Delay control in attosecond pump-probe experiments

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Abstract: The time delay between the pump and probe pulses in attosecond time-resolved experiments, such as attosecond streaking, is commonly introduced by splitting and recombining the two pulses in an interferometer. This technique suffers from instability in the optical path lengths of the two arms due to mechanical vibration of the optical elements and fluctuating environmental conditions. We present a technique with which the instability of the unconventional interferometer is suppressed while at the same time the time delay is controlled to within 20 as RMS using a feedback loop. Using this scheme, the streaked spectrogram of an attosecond pulse was measured.

OCIS codes: (320.7150) Ultrafast spectroscopy; (120.3180) Interferometry.

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1. Introduction

Single isolated attosecond pulses have been characterized by the attosecond streaking technique [1] using Mach-Zehnder [2,3] and collinear [4] interferometer setups. Such measurements rely on the ability to scan the delay between the extreme ultraviolet (XUV) attosecond pulse and the femtosecond near infrared (NIR) streaking laser pulse in steps comparable to the duration of the attosecond pulse to be measured [5]. Furthermore, electron dynamics have been studied on attosecond time scales in gaseous atoms [6–10], molecules [11], and solids [12] using attosecond and femtosecond pulses in such configurations. With the advent of gating schemes which are scalable to high-power multi-cycle lasers [13,14], pump-probe experiments using two attosecond pulses are on the horizon.

The shortest attosecond pulses generated thus far were measured using a collinear interferometer configuration [4], which can achieve good stability of the pump-probe optical path lengths. However, this technique suffers from the low reflectivity and narrow reflection bandwidth of the multilayer mirrors (such as Mo/Si) used to focus the XUV light and introduce the pump-probe delay. The multilayer mirror used in that experiment, for example, had a peak reflectivity of only 3% within its 30 eV bandwidth. XUV supercontinua with broader bandwidths have been recently demonstrated [15,16], but such broadband pulses could not be characterized using currently available multilayer mirrors. The Mach-Zehnder interferometer configuration allows for the use of grazing incidence metal-coated focusing mirrors, which offer reflectivity of more than 80% over a much broader bandwidth [2,17]. Furthermore, this configuration offers the flexibility to shape the amplitude and phase of the pump and probe arms independently of one another. However, stabilization of the pump-probe delay is necessary in the Mach-Zehnder configuration, for which fluctuations in the environmental conditions as well as mechanical vibrations are likely to affect the optical path length difference between the two interferometer arms.

Interferometric stability can easily be obtained in conventional Mach-Zehnder optical interferometers by propagating a continuous wave (CW) laser through both arms of the interferometer and stabilizing the spatial interference pattern [13,18]. However, sending a CW

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beam through the XUV attosecond generation arm is non-trivial, as there are no beam-splitting optics available to combine the XUV and NIR pulses while allowing a reference beam to pass through for stabilizing the interferometer. Furthermore, the attosecond beam is generated in a gas target and any residual IR must be removed with a metal film, which also blocks the CW pilot beam. To date, all pump-probe experiments with attosecond pulses have relied on passively "stable" interferometers to measure dynamics on time scales of ~1 fs (0.3 μm optical path). However, active delay stabilization and control is necessary to measure true attosecond dynamics, which require delay steps of only a few nanometers. Here, we demonstrate a technique to stabilize and control the delay in such special interferometers.

2. Experiment

Our pump-probe setup shown in Fig. 1 was used to measure the streaked spectrogram of a single isolated attosecond pulse generated by double optical gating (DOG) [13]. The experiments were carried out using the Manhattan Attosecond Radiation Source (MARS) [19] laser system. The 1 mJ, 8 fs pulses centered at 790 nm were split into two arms by a fused silica window which transmitted ~90% of the input pulse energy. The 0.9 mJ pulse was then sent through the collinear DOG optics [20] and focused by a mirror (f = 300 mm) to an argon gas cell for generation of the attosecond pulses. The gas cell was a glass tube with inner diameter of 1.4 mm and outer diameter of 1.5 mm. The beam then passed through an aluminum (Al) filter with thickness of 300 nm for compensation of the intrinsic XUV chirp and for filtering out the residual NIR. The XUV light was focused by a grazing incidence gold-coated toroidal mirror (f = 500 mm) through the hole (d = 5 mm) in a drilled dielectric mirror (670 – 1010 nm high reflection bandwidth) to another argon gas jet located in a velocity map imaging (VMI) electron spectrometer for attosecond pulse detection. The 0.1 mJ pulse was sent through an equal optical path length and focused by a lens (f = 420 mm) to the same gas jet.

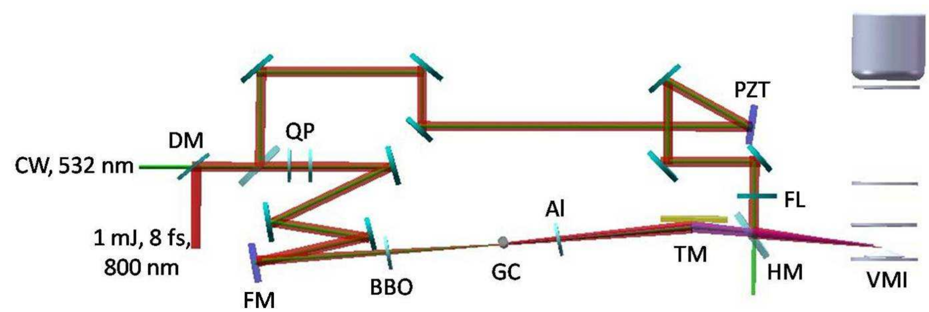

Fig. 1. Interferometer configuration for attosecond pump-probe experiments. DM: dielectric mirror, QP: quartz plates, FM: focusing mirror, BBO: second harmonic crystal, GC: gas cell, Al: aluminum filter, TM: toroidal mirror, PZT: piezo-mounted mirror, FL: focusing lens, HM: hole-drilled dielectric mirror, VMI: velocity map imaging spectrometer.

Measurement of attosecond dynamics requires much finer delay control than is afforded by conventional translation stages. However, piezoelectric stages suffer from hysteresis and the motion is not repeatable. Therefore, the field oscillations of a reference laser must be used as a "ruler" to stabilize the interferometer and control the delay. To accomplish this, a weak 532 nm CW laser copropagated through both the attosecond pump arm and the NIR probe arm of the interferometer. In the attosecond generation arm, the Al filter had a diameter of 3 mm and was mounted on a hole-drilled fused silica plate. The filter was chosen to allow a portion of the CW laser to pass around the edge of the metal film while blocking the NIR. The two arms were recombined at the drilled dielectric mirror. Because the mirror was chosen to have high reflectivity for NIR wavelengths, it also served to block any NIR that passed around the Al filter. The interference pattern of the CW laser was detected by a CCD camera,

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and the dielectric mirror had sufficient reflectance and transmittance at the 532 nm wavelength for high-contrast fringes to be detected.

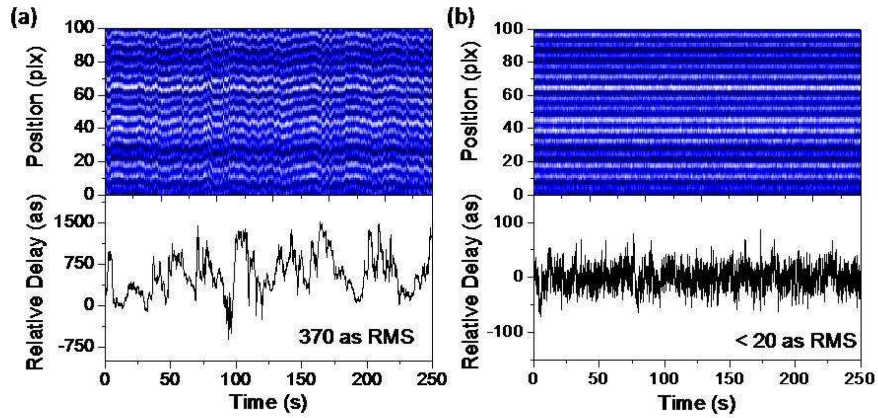

Fig. 2. (a) Top: Interference fringe measurement for the free-running interferometer. Bottom: Relative delay extracted from the fringes. (b) Top: Interference fringe measurement for the locked-interferometer. Bottom: Relative delay extracted from the fringes.

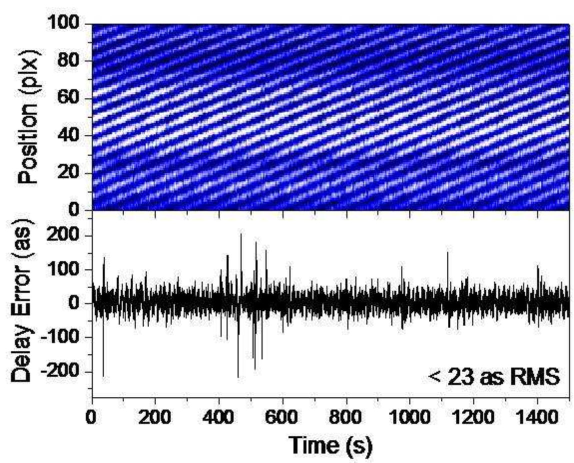

Fig. 3. Top: Interference fringe measurement with the delay scanned over a range of more than 29 fs in steps of ~280 as for an attosecond streaking measurement. Bottom: Relative delay error indicating that the interferometer was stable to within 23 as RMS over the entire measurement, including several instances where the locking was briefly disturbed.

The relative phase and time delay were extracted from the shifts of the interference fringes using Fourier-transform interferometry [21]. Home-built computer software was used to extract relative delay shifts from the interference fringes and generate an error signal used to control a mirror mounted to a piezoelectric translation stage in the NIR probe arm of the interferometer. The software locking was able to overcome the slow delay drifts below ~20 Hz. Figure 2(a) shows the interference fringe measurement and relative time delay drift of the free-running interferometer, and Fig. 2(b) shows the interference and relative time delay when the interferometer was locked. The relative delay between the two arms was locked to within 20 as RMS for the entirety of the measurement.

To control the relative delay between the two arms, only a modification of the feedback loop was required. When the delay was set to a new value, the feedback loop was used to scan the PZT to the new locking point, as determined from the interference fringes, and then stabilize the interferometer at that position. The interferometer thus maintains stability even as

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the delay is changed. The piezoelectric stage used had a full extension range of 15 μm, giving delay control with a full range of 200 fs. This technique thus allows for fine control of the delay between the two arms over a large range, such that fast electron dynamics can be accurately measured within the time scale of a multi-cycle laser pulse. Figure 3 shows the interference fringes as the relative delay was scanned in steps of ~280 as over a range of more than 29 fs for the measurement of an attosecond streaking spectrogram. Figure 3 also shows the delay error, indicating that over the entire measurement, the interferometer was stabilized to within 23 as RMS.

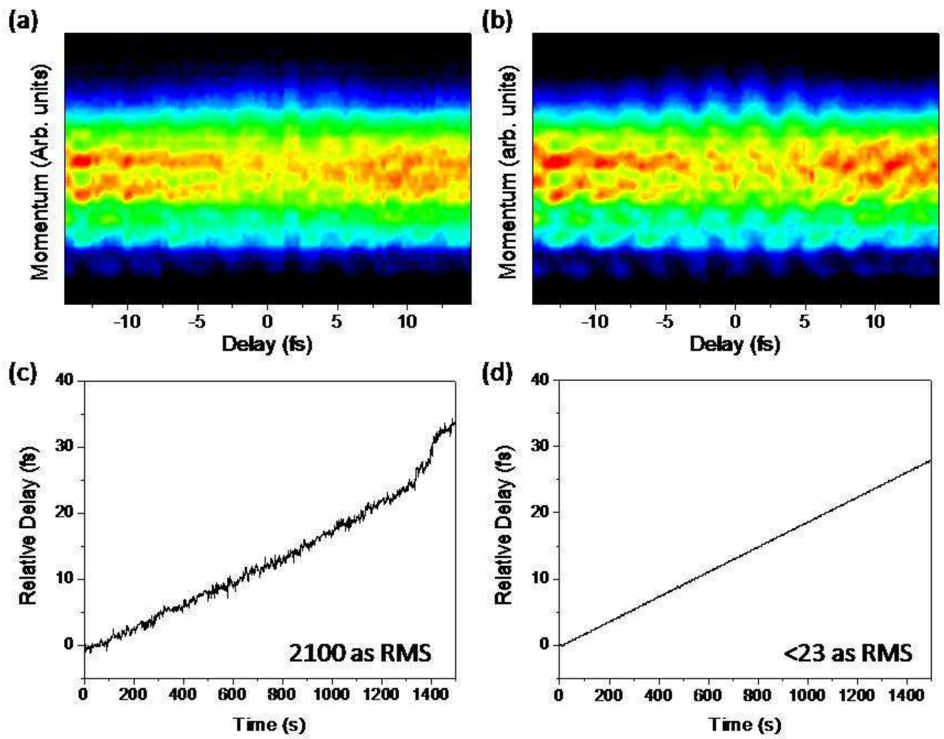

Fig. 4. (a) Streaking trace measured with interferometer unlocked and manual control of the pump-probe delay. (b) Streaking trace measured with interferometric delay stabilization and control. (c) Delay scan corresponding to the trace in panel (a). (d) Delay scan corresponding to the trace in panel (b).

Figure 4(a) shows the attosecond streaking trace measured with no locking and manual control of the delay, and Fig. 4(b) shows the streaking trace taken under the same conditions but with the interferometer used to stabilize and control the delay. The relative delay extracted from the interference fringes is plotted in Fig. 4(c) and Fig. 4(d) for the unlocked and locked cases, respectively. The VMI image was reconstructed using an iterative algorithm [22]. Argon gas was used as both the attosecond generation and the detection gas, and the spectrum supported transform-limited pulses of 180 as. The streaking field had insufficient intensity to obtain an accurate measurement of the attosecond pulse duration from CRAB [23], so the reconstruction was not performed. However, the interferometer locking is evident in the experimental trace in Fig. 4(b). The integration time at each delay step was 10 s, and the measurement took roughly 20 minutes. Even with such a short time required for the measurement, the delay drift was enough to smear out many features of the trace. Without interferometric control, such a streaking trace could be accurately measured only if all air

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fluctuations and mechanical vibrations were adequately damped. Since measurements with attosecond XUV pulses require that the beam propagates under vacuum, isolating the vibration is not only difficult, but also can be quite costly. Furthermore, measurements requiring longer integration times, such as those using gas targets with small XUV photoabsorption cross-sections, would be impossible.

3. Conclusion

Measurement of true attosecond dynamics requires active stabilization and control of the pump-probe delay in unique interferometer configurations. We have for the first time demonstrated a method to copropagate a CW laser with an attosecond XUV beam to both stabilize and control the relative delay between the pump and probe arms to within 20 as RMS in such a special interferometer. This technique overcomes the problems of passively "stable" interferometers, allowing for easily repeatable experiments and making measurements requiring long integration times possible. Furthermore, the method is economical, simple to implement and requires little modification to existing setups.

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