High flux coherent supercontinuum soft X-ray source driven by a single-stage Ti:sapphire-pumped OPA

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Abstract: We demonstrate the highest flux tabletop coherent soft X-ray source to date, using high harmonics driven by a single-stage Ti:sapphire-pumped OPA at 1.3μm. The spectrum extends to 200eV, with a flux of $>10^6$ photons/pulse/1% bandwidth.
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Introduction

High-order harmonic generation (HHG), which upconverts light from a tabletop femtosecond laser while retaining full spatial and temporal coherence when implemented in a phase matched geometry [1-2], has been used to uncover new understanding of coupled electron, phonon and spin dynamics in molecules and materials. Most implementations of HHG to-date have used Ti:sapphire driving lasers at a wavelength around ~0.8 μm, which limits bright HHG to the extreme ultraviolet region of the spectrum, at photon energies ≈ 100 eV. A recent breakthrough extended phase matching of HHG to much higher keV photon energies, by using *longer* wavelength mid-infrared driving lasers at wavelengths from 1.3 to 3.9 μm [2-4]. However, these preliminary studies were done using lasers operating at 10 Hz repetition rates, with a total photon flux of 10^6 photons/s in 1% bandwidth, which is not sufficient for application in spectroscopy or imaging. To fully harness the unique properties of soft X-ray HHG, kHz or higher repetition-rates are required in a phase matched geometry. This goal requires the development of stable, compact, high pulse-energy, ultrafast laser amplifiers that can generate multi-millijoule, < 10 cycle, mid-infrared laser pulses required for efficient HHG.

In this work, we demonstrate the highest pulse energy (10 mJ), single-stage, ultrafast (45 fs) Ti:sapphire amplifier to date, with a repetition rate of 1 kHz. We then use this laser to pump an optical parametric amplifier system and generate 1.3 μm, 30 fs pulses with sufficient energy (> 2 mJ) for optimally-efficient, phase matched HHG conversion. This allows us to demonstrate the highest flux, soft X-ray HHG source to date with > 10^6 photons/pulse/1% bandwidth at 1 kHz (corresponding to > 10^9 photons/s/1% bandwidth) in a broadband, continuum, spectrum extending to 200 eV. This photon flux represents an approximately 3 orders-of-magnitude increase compared with past work [3, 5]. Using this unique bright *supercontinuum* HHG soft X-ray source, we demonstrate soft X-ray absorption spectroscopy with 0.25 eV spectral resolution, and with the ability to resolve near edge fine structure with high fidelity.

Experiment

The maximum output pulse energy obtainable from an ultrafast Ti:sapphire regenerative amplifier is primarily limited by thermal lensing and optical damage. By employing closed-cycle cryogenic cooling of the Ti:sapphire crystal, the thermal lens present in the laser crystal is greatly reduced. By increasing the cavity length and temporally stretching the pulse using high groove density gratings, the amplified pulse intensity is kept below the damage threshold of commercial broadband optics. The maximum output (compressed) pulse energy is 10.6 mJ when pumped with 50 mJ of 527 nm light from a Nd:YLF pump laser, with a 45 fs pulse duration, and a near perfect Gaussian profile output mode with M^2 values for both x- and y-axes of ~1.1. To obtain high-quality 1.3 μm pulses to drive soft X-ray HHG, we use a homebuilt 3-stage OPA that achieves greater than 40% conversion of the 0.8 μm light into 1.3 μm (signal) and 2 μm (idler) beams, so that 1.3 μm pulse energy is greater than 2 mJ. Additionally, the broad spectrum of the 1.3 μm light supports an ultrashort pulse duration of 30 fs.

Bright soft X-ray harmonics are generated by focusing the 2 mJ pulse energy, 1.3 μm, OPA signal output into a high pressure gas-filled hollow waveguide (length 1 cm and diameter 150 μm) using an f = 25 cm lens. By controlling the gas pressure inside the waveguide and the 1.3 μm pulse energy before the waveguide, we can optimize the HHG flux at the phase matching pressure for each gas, as shown in Fig. 1. Since numerous experiments

have been implemented utilizing 0.8 μm-driven harmonics, we also compared the optimized fully phase matched HHG flux driven by 0.8 μm light with the HHG flux driven by 1.3 μm light (see Fig. 1).

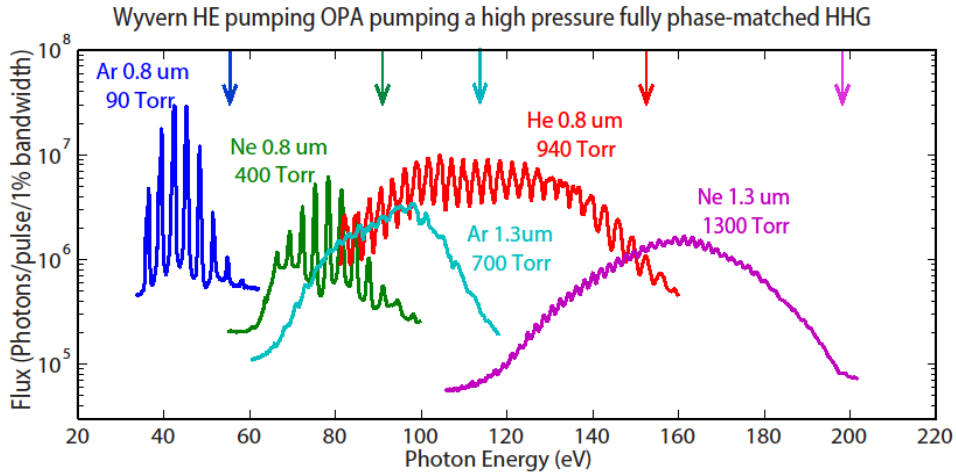

Fig. 1. Flux characterization and comparison of the optimized fully phase matched HHG emission from a 1 cm long, 150 μm diameter, waveguide driven by 1.3 μm and 0.8 μm light in various noble gases (Ar, Ne, and He). The vertical arrows indicate the maximum phase matched HHG energy cutoff for each gas and laser wavelength.

The high-flux, broadband HHG spectra provide a unique opportunity to measure soft X-ray absorption spectra from multiple orbitals of an element or multiple elements simutaneously, with the ability to resolve x-ray absorption near-edge fine-structure (see Fig. 2). Additionally, the kHz repetition rate ensures high quality spectra in a short data acquisition time. The long term stability test demonstrates that the HHG flux is stable over many hours, which is important for time-resolved experiments.

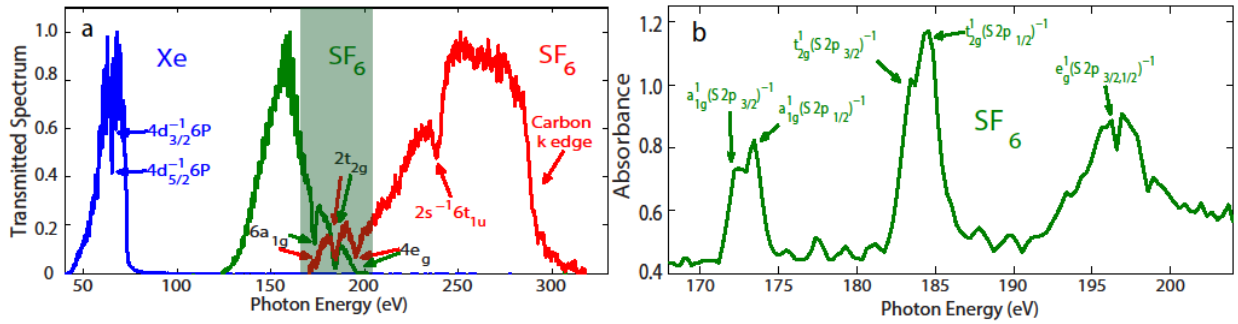

Fig. 2(a) Normalized raw data for 1.3 μm-driven HHG from Ar (blue), Ne (green), and He (red) after transmission through a 2 mm long sample cell of Xe (HHG from Ar), and SF_6 (HHG from Ne and He). (b) NEXAFS spectrum of SF6 Sulfur L-edge 2p orbital.

Conclusion

We demonstrate the highest pulse energy signal-stage Ti:Sapphire amplifier, and use it to produce the highest coherent tabletop soft X-ray photon flux to date, as well as applications in soft X-ray spectroscopy.

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