Approaching the Atomic Unit of Time with Isolated Attosecond Pulses

Zenghu Chang
Institute for the Frontier of Attosecond Science and Technology (iFAST),
CREOL and Department of Physics,
University of Central Florida, Orlando, FL, USA
Zenghu.Chang@ucf.edu

Abstract: Isolated attosecond pulses are powerful tools for studying electron dynamics in atoms, molecules and condensed matter. The shortest pulses achieved so far are 67 as. Challenges and approaches for further shortening such pulses are introduced.

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1. Introduction

Attosecond light sources, first successfully demonstrated in 2001, have been built upon the high-order harmonic generation process first discovered in late 1980s. When a femtosecond Ti:Sapphire laser beam with an intensity of 10^{14} to 10^{15} W/cm^2 interacts with noble gas atoms, a broad plateau appears in the harmonic spectra, where the intensity of the harmonics does not change much with the harmonic order. It is now known that such a spectrum corresponds to a train of attosecond pulses [1, 2]. Attosecond pulses have enabled the study of electron-electron interactions on their natural time scale for the first time. For pump-probe experiments to time-resolve the electron dynamics, single isolated attosecond pulses are desirable. Ideally, two isolated attosecond pulses with variable delay should be used, which is yet to be demonstrated. Various switching methods have been invented to extract a single attosecond pulse from an attosecond pulse train [3, 4]. We demonstrated that the Double Optical Gating is a powerful sub-cycle optical switch for generating broadband single isolated attosecond pulses, which takes advantage of the plateau region of high harmonic spectrum [5]. Pulses as short as 67 as have been generated with this method [6]. Further shortening the attosecond pulses faces three major challenges: generating even broader high harmonic spectrum, reducing or compensating the atto-chirp and accurately characterizing the pulses. Isolated attosecond pulses with shorter duration and broader spectrum will enhance the power of attosecond transient absorption for studying more states in matter [7, 8].

2. Polarization Gating and Double Optical Gating

The cutoff photon energy is proportional to the square of the driving laser wavelength before the atom is fully ionized. Dramatic extension of the cutoff using an Optical Parametric Amplifier (OPA) with 1.51 μm center wavelength as compared to a Ti:Sapphire laser was first demonstrated in 2001[9]. Therefore increasing the wavelength of driving lasers is one of the approaches to generate broader spectrum to support shorter pulses.

High-order harmonics can be generated efficiently when the driving laser field is linearly polarized. The conversion efficiency drops rapidly when the driving laser is elliptically polarized [10]. Two examples are given in Fig. 1. It is clear that the harmonic yield drops much faster for a 2 μm driving laser than a 0.8 μm. If a driving laser can be produced with linear polarization during only a single half-cycle, with all other portions elliptically polarized, then a single attosecond pulse can be isolated, which is the foundation of polarization gating [11]. The driving pulse for polarization gating can be produced by combining two counter-rotating circularly polarized few-cycle laser pulses with a certain delay between them. For a properly chosen delay, the resulting pulse can be linearly polarized only during the central half-cycle, with elliptically polarized leading and trailing edges. Polarization gating is more effective for a long wavelength driving laser. Numerical simulations predict that 25 as pulses can be generated by polarization gating with a 1.6 μm few-cycle laser[1].

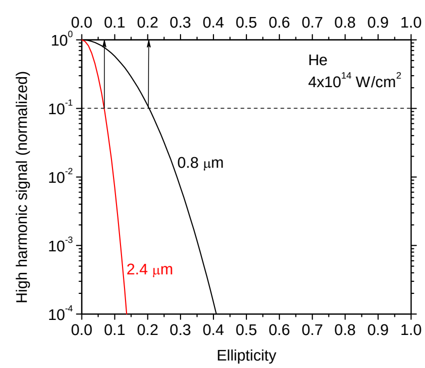

Fig. 1. Dependence of high harmonic generation yield on ellipticity of the driving laser.

For Ti:Sapphire lasers, the less-effective polarization gating can be improved with the addition of a linearly polarized second-harmonic field to the driving field with time-dependent ellipticity. This technique is known as Double Optical Gating as it is combination of polarization gating and two-color gating. The DOG technique allows a significant increase in the driving laser intensity when compared with polarization gating for the same pulse duration. For multi-cycle driving lasers, the cutoff photon energy of the attosecond pulses is determined by the ionization saturation intensity of the target atom. It is advantageous to start with elliptically-polarized pulses to reduce the depletion of the ground state population of the target atom by the leading edge. Such pulses can be obtained by passing a linearly-polarized pulse through a birefringent quartz wave plate, a Brewster window, and a quarter-wave plate. At the Ti:Sapphire wavelength, pulses up to 28 fs in duration have been used successfully to generate isolated attosecond pulses using this method. Double Optical Gating works well simultaneously across the whole plateau and cutoff region of the high harmonic spectrum. We have demonstrated the generation of attosecond continua down to 12 eV and up to more than 500 eV, which support 16 as transform-limited pulses [12]. New techniques have yet to be developed for compensating the atto-chirp of such broadband pulses.

Double Optical Gating can be implemented with multi-cycle lasers directly from Ti:Sapphire chirped pulse amplifiers. It is worth mentioning that only the driving laser energy inside the polarization gate has the potential to be converted to photons of isolated attosecond pulses. Therefore, the shortest possible pulses should be used for driving the Double Optical Gating. We have developed a Ti:Sapphire laser system that is capable of producing sub-14 fs pulses with more than 300 mJ energy at 800 nm. Attosecond XUV continua with more than 100 nJ at 35 eV have been generated by driving the Generalized Double Optical Gating with this laser [13]. We are currently constructing a 200 TW driving laser to increase the XUV energy by another order of magnitude for demonstrating attosecond pump-attosecond probe experiments.

3. Characterization of broadband pulses with Phase Retrieval by Omega Oscillation Filtering (PROOF)

While the FROG-CRAB algorithm has been widely applied to the characterization of isolated attosecond pulses and attosecond pulse trains, it is not suited for the characterization of ultrabroadband attosecond pulses, as the reconstruction algorithm relies upon a narrow-bandwidth approximation called the “Central Momentum Approximation.” The PROOF technique was developed for characterizing ultrabroadband attosecond pulses [14], which has already been used to evaluate isolated attosecond pulses generated with Ti:Sapphire lasers. PROOF relies on the measurement of electrons photoionized by an attosecond pulse under the perturbation of a dressing laser field. An attosecond streak camera with high energy resolution will be required to accurately assess the duration and contrast of the attosecond pulses generated with long wavelength driving lasers.

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4. References

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