough intensity to go beyond the perturbative regime. The atoms can then up-convert the frequency of the laser, by combining large numbers of laser photons into single high-frequency harmonics. In contrast with "standard" harmonic generation, where the efficiency of the conversion drops exponentially with the number of laser photons being combined, L'Huillier and her team at CEA in Saclay, France, observed a long, flat plateau of harmonics (somewhat reminiscent of an optical frequency comb[15,16]) emitted with roughly equal intensities,[8] which had first been glimpsed one year previously.[17]

The process is best understood via the so-called three-step model, discovered independently by Paul Corkum and by Kenneth Kulander et al. in 1993,[18,19] building on previous work by Brunel[20,21] and Kuchiev.[22] The driving laser first rips an electron off an atom via tunnel ionization,[23] then accelerates it in the continuum through the oscillations of the electric field of the laser over one cycle, and finally clashes it with its parent ion at high speed, where it recombines with the hole it left behind and emits its considerable kinetic energy as a burst of light. This process then repeats every half-cycle of the driving laser, producing a train of pulses. The simple classical picture was confirmed by experiments correlating HHG to recollision-induced double ionization,[24] and it was shortly followed by an analytical quantum-mechanical theory developed by Maciej Lewenstein and co-workers.[25]

The bursts of radiation emitted by the recolliding electron occur on a subcycle timescale. It was realized early on after the discovery of the HHG plateau that, if the harmonics are locked in phase with each other, then their broad bandwidth would support subfemtosecond pulses.[26–28] Moreover, the identification of the subcycle – attosecond – electron dynamics in the continuum that produces these flashes of light opened the door to the design of methods to confine the attosecond-pulse emission to only one half-cycle of the driving laser, thus singling out one of the attosecond pulses in the train as an isolated attosecond pulse,[29–31] a family of methods now known as "gating."[32]

The scent of the attosecond world fired the starting pistol, and the race was now on: to generate attosecond pulses of light from HHG, either in a train or in isolation. This race presented significant challenges, in the design and implementation of methods for temporal characterization which could reach such an unprecedented and demanding time resolution, and – for the generation of isolated pulses – the development of gating methods practical enough to be built in the lab.

The first to cross the finish line was Pierre Agostini and his team, also at CEA in Saclay, who built a stable HHG setup and successfully timed the duration of the radiation bursts in the train down to a quarter of a femtosecond,[9] i.e., 250. To do this, they used a frequency-domain technique now known as RABBITT[33,34] (attosecond science acronyms have a distinct "animal" theme[35–37]), which looks at interference patterns in electrons ionized by the attosecond pulse train in the presence of a mid-intensity replica of the driving laser.

Almost simultaneously, Ferenc Krausz and his team at the Technical University of Vienna reported the generation of the first isolated

Fig. 1 An artist's idea of weak and strong fields interacting with matter: the weak field produces linear and low-order harmonic response to the laser field, while the strong one breaks the matter and generates high-order harmonics. Applying some imagination, one can even attribute the sound of breaking strings to attosecond pulses. Courtesy of Vasily Strelkov and Inna Midzyanovskaya.

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attosecond pulse.[10] This was the result of a long development campaign to push the duration of strong laser pulses down to the few-cycle (and even single-cycle) regime. With such a short driver, the HHG emission is effectively confined to only a single burst, a technique now known as amplitude gating. Moreover, Krausz and his team managed to measure the duration of the emission down to 650 attoseconds, using a cross-correlation technique[38] closely related to a now-standard method called the attosecond streak camera.[39,40]

In the two decades since these breakthroughs, the floodgates for discovery have opened wider and wider, allowing for a number of measurements and observations that were considered impossible or unthinkable for decades. Attosecond pulses have been used to track nuclear motion,[41] and later to observe faster and faster dynamics, including direct tracking of valence electron motion in atoms[42] and biomolecules,[43] and of the even faster motion of core-shell electrons.[44] They have also provided views of the time-resolved build-up of quantum interference patterns in atomic spectra,[45] the interference of the various quasi-classical electron trajectories that produce HHG,[46] and of the time-resolved coupling of electron and phonon degrees of freedom.[47]

One standout example is the application of the attosecond streak camera to directly observe the oscillations of the electric field of a light wave,[48] providing a new and fresh answer to the question of optical coherence.[49,50] This possibility breaks many of the assumptions held during the construction of quantum mechanics in the 1920s and 1930s, similarly to the prospects offered by the manipulation of individual quantum systems.[51,52]

The attosecond pulses produced via HHG have also been joined by pulses from additional sources, including other high-order parametric processes,[53] oscillating relativistic plasma mirrors,[54] and, most importantly, facility-scale X-ray free-electron lasers.[55] XFELs also offer attosecond capabilities, at high brightness, and with a nuanced set of trade-offs regarding coherence and timing precision. This wider set of sources promises to further enrich our ability to probe the microscopic world and its attosecond dynamics.

Looking forward, the future of attosecond science promises significant and inspiring innovations, both in the advance of attosecond sources and in their applications. On the side of the sources, it has recently become possible to use attosecond pulses to both pump and probe ultrafast phenomena in the same experiment,[56] and attosecond interferometry has reached extreme levels of precision,[57] both of which hold significant promise in reaching new regimes and dynamics. The optical control of attosecond pulses continues to increase, including the use of structured light,[58] tailored polarizations,[59] chiral states of light,[60] attosecond frequency combs based on femtosecond enhancement cavities,[61] and detailed control of the HHG wavefronts.[62]

Attosecond science also continues to expand the range of systems it can study, from HHG in solids[63] to liquids[64] and nanostructures.[65] Its growth has also made it possible to build fertile interfaces with other branches of physics, including quantum optics,[66] and quantum information processing,[67] which hold substantial promise of innovation for this young – but now mature – and dynamic discipline.

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