High Photon Flux 70 eV HHG Source for Applications in Molecular and Solid State Physics

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Abstract: We present a high harmonic generation source driven by a nonlinearly compressed 50-100 kHz fiber laser enabling record high \(10^{11}\) photons/s in single harmonics between 55-73 eV. The unique capabilities of the source are underlined by using it for coincidence experiments in gas phase CH$_3$I and measurements of the transversal magneto optical Kerr effect in nickel and iron samples.

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Coherent extreme ultraviolet sources with ultrashort pulses durations are incredibly important for a multitude of applications aiming at the understanding of physical, biological and chemical processes at atomic length and time scales. In that regard sources based on high harmonic generation (HHG) of table-top femtosecond lasers have emerged as an interesting complement to large-scale facilities such as free electron lasers. This is even more true when considering the recent advances in HHG with high average power fiber lasers [1]. The combination of high photon flux and repetition rate holds promise to enable a multitude of applications, e.g. in coincidence experiments, femtosemagnetism and photoelectron spectroscopy and microscopy.

Here we present HHG of a nonlinearly compressed fiber laser operated between 50-100 kHz. Due to the high average power of the laser system more than \(10^{11}\) photons/s can be generated in a single harmonic at 68.6 eV, which is an order of magnitude higher than previous demonstrations [2]. The unique combination of high repetition rate and photon flux is utilized in coincidence experiments on CH$_3$I molecules paving the way towards novel experiments in imaging molecular dynamics. Additionally, we use the HHG source to measure element specific magnetic properties of nickel and iron samples.

The frontend used for high harmonic generation is a fiber chirped pulse amplifier (FCPA) with subsequent nonlinear compression. The FCPA system is operated with 1 mJ, 300 fs pulses at repetition rates between 50 kHz-100 kHz (50-100 W average power), respectively. Subsequently, the pulses are shortened to 30 fs in a gas-filled capillary to achieve efficient high harmonic generation, in particular, around 70 eV photon energy. The pulse energy after temporal compression is 0.5 mJ resulting in 25-50 W of available average power for HHG. These pulses are then sent into a vacuum chamber (Fig. 1 a)), where they are focused to an intensity of \(~2\cdot10^{14}\) W/cm$^2$ to generate harmonics in a 150 µm argon gas jet. The co-propagating harmonics and infrared laser impinge on a chicane of two-grazing incidence plates that have an antireflection coating for the laser and a top layer of SiO$_2$ that reflects the harmonics with sufficient efficiency (40% after 2 reflections). Afterwards, an additional 200 nm thick aluminum filter further suppresses the infrared laser and separates the harmonic chamber (>10$^2$ mbar) from the experimental chambers (10$^{-6}$-10$^{-9}$ mbar). After this filter either a flat-field grating spectrometer analyzes the harmonics or the HHG beam is sent to the specific application. Fig. 1 b) shows the spatial spectral characteristics of the generated harmonics between 55-73 eV under optimized conditions showing Gaussian like spatial profiles (right lineout) as expected for phase-matched generation.
Fig. 1 a) Experimental setup of the high harmonic generation at 70 eV (GIP-grazing incidence plates). b) Spatial spectral lineout of the generated harmonics. The right side shows the spatial beam profile of the H57 at 68.6 eV.

Using the known detection efficiencies [1] allows calculating the photon flux to be more than $10^{11}$ photons/s per individual harmonic at 100 kHz. This value constitutes an order of magnitude improvement over previous results [2] making this source highly attractive for applications.

Fig. 2 a) Ion-ion coincidence spectrum of CH$_3$I as obtained in the coincidence experiment. b) Asymmetry parameter for a nickel on gold sample.

One of the most benefiting applications for a high repetition rate HHG source are coincidence experiments, e.g. aimed at imaging molecular dynamics [3]. Additionally, the (sub)-femtosecond pulse duration and low timing jitter with respect to the driving laser is ideally suited for ultrafast pump-probe measurements. We performed such an experiment by using the 57th harmonic (68.6 eV) and focus it on a molecular jet of CH$_3$I molecules. In a first successful test the above-described laser system could be operated for more than 20 hours enabling the recording of electron-ion-ion coincidences for the photoionization of CH$_3$I molecules by the HHG pulses (Fig. 2 a). The coincidences are identified by diagonal lines, which appear due to momentum conservation. Another potential application is the study of femtomagnetism via the transversal magneto-optical Kerr effect, which relies on measuring the reflection of the HHG radiation from a magnetic sample [4]. Since this is done near Brewsters angle resulting in reflection coefficients of $10^{-3}$-$10^{-4}$ the high photon flux delivered by the source is beneficial to reduce integration times and improve the signal to noise ratio. Fig. 2 b) shows the asymmetry parameter obtained from the reflectivity measurements for a nickel gold sample, which is found to be consistent with earlier measurements [4].

In summary, the highest photon flux HHG source at around 70 eV has been demonstrated by utilizing high average power fiber lasers. The unique capabilities of this source are underlined by addressing very demanding application such as coincidence experiments aiming to image molecular dynamics and femtomagnetism studies in solid state physics. Further potential of source development towards new wavelength regimes, e.g. at 90 eV, 150 eV and water window, will be discussed with regard to addressing new classes of experiments will be discussed.