Characterization of Tunable Few-fs VUV Pulses

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The extension of few-cycle pulse technology into the deep- and vacuum-ultraviolet (DUV: 200-300 nm and VUV: 100-200 nm) spectral range will allow time-resolved measurements of valence electron dynamics at an unprecedented temporal resolution. Such experiments promise to answer a manifold of fundamental questions in chemistry and physics. Recently, µJ-level, few-fs-long pulses tunable across the UV became available by resonant dispersive wave (RDW) emission during soliton self-compression in capillaries [1]. Beyond the pulse generation, beam handling and pulse characterization are also essential for successful time-resolved experiments. These are, however, very challenging due to the vicinity of electronic resonances in virtually every material in the UV inducing severe absorption and dispersion. Especially, reliable characterization of the few-fs RDW pulses proved to be difficult. Recently, sub-3 fs RDW pulses in the deep UV (200-300 nm) could be measured by an all-reflective self-diffraction FROG placed directly in the vacuum beamline [2]. However, such all-optical schemes break down towards even shorter wavelengths of the VUV, as the material dispersion rapidly increases there.

Here, we report on the first in-situ full characterization of sub-3 fs long pulses tuned in the VUV between 160 and 190 nm [3]. We apply a special version of FROG, where we use two-photon ionization of noble gases as second-order nonlinearity. The kinetic energy distribution of the photoelectrons provides the spectral information of the autocorrelation signal. The dilute noble gas target induces virtually no dispersion even very close to resonances, at the same time the photo-electrons can be very sensitively detected by standard technology, e.g. by time-of-flight or velocity map imaging spectrometers. However, the understanding of the electron FROG traces is not as straightforward as by optical FROG traces, because they have an imprint of the atomic structure of the detector gas. We developed a new FROG retrieval code taking the structure of the trace into account. We validated the pulse retrieval by ab initio quantum mechanical calculations. Our measurements reveal that the generated VUV pulses have a duration of sub-3 fs proving that RDW pulses have a few-fs duration not only in the DUV, but also in the much more challenging VUV spectral range. The few-cycle VUV pulses were utilized for studying valence electron dynamics in small molecules, which will also be presented.

References

[1] J C Travers, T F Grigorova, C Brahms and F Belli, Nat. Photonics **13**, 547 (2019)

[2] M Reduzzi, M Pini, L Mai, *et al.*, Opt. Express **31**, 26854 (2023)

[3] J R C Andrade, M Kretschmar, R Danylo, *et al.*, arXiv:2411.11769 (2024)