Wavelength-scaled High Harmonics for Probing Dynamics on a sub-Atomic Time Scale

Erik P. Power

Recent years have witnessed a revolution in the generation of ultra-short bursts of coherent radiation. Attosecond (1 as = 10^{-18} s) physics, made possible by high harmonics created through the nonlinear interaction of intense ultrafast laser pulses with atoms [1], offers the promise of major advancements in coherent control applications, compact x-ray sources, and accelerated electronic / magnetic storage schemes. Perhaps one of the most intriguing experiments in the attosecond domain is a time-resolved study of electron wave packet dynamics during the Auger decay process. XUV or x-ray radiation ionizes an inner shell electron; the resulting hole is quickly filled by an outer shell electron, and a third electron is ejected into the continuum to fulfill energy conservation. The dynamics of the shakeup, or electron redistribution following the initial scattering event are, as yet, unobserved. In 2007 Uiberacker, et al. demonstrated a 400 as rise time in the buildup of Ne^{2+} [2], demonstrating that the shakeup occurs on a timescale too fast for current tools to measure (80as is the current record pulse length, but it is not widely available). In order to directly resolve the wave packet dynamics during the shakeup, I propose to exploit scaling laws that favor harmonic production using mid-infrared (1.9 μm $-$ 5 μm wavelength) laser pulses to generate XUV bursts with durations less than one atomic unit of time (1 a.u. $\sim$ 24 as). An XUV pump will ionize an inner shell electron, and a sub-24 as probe will fragment the ion at various time delays. The fragments will be observed using an electron streak camera or a Cold Target Recoil Ion Momentum Spectroscopy (COLTRIMS) [3] setup, which allows for complete reconstruction of the momentum vectors for all fragments.

The primary hurdle to breaking the 24 as barrier is development of a high energy, few cycle laser in the mid-IR. I will detail my plans for a difference frequency generation optical parametric amplifier (DFG OPA) specifically designed to provide sufficient pulse energy and duration for ionization saturation in argon. My design will produce $\lambda > 3$ μm light with 20$\times$ more pulse energy and duration nearly two times shorter than the current state of the art. An unfortunate drawback to using mid-IR light to drive HHG is the loss of XUV / x-ray flux: simulations show that harmonic generation efficiency scales as $\lambda^{-9/2}$ [4], however experimental results indicate that macroscopic phase matching conditions can be optimized to provide superior XUV flux at shorter wavelengths [5]. I will examine improvements that can be made to enhance the generated XUV / x-ray flux, most notably by adding an optical parametric chirped pulse amplifier (OPCPA) booster to the end of the laser chain. Alternative schemes will also be discussed, such as generating harmonics from relativistic laser-plasma interactions using solid targets. In 2009 Nomura, et al. measured sub-femtosecond (1 fs = 10^{-15} s) bursts from relativistic solid target harmonics [6]. Relativistic harmonics from solids are attractive because they can offer orders of magnitude higher conversion efficiency than gas-generated harmonics. As a low frequency alternative, I will discuss synthesizing sub-fs VUV pulses from below-threshold harmonics, i.e. harmonics with photon energy less than the ionization potential. I have shown these harmonics exhibit non-perturbative character (in particular, negative dispersion) [7] consistent with the standard semi-classical model for above-threshold harmonic generation [8]. Finally, I will present a brief overview of my current project: the design and fabrication of a chip-based continuous-wave (CW) atom laser [9].

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