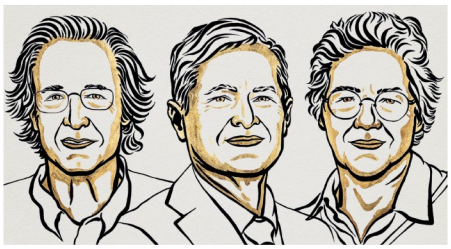
https://doi.org/10.1038/s41567-023-02305-y

Light as fast as electrons

The 2023 Nobel Prize in Physics has been awarded 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".



If scientists got to pick a superpower, many physicists and chemists would want to be able to directly watch electrons moving around atoms, molecules and condensed-matter systems. The generation of attosecond light pulses, which was awarded this year's Nobel Prize in Physics, has brought science closer to this goal than ever before.

For a long time, femtosecond pulses, enabled by the chirped pulse amplification that won half of the Nobel Prize in Physics in 2018, seemed to be the shortest pulses experimentalists were able to produce. However, the natural timescale of electron dynamics is on the order of 20–200 as, which is too fast to be captured even by the 5-fs pulses that held the world record for many years. But sources using chirped pulse amplification do not only emit short pulses, they can also generate sufficient peak powers for highly nonlinear processes, such as high-harmonic generation (HHG)[1]. Exploiting HHG became crucial to the generation of attosecond pulses.

Harmonic generation is a nonlinear process in which N incident photons with the same frequency combine to the N-th multiple of the original (fundamental) frequency. The simplest picture of nonlinear optics suggests that the efficiency of this generation falls rapidly with the number of photons combined, but early HHG experiments with infrared lasers[1] found that this is only the case up to a certain point, above which the intensity of the harmonics remains roughly the same until it begins to fall again.

This unexpected behaviour required a new theory, which was found in the three-step model[2] – a semiclassical description that has served the attosecond community well. First, the laser creates tunnelling ionization, after which the laser field accelerates the electron away from the ion until the field reverses in the next half cycle of the light field and the electron comes back. The recombination of ion and electron releases the kinetic energy gained by the electron while free as electromagnetic radiation, typically in the extreme ultraviolet range.

But how does HHG help to create attosecond pulses? Simply put, in order to compress a pulse in the time domain one needs to stretch it in the spectral domain (because of the uncertainty relation between energy and time). The plateau in the HHG spectrum provides a sufficient number of phase-locked frequency components to form attosecond pulses[3]. Of course, technical finesse is needed to produce such short pulses, typically from gas-filled fibres, which today can be of a duration shorter than a single optical cycle.

Although technical advances were invaluable in the development of attosecond pulses, it also required significant insights into the physics at play at the intrinsic timescale of atoms and molecules. Interestingly, attosecond science had its eyes on using the short pulses to explore fundamental science from the start[4] – allowing the study of questions that were previously inaccessible.

For example, before the establishment of attosecond science, it was commonly assumed that the photoelectric effect – the emission of electrons from a material hit with light – occurred instantaneously, and most research focused primarily on the energetics of the process.

The possibility of a delayed response was first proposed in the 1950s, but the timescale was so short that it took until the advent of attosecond pulses before researchers could delve into the problem. In 2010, some groundbreaking measurements of time delays between photoemission processes in neon atoms[5] not only confirmed that photoemission is not immediate, but showed a small (20 as) time delay between the emission of electrons from different orbitals using the same ultrafast pulse. This result established photoemission as the result of complex multielectron correlations rather than a single-electron process.

The ability to probe attosecond time delays has been instrumental in the characterization and reconstruction of quantum mechanical wave packets in photoionization processes. Molecular pump–probe experiments provided the first time-resolved detection of electronic charge localization in simple diatomic molecules[6], and isolated attosecond pulses opened the door to the monitoring and control of ultrafast electron dynamics in more complex molecules[7]. This propelled the nascent field of attochemistry into uncharted territory.

Significant progress has also been made in the generation of extreme ultraviolet radiation in nongaseous media. In particular, the realization of HHG in solids[8] spurred a lot of interest in elucidating its underlying mechanism, which might be crucial for developing compact sources of attosecond light and metrological applications. Moreover, attosecond pulses offer a means to control the electronic properties of materials on an ultrafast timescale. For instance, attosecond spectroscopy has enabled the manipulation of the electronic structure and polarizability of dielectrics in a reversible fashion[9].

This year's Nobel Prize rewards a field that produced a large number of fundamental contributions, of which we are only starting to see the potential for applications. The future looks bright for attosecond science, and it might be coming in much less than a blink of an eye.

Published online: 9 November 2023

References

1. Ferray, M. et al. *J. Phys. B: At. Mol. Opt. Phys.* **21**, L31 (1988).
2. Corkum, P. B. *Phys. Rev. Lett.* **71**, 1994–1997 (1993).
3. Paul, P. M. et al. *Science* **292**, 1689–1692 (2001).
4. Hentschel, M. et al. *Nature* **414**, 509–513 (2001).
5. Schultze, M. et al. *Science* **328**, 1658 (2010).
6. Sansone, G. et al. *Nature* **465**, 763–766 (2010).
7. Calegari, F. et al. *Science* **346**, 6207 (2014).
8. Ghimire, S. et al. *Nat. Phys.* **7**, 138–141 (2011).
9. Schultze, M. et al. *Nature* **493**, 75–78 (2013).