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PERSPECTIVE

Hearing the Heartbeat of Atoms: Unveiling Attosecond Horizons

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The 2023 Nobel Prize in Physics spotlights the techniques to generate attosecond light pulses. The generation of attosecond pulses heralds a new era in understanding electron dynamics. This perspective traces the evolution of ultrafast science, from early microwave electronics to the recent breakthroughs in attosecond pulse generation and measurement. Key milestones, such as high harmonic generation, the RABBITT method, attosecond streaking camera, etc, illuminate our journey toward capturing the transient electron motions in atoms. Recent discoveries, including zeptosecond delays in H_2 single-photon double ionization and the potential of attosecond "electron" pulses despite challenges, etc., hint at an exciting future for ultrafast studies.

Unveiling the Dance of Electrons: The Birth of Attosecond Science

The Nobel Prize in Physics for 2023 was awarded to Pierre Agostini, Ferenc Krausz, and Anne L'Huillier for pioneering methods to generate attosecond light pulses [1]. These groundbreaking techniques offer unprecedented time resolution to access ultrafast dynamics of electrons in materials, marking a new chapter in the exploration of the "world of electrons".

Our world thrives on diverse material properties, such as conductivity, insulativity, magnetism, superconductivity, mechanical strength, etc. These properties, however, hinge on the dynamics of electrons. At the atomic level, electrons orbiting nuclei determine the very nature of chemical bonds and, subsequently, the macro properties of matter. Given this, to grasp the electron dynamics in atoms, we need an immensely high-resolution temporal "camera". With the Bohr model, it is deduced that an electron's journey around its 1s orbit in a hydrogen atom roughly takes about 150 as [2]. Attosecond represents a billion-billionth of a second—a truly atomic time scale. Tracking such evanescent electron motions demands the probes that operate in the realm of attoseconds.

In the early 1960s, microwave electronics was central to ultrafast science [3]. However, due to Coulomb interactions, the time scale measurable is limited to the picosecond range. On the other hand, the ultrashort laser pulse technique has seen very impressive advances. The journey of ultrashort pulse generation has its origins in the development since mode-locking techniques in the 1960s [4]. Early mode-locked lasers, using active devices like acousto-optic modulators, produced picosecond pulses. After that, it was the advent of passive mode locking with saturable absorbers that led to the generation of subpicosecond pulses in the 1970s [5]. In 1974, the dye lasers mode-locked with Kerr lens was proved to be instrumental in producing femtosecond pulses [6]. The transition from picoseconds to femtoseconds was further enhanced with the introduction of chirped pulse amplification by Strickland and Mourou in 1985 [7], which help great enhance the peak power and intensity of femtosecond lasers. The discovery eventually earn them the winners of 2018 Nobel Prize in Physics [8]. However, the achievable shortest laser pulse is about 4 fs via the laser-optical technique [9,10]. To generate even shorter pulses, one needs to shift the paradigm. These efforts yielded limited results until 2001 when attosecond science, made possible by "lightwave electronics", became the Nobel-prized innovation.

The essence of lightwave electronics is using a controllable strong laser field to manipulate the electron–atom interactions, producing shorter light pulses [3]. In 1979, Pierre Agostini et al. discovered a benchmark strong-field phenomenon called "above threshold ionization (ATI)" [11]. Here, the electrons in atoms absorb multiple photons, being ionized beyond the atomic ionization energy. Another important discovery in 1987 by A. L'Huillier et al. revealed that the rare gas atoms (like Xe, Kr, and Ar) exposed to an infrared light field exceeding 10^{13} W/cm^2 would yield the high-order harmonic, termed "high harmonic generation (HHG)" [12]. This highly nonlinear process, along with ATI, was very intuitively tied to attosecond pulse generation, leading researchers to delve deeper into the theoretical understanding. In the early 1990s, a collaboration involving Kulander, Anne L'Huillier, and Kenneth Schafer worked on the theoretical models for HHG [13]. Kulander introduced a semiclassical model termed "the re-collision model" at a conference in Belgium [14], which was published in 1993 [15]. Around the same time, Paul Corkum proposed the 3-step model to detail the description of the ATI and HHG process [16]. In 1994, a

quantum theory was developed, confirming these earlier semi-classical explanations [17]. The re-collision model provides a very clear picture to calculate the cutoff frequency for HHG, namely, $E_c = I_p + 3.17U_p$, where I_p and $U_p \propto \lambda^2 I$ are the ionization potential of atoms and ponderomotive energy in the oscillating driving field, respectively [18].

Upon theoretically unveiling the physics behind HHG, researchers then began to explore the ideas for generating attosecond pulses. Remarkably, Farkas and Toth mentioned the possibility of generating attosecond pulses using HHG for the first time in 1992 [19]. L'Huillier and Lewenstein conceived the first proposal to generate the attosecond pulse train in 1996 [20]. Following the original ideas of the 2-color photoionization by Cionga et al. and Véniard et al. [21,22], Agostini's team demonstrated the so-called "Reconstruction of Attosecond Beating by Interference of Two-photon Transitions (RABBITT)" method experimentally to measure the temporal width of attosecond pulses in these trains [23]. Aiming for single attosecond pulses, Corkum, Burnett, and Ivanov suggested constraining HHG within a single cycle [24]. Concurrently, Schafer and Kulander proposed harnessing the cutoff region harmonics to achieve the isolated attosecond pulse [25]. In the same year, Kapteyn–Murnane's group demonstrated the possibility of generating isolated attosecond pulses using the cutoff of the harmonics generated by a few-cycle pulse [26]. Experimental techniques advanced soon after, with teams led by Krausz in Vienna and Nisoli in Milan making the breakthroughs [27,28]. Finally, in 2001, Agostini's group in Paris-Saclay generated a 250-as pulse sequence, whose pulse length is characterized by the RABBITT method [29]. Meanwhile, in Vienna, Krausz's team produced a 650-as isolated pulse based on the theory proposed by Shafer and Kulander. Since then, the era of attosecond is coming [30].

From Mystery to Half-mastery: Rapid Advancements in Attosecond Techniques Since Its Birth

Before and after the first successful generation of isolated attosecond pulses, other experimental methods, such as polarization gating [24,31,32], double optical gating [33,34], ionization gating, and photonic streaking [35,36], have progressively emerged. Notably, the amplified few-cycle femtosecond pulses with the stabilized and controlled carrier-envelope phase were realized in 2003 [37], leveraging the contribution from another Noble Prize in Physics in 2005 [38].

Around 2010, a palpable improvement occurred. Until then, the Ti:sapphire laser was the go-to driving light source, constraining the HHG cutoff energy to roughly 100 eV and a repetition rate of ~1 kHz [39]. To generate the shorter attosecond pulses, the mid-infrared driving lasers rise in prominence. Thanks to the optical parametric amplifier [40], the harmonic photon energy, can be extended beyond 300 eV, nearly accessing the water-window spectral region [41]. Other methods like optical parametric chirped pulse amplification [42], and frequency-domain optical parametric amplification [43] also arose. Yet, this was not without challenges. The conversion efficiency $\eta \sim \lambda^{-6}$ faced a sharp decline as the driving wavelength increased [44,45]. Researchers attributed this to electron wavepacket spreading and the increasing harmonic order. Significant efforts were made to optimize the waveform of the driving laser pulse [46,47], increasing its energy [48] and the repetition rate [49]. While simply amplifying the driving laser's power seemed an obvious choice to intensify the attosecond pulse, the ionization would pose limitations. Researchers then veered toward a loose-focusing methodology to expand the active area [50,51]. Due to the higher electron density and harmonic generation efficiency, HHG via liquids and solids, together with its optical properties also attracts much attention [52–57]. Beyond HHG, other methods such as x-ray free electron laser and nonlinear Compton scattering were developed to generate attosecond pulses in x-ray [58–60] or γ-ray range [61]. To date, the isolated attosecond pulse has been shortened to a pulse duration of approximately 50 as [32,62]. A maximum repetition rate of a few MHz and a maximum average flux of about 4.4×10 photons/s/1%Bw are accessible [63]. On another track, the proposals and experiments have been advanced for generating attosecond pulses with distinct spin and orbital angular momentum structures [64–77]. These structural light fields enhance the potential functionalities of attosecond pulses.

With the advancement of attosecond pulse generation technology, the measurement techniques for these pulses have significant progress. In addition to RABBITT and attosecond streak camera [78,79], U. Keller and colleagues from ETH Zurich demonstrated the "attosecond angular streaking method" based on ionization in few-cycle elliptically/circularly polarized light fields in 2008 [80]. This method is also referred to as the "attoclock". In 2018, this technique was further developed into the so-called "dual-pointer" attoclock by our group from PKU China [81]. The RABBITT approach can be integrated with the streaking method, leading to the FROG-CRAB method, which is capable of not only reconstructing attosecond pulses but also the driving infrared light [82]. Besides these "ex situ" techniques, methods that do not require a second gas target ("in situ" methods) have been introduced. For example, utilizing a 2-color light field breaks the symmetry of electron trajectory, leading to the generation of even-order high harmonics [83]. The same analysis akin to the RABBITT method would provide temporal information about attosecond pulses. Another promising technique using a 2-color field is the "petahertz optical oscilloscope" [84]. The "photonic streaking" method, also known as the "attosecond lighthouse", has garnered much attention due to its capability to select single pulses from an attosecond pulse train [85–87]. Lastly, attosecond transient absorption spectroscopy, one of the attosecond measurement techniques involving the pump-probe process, has been experimentally realized in several laboratories [88–92].

Since its infancy, the technique of attosecond generation has been applied to the study of various ultrafast dynamical processes [93–95], most typically trying to address questions concerning "ionization time" and "tunneling time" [96]. Concepts such as "Wigner time delay", and "continuum-continuum time delay" have been studied, which are crucial for understanding the fundamentals of quantum physics [97–101]. Beyond the study of ionization phenomena in atomic gases [102], attosecond science has also been employed in the research of dynamics in molecules [103–108], liquids [109,110], and other condensed matter systems [109,111–115]. On another front, attosecond pulses are also employed to comprehend and modify the functioning of chemical–biological systems [116–118].

Recently, the fusion of attosecond science and quantum optics has become a focal point of recent research [119–121]. Here, the driving light is interpreted quantum-mechanically, shedding light on quantum effects that had been previously neglected. Pioneered by Gorlach and his team, a theoretical framework where HHG through quantum light source exhibits marked differences from those generated classically was proposed [122]. Particularly, the

resulting harmonic spectrum extends its cutoff by an impressive factor when irradiated using this quantum source, marking a notable stride in the realm of quantum optics. Parallelly, Andrea Pizzi and colleagues unveiled that when quantum-correlated atoms are subjected to intense laser fields, the emitted harmonics, too, bear the quantum correlations [123]. These harmonics exhibit exotic quantum features, deviating significantly from classical expectations. These studies not only advance our comprehension of HHG within a full-quantum framework but also pave the way for novel experimental methodologies that may redefine quantum optics with attosecond photons. As we continue to perceive quantum worlds, such pioneering research offers promising horizons.

The Flash Future: Attoseconds Boosting Ultrafast Science

The winding evolution of attosecond science, from infancy to thriving, undeniably opens a new page on contemporary physics. The past 2 decades have witnessed this field transcend from mere theoretical possibilities to fruitful experimental outcomes, greatly furthering our grasp of ultrafast electron dynamics. Each challenge overcome becomes a stepping stone to the next horizon.

The present breakthrough promises even more exciting vistas. As we further refine and perfect attosecond pulse generation and measurement techniques, our capability to explore and manipulate the ultrafast world will be enriched. This, in turn, has profound implications for diverse domains, enriching our collective knowledge base. Thus, it can be confidently asserted that attosecond pulse technology will be pivotal in advancing tomorrow's science. For instance, compressing pulse duration even further and achieving higher repetition rates, ultimately realizing true attosecond lasers, will vastly enrich the tools available for studying atomic-molecular physics, condensed matter physics, and material science. Pushing pulse compression to atomic characteristic time (~24 as) will facilitate answering fundamental questions in quantum physics. Structural attosecond light can be instrumental in understanding and modifying biological and chemical chiral molecules. Additionally, the pursuit of higher power and photon flux in attosecond pulses will refresh the field of high-energy physics. Last but not least, as the integration of attosecond science and quantum optics rises, we earn a deeper understanding of light–matter interactions in the quantum domain.

People never stop probing into the material world on increasingly finer temporal and spatial scales. In 2020, R. Dörner's group from the University of Frankfurt reported the observation of a time delay of 247 zs (1 zs = 10^{-21} s) between electrons emitted from different centers of the H_2 molecule [124]. This delay essentially reflects the time taken for light to traverse from one end of an H atom to the other in the H_2 molecule. As attosecond physics flourishes, the emergence of zeptosecond physics beckons.

Electron pulses, much like their optical counterparts, can be compressed into an atomic time scale [125–129]. After interacting with an optical near field, these free electrons, upon traveling a certain distance, evolved into a train of temporal attosecond pulses. This is achievable in 4-dimensional ultrafast transmission electron microscopy [130,131]. In the realm of free electrons, there exists a method analogous to RABBITT for tomography [132]. Nowadays, many proposals and experiments regarding attosecond electron pulses have been demonstrated [133–139]; however, due to the Coulomb repulsion among electrons, their practical application remains a challenge.

The road ahead, while challenging, gleams with the promise of further revelations and deeper insights, ensuring that the "world of electrons" persists in the dynamic and ever-evolving frontier of ultrafast science.

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