

# Attosecond Optics-from Genesis to Revelation

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## Abstract

Attosecond optics emerged in 2001. It deals with the generation, characterization and application of extreme ultraviolet pulses shorter than one optical cycle of visible light. The duration of isolated attosecond pulses has recently reached 67-as.

## I. INTRODUCTION

Since the first demonstration of the laser in 1960, the duration of coherent light pulses has been dramatically reduced to improve the temporal resolution of ultrafast imaging and spectroscopy. Laser pulses less than 10 fs could be generated by the mid-1980s [1] and became important probes for studying rotational and vibrational dynamics in molecules. Attosecond pulses are needed to “freeze” electronic motion in atoms, molecules and condensed matter. However, generating such extremely short pulses directly from visible lasers was a grand challenge at that time because of the limited spectral bandwidth. For about 15 years, only minor progress was made in reducing the laser pulse duration, as depicted in Fig 1.

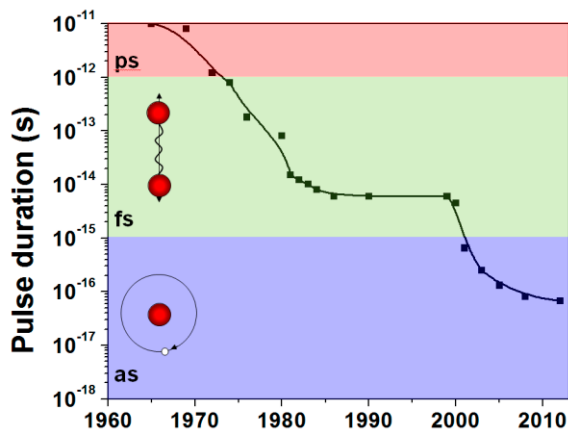


Fig. 1. Evolution of the ultrafast lasers in 50 years.

## II. THE ATTOSECOND REVOLUTION

The attosecond barrier was broken in 2001 by using a nonlinear optical process called high-order harmonic generation [2, 3]. High harmonic generation was discovered in the late 1980s [4, 5]. When the intensity of a short pulse laser interacting with noble gases reaches

$10^{13}$  to  $10^{15}$  W/cm<sup>2</sup>, multiple high-order harmonics of the laser with comparable intensity can be emitted. This is drastically different from the perturbative harmonic generation process at lower laser intensities. By 1997, the spectral range covered by the high harmonic radiation already extended to soft x-rays [6, 7], which can support extremely short attosecond pulses.

### A. Three-step Model and Strong Field Approximation

Another prerequisite of attosecond pulse generation is that the spectral phase of the high harmonic light must be well behaved. This is indeed the case as explained by a semi-classical model [8, 9]. The model vividly describes how an electron in an atom moves under the influence of strong external field during one laser cycle, as illustrated in Fig. 2. First, the electron tunnels out the potential barrier formed by the superposition of the laser field and the atomic Coulomb field. Then the freed electron gains a large kinetic energy (hundreds of electron volts) from the laser field. Finally, it recombines with the parent ion that leads to the emission of extreme ultraviolet or soft x-rays. The semi-classical model shows that only electrons ionized during a fraction of laser cycle can return, thus the emission time is shorter than the laser cycle. This picture was validated by a more comprehensive quantum model based on the strong field approximation [10, 11].

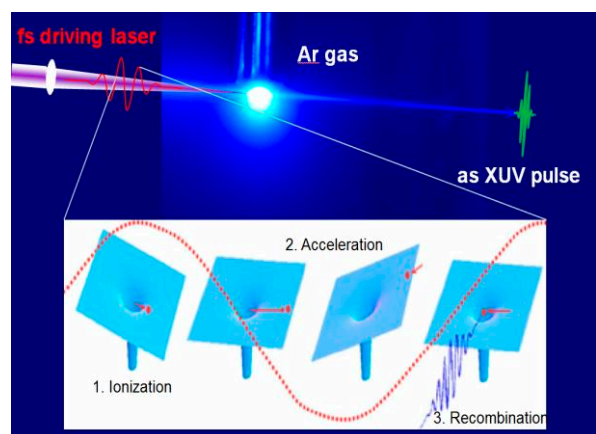


Fig. 2. Semi-classical model of attosecond pulse generation.

### B. Isolated Attosecond Pulses and Temporal Gating

The three-step model reveals that the high harmonic frequency comb generated with a multi-cycle

femtosecond laser results from the interference of many attosecond pulses separated by half a laser cycle, which was later confirmed by experiments [2]. For attosecond pump-attosecond probe experiments, it is desirable to use single isolated attosecond pulses so that the starting time of the process as well as the time that the system is probed are well defined. Various gating schemes have been invented to select a single attosecond pulse from the pulse train, such as amplitude gating, polarization gating and Double Optical Gating (DOG) [12]. The shortest light pulses, 67 as, was generated with the DOG method [13], as shown in Fig. 3.

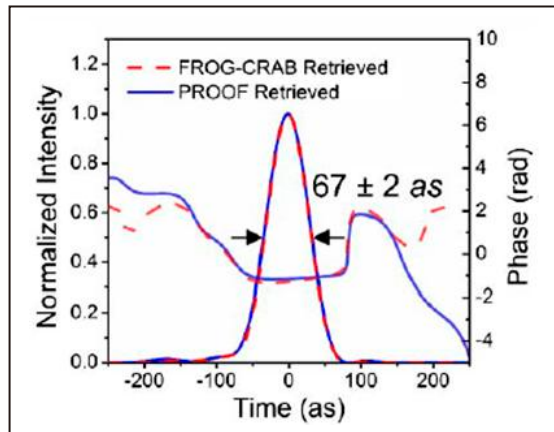


Fig. 3. Isolated attosecond pulse generated by DOG.

### III. NEW CHALLENGES

Over the last ten years, attosecond pulses have already been demonstrated to be a powerful tool for studying electron dynamics [14]. However, due to the low conversion efficiency of the high harmonic generation process, the photon flux of the isolated attosecond pulses is rather low (pJ to nJ). As a result, almost all attosecond time-resolved experiments have been conducted by synchronizing a strong femtosecond laser with the weak attosecond light source in the target medium. Attosecond streaking and attosecond transient absorption that make use of such combinations have been developed [14-16]. Although interesting sub-cycle physics has been uncovered, the interpretation of the data can be rather difficult [17].

Much higher flux ( $\mu\text{J}$  per pulse) is required for conducting true atto pump-atto probe experiments [18]. Generating such high power isolated attosecond pulses is one of the main thrusts in attosecond optics research. It has been demonstrated that isolated attosecond pulses can be generated from mJ level, 20 fs laser pulses directly from chirped pulse amplifiers by using Generalized Double Optical Gating. We are therefore optimistic that isolated attosecond with sufficient energy can be obtained by implementing this or other gating schemes with  $\sim 100$  TW level driving lasers in the next few years.

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