

Phase-sensitive measurements, central for gaining insight of light-matter interactions, require phase-controlled electric fields. Combining phase-control with shortwavelength laser pulses provides opportunities to trace electron dynamics with unprecedented spatial and temporal resolution. Thanks to a special mirror developed in collaboration between projects A2 and C3 we can control the phase of XUV and soft X-ray pulses from free-electron lasers (FELs) with attosecond precision. This paves the way towards timedomain interferometry in the soft X-ray range, thus increasing the amount of information retrieved from nonlinear optics experiments even at partially coherent FEL sources.

In optics, direct information about lightwave phase is obtained by interferometric techniques. In the last decade, analogous methodologies have been developed to derive similar information on ultrafast electron wave packet dynamics in atoms, molecules, clusters and solid-state materials. It has been shown that full control over the light phase in the shortwavelength limit allows for a new class of light-phase sensitive experiments, such as XUV nonlinear wave mixing and attosecond coherent control.

Interferometry in the time domain is established in the optical spectral range for decades but is still in its infancy for XUV and soft X-ray photon energies. The progress in short-wavelength interferometry is impeded by the usage of reflective optics and harsh precision requirements for control of the experimental equipment. The workhorse of XUV pump—XUV probe studies is a doublemirror split-and-delay unit (SDU) that generates two pulse replicas with a variable delay (Fig. 1a). The two-element mirror produces two spatially-separated pulses which can be superimposed only at a skew angle. This greatly reduces the phaseresolution of the recorded pump-probe signal as illustrated in Fig. 1a.

Fig. 1: Comparison of a conventional double-mirror SDU (a) and a lamellar-grating SDU (b). The insets show simulated light intensity distributions in the focal (XY) and the propagation (YZ) planes for nine different phase delays ranging from 0 to 2 π . (a) The double-mirror SDU generates two spatially separated beams, which are superimposed by the focusing optics at a skew angle with their wavefronts tilted in opposite directions. As a result, their relative phase varies continuously along the Y-axis. Hence, a detector with infinitely high resolution would be required to discriminate single interference states. (b) Each grating of the SDU diffracts the incident beam. Partial beams from the two gratings propagate collinearly in every diffraction order as justified by the complete destructive interference in the zeroth order for $\Delta \phi = \pi$. Spatial selection of a single order for detection allows for high-contrast interferometric measurement.



Bringing together the expertise from research areas A and C made it possible to develop an experimental apparatus that overcomes this limitation. A smart split-anddelay unit developed in the DESY group of Laarmann (project A2) in close Tim collaboration with Christoph Becker from ILP (project C3) splits the wavefront of the incoming short-wavelength pulse uniformly across the beam profile by two interleaved lamellar gratings (Fig. 1b). The two pulse replicas produced by the lamellar geometry propagate collinearly and thus have a constant phase difference across the beam profile. This property makes the SDU an all-Michelson reflective analog of interferometer and allows for measurement of signals with maximum interferometric contrast. The precise control over the temporal phase of the light wave on the subcycle attosecond time scale is a prerequisite for interferometry in XUV and soft X-ray domains. Although, phase control is an established technique in optics, soft X-rays oscillate a hundred times faster than visible light, requiring a hundred times better precision. The developed SDU is paired with a diagnostics system based on an in-vacuum white light interferometer (WLI). The WLI monitors the topography of the SDU in real time and provides the precise feedback about the generated twin-pulse delay for every laser shot. This way, the question how precisely one can control the phase is transformed into how precisely one can measure the delay for each pulse pair. The WLI system has precision of 1 nm which translates in the delay uncertainty of just 3 attoseconds in our experimental geometry.

Recently, we demonstrated attosecond phase control of XUV light waves by generating two phase-locked replicas of selfamplified spontaneous emission (SASE) pulses from FLASH FEL with the lamellargrating SDU and observing their interference directly in time domain. With this achievement, the developed experimental setup allows realization of novel phasesensitive spectroscopic studies with attosecond resolution even at partially coherent XUV and soft X-ray FEL sources.

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Original publications:

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