Attosecond Science: an emerging field brought to light by the Physics Nobel Prize

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The 2023 Nobel Prize in Physics has been awarded to Anne L'Huillier, Pierre Agostini and Ferenc Krausz "for experimental methods that generate attosecond pulses of light for the study of electron dynamics in matter". In this article, we contextualize their seminal works and their central roles that led to the emergence of a global ultrafast community exploring the frontiers of electron dynamics in dilute and condensed matter.

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In Niels Bohr's model of the hydrogen atom, the electron completes its orbit around the nucleus in just 150 attoseconds (1 as = 10^{-18} s). This astonishingly tiny unit of time is the natural time scale for the movement of electrons in matter, relevant to many processes on the microscopic level, from the first stage of chemical reactions to electrical currents and photovoltaic processes. For a long time, these ultrafast phenomena were inaccessible to experimental study because of the lack of light flashes short enough to freeze electronic movements and record a sequence of events like a stroboscope. The discoveries and experiments of Anne L'Huillier, Pierre Agostini and Ferenc Krausz in the 1980's to early 2000 revolutionized ultrafast physics and opened up the field of attosecond science.

Soon after the invention of the pulsed lasers, in the 1960's, a laboratory was settled at the "Commissariat à l'Énergie Atomique" (CEA) in Saclay (France), headed by Gérard Mainfray, to study the interaction between intense lasers and matter (now the Lasers Interactions & Dynamics Laboratory - LIDYL). **Pierre Agostini**, who had joined the laboratory in 1968, investigated multiphoton ionization of atomic gases. An important breakthrough occurred in 1979, when his team observed for the first time "above-threshold ionization", where an atom absorbs more laser photons than required to eject an electron into the continuum [1]. This first observation of continuum-continuum transitions opened up a whole new field of research. Numerous other pioneering studies coordinated by Pierre Agostini followed, laying the foundations for the temporal characterization of attosecond pulses that would be achieved 20 years later.

But before that, and soon after this first landmark, in 1982, **Anne L'Huillier** joined the CEA laboratory, first for her doctorate on multiple ionization of atoms by intense picosecond laser pulses, then as a full researcher. While, in the mid-1980s, the trend of the community was to detect and study the charged particles, ions and electrons, produced through that interaction, she decided to measure also the emitted light. Expecting incoherent ●●●

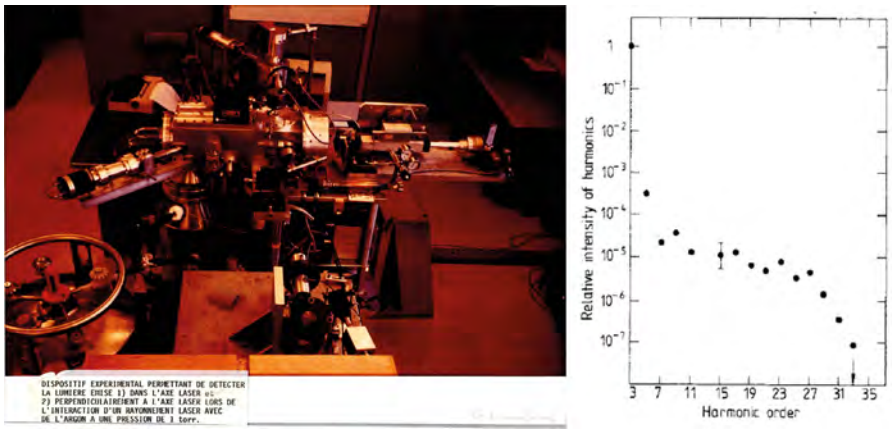
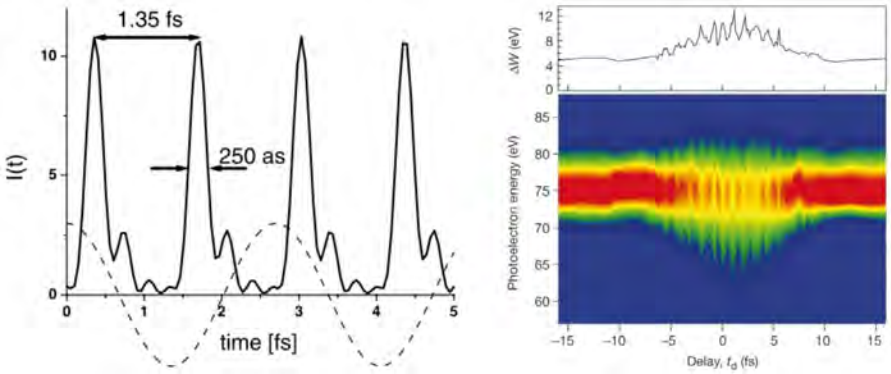


Figure 1. (left) Picture of the first spectrometer used by Anne L'Huillier and coworkers to measure their first published HHG spectrum (right).

fluorescence that would inform about involved excited atomic states, she discovered a neat light beam with a wide range of new frequencies that are multiples of the laser fundamental frequency. This process, known today as High Harmonic Generation (HHG), converts the infrared pulses into the extreme ultraviolet (XUV). In the pioneering publication [2], a spectrum extending up to the 33rd harmonic was detected, spanning 10s of eVs in photon energy. Anne L'Huillier immediately realized the potential of this new XUV light source to complement the existing synchrotrons, stating in the article's conclusion: "Therefore, such an XUV generator [...] could be efficiently used for the general purpose of XUV spectroscopy".

It was soon recognized that the extreme XUV bandwidth from this process could sustain attosecond pulses – if it were coherently generated, and if all the frequencies could be synchronized to constructively interfere in time. Indeed, the minimum duration of a light pulse ($\Delta\tau$) is determined by the width of its spectrum (ΔE, in photon energy) through the time-bandwidth product, $\Delta\tau \cdot \Delta E \geq \hbar$. This fundamental limit states that a 100-as pulse duration requires a minimum bandwidth of 18 eV, the equality being valid for a Gaussian spectral shape, with equal phases for all spectral components. The question of the synchronicity of the XUV high-harmonics then became central to the emerging community. But the answer was not straightforward as the process was poorly documented, and the theory was not developed. Anne L'Huillier and her group thus undertook systematic experimental as well as numerical studies of the phenomenon. The XUV spectrum was extended up to the 135th harmonic, and the yield of the process was optimized, notably through the crucial role of phase-matching of the very many single-atom-emitters in the gas medium. The experimental and theoretical knowledge of HHG rapidly progressed in the early 1990's, attracting ever more research groups and leading to a theoretical breakthrough. In 1993, the first model of HHG was put forward simultaneously by Kenneth Kulander *et al.* [3], and by Paul Corkum [4]. They reckoned that, during the duration of a single oscillation cycle of the strong driving laser field, an outermost electron of the atom is tunnel ionized, subsequently accelerated by the laser field in the continuum, and finally driven back to re-collide and recombine with its parent ion. During the recombination, the electron's energy is released as an XUV photon, responsible for the observed HHG spectra. Just a year later, the derivation of the quantum version of this model [5] was led by Maciej Lewenstein, who at the time was on a sabbatical with Anne L'Huillier at CEA-Saclay. With further refinements over the years, this remains today the most popular framework to model and understand HHG. Building on this model, in 1996, Anne L'Huillier and co-workers could compute the expected phases of the different harmonics, and showed that, in specific generation conditions, attosecond pulses should indeed be obtained [6]. The race towards their first observation was then open, and marked by three parallel key developments. From 1994 onwards, **Pierre Agostini**, in collaboration with Harm Muller (FOM Institute in Amsterdam), built an experimental program aimed at characterizing the duration of ultrashort XUV light sources. With

Figure 2. First measurements of attosecond pulses. A. Attosecond pulse train reconstructed using the RABBIT technique by Pierre Agostini and coworkers [8]. B. Photoelectron Streaking trace measured by Ferenc Krausz and colleagues [10] showing fast oscillations with respect to XUV-IR delay evidencing an isolated attosecond pulse.



colleagues at LOA (Laboratoire d'Optique Appliquée) in Palaiseau, they superimposed XUV light from the new HHG source with a fraction of the fundamental laser inside a photoelectron spectrometer. In addition to electrons ionized via the absorption of single XUV photons, they also detected electrons with their energy shifted by the laser-induced continuum-continuum transitions that Pierre Agostini had discovered 15 years earlier. Since these electrons only appear when both light pulses interact simultaneously with the ionizing atoms, they encode a cross-correlation of the XUV and laser pulses on the femtosecond time scale.

The step onto the attosecond scale came with the second development, initiated in 1996 by a team of theorists from the Laboratoire de Chimie-Physique-Matière et Rayonnement (LCPMR) at Sorbonne University, led by Alfred Maquet. Based on this theoretical work, it was realized that, within this cross-correlation trace, the number of energy-shifted photo-electrons is modulated by the laser-XUV delay, with a period of half a laser cycle [7]. This changed the perspective from modulating XUV-ionized photoelectrons with a laser pulse of femtosecond duration, to modulating with the laser *field*, which varies on the attosecond time scale. These modulations result from interference between different ionization pathways involving successive XUV harmonic orders, and therefore depend on their relatives phases. This led Muller and Agostini to the development of a method called RABBIT ('Reconstruction of attosecond beating by interference of two-photon transitions') that measures via a fine scan of the XUV-laser delay the XUV spectral amplitudes and phases. A Fourier transform then yields the attosecond XUV pulse profile. Implementing this electron interferometry required great stability and a high repetition rate, which Agostini found at LOA, who had just built one of the most advanced femtosecond laser systems of the time and an associated HHG source. This collaboration demonstrated the first measurement of a train of 250-attosecond pulses [8].

The third key development was the rapid advancement of femtosecond laser technology to its ultimate limit, *i.e.* light pulses containing barely a single field oscillation cycle. **Ferenc Krausz**, then at TU Vienna and at the forefront of this work, built lasers producing intense pulses containing only 10 cycles of red light. He invited Mauro Nisoli from Politecnico di Milano, who in 1996, together with Orazio Svelto and Sandro de Silvestri, had shown how to compress such pulses further. They had spectrally broadened the laser bandwidth through nonlinear propagation in a gas-filled hollow-core fiber, before resynchronizing the newly formed spectral components, initially with a prism compressor. In Vienna, Krausz proposed to replace the prisms by so-called "chirped mirrors", which allowed a more fine-tuned compression and led to pulses of 4.5 fs, *i.e.*, less than 2 cycles [9]. With only a single oscillation cycle strong enough to drive HHG, this allowed generating the first isolated attosecond pulses in Vienna. In 2001, Ferenc Krausz's team then met the challenge to characterize them with the help of • • •

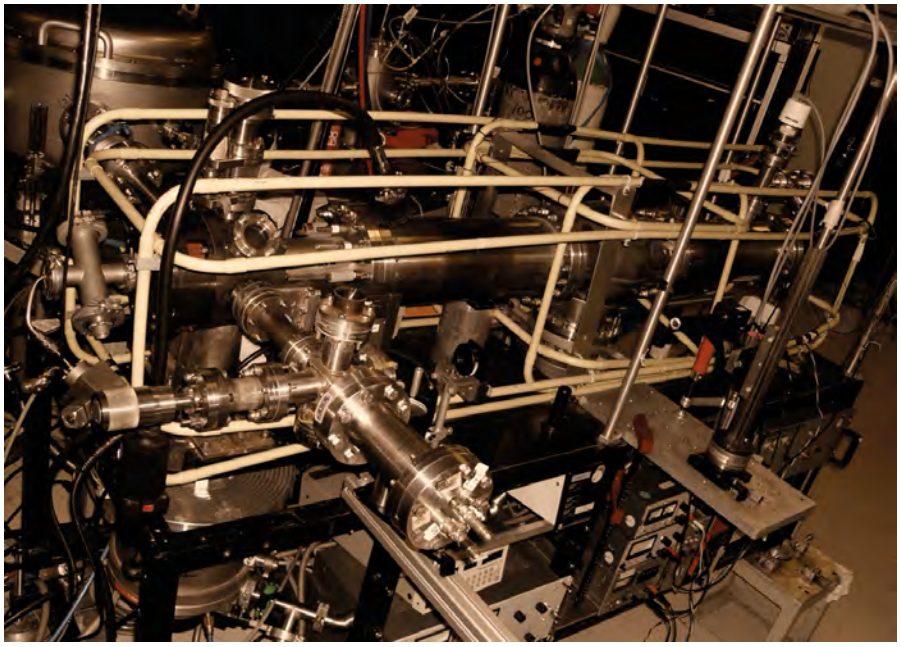


Figure 3. Picture of the magnetic bottle electron spectrometer used by the team of Pierre Agostini to detect the first attosecond pulse trains. This spectrometer is still currently operated on the ATTOLab facility of LIDYL.

Paul Corkum, by an approach related to RABBIT but using a much stronger laser field to act like a streak camera on the photoelectrons ionized by the attosecond pulse. This resulted in the first measurement of an isolated attosecond pulse of 650 as duration [10]. Such pulses became fully reproducible 2 years later when the laser field oscillation inside the pulse envelope was fully controlled by stabilization of the carrier-envelope phase [11], using the frequency-comb technique pioneered by another Nobel Prize winner, Theodor Hänsch.

This attosecond metrology had an immediate impact, as it paved the way both for the continued optimisation of attosecond sources and for their applications. Among the latter, the Nobel Committee highlighted the conceptual advance of time-resolving the photoelectric effect, a process that Einstein had theorized in 1905 through the absorption of a quantum of light (the photon) and the almost simultaneous emission of an electron. The RABBIT and the Streaking methods made it possible to measure tiny emission delays of a few tens of attoseconds. This provided valuable information about the cohesion of matter on a microscopic scale and the complex many-body quantum dynamics, whose theoretical understanding remains one of contemporary physics' major challenges.

The heirs of Anne L'Huillier, Pierre Agostini and Ferenc Krausz have extended the field opened by these pioneers to chemistry, solid-state physics, plasmas and biology. Today, on an international scale, a whole new field, attosecond science, is taking shape with exponential growth. Major investments are being made in the construction and operation of large scale facilities, such as the Hungarian pillar of the European Extreme Light Infrastructure - Attosecond Light Pulse Source (ELI-ALPS) in Szeged-Hungary. At the national scale, attosecond beamlines are already open to users at LIDYL (ATTOLab), ILM-Lyon and CELIA-Bordeaux, within the 'Ultrafast' platform of the national LUMA program, as well as at LOA. The proliferating tools of attosecond science allow observing and controlling the electronic dynamics in strongly correlated materials of interest for future electronic devices, the spin dynamics in magnetic matter, the electronic dynamics in chemistry and biology-relevant molecules, the collective dynamics of plasmas, and are of interest for photovoltaic applications or molecular screening. To fuel these new research paths, source developments are pursued, to industrialize gas phase harmonics, but also to increase their variety through the development of solid state HHG, plasma HHG, or x-ray Free Electron Laser (XFEL) facilities that are now reaching the attosecond frontier. Attosecond science, which began in a handful of laboratories in the 90s, is now emerging as a burgeoning area of research, which has not yet reached its full potential but is developing at a fascinating pace worldwide, spreading into many unexpected areas. ●

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