Attosecond interferometry with free-electron laser pulses

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Synopsis Light-phase-sensitive techniques are well-established in a broad spectral range, spanning from radio-frequencies in nuclear magnetic resonance spectroscopy to nonlinear optics with table-top fs lasers. In the present contribution, attosecond (as) phase control is achieved in a Michelson-type all-reflective autocorrelator in the limit of short wavelength. The observed fringe contrast monitors the field interference, i.e. the free-electron laser (FEL) light wave oscillation with a period of 129 ± 4 as at 38 nm wavelength. The transfer of a powerful table-top method towards FEL science paves the way to use advanced nonlinear soft X-ray techniques for watching the transformation of electronic orbitals in real time.

Science with X-ray free-electron lasers has enabled multiple breakthroughs covering the broad range from basic research in life sciences to applications in material science and catalysis. Particularly, the high degree of spatial coherence of the light field allows for key applications such as serial fs X-ray crystallography using the well-established and robust self-amplified spontaneous emission (SASE) of FELs [1]. Recently, temporal coherence provided by seeded FELs moved into the focus of interest [2-4]. It has been shown that full control over the light phase allows for a new class of light-phase sensitive experiments in the short-wavelength limit [5-7], such as non-linear four-wave mixing [8] and coherent control [9]. These give novel opportunities to study and possibly control ultrafast electron dynamics in molecular systems of increasing complexity with unprecedented spatial and temporal resolution.

The present study has been performed at the monochromator beamline of the soft X-ray FEL in Hamburg, FLASH. The FLASH pulse exhibits many coherent longitudinal modes. It is important to note that the selected spectral bandwidth is significantly smaller than the width of individual modes. Thus, the radiation field comprising approximately 10^{10} photons per pulse passing the exit slit possesses a high degree of spatial and temporal coherence. A reflective split-and-delay unit (SDU) plays the key role for phase-resolved one-color pump-probe spectroscopy discussed in the present contribution. It splits the wavefront of the incoming FEL pulse uniformly across the beam profile by two interleaved gratings and provides two pulse replicas with a variable delay.

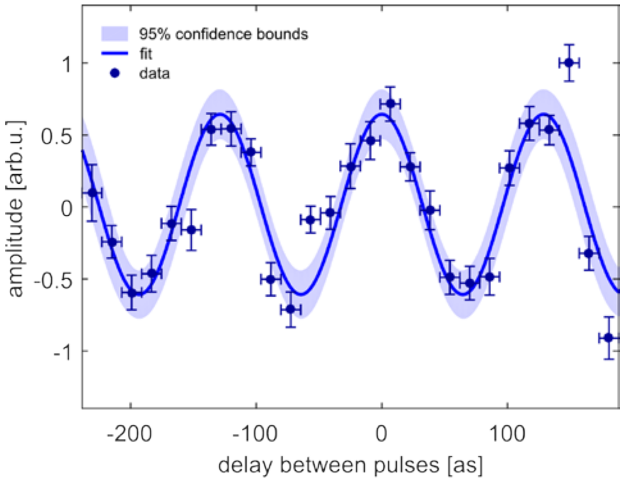

Figure 1. Interferometric autocorrelation recorded with FEL (SASE) pulses.

The above geometry provides collinear propagation of both pulse replicas and thus constant phase difference across the beam profile. This enables to record phase-resolved autocorrelation signals with maximum contrast as shown in Fig. 1. Our study opens up the door for high-contrast time-domain interferometry even at partially coherent SASE FEL sources. It makes the local electronic structure and dynamics accessible, i.e. controllable.

References

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