

Physics Nobel 2023: the study of physical phenomena with Attosecond time resolution

Introduction

Recent advancements in coherent light sources have enabled the generation of incredibly brief light pulses, clocking in at durations as fleeting as just over 100 attoseconds ($1 \text{ as} = 10^{-18} \text{ s}$) within the extreme ultraviolet (EUV) range of the electromagnetic spectrum. This significant stride in laser technology has opened avenues for directly influencing the rapid movement of electrons within molecules. Attosecond science has produced a number of significant discoveries in the realms of atomic, molecular and solid-state physics. Noteworthy strides in creating and employing sub-femtosecond extreme-ultraviolet pulses, implementing experimental methods capable of tracing electron dynamics in quantum

systems across time, and crafting advanced theoretical frameworks for deciphering experimental findings have fuelled a sustained and growing interest in attosecond phenomena. The 2023 Nobel Prize in Physics has been granted to Pierre Agostini of Ohio State University (USA), Anne L'Huillier of Lund University (Sweden), and Ferenc Krausz of the Max Planck Institute of Quantum Optics, Garching and Ludwig-Maximilians-Universität, Munich (Germany), in acknowledgement of their extensive contributions to attosecond science.

Understanding the importance of light pulses in studying time-domain phenomena arises from realizing that swift events seamlessly merge when observed by humans, much like the continuous flow perceived in a cinematic film composed of static im-

ages. To effectively examine fast events, specialized tools such as light pulses become essential, which can conveniently be derived from laser systems. The advent of the first functional laser by Theodore Maiman in 1960, utilizing a synthetic ruby crystal as the amplifying medium, signalled a pivotal moment in the interaction between light and matter. This breakthrough sparked an upsurge in research and experimentation, exploring a wide array of materials and techniques to generate laser light. The development of Q-switching around 1962 enabled the creation of nanosecond (10^{-9} s) laser pulses, while subsequent breakthroughs in mode-locking allowed for the generation of even briefer pulses, reaching picoseconds (10^{-12} s) and femtoseconds (10^{-15} s). It is worth noting that fast electronic devices operate within the nanosecond timeframe, molecular vibrations occur in the picosecond realm, and electron-electron thermalization in an excited lattice happens in the femtosecond domain.

The Ti : Sapphire laser, frequently utilized to create femtosecond laser pulses, acts as an excellent pump laser for high harmonic generation in gas media, allowing the production of attosecond light pulses. It is significant to highlight that an attosecond is incredibly brief, with as many attoseconds in a second as there have been seconds since the universe's inception. In the electron domain, alterations happen within attoseconds, and the light pulses resulting from the Nobel laureates' research have played a pivotal role in capturing visuals



Pierre Agostini



Anne Geneviève L'Huillier



Ferenc Krausz

of such electronic processes taking place within atoms and molecules.

High harmonic generation and attosecond pulse generation

The simple semi-classical approach, initially presented by Corkum¹ and commonly referred to as the three-step model, is frequently employed to elucidate the fundamental physical mechanisms behind high harmonic generation (HHG). According to this model, when a femtosecond laser pulse, known as the 'driving pulse', is concentrated onto a gas target with a peak intensity ranging from 10^{13} to 10^{15} W/cm², the electric field of the laser significantly alters the Coulomb potential felt by the electrons in the outer shell. This alteration forms a potential barrier that allows an electron to tunnel through. Tunnelling of electrons primarily occurs within a narrow time frame around each peak of the electric field because the rate of tunnel ionization exponentially depends on the field's strength. Thus, the initial stage in HHG involves the purely quantum mechanical phenomenon of tunnel ionization.

Following tunnel ionization, the released electron navigates through the continuum, influenced by the electric field of the driving pulse. Its movement can be reasonably explained using classical principles, primarily Newton's second law, serving as an initial approximation. Initially accelerated away from the parent ion by the linearly polarized driving field, the freed electron decelerates as the electric field shifts direction. Eventually, it reverses its trajectory and returns to its original position, which is termed recollision. The electron's duration in the continuum between ionization and recollision varies based on its specific path. When the recolliding electron recombines with the parent ion, the kinetic energy gained during its continuum journey is discharged as a burst of high-energy photons with subcycle durations. This photon burst's spectrum lies in the EUV region, and it can span energies from a few electronvolts to several hundred electronvolts, corresponding to multiple higher-order harmonic frequencies of the driving field. The entire sequence, comprising ionization, continuum motion and recombination, recurs every half-optical cycle since electrons can be liberated near any of the field's peaks. HHG forms the foundation for producing attosecond light pulses, which are temporally separated from each other by half an optical cycle of the driving radiation.

The Nobel Laureates

Born on 23 July 1941 in Tunis under the French protectorate of Tunisia, Pierre Agostini is a distinguished French experimental physicist, currently serving as Emeritus professor at Ohio State University. Renowned for ground-breaking contributions to strong-field laser physics and attosecond science, Agostini commenced his research journey at CEA Saclay in 1969 after his doctoral studies, where he remained until 2002. During this time, Agostini collaborated with Gérard Mainfray and Claude Manus in their laboratory, exploring multiphoton ionization using high-powered lasers. A significant achievement occurred in 1979: the observation of above-threshold ionization (ATI) in Xenon gas². Continuing his research pursuits, Agostini reached a major milestone in 2001 by generating a sequence of attosecond pulses through HHG³. He is also credited with developing the RABBITT (reconstruction of attosecond beating by interference of two-photon transitions) technique for characterizing attosecond light pulses³.

Anne Geneviève L'Huillier, born on 16 August 1958, is a French-Swedish physicist and a professor of atomic physics at Lund University. Leading a research team in attosecond physics, she focuses on observing electron movements in real-time to comprehend chemical reactions at the atomic scale. Widely acclaimed for her contributions in both experimentation and theory, L'Huillier is recognized for laying the groundwork for the emerging field of attocchemistry. In 1987, L'Huillier was instrumental in the ingenious discovery of high harmonic generation of 1064 nm laser light in rare gases. This phenomenon stemmed from the interaction between laser light and atoms in the gas⁴. Subsequently, she continued her exploration of HHG, paving the way for subsequent breakthroughs. L'Huillier explained the observed Plateau in HHG using numerical TDSE (time-dependent Schrödinger Equation) with the Single Active Electron (SAE) approximation. Collaborating with Lewenstein and Corkum, she also played a crucial role in developing the comprehensive quantum theory of the process. In 2003, L'Huillier's research group set a world record by generating the shortest light pulse, lasting 170 attoseconds.

Ferenc Krausz, born on 17 May 1962, is of Hungarian–Austrian descent. Currently, he serves as a director at the Max Planck Institute of Quantum Optics, Garching, and holds a professorship in experimental

physics at the Ludwig Maximilian University in Munich. Krausz earned his Ph.D. in physics from Eötvös Loránd University in Budapest, Hungary. Since then, his primary focus has been on advancing laser technology to generate short bursts of light. Krausz has made significant contributions in this domain, particularly in developing few-cycle laser pulses, broadening the spectrum of high harmonics, and producing the first isolated attosecond light flashes (IAP). His pioneering work involved measuring IAP using cross-correlation and atto-streaking⁵. Additionally, he expanded these methodologies into attosecond metrology, using attosecond pulses to observe electron dynamics in diverse systems, including biological samples. Krausz has played a crucial role in advancing research on refining laser sources and techniques for controlling the waveform of light pulses. His endeavours have led to the development of laser sources characterized by unparalleled precision and control over temporal properties, enabling the exploration of timescales previously beyond reach.

Outlook

Announcing the prize, Eva Olsson, Chair of the Nobel Committee for Physics, said: 'We can now open the door to the world of electrons. Attosecond physics gives us the opportunity to understand mechanisms that are governed by electrons. The next step will be utilising them.' Obviously, attosecond physics offers several new avenues of research into hitherto uncharted territories, and it can be expected that future results from some of these areas will be fascinating and non-conventional.

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Reji Philip, Light and Matter Physics Group, Raman Research Institute, Bengaluru 560 080, India.
e-mail: reji@rri.res.in