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XUV-FEL chirp characterization by Transient Absorption Spectroscopy

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Having direct access to the spectro-temporal pulse characteristics of FEL pulses is a key ingredient for state-selective multidimensional spectroscopy at high photon energy which now comes into reach with next-generation XUV and x-ray FEL radiation sources. While the characterization of visible and infrared laser pulses is nowadays a matter of routine, a direct transfer of these concepts to XUV/x-ray FELs, which are based on the self-amplified spontaneous emission (SASE) process, remains challenging. One of the major difficulties is the lack in temporal/spectral coherence which complicates their synchronization to external timing references.

Here, we present a new self-referencing method for the spectro-temporal characterization of high-frequency SASE FEL radiation pulses. Without requiring any additional external fields, it is based on the technique of all-XUV split-beam pump-probe transient absorption spectroscopy and provides a combined high temporal and spectral resolution (≤1 fs relative timing stability with spectral resolving power E/ΔE ˜1500) over a 1-2 eV broad spectral bandwidth of probing photon energies. For this XUV-frequency sensitive timing tool we utilize sequential double ionization in neon via two XUV photons derived from the pump pulse and probe the ultrafast ionization dynamics in real time. We employ the near-instantaneous response of the target system due to ionization as an ultrafast temporal gate on the spectrally resolved probe pulse. This allows to extract the group-delay dispersion, that is, the time-delay vs. photon-energy relationship from a measured two-dimensional transient-absorption spectrogram. Thereby, our measurement revealed a pronounced linear chirp of about 30 fs^2 [**1**], while the method is also sensitive to higher orders of nonlinear chirp.

The characterization method presented here is not limited to the XUV domain nor to femtosecond time scales; it can be straightforwardly extended even to hard x-ray and attosecond pulses, and only requires an x-ray intensity-induced change of the absorbance properties of a moderately dense target medium, e.g., through nonlinear ionization of rare gas atoms [**2**].

[**1**] T. Ding, *et al.*, *Measuring the frequency chirp of extreme-ultraviolet free-electron laser pulses by transient absorption spectroscopy*, Nature Communications, accepted (2021).

[**2**] L. Young, *Femtosecond electronic response of atoms to ultra-intense X-rays*, Nature **466**, 56–61 (2010).

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