Yb-laser Driven High Harmonic Generation and Applications in Magnetic Imaging and to monitor the Electron Dynamics in complex systems

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High harmonic generation (HHG) in gases, driven by Yb-laser, is meanwhile widely used for many challenging experiments such as resonant magnetic diffraction imaging and to investigate complex electron dynamics in solids. By using a Yb:CaF₂ laser amplifier at 1030 *nm* delivering sub-picosecond laser pulses of up to 6 *mJ* at 500 *Hz* and exploiting the processes of Self-Phase Modulation and Stimulated Raman Scattering in a gas-filled hollow-core fiber [1] in combination with chirped mirrors, it is possible to produce strong laser pulses compressed to the femtosecond time-scale. These post-compressed fs-pulses are exploited to drive the HHG-process in a gas medium to obtain a broadband spectrum of harmonics reaching up to the extreme ultraviolet (EUV) or the soft X-ray regions, optimised to cover the energy interval 50 - 220 eV. This spectral range covers the ferromagnetic N and M absorption edges in magnetic targets as well as the inner-shell absorption edges of some metals such as Al and Cu, thus making it ideal for coherent magnetic domain imaging [2] and to follow the complex electron dynamics in metal surfaces.



Figure 1. Continuously tunable, EUV – X-ray high harmonics from He, generated by a tunable VIS driving laser with wavelengths spanning the 480 - 550 nm range. Left – Schematic of the setup, Right - A. Blue-shift and red-shift wavelength-tunable harmonics. The arrows indicate the shift direction when atomic Ar (blueshift) or molecular N_2 gas (red-shift) is employed for spectral broadening (the color-coding denotes the corresponding pressure required for broadening). B. and C. Experimental pressure dependence of the high harmonics extending into the soft X-ray region for Ar-filled waveguide at 400 *mbar*. D. Single-atom estimate of the harmonic yield from neutral He atoms and He⁺ ions at an intensity of >10AB W/cm^2 .

In a recent work, we generated high harmonics from He employing a tunable driving laser in the visible range, with wavelengths spanning between 480 - 550 nm, using a BBO type I crystal before the HHG gas medium. This approach allows to fine-tune the spectrum of harmonics, shifting it towards the "red" or "blue" wavelengths in the EUV regime including the generation of attosecond pulse trains (APT) exhibiting a characteristically low attosecond chirp (Fig. 1).

In another ongoing work [3], the same type of HHG-set-up is used to generate APT in the EUV-regime, suited for the temporal investigation of the photoelectron emission processes in Aluminium. The main objective of this ongoing project is to resolve the complete dynamics of a bulk plasmon using RABBITT (Reconstruction of Attosecond Beating By Interference of Two-photon Transition) and Attosecond Streaking techniques. In a first step, a comb of EUV-harmonics around 150eV is sent as a pump-signal onto the Al target, thus inducing photoelectron emission from the L-shell. The near-infrared seeding pulse (NIR) is used as a probe-signal reaching the Al-surface at various time-delays. The ejected photoelectrons (PEs) can either be directly detected or they can additionally undergo inelastic scattering events, thus transferring part of their energy to the surrounding solid-state electrons prior to their escape above the surface barrier. These inelastically scattered PEs are intimately linked to the excitation of the bulk plasmon in Al, exhibiting a characteristic resonant energy of $\hbar \omega_{pl} = 15 \text{eV}$. By monitoring the RABBITT traces of these two PE-signals it will be possible to determine the time delay intercurring in between the direct emission of PEs and the rise of a bulk plasmon. To actually monitor the complete plasmon dynamics, it is necessary to detect – along the same time-scale – the so-called "secondary" electron, which is emitted from the target as a consequence of the bulk plasmon decay.

References

[1] P. A. Carpeggiani, et al., "Extreme Raman red shift: ultrafast multimode nonlinear space-time dynamics, pulse compression, and broadly tunable frequency conversion," Optica 7, 1349-1354 (2020).

[2] D. Popmintchev, P. Carpeggiani, V. Shumakova, A. Imani, S. Wang, J. Yan, W. Brunner, E. Kaksis, T. Flöry, A. Pugžlys, A. Baltuška, and T. Popmintchev, "Continuously Wavelength-Tunable Coherent EUV and Soft X-ray Light for Dynamic Magnetic Imaging and Metrology" in Frontiers in Optics + Laser Science 2022 (FIO, LS), Technical Digest Series (Optica Publishing Group, 2022), paper JTu5B.22.

[3] A. Bellissimo "Time-Resolved Electron Spectroscopy: a Challenging Highly Innovative Collective Excitation Study" – TRES-CHIC-ESt – H2020 MSCA-IF (GA.No. 101022318).