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Scientific Background to the Nobel Prize in Physics 2023

"FOR EXPERIMENTAL METHODS THAT GENERATE ATTOSECOND PULSES OF LIGHT FOR THE STUDY OF ELECTRON DYNAMICS IN MATTER"

The Nobel Committee for Physics

The Royal Swedish Academy of Sciences has decided to award
the Nobel Prize in Physics 2023 jointly to

Pierre Agostini, Ferenc Krausz and **Anne L'Huillier**

"for experimental methods that generate attosecond pulses of light for the study of electron dynamics in matter"

Historical background

The 2023 Nobel Prize in Physics is for the study of the movement of electrons in atoms, molecules and matter in the condensed phase by means of attosecond spectroscopy.

When Werner Heisenberg formulated the new quantum mechanics in 1925 [1], his main argument was that the old quantum mechanics forced physicists to use quantities that were, in principle, unobservable, such as the position and period of revolution of the electron in the hydrogen atom. Heisenberg argued that a new theory should be based on "observables", such as the frequencies of quantum transitions.

Heisenberg's intuitive paper of 1925 is one the most important in physics from the 20th century, but what he could not anticipate was that what was once "in principle" unobservable is now becoming accessible in laboratory experiments. We may not yet be able to observe, in the strict sense, the position and revolution of an electron around a nucleus, but today, we can "see" the dynamics of electrons in atoms, molecules and matter in the condensed phase in laboratory experiments.

How is this possible? Simple arguments based on a comparison of the intrinsic atomic unit of time, about 24 attoseconds (as), and the timescale of a single-cycle optical pulse, about one femtosecond (fs), would suggest that it would never be possible to probe electron dynamics in real time. In fact, for quite some time, the shortest pulse produced by laser laboratories was about 6 fs. The experimental development of short optical pulses has been intimately related to technical developments in laser technology, such as mode locking and light-pulse duration measurements. This made it possible to probe how atoms move in a molecule, and, in particular, the possibility to study the elusive transition states in chemical reactions [2], studies for which Ahmed Zewail was awarded the Nobel Prize in Chemistry in 1999.

All developments in the possibility of measuring the briefest time intervals since around the year 1600 (about one second) were based on the advancement in technology. But breaking the 1-fs barrier required a paradigm shift based on fundamental physics.

The long road to attosecond pulses of light

During the 1980s, several research groups produced and studied highly charged atomic ions or atoms with no or only a few electrons. Researchers relied on different approaches, such as the use of advanced ion sources and high-power lasers. They demonstrated how multiphoton ionization processes could produce multiply charged ions [3].

A natural question to ask was "at what wavelengths are photons produced in these multiphoton ionization processes?" Multiply charged ions are quite easy to detect; spectrally resolved photons are more difficult.

The first experiment using infrared photons [1064 nanometer (nm) from a powerful laser] was performed in a research centre in Paris-Saclay in France [4] and gave very surprising results. Many high-order harmonics were produced when rare gases were exposed to infrared photons of intensity 10^{13} W/cm^2, corresponding to coherent radiation emissions at frequencies that were multiples of the laser frequency. What was surprising was that the emission intensity of the odd harmonics first decreased rather sharply, then were essentially constant from the 5th harmonic up to about the 33rd harmonic for argon, and then decreased again. This was not the first observation of high-harmonic generation (HHG), but it was the first for which a very clear plateau was observed. The question remained as to what physical process was responsible for the formation of the plateau and how it could be exploited. Figure 1 shows generically the emission intensity as a function of frequency in HHG.

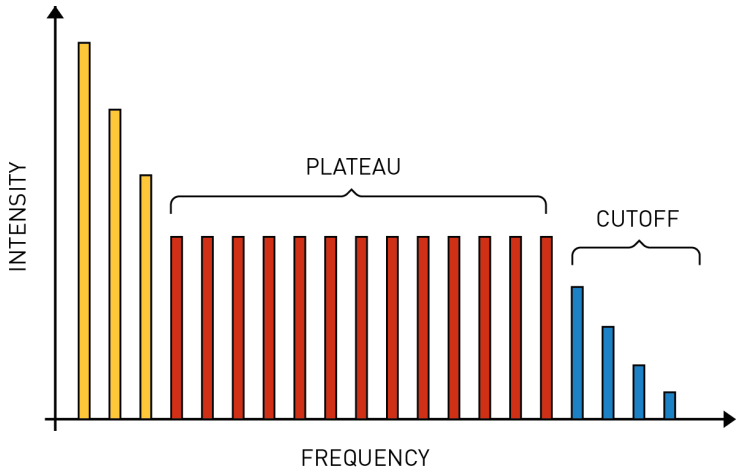

***Figure 1**. A generic high-harmonic generation (HHG) spectrum with its three characteristic features: the initial intensity fall-off, the plateau and the cutoff.*

Several groups suggested that the plateau provided the bandwidth needed to produce very short pulses [5–7], but their proposals were framed as "it should in principle be possible". In order to make practical use of the HHG plateau, an understanding of the mechanism for HHG was required.

In a paper published in 1991, Anne L'Huillier, Kenneth Schafer and Kenneth Kulander [8] presented results from a numerical solution of the time-dependent Schrödinger equation (TDSE), and they provided a clear understanding of the HHG process. They correctly predicted the general shape of the HHG spectrum (Figure 1), realized that HHG is a single electron effect (SAE, single-active electron approximation), and provided the first ever discussion of macroscopic phase matching, which required solving Maxwell's equations.

Later, Kulander's group used TDSE SAE to derive a simple formula for the cutoff energy for inert gas atoms [9]:

$$E_c = I_p + 3U_p$$

where I_p is the ionization potential of the atom and U_p is the pondermotive potential of the laser field (proportional to the product of the laser intensity and wavelength), or in other words, the mean kinetic energy acquired by an electron oscillating in the laser field. In early 1993, at a conference in Belgium, Kulander gave an oral presentation of the team's newly formulated rescattering model, which shows how short pulses in the extreme ultraviolet (XUV), in the range of about 10 to 120 eV, are produced by HHG [10]. Figure 2 shows how to conceive of the process by means of the rescattering model.

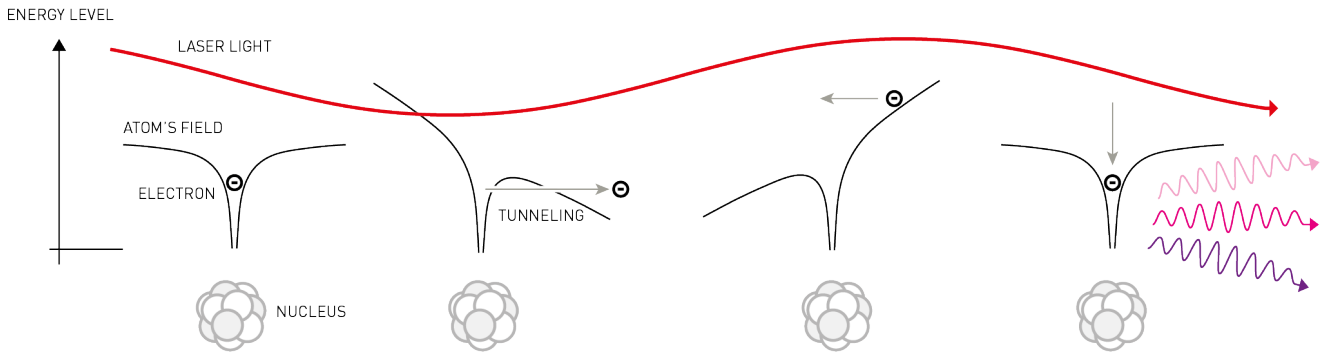

Figure 2. *The rescattering, or three-step semiclassical model of HHG. In the first step, the laser field causes tunnelling ionization; in the second step, the laser field accelerates the electron. When the field reverses in the next half-cycle, the free electron may return back to the ion and*

recombine. In the third step, the recombination process converts the kinetic energy of the electron to an emitted extreme ultraviolet (XUV) photon.

The exciting finding of Kulander and co-workers grew out of their theoretical work on the related process of above threshold ionization (ATI), in which a strong laser field drives free-free electron transitions. In other words, the electron is driven in the continuum above the first ionization limit in steps corresponding to the laser wavelength. The process of ATI was first discovered experimentally by Pierre Agostini and coworkers in 1979 [11], and ATI was at the centre of attention for strong-field laser atomic physics for a decade.

Around the same time that Kulander and his team were working on the rescattering model [10], Paul Corkum [12], who also had a background in strong-field atomic physics, formulated an alternative to that model, which became known as the three-step model. In both the Kulander and Corkum models, the important component is the action between the electron and the ion in the rescattering process. In ATI, only the interaction of the laser field and the electron is important. In the rescattering model, as illustrated in Figure 2, the electron is not excited to the point where it escapes. Instead, it returns, driven back by the laser field to the singly charged ion that it left behind. In the recombination process, the kinetic energy of the electron gets converted to an XUV photon when the electron returns to the ion.

Figure 2 also illustrates why the discovery of the plateau of the HHG spectrum (Figure 1) was facilitated by the use of a powerful Nd:YAG laser at 1064 nm, instead of the 248-nm radiation used by the group of C.K. Rhodes [13]. The electron is forced by the 248-nm laser radiation to return to the ion much faster than at 1064 nm, with much less kinetic energy at its disposal; the possibilities for the electron to develop a HHG plateau are more limited in this scenario.

The models of Kulander and Corkum were semiclassical, and in 1994, Lewenstein, L'Huiller and Corkum, with several other co-authors, presented a full quantum theory [14] that confirmed the semiclassical interpretations of Kulander and Corkum. Armed with a solid understanding of the rescattering process, from both semiclassical and quantum points of view, L'Huillier and co-workers could now proceed to move the observation and understanding of HHG towards its use to generate attosecond pulses, laid out in a series of seminal papers in the mid- to- late 1990s [15–17].

The generation of attosecond pulses of light

The road map towards the generation of attosecond pulses had been outlined and the theoretical steps had been experimentally verified. The next requirement was to develop a metrology to measure the duration of attosecond pulses and the use of suitable laser systems.

One important step was taken by Agostini and co-workers in 1994 [18], where they investigated the principle of frequency modulation in a two-colour photon field. This principle was later developed to the metrology technique named RABBIT (reconstruction of attosecond beating by interference of two-photon transitions). The RABBIT technique makes it possible to measure the pulse duration of a train of attosecond pulses by focusing the XUV pulse and light from the drive laser onto a rare gas target and analysing the photoelectrons emerging from the target.

Important insight was also provided by Corkum, Burnett and Ivanov [19] and later by Schafer and Kulander [20]. In the first paper [19], theory was used to propose how a drive-field with time-dependent polarization could result in confining the harmonic emission to a single cycle. Schafer and Kulander [20] suggested, also based on theory, that an isolated attosecond pulse could be formed by harmonics near the cutoff by using a few-cycle pulse. This method is exactly what Ferenc Krausz exploited when producing isolated attosecond pulses for the first time.

The path to isolated attosecond pulses required technical developments that Krausz explored with his research group in Vienna in collaboration with Mauro Nisoli's group in Milan [21, 22]. The Milan–Vienna collaboration resulted in production of what were then the shortest pulses ever: 4.5 fs, using krypton filling a hollow fibre, and 5 fs with argon as the fibre-filling gas. The group in Milan had pioneered the technique of compressing a laser pulse using a gas-filled hollow core fibre. In Vienna, the Krausz group generated a broadened HHG spectrum with a cutoff at about 300 eV [22].

Everything was now set for producing attosecond pulses. In 2001, 13 years after the first HHG spectra driven by an IR laser were observed [4], attosecond pulses were demonstrated in Paris-Saclay and Vienna. In Paris-Saclay, at the same research centre where HHG was discovered [4], the Agostini group produced a train of pulses with a duration of 250 as [23], as measured with the RABBIT metrology using argon as the target gas. In Vienna, the Krausz group produced isolated pulses of duration 650 as [24]: to do this, they used spectral filtering to select relevant harmonics with a multilayer XUV mirror. Then they measured the kinetic energy spectrum of 4*p* photoelectrons ejected from krypton atoms under simultaneous irradiation by 90 eV photons and the light pulses at 750 nm from the drive laser generating the harmonic radiation (streaking).

They had opened the window to the study of electron dynamics in atoms, molecules and matter in the condensed phase.

Delayed photoemission

The opening of a new time window made it possible to answer questions that were previously impossible to address. In 1905, Albert Einstein published the first explanation of the photoelectric effect, but at the time, it was impossible to resolve the timescales that were relevant for this effect. For a long time, physicists assumed the effect was instantaneous.

Einstein was eventually awarded the 1921 Nobel Prize in Physics "for his services to Theoretical Physics, and especially for his discovery of the law of the photoelectric effect". Parenthetically, when he gave his Nobel lecture, it was not in Stockholm in December 1922 (Einstein was in Japan at that time), but in the middle of the summer of 1923, in Gothenburg – a unique event in the history of the Nobel Prize. His talk did not concern the photoelectric effect but the theory of relativity, the theory for which he was never awarded a Nobel Prize.

The fundamental question that this year's Laureates made it possible to pose was "what is the timescale for the photoelectric effect?" When an atom or a surface absorbs sufficient energy from incoming light, it can transfer that energy to an electron, which is then emitted with a kinetic energy equal to the photon energy minus the binding energy of the electron. The complex dynamics of atomic photoemission results in a small time delay, and the question is how small this time delay is. Before the window to attosecond science was opened, one could assume that the process occurred instantaneously, and so the research focus was on the energetics. This is the foundation of photoelectron spectroscopy.

The Krausz group [25] found in a pioneering experiment that when the neon atom is ionized by 100-eV photons, there is a time delay between the emission of electrons from 2*s* and 2*p*. The 2*p* electron is slower to leave the neon atom than the 2*s* electron by 21 as. This can be compared with the natural orbit time of 100 as for a 2*p* electron in neon. The photoemission delays are a signature of collective dynamics of the electron cloud. The Krausz group used isolated attosecond pulses and the streaking technique, which records the electron kinetic energy as a function of the timing offset between the XUV and IR pulses.

Theoretical calculations [26–28] were unable to reproduce the experimental result, and they gave results about a factor of two shorter. Although these are complicated calculations, different theoretical groups agreed on the calculated time delays.

L'Huillier's group addressed the discrepancy [29] by making use of the HHG spectrum obtained by phase-stable interferences between attosecond pulses in a train. In this way the researchers could circumvent the natural trade-off between temporal and spectral resolution, as had earlier been demonstrated [30]. Figure 3 shows the experimental setup the team constructed in Lund, Sweden. In order to avoid overlap between the ionization signals from 2*s* and 2*p*, they applied spectral filtering in order to select harmonics spanning less than 27 eV. The binding energy of 2*s* exceeds that of 2*p* by 27 eV. The results are shown in Figure 4.

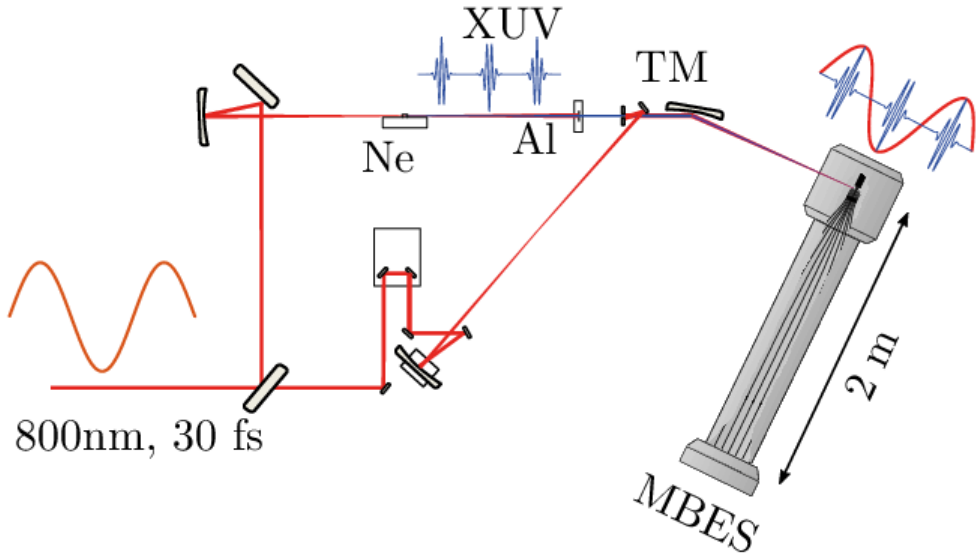

Figure 3. *Schematic illustration of the experiment in Lund. It uses the technique of "reconstruction of attosecond beating by interference of two-photon transitions" (RABBIT). The drive laser at 800 nm, 30 femtosecond (fs), is sent into two different arms by a beam splitter. High harmonic generation (HHG) in a gas jet of neon gives a train of XUV attosecond pulses. The IR pulses from the drive laser and the XUV pulses (typically with a 10–20 eV bandwidth) are overlapped and focused by a toroidal mirror on the neon gas target. The XUV photons ionize the target gas, and the photoelectrons are analysed by a magnetic bottle spectrometer. The overlapping IR/XUV pulses give rise to sideband signals that can resolve shake-up electrons (with 2p being ionized while also promoting another 2p electron to 3p) from 2s electrons despite an energy difference of only 7.4 eV. (Figure uploaded on ResearchGate by David Busto, Lund University.)*

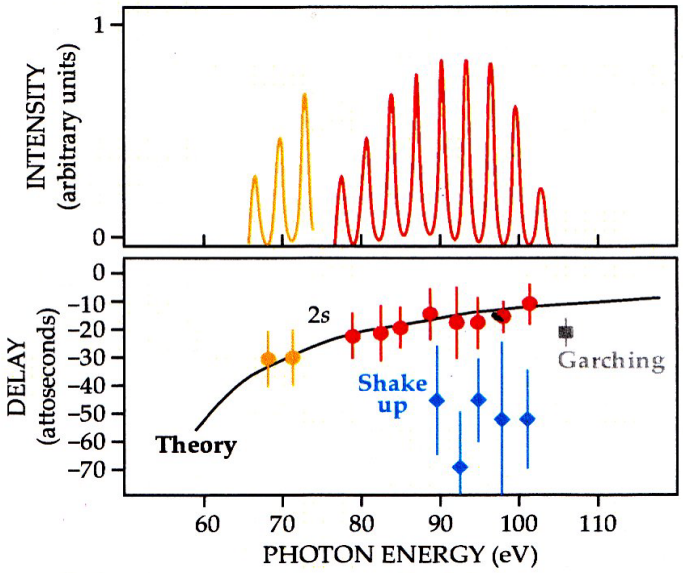

Figure 4. *Time delays in photoionization [29] of the 2p electron with respect to the 2s electron in the neon atom [DELAY = τ(2s) –τ(2p)]. In the upper part, the HHG spectrum used in the experiment is shown. Note that neon was used both for HHG and as the target in the magnetic bottle spectrometer marked MBES in Figure 3. The negative time delays for ionization of 2p with respect to 2s are shown as yellow and red dots. The experimental result from Krausz group in Garching is shown as a square [25]. The blue diamonds are measured time delay differences between shake-up and 2p ionization [DELAY = τ(2p→3p shake-up) – τ(2p)]. The black solid line is theoretical with many-body perturbation theory [29]. (Figure reproduced from ref. [31].)*

The very good agreement between the Lund experiment and theoretical calculations by many-body perturbation theory suggest that the pioneering result from the Krausz group [25] was affected by shake-up. The energy difference between a single photon removing an electron from 2*p* while at the same time promoting another 2*p* electron to the 3*p* subshell is only 7.4 eV more energy than direct ionization of a 2*s* electron. In the Lund experiment, the sideband signals from shake-up and 2*s* could be separated [29]. This would have gone unresolved in the Garching experiment. [25]

Applications in materials science

Studies of light-matter interaction in the attosecond time domain have recently expanded to new vistas, examining molecules, liquids and solids. As an example, we describe briefly an experiment that illustrates in a quantitative way how electron dynamics depend on the chemical environment

[32]. Figure 5 shows a setup of an attosecond interferometry experiment designed to measure the time delay between the photoemission from liquid water and from gaseous water.

At attosecond timescales, all types of structural dynamics are frozen, except that of the electrons, and the experiment hence allows the study of electron dynamics. As Figure 5 shows, an attosecond pulse train superimposed with a near-IR femtosecond laser pulse was allowed to interact with water in liquid phase as well as in gas phase. The photoelectrons were simultaneously emitted from water molecules in the liquid and gas phases, and the experiment showed a time delay of 50 to 70 attoseconds (as) between the photoelectrons from liquid water and those from gaseous water. This experiment hence enables a quantitative measure of the fact that electrons from liquid water arrive at the photoelectron detector later than electrons from gaseous water.

The slower motion of electrons from liquid water may feel intuitively reasonable, since these electrons have to travel through a more complex potential landscape, as compared to water molecules in gas phase. The experiment in Figure 5 showed this phenomenon in detail: the measurements could isolate the effect of solvation – water molecules interacting with nearby water molecules – as the dominant one for the time delay in the attosecond photoionization dynamics.

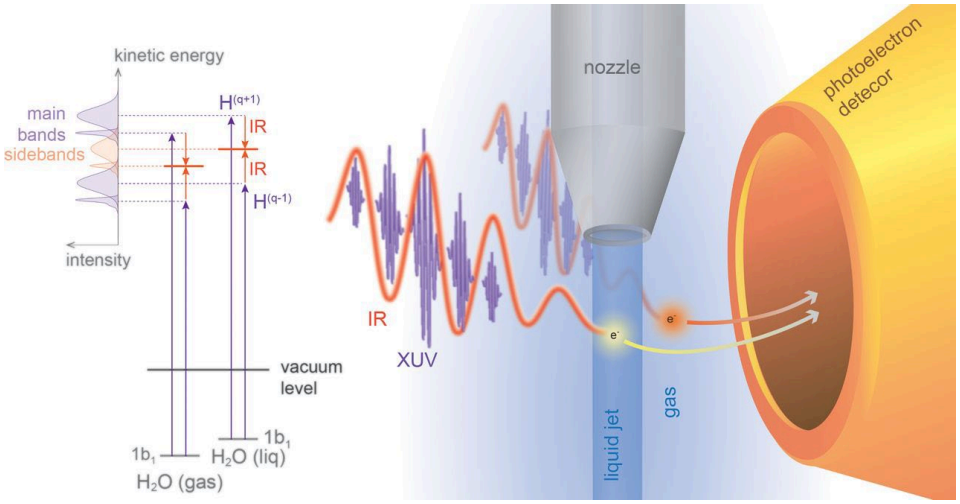

***Figure 5**. An attosecond pulse train (blue) composed of a few high-harmonic orders superimposed with a near-IR femtosecond laser pulse (red), interacting with a microjet of water. Photoelectrons are simultaneously emitted from the liquid and the surrounding gas phase and detected in the photoelectron detector. (Figure reproduced from ref. [32].)*

For solids, attosecond spectroscopy is expected to reveal a plethora of complex electron interactions, e.g. processes that include charge transfer and charge screening effects, image charge creation, and electron-electron scattering, as well as collective electronic motion. One such experiment with tungsten [33] addressed the complexity of electron interactions in solids. In a direct time-domain photoelectron emission study, with attosecond resolution, researchers could demonstrate a delay of approximately 100 as between the emission of photoelectrons that originate from localized atomic-like *4f* states, compared with those that come from itinerant conduction-band states. They used a so-called pump-probe experiment, where an initial light pulse triggers the dynamics of the tungsten metal, followed by a second light pulse that probes the transient state via a photoemission process.

This experiment [33] used the fact that the binding energy of the atomic-like *4f* states is distinctly different from that of the itinerant valence band states; this difference allowed the researchers to follow the distinct timescales of these unique types of electron states. They found that on average, photoelectrons originating from the localized *4f* states emerge from the tungsten surface approximately 100 as later than those originating from the delocalized conduction band. The observed delay effect in tungsten metal and in similar materials occurs during transport of the excited photoelectrons to the surface, which illustrates the possibility to directly observe features of electron wave packet propagation with attosecond precision.

The ability to shake the electrons in pump-probe experiments and to study the response on attosecond timescales offers tremendous opportunities to explore completely new physical phenomena. In materials science, so far, only the pioneering initial investigations of attosecond physics come to fruition, but thanks to the groundbreaking research of L'Huillier, Krausz and Agostini, one may anticipate many surprising and non-intuitive results to appear in the future. To imagine the surprises that attosecond dynamics of electrons may hold, it is instructive to rely on some of the unexpected dynamic responses we know that everyday objects have, for example, the Kapitza pendulum, with its seemingly miraculous ability to oppose gravitation (see e.g. https://www.youtube.com/watch?v=GgYABmG_bto).

Concluding remarks

This Scientific Background cannot give justice to the breadth of attosecond science. What started as the fairly narrowly focused field of multiphoton processes in atomic physics has now expanded towards many frontiers in molecular physics, physical chemistry, condensed matter physics and applied fields such as light-generation technology. And the first steps towards biological applications have been taken by the Krausz group in Garching.

By combining broadband optics, ultrafast laser sources, and precision femtosecond-attosecond field resolving technologies, the Krausz group has developed electric-field molecular fingerprinting that can detect changes in molecular composition of biofluids. This holds promise as a new *in vitro* diagnostic analytical technique to detect characteristic molecular of traces of diseases in blood samples [34]. The great advantage is that many molecules can be monitored at the same time, and the radiation is non-ionizing and therefore not harmful.

In further expanding the basics of attosecond science, important work has also been carried out by other groups. See those of Margaret Murnane and Henry Kapteyn at the University of Colorado, Boulder (for example, [35]), and of Ursula Keller at ETH Zurich [36].

A recent and comprehensive review article by Rocio Borrego-Varillas, Matteo Lucchini and Mauro Nisoli describes the Laureates' research and what it has catalysed [37]: in harnessing powerful laser effects to shift time to attosecond scales, they could see electrons move in atoms, molecules and matter in the condensed phase. This year's Nobel Prize in Physics opens windows that were unimaginable to Heisenberg, to explore phenomena that were previously impossible to observe.

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