A bright future for attosecond physics

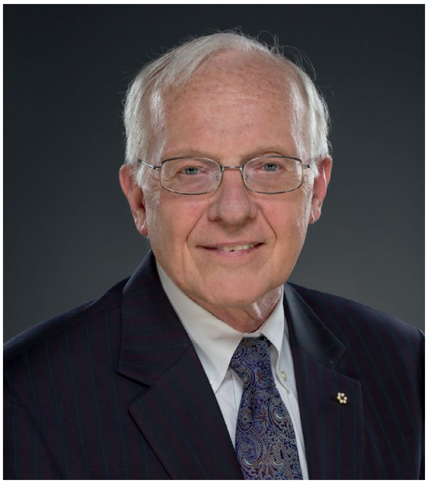
The 2022 Wolf Prize in Physics has been awarded to Paul Corkum, Anne L'Huillier and Ferenc Krausz for their pioneering contributions to ultrafast laser science. *Nature Photonics* spoke to them about the milestones, challenges and future opportunities for the field.

Can you give us a brief overview of what is attosecond physics?
FK: Attosecond physics is the science of atomic-scale electronic motions, often triggered by light. The force that visible/infrared light waves exert on charged particles varies within hundreds of attoseconds. Electrons can respond to these hyperfast variations by changing their state (position, velocity) on the same time scale.

PC: Attosecond physics originated by adapting ideas that were well-developed in plasma physics. This adaptation led directly to the recollision model that allowed us to unify previously poorly understood aspects of atomic physics (such as above-threshold ionization and high-harmonic generation). 22 years after the first experimental demonstration, attosecond physics has become a short-hand for many aspects of intense light interacting with matter, for extreme nonlinear optics, for high harmonics extending into soft X-rays, and for the fastest measurements that we, as humans, can make.

Why is it important to study physical phenomena at such short timescales?
ALH: Both the time scale and the energy of the attosecond pulses match the timescale of the interaction of light with matter, and especially the motion of the electrons in matter induced by photoabsorption. Studying these phenomena in time provides new insights on the physics of the interaction, which is of interest in atomic and molecular physics, in chemistry as well as in condensed-matter physics.

FC: Atomic-scale electronic motions are responsible for the generation of light, for the formation and break-up of chemical bonds, which changes the structure of biomolecules and their function in living systems, as well as for the fastest possible processing of information. If we wish to understand the origin of diseases at the most fundamental level or advance information processing to its ultimate speed limit, we need to understand the microscopic (better, nanoscopic) motion of electrons. These vital dynamics typically unfold within tens to hundreds of attoseconds and often evolve over tens to hundreds of femtoseconds.


Paul Corkum. Credit: Daniel Gamache


Anne L'Huillier. Credit: Kennet Ruona

PC: Not only are electrons fast, they are also important. Chemistry is governed by electrons while technology is often based on electrons, which enables us to influence these very important areas of technology through the measurements we make, as well as through the new way we learn to control electrons.

What made it possible to probe such short timescales?
PC: It's back to electrons again! Basically, to construct an attosecond pulse, we make use of the cycle time of a light wave, which is about 2,000 attoseconds. The light wave is a wave of force on a charged particle, and the electron, having low mass, can therefore respond on this time scale. When ordinary matter is ionized, we always know where the electron comes from, relative to the ion from which it was produced, so we can predict, if, when, and how the electron recollides with its parent ion. It is during this brief interaction between the newly born electron and its associated ion that the attosecond pulse is born. Since every atom is alike, the signal from each atom strengthens the signal, making a strong pulse — the attosecond pulse.

FK: To resolve processes in the attosecond domain we need to have attosecond-duration pulses at our disposal. High-order harmonic generation in atomic gases, pioneered experimentally by ALH and theoretically by PC, have spawn coherent extreme ultraviolet radiation. If properly controlled, the process generates a series of bursts of attosecond duration or, under special circumstances, as an isolated attosecond pulse. Many colleagues, including Pierre Agostini, Misha Ivanov, Maciej Lewenstein, Kenneth Kulander and Kenneth Schafer, have made seminal contributions to these advances.

What have been the key milestones in attosecond physics since the birth of the field?
ALH: I think the first milestone was 1987, which was the discovery of high-order harmonics. The second step was the understanding of the atomic physics part of the problem, in 1993/94, with the three-step model by PC and Ken Kulander and co-workers and then the quantum mechanical formulation by Maciej Lewenstein. The third step was 2001, where it was shown in the laboratory that indeed very short light pulses could be produced, both in a train, in the work by the group of Pierre Agostini, and then isolated, which was observed by the group of FK. 2010 is also an important date because it's when we

Ferenc Krausz. Credit: Peter Seidel

started measurements of photoionization time delays, which is such a nice application of attosecond physics.

PC: The field developed with the discovery of above-threshold ionization in the late 1980s, which was soon followed by high-harmonic generation. In the early 1990s, the recollision model explained and gave a prescription for how to generate and measure attosecond pulses. According to the model, underlying each attosecond photon was an attosecond electron of about the same energy recolliding with its parent ion. One could think of the photon as a proxy for the associated electron. In the late 1990s, laser technology reached the point where the carrier-envelope phase was stabilized, which allowed the production of identical laser pulses every time the laser fired. Every laser pulse could create an identical attosecond pulse. In the early 2000s, the proposed method for producing and measuring isolated attosecond pulses (and trains of attosecond pulses) was demonstrated. In the mid 2000s, ångström-scale imaging was added to the ability to make accurate time measurements, and in the early 2010s, high-harmonic generation was discovered in transparent solids. This opened a new method to study solids, their band structure, bonding network, and topology.

FK: Since the invention of attosecond tools and techniques, a great number of spectacular experiments have been performed with them. It is hard to assess objectively which of them deserves the attribute ‘milestone’. The new technology has provided, for the first time, direct, time-domain access to phenomena as fundamental as the decay of inner-shell vacancies in atoms, electron tunnelling through the barrier imposed by the atomic Coulomb potential, migration of electrons within molecules, optical-field-induced ionization and subsequent recollision of the freed electron with its parent ion, or intra-atomic and inter-atomic electron correlations. The listing could continue. Novel prospects have been opened by using the attosecond-controlled motion of recolliding electrons to combine atomic-scale temporal resolution with atomic-scale spatial resolution.

What are some of the fundamental questions still unanswered where attosecond techniques can contribute?

ALH: This is a difficult question. I think that attosecond techniques have only scratched the surface of the ultrafast response of matter to light excitation. Only a few systems have been studied, simple systems, like atoms. In molecules and condensed matter there’s still a lot to do. It’s only the beginning as I see it. We are still developing techniques and extending the parameter space of what we can study. Another direction that we are now taking is towards the measurement of the coherence of photoelectrons, thanks to attosecond techniques.

FK: How do electrons interact with each other, inside atoms, molecules, and — most importantly — inside solid matter? How do electron correlations affect processes underlying classical and quantum information processes? What is the ultimate limit of signal processing rate in classical computation? How can electron–electron and electron–lattice collisions be reduced or suppressed, thereby extending the time and spatial scale of ballistic charge transport for dissipation-free electronics? What is the maximum information content that electromagnetic waves from the infrared to the ultraviolet can deliver about the atomic structure and composition of large biomolecules?

PC: I believe that the most important problem that we can address is: “What is the ultimate time response of electronic matter?” Although the ultimate time response is a fundamental characteristic of electronic matter, we have not even yet attempted to measure it! When textbooks are written 100 years from now, the ultimate time response, and how it is measured, will certainly be discussed.

What practical applications are expected for attosecond science beyond their fundamental importance?

ALH: I can see a real push now for applications of high-order harmonics, as these provide a broadband, coherent source with good spatial properties.

This is interesting for example for the semiconductor industry, which needs a coherent source in the extreme ultraviolet for metrology or imaging. Laser companies also start selling high-order harmonic sources, as an addition to their lasers.

I remember at the end of the 90s we had understood that high-order harmonics would correspond in the time domain to short attosecond light pulses, but how to measure them and how to use them seemed completely out of reach. I think that this field of research is evolving in a very promising way.

PC: We can cover the spectrum from THz to 100 PHz, and even beyond, with synchronized beams, and in the visible and IR the pulses can be intense. This is an unrivalled toolbox for studying nature! One forefront of attosecond science is to combine the surface-control methods of modern electronics with intense light. This will lead to focussed attosecond pulses where the focus can be on the 100-nm scale. If hybrid attosecond pulse generation can be developed, then, focussing might be extended to shorter wavelengths and smaller sizes, perhaps to 10 nm. This would give a new tool for interacting light and matter, where we can obtain spectroscopic information, and even exercise control, on the nanometre scale.

FK: The first generation of attosecond metrology (AM1.0), based on extreme UV light, relies on multi-metre-scale vacuum beamlines. The cost and complexity of such systems have severely hampered their proliferation and prohibited widespread use of this powerful measurement technology up to the present day. More fundamentally, extreme-UV pulses are unable to penetrate condensed matter more than a few nanometres and pulses with photon energies of >>10 eV cannot directly trigger/probe valence–conduction band transitions.

Next-generation attosecond metrology (AM2.0) overcomes all these shortcomings. AM2.0 is based on the attosecond-varying controlled force exerted on electrons by the controlled electric field evolution of visible–infrared waveforms. (Near-)octave-spanning laser pulses can carry a substantial fraction of their energy in a single half cycle of their field oscillations. Such an isolated field peak is able to inject mobile charge carriers into the valence and conduction bands within less than one femtosecond. Sub-fs carrier injection opens a new frontier in interrogating ultrafast electron dynamics in solid matter, including all those that underlie contemporary electronics and optoelectronics. AM2.0 will play the role for future THz-to-PHz electronics that oscilloscopes have been playing for contemporary electronics. High-fidelity and -sensitivity sampling of electric fields by AM2.0 also holds promise for opening new routes to probing the molecular composition of biological systems, thereby probing changes in the physiological states of living organisms for health monitoring and disease detection.

What are the outstanding problems, either fundamental or technical, that the field is still facing?

FK: The laser technology underlying the attosecond-shutter-speed 'camera' is still far too complex to allow the fastest metrology in the world to proliferate into all those, either basic-science or real-world, applications. Moreover, improved temporal resolution has generally come at the expense of reduced sensitivity in measuring physical observables. Single-cycle waveforms from mode-locked laser oscillators will pave the way towards attosecond metrology at MHz pulse repetition rates (AM3.0) with much increased sensitivity and reduced complexity. Petahertz oscilloscopy is still in its infancy, but on the horizon.

ALH: If I'm talking about attosecond pulses by high-order harmonic generation, one fundamental problem is that the conversion efficiency from the IR to the extreme UV or X-ray is low, and this is a fundamental limit. It's due to the single-atom response. These pulses are not intense enough to do nonlinear processes, except at low photon energy, and it really limits what you can do with them. It is a fundamental issue, inherent to the physics of the process. Then of course you always have technical issues related to stability and to the properties of the pulses. If you take the next step, you want to focus these pulses, but then you realize that you get a lot of chromatic aberrations. This is inherent to the process, so in a way it's fundamental and it has to be solved to achieve a small focus and still conserve the pulse duration. There are still many things to solve.

Interviewed by Giampaolo Pitruzzello

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