Improved stabilization scheme for extreme ultraviolet quantum interference experiments

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Abstract
Interferometric pump–probe experiments in the extreme ultraviolet (XUV) domain are experimentally very challenging due to the high phase stability required between the XUV pulses. Recently, an efficient phase stabilization scheme was introduced for seeded XUV free electron lasers (FELs) combining shot-to-shot phase modulation with lock-in detection Wituschek et al (2020 Nat. Commun. 11 883). This method stabilized the seed laser beam path on the fundamental ultraviolet wavelength to a high degree. Here, we extend this scheme including the stabilization of the XUV beam path, incorporating phase fluctuations from the FEL high gain harmonic generation process. Our analysis reveals a clear signal improvement with the new method compared to the previous stabilization scheme.

Keywords: extreme ultraviolet, high harmonic generation, free electron laser, interferometry, lock-in detection, phase stabilization, ultrafast spectroscopy

(Some figures may appear in colour only in the online journal)

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1. Introduction

Interference phenomena can be exploited to control quantum pathways with high precision [1] and to improve the attainable resolution of experiments with respect to temporal, spectral and spatial information [2–6]. In order to observe interference between two optical pulses, sub-cycle phase/delay stability between the pulse replicas is required. This explains why techniques exploiting the interference between multiple laser pulses/beams are very challenging to perform at short wavelengths in the extreme ultraviolet (XUV) to x-ray regime where only a few such experiments have been reported to date [7–19].

At visible wavelengths several phase stabilization methods have been developed to solve the phase jitter issue [20–24]. However, implementing these stabilization concepts in the XUV and x-ray domain is very challenging, due to the lack of suitable optics for short wavelengths and/or the required complex optical setups. Among the established stabilization schemes, one very efficient concept is the phase modulation (PM) technique [24]. This method features extraordinary sensitivity to probe highly dilute samples in the condensed and gas phase [25–27], which can be further improved using selective detection schemes [28–31], and can be transferred to shorter wavelengths using frequency up-conversion [32]. Recently, this method was successfully implemented in the XUV domain using high-gain harmonic generation (HGHG) [33] at the seeded XUV free electron laser (FEL) FERMI [15] where it enabled the measurement of the dephasing of a Fano resonance in the time domain [9]. It was also implemented in tabletop high harmonic generation (HHG), which enabled the spectral characterization of narrow-band harmonics with very high spectral resolution (<0.7 meV, ΔE/E ~ 10−5) [10].

In these experiments, the timing and phase control, including the phase stabilization, was implemented on the fundamental wavelength before the harmonic generation was performed. This is advantageous, since no modification of the XUV beampath is necessary, which greatly simplifies the experimental implementation. However, this approach does not stabilize phase fluctuations introduced in the harmonic generation process itself or the phase jitter introduced in the XUV beampath. The same applies to other demonstrated concepts stabilizing the pulse sequences on the fundamental wavelength [8, 17, 34]. In the tabletop-approach based on HHG in gases, this problem is usually not significant, since the phase jitter introduced between the pump and probe pulses can be kept small [18]. In contrast, in HGHG the phase jitter can be substantial [9, 35] and, hence, including the XUV beampath in the stabilization method would be desirable. The PM technique is a passive stabilization scheme, which tracks the phase fluctuations and corrects them in the signal detection, in contrast to most other stabilization methods. This opens up the possibility to track and correct the phase fluctuations of the harmonic generation and the XUV beampath, even if only the beampath of the fundamental wavelength is controlled.

Here, we extend the PM technique in order to track and correct phase fluctuations in the XUV beampath at the FERMI FEL-1 source. This is not possible with other concepts actively stabilizing the interferometer at the fundamental wavelength. Our approach clearly improves the signal quality compared to the same measurement stabilizing only the beampath before the harmonic generation.

2. Method

The interference experiments are based on XUV electronic wave packet interferometry (WPI) (figures 1(a)–(c)). Here, pump and probe pulses each excite a coherent superposition of electronic ground and excited states in the system, giving rise to constructive/destructive interference in the excited state population as a function of the relative phase between the pulses/excitation pathways (figure 1(a)). Detecting the excited state population, e.g. by fluorescence or photoionization, while sweeping the temporal pump–probe delay results in an interferogram in the time domain (figure 1(b)). This signal reflects the free polarization decay of the induced dipole transition between ground and excited electronic states. Accordingly, the oscillation frequencies (ωeg) correspond to the energy difference between the involved states (ωeg = (Eg − Eg)/h), and a Fourier transform yields the linear absorption spectrum of the sample (figure 1(c)). The attainable frequency resolution is given by the scan range in the time domain and is therefore decoupled from the spectral width of the XUV pulses, thus facilitating high resolution spectra even with ultrashort and broadband pump and probe pulses.

In order to detect clean interferograms, the phase fluctuations between pump and probe pulses must be kept small: δφ < 2π/50 [36], which is increasingly difficult to achieve at short wavelengths. In the PM technique, this issue is solved by removing the phase jitter from the signal by heterodyne detection with a reference signal exhibiting the same phase fluctuations. To this end, the relative carrier-envelope phase (CEP) between pump and probe pulses is modulated at a frequency of Ω on a shot-to-shot basis. This characteristic modulation enables lock-in amplification to efficiently extract the interference signal from background contributions. At the same time, the optical interference between pump and probe pulses is detected to track the phase changes and fluctuations in the interferometer. Using this signal as reference for the lock-in amplification returns as output the phase difference between the input signal and the reference: φout = φsig − φref. This leads to cancellation of the correlated phase jitter of the signals and down-shifts the signal frequencies by orders of magnitude to a lower frequency regime (rotating frame sampling) [24]. Accordingly, the residual signal phase at the lock-in amplifier (LIA) output is

φout = Δωτ + δφsig−ref, (1)

where Δω = ωeg − ωref denotes the frequency difference between the optical resonance ωeg and the optical frequency of the reference signal ωref. τ denotes the pump–probe delay and δφsig−ref the uncorrelated phase noise between signal and reference.

A detailed description of the PM technique and of the experimental setup are given in references [9, 24, 28, 37, 38]. Figure 1(d) gives an overview over the experimental setup.
Figure 1. Experimental scheme. (a) Principle of electronic WPI. Pump and probe pulses each excite a quantum pathway (green, brown) in the system. The interference of the excitation pathways gives rise to interference fringes as a function of the relative phase between the pathways. (b) Excited state population as a function of the pump–probe delay, reflecting the constructive/destructive pathway interference. (c) Fourier transform of (b) reveals the linear absorption spectrum of the system. (d) Experimental setup: the UV seed pulses are split-and-delayed and phase-modulated in the PM interferometer setup. Subsequent HGHG generates phase-modulated XUV pump–probe pulse pairs used to excite the sample. A portion of the XUV light is branched off and analyzed in the XUV beamline spectrometer. The reference for lock-in demodulation is retrieved either from the seed laser interferometer (Ref1) or from the XUV spectrometer (Ref2). \( \phi_{21} \): phase difference between seed laser pump and probe pulses. \( \delta \phi \): fluctuations of \( \phi_{21} \). \( \delta \alpha \): accumulated phase fluctuations from HGHG and subsequent XUV beam path. With a highly stable, monolithic PM interferometer setup [38] in the seed laser beam path, phase-modulated pump and probe pulses are generated. Upon generation of the \( n \)’th harmonic of the seed laser pulses, the phase of each pulse increases by an integer factor of \( n \) (figure 1(d)). The XUV pulses excite and ionize the sample and the produced ions are mass-resolved with an ion time-of-flight (TOF) detector. The PM is introduced on the seed laser and is reflected in the ion yield and is demodulated with a LIA. In addition, phase fluctuations \( \delta \alpha \) introduced in the HGHG process and the XUV beamline contribute to the signal. Here, the timing jitter between the seed laser pulses and the electron bunch is identified as the major contribution to \( \delta \alpha \) [9]. In the previous works [9, 37], the signal (sig) was demodulated with Ref1, tracking the phase changes and fluctuations in the seed laser setup. To this end, Ref1 was digitized and its \( n \)’th harmonic was computed inside the LIA. In that implementation, the XUV phase fluctuations \( \delta \alpha \) were not tracked and therefore did not cancel upon lock-in amplification. In the current work, we extend the scheme. Instead of using a commercial LIA for the signal detection, we employ a software-based universal lock-in amplifier (ULIA) [39], which provides more flexibility in the signal processing of the experimental data and the reference signal.

While commercial LIAs require smooth, single-channel analog signals as inputs, the ULIA can process pulsed (single-shot-resolved) multidimensional detector signals [31]. For the XUV experiments, this has two advantages. First, entire photoelectron/ion TOF traces can be processed on the single-shot level for the ULIA signal input, and, thus, interference signals from many different ion/electron contributions can be extracted, simultaneously in a single measurement. To this end, the TOF traces are digitized with a fast analog-to-digital converter and each time bin is demodulated with the ULIA algorithm, similar to the procedure in reference [31].

Secondly, extraction of the reference signal from the XUV beamline spectrometer (2D CCD array detector) becomes possible, which allows tracking the XUV phase fluctuations \( \delta \alpha \) on the single-shot level (figure 1(d)). Figure 2(a) shows the XUV beamline spectrometer signal for two consecutive shots, revealing a clear interference fringe spectrum of the XUV pump–probe pulses. In the Ramsey-type interference fringes, the fringe spacing is inversely proportional to the pump–probe delay \( \tau \), while the phase-offset corresponds to the CEP difference between both pulses. Shot-to-shot modulation of the relative phase between the seed laser pulses leads to a modulation of the XUV interferogram at \( n \)-times the modulation frequency (figure 2(b)). Hence, while keeping \( \tau \) fixed, extracting the amplitude of the spectra at one specific photon energy \( h \omega_{ref} \) for multiple consecutive laser shots, returns a periodic
For a direct comparison of the signal demodulation using the commercial LIA with Ref1 and the ULIA with Ref2, we apply both methods to the same data sets, that is the \(3s^23p^6 \rightarrow 3s^43p^4\) Fano resonance in argon atoms and the \(1s^2 \rightarrow 1s4p\) transition in helium, as probed in reference [9]. In the experiment, the FEL was operated on the 6th/5th harmonic of the seed laser (28.53 eV/23.74 eV), respectively and the XUV pump–probe delay was scanned from 150 fs to 600 fs in 2 fs steps. The PM frequency was \(\Omega = 18.24\) Hz (at the 6th harmonic), which was chosen close to the Nyquist limit of 25 Hz for the FERMI FEL-1 source operated at a repetition rate of 50 Hz. At each delay the signal was demodulated over 800 laser shots when using the commercial LIA and Ref1, and 700 laser shots when using the ULIA and Ref2, both at a repetition rate of 50 Hz. The FEL pulse duration was \(\approx 57\) fs, pulse energies were 10 \(\mu\)J (for argon) and 30 nJ (for helium) and sample densities were \(\approx 10^9\) cm\(^{-3}\) (argon) and \(10^{13}\) cm\(^{-3}\) (helium). In the previous method, the XUV interference data was extracted by gating the ion-TOF spectra on the Ar\(^+\) mass with a boxcar integrator yielding a sufficiently smooth signal for demodulation with the commercial LIA. The reference was extracted from the optical interference in the seed laser interferometer (Ref1 in figure 1(d)). In the current work, we use the same data set, but demodulate the data with the ULIA algorithm and use the XUV beamline spectrometer data for the reference (Ref2 in figure 1(d)).

3. Results

First, we demonstrate the principle of rotating frame detection. The excited Fano resonance in argon is at a photon energy of 28.51 eV, which corresponds to an interference fringe spacing of 145 as in the time domain signal and would require an extremely fine sampling with \(\tau\)-increments of \(<145/2\) as. In the PM technique, beating the signal with the reference leads to a down-shifted frequency of \(\Delta \omega = \omega_{eg} - \omega_{ref}\), which depends on the reference frequency \(\omega_{ref}\). In the current work, \(\omega_{ref}\) can be freely chosen within the bandwidth.
Figure 4. Comparison between the phase stabilization methods using Ref1 (blue) and Ref2 (orange) (cf figure 1(d)) for demodulation. (a) Time-domain interferograms of the Fano resonance. Both signals exhibit strongly down-shifted frequencies. Due to the different reference signals, the oscillation periods differ and are 7.5 fs (blue trace) and 491 fs (orange trace), respectively. (b) Residual phase fluctuations in the demodulated time-domain signals. (c) and (d) Fourier spectra (real part) of the time domain signals along with a steady-state absorption spectrum obtained with synchrotron radiation [41] (black).

Figure 5. Fourier spectra of the $1s^2 \rightarrow 1s4p$ coherence excited in He atoms for demodulation using Ref1 (a) and Ref2 (b). The overall SNR in (b) has been improved by a factor of 3.1 compared to (a). Moreover, in (a) the spectral peak exhibits side lobes as a typical sign for discontinuities in the temporal phase of the signal. These effects are well compensated when using Ref2 for the demodulation.

This flexibility was not available in the previous detection method [9], where $\omega_{\text{ref}}$ was defined by a continuous-wave (cw) metrology laser tracing the seed laser interferometer and was therefore not tunable. The cw laser was necessary to obtain a smooth waveform for the lock-in demodulation with the commercial LIA [40]. There, a frequency down-shift by a factor of 51 was achieved, almost two orders of magnitude less than in the current work. This clear difference between the two demodulation methods using the fixed Ref1 and the flexible Ref2 is shown in figure 4. Figure 4(a) compares the corresponding down-shifted signal frequencies in the time domain. For a quantitative comparison of the signal quality, the phase noise is shown in figure 4(b). For a better visualization, the theoretically expected linear phase slope is subtracted from the data revealing the phase fluctuations relative to an ideal harmonic oscillation. At a delay of >350 fs where the signal amplitude has decayed to $\approx 18\%$ the residual phase jitter clearly increases. In this region, intensity fluctuations of the FEL start to dominate the signal and a comparison of the residual phase noise from both demodulation methods is not reasonable. In the region <350 fs (gray shaded), the phase noise is low for both methods, meaning that the phase for these delays is stable. However, the RMS fluctuations in this region are a factor of 1.4 smaller for the ULIA demodulation. This suggests that using the XUV reference (Ref2, cf figure 1(b)) for the signal demodulation, indeed improves the signal quality.

Next, we evaluate the Fourier spectra of both signals. The interfering ionization pathways at a Fano resonance alter the absorption line shape between absorptive and dispersive character depending on the phase shift between the direct and indirect ionization path [42]. The Fano line shape is recovered in the real part of the Fourier transforms of the WPI signals. Accordingly, figures 4(c) and (d) compare the Fourier spectra of both signals, up-shifted by the corresponding $\omega_{\text{ref}}$-values to
obtain the absolute frequency axis. To recover the line shape in the lock-in detection correctly, a calibration of the absolute phase shift between signal and reference has to be performed. While our new approach conveniently omits phase shifts from analog electronics (amplifiers, filters, etc), optical chirp on the XUV pulses leads to a phase slope in the spectral interferogram used to extract the reference signal. This phase function was not characterized in the current work. Instead, in figure 4(d) the phase parameter was optimized by fitting the data to the reference spectrum taken from a static absorption spectrum measured with synchrotron radiation [41]. For a better comparison between both demodulation methods, the same fit procedure was applied to the data in figure 4(c).

Comparing both demodulation methods with the synchrotron data, a better qualitative match is found for the phase stabilization using the XUV reference Ref2 (figure 4(d)). For a quantitative comparison, we calculated the RMS deviation from the synchrotron data in the gray shaded spectral region. Outside of this region, no significant signal is expected and the spectra are dominated by statistical noise. The RMS deviations are 4.93 \times 10^{-3} (figure 4(c)) and 0.96 \times 10^{-3} (figure 4(d)), respectively, implying a signal improvement by a factor of 5.1 when using Ref2 for the signal demodulation. This is in accordance with the time domain evaluation, showing the same tendency of an improved signal quality when using the new stabilization scheme. The result is also in agreement with another experiment (figure 5), where we measured the coherence between two bound states in helium (1s^2 \rightarrow 1s4p, 23.74 eV) [9]. Here, we compare the signal-to-noise ratio (SNR) obtained with the two stabilization methods, yielding a SNR of 14.0 using Ref1 and 43.3 using Ref2 for demodulation, which is a remarkably high SNR for interference experiments in the XUV domain. For the SNR determination we defined the noise floor as the mean value of the background signal plus two times its standard deviation.

In general, using the XUV reference (Ref2) for demodulation one may expect an even higher improvement of the signal quality than observed in our study whose limited performance can be explained by two reasons. First, the shot-to-shot amplitude fluctuations of the FEL output introduces noise which is not corrected by either of the two stabilization methods. Second, the correlation of the phase fluctuations \( \delta \alpha \) is small between consecutive laser shots. Hence, the bandwidth of the lock-in-based stabilization method is insufficient to correct these high-frequency phase fluctuations. The correction of such high-frequency noise contributions requires single-shot self-referencing methods, e.g. as implemented in references [43, 44]. We investigated the possibility of single-shot self-referenced data correction using the XUV spectrometer data and adapting the ULIA algorithm accordingly. However, this did not improve the data quality further due to the large uncertainties for extracting the required amplitude, phase and delay values from the XUV spectra.

4. Conclusion

We introduced a new stabilization scheme for interferometric XUV pump-probe experiments with seeded FELs. The method is based on a previous stabilization scheme combining PM and lock-in amplification to stabilize the seed laser interferometer at the fundamental wavelength [9]. In the current work, we have extended the method to include the stabilization of the XUV beampath, in particular of phase fluctuations introduced in the HGHG process. The new method simplifies the optical setup by omitting a cw metrology laser. Applying both methods to track the evolution of electronic coherences in argon and helium atoms, we find an up to five-fold signal improvement with the new scheme. Further improvement of the signal quality is limited by high-frequency noise introduced in the HGHG process which cannot be suppressed with the limited bandwidth of the stabilization method, and by the amplitude fluctuations which are not corrected in the stabilization scheme. In general, the possibility of tracking and stabilizing phase fluctuations in the XUV generation and the XUV beampath opens up the extension of interferometric spectroscopy schemes to shorter wavelengths, potentially to the x-ray domain, where recently first coherent control experiments were demonstrated [45].

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Conflict of interest

The authors declare no conflicts of interest.

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Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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