Self-Referencing, Spectrally, or Spatially Encoded Spectral Interferometry for the Complete Characterization of Attosecond Electromagnetic Pulses

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We propose a method for the complete characterization of attosecond duration electromagnetic pulses produced by high harmonic generation in an atomic gas. Our method is based on self-referencing spectral interferometry of two spectrally sheared extreme ultraviolet pulses, which is achieved by pumping the harmonic source with two sheared optical driving pulses. The resulting interferogram contains sufficient information to completely reconstruct the temporal behavior of the electric field. We demonstrate that such a method is feasible, and outline two possible experimental configurations.

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Recent advances in laser science have enabled dynamics to be observed in physical systems on the time scale of attoseconds [1]. This heralds a new era in the time-resolved spectroscopy of matter for which improved metrology of soft-x-ray extreme ultraviolet (XUV) attosecond pulses will be a key technology in the development of sources and experiments. In the past decade several robust and reliable methods for characterizing the temporal structure of ultrafast optical pulses have been demonstrated, primarily based on spectrography and self-referencing spectral interferometry [2,3]. Each method requires a time-dependent filter which is most often implemented using a nonlinear interaction [4], which for XUV pulses is normally photoionization of an atom by the pulse and the acceleration of the liberated electron by a synchronized optical field. This phenomenon has been used in several schemes for estimating the duration of an XUV pulse [1,5–8] and also to measure vacuum ultraviolet pulses by means of spectrography [9]. This method may be adapted for attosecond duration pulses with some modification [10,11]. Another technique uses this nonlinear process as a frequency modulator in order to measure the average temporal shape of individual attosecond pulses in a pulse train by means of spectral shearing interferometry [11–13]. In all cases, the optically accelerated photoelectron spectra require significant averaging to obtain adequate signal to noise ratios, making accurate reconstruction of individual XUV pulses difficult. Moreover, reconstructions are susceptible to the inevitable fluctuations of the optical pulse energy and shape, as well as the noninstantaneous response of the atoms to the fields, which must be accounted for in extracting information about the field from the measured photoelectron spectra. Therefore it is worth considering whether there is an alternative to measuring photoelectrons that could also provide the spectral phase information needed to completely characterize the electric field of an attosecond pulse.

In this Letter, we outline a technique for the complete characterization of both isolated attosecond XUV pulses and pulse trains based on spectral phase interferometry for direct electric field reconstruction (SPIDER) [2], where the signal is measured by directly detecting the XUV photons, rather than photoelectrons. The advantage of this method is the possibility of larger signal to noise ratio, and therefore of single-shot measurement, as well as higher accuracy from data accumulated over multiple shots. It also obviates the need for a complex and costly photoelectron spectrometer. Adapting standard optical pulse characterization methods to the XUV cannot be accomplished by known nonlinear mechanisms. Therefore we propose a different approach: synthesizing an appropriate XUV spectrum by manipulation of the driving pulse field, rather than the XUV harmonics themselves. We show in this Letter that it is possible in principle to generate pairs of pulses that are identical except for a frequency shear, and that the simultaneous detection of these pulses in an XUV spectrometer yields an interferogram from which the spectral phase may be extracted directly. This offers the possibility of single-shot characterization of attosecond pulses with very rapid update rates, and therefore provides new experimental capabilities for attosecond spectroscopy. Moreover, the technique can be adapted to provide information about the spatial character of the field in addition to its temporal character [14]. This means that experiments to examine the complete space-time response of structures on the submicron scale with attosecond temporal precision are possible.

The basis of the new method is the realization that the XUV radiation produced when an optical pulse is incident on an atom depends on the details of the electric field of that pulse, in particular, its mean frequency. For pulses that are >3 optical cycles in duration, the generated XUV radiation has a spectrum that consists of several peaks, each at an odd harmonic of the fundamental pump pulse frequency. Thus a change in the fundamental frequency results in a scale change in the harmonic spectrum. This

means that a harmonic pulse train generated by an optical pulse of mean frequency ω and one generated by a pulse of mean frequency $\omega + \delta\omega$ will be spectrally sheared with respect to one another by $n\delta\omega$ at the nth harmonic. This allows the SPIDER method to be used to extract the electric field of the harmonic pulses. The advantages of this interferometric approach are that the inversion is direct, rapid and robust with respect to noise, as well as to imperfections in or nonuniformity of the spectrometer response. This robustness is a consequence of the spectral phase being encoded in the spacing of the fringes of the XUV spectrum, not their amplitude. Therefore only the wavelength calibration of the spectrometer is needed. This is in contrast to spectrographic methods for which the detector response must be normalized; for example, in photoelectron-based methods the atomic response function must be deconvolved from the measured spectrogram.

In order to demonstrate that this mechanism works in principle, we have performed numerical simulations corresponding to the experimental setup drawn in Fig. 1(a). The principle applies to very short wavelengths and very short pulses, but the parameters we use demonstrate the phenomenon effectively. The nonperturbative model used has been applied previously to simulate above threshold ionization of gases and gives excellent agreement with experiments [15,16]. It involves the numerical integration of the 3D time-dependent Schrödinger equation (TDSE) for argon with optical pulses incident so as to implement

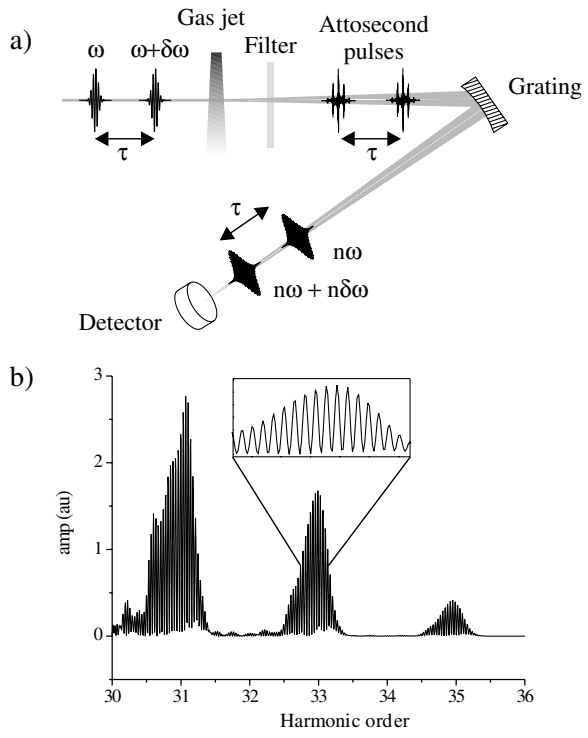

FIG. 1. (a) Schematic diagram of the arrangement for measuring XUV pulses using SPIDER, where n denotes harmonic order; (b) Simulated XUV-SPIDER signal generated in argon for $\lambda_1 = 800$ nm, $\lambda_2 = 804$ nm, and $I = 1.7 \times 10^{14}$ W/cm^2.

this technique (called XUV-SPIDER). Two replicas of the optical pulse are delayed from each other by a time τ. They have temporally identical optical fields, except that one has a slightly different mean frequency from the other [2]. Each of the replicas generates an XUV pulse with a different frequency. These are identical, except the frequencies are scaled by the spectral shear Ω, that is, the spectra are given by $\tilde{E}(\omega)$ and $\tilde{E}(\omega + \Omega)$, where $\Omega = n\delta\omega$ is the shear of the nth harmonic if the driving pulse mean frequencies differ by $\delta\omega$. The pair of XUV pulses is incident on a spectrometer, giving rise to interference fringes in the XUV spectrum as illustrated in Fig. 1(b). The interferogram is related to the input x-ray pulse fields by

$$S(\omega) = \left| \tilde{E}(\omega) + \tilde{E}(\omega + \Omega)e^{i\omega\tau} \right|^2. \quad (1)$$

It is clear that the interference fringes are determined by the spectral phase difference function $\varphi(x, \omega) - \varphi(x, \omega + \Omega)$. The phase difference between the spectral components of the x-ray pulse at frequencies ω and $\omega + \Omega$ may be extracted using standard Fourier processing techniques, enabling the spectral phase $\varphi(\omega)$ to be reconstructed. Since the x-ray pulse exists only within a temporal window of duration T, the Whittaker-Shannon theorem allows the complete reconstruction of the pulse field by sampling of the pulse spectrum at frequency intervals that are multiples of $2\pi/T$. Therefore, the spectral shear Ω must be less than this interval. The spectral phase, when combined with the spectrum, completely characterizes the pulse whose temporal field can be obtained via an inverse Fourier transform. The case of an isolated attosecond pulse is straightforward. However, for a long train of pulses there may be a limitation to this technique. In particular, if the spectrum consists of a set of distinctly separated harmonics, i.e., when the spectral intensity between two adjacent harmonic peaks falls below the detection noise floor, it is not possible to extract the relative phase between the two. Nevertheless, we are able to reconstruct the complete spectral phase from a single XUV-SPIDER interferogram across many harmonics for driving pulses of comparable duration to the longest currently used in experiments generating attosecond pulse trains.

Figure 1(b) shows the spectrum obtained when a pair of 30 fs pulses, temporally separated by 77 fs and with mean wavelengths of 800 and 804 nm, are incident on gaseous argon. The peak intensity of each driving pulse is about 1.7×10^{14} W/cm^2, giving a total ionization yield of $\sim 10^{-3}$. We have checked that harmonics generated with 800 and 804 nm pulses are almost identical, but frequency sheared, replicas. The interferogram has a signal to noise ratio of $\sim$40 dB and can be inverted to give the spectral phase of the XUV pulse, as shown in Fig. 2. It can be seen from the figure that the reconstruction is excellent for both the individual harmonics and the relative phase between them. The XUV pulse is reconstructed using conventional

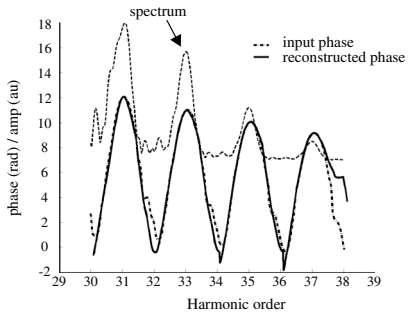

FIG. 2. The spectral phase retrieved from the interferogram in Fig. 1.

SPIDER inversion methods. This inversion can be both rapid and accurate—1 kHz repetition rates for SPIDER have been demonstrated [17]. The ability to measure pulse fields with a single shot will be critical for low-repetition rate sources since harmonic generation is a high-order nonlinear process, and very sensitive to changes of the driving pulses from shot-to-shot.

The above scheme is applicable to pulses with almost arbitrarily short duration, as long as a shift in frequency of the pulse can be achieved by a shift in the pump pulse frequency [18]. The requirements for implementing XUV-SPIDER in the conventional manner described above are not stringent. First, it is necessary to generate two replicas of the pump pulse that are identical except for a frequency shift. This is possible using conventional optical pulse shaping methods [19]. Second, the resolution of the XUV spectrometer must be sufficient to allow sampling of the fringes at the Nyquist limits, which requires a 0.01 nm resolution for an x-ray photon at 25 nm. Third, the phase difference between the two replicas can be extracted directly using Fourier processing only if there is a time delay between the pulses, since it allows the useful interferometric component in the XUV-SPIDER signal to be separated from the other information. For proper pulse reconstruction it is important to accurately calibrate the delay between the interfering pulses. This is easily measured to the required precision by extracting the phase from an interferogram resulting from harmonics created with the same delay but without a spectral shear between them [20,21].

The major drawback to this configuration is that ionization due to the first driving pulse should not significantly distort the medium seen by the second driving pulse, which limits the intensity of the first pulse and hence reduces the maximum order of the harmonics that can be characterized. This difficulty can be avoided if the geometry of the nonlinear interaction is altered, so that the interferogram has a spatial, rather than spectral, carrier [14,22]. The setup for attosecond pulse characterization using the spatially-encoded scheme is shown schematically in Fig. 3(a). The two driving pulses are now separated in space but not delayed in time. The two spectrally sheared harmonic pulses are generated in spatially separated regions at the same time, thus avoiding the ionization problem. The harmonic radiation propagates to a spectrometer with a toroidal grating, which records the spatial interference pattern [23–25] as a function of XUV wavelength. The encoding of the phase information in this geometry is achieved by interfering the energy-shifted XUV pulses after they have propagated away from the generation region so that they overlap due to diffraction, leading to spatial fringes at the output of the spectrometer. The spatial fringes allow the useful interferometric component to be separated using the same kind of Fourier processing techniques as in conventional SPIDER [22]. This configuration relaxes the constraint on the frequency resolution of the spectrometer by about an order of magnitude and since there is no time delay between the interfering pulses, the calibration is also simplified.

The interferogram measured in this spatially-encoded arrangement for SPIDER (SEA-SPIDER) [22] has the form of a spatially and spectrally resolved XUV pulse spectrum:

$$S(x,\omega) = \left| \int dx' [\tilde{E}(x',\omega) + \tilde{E}(x',\omega + \Omega) e^{i\kappa x'}] e^{i(kx'^2/2L) + i(kx'x/L)} \right|^2. \quad (2)$$

Here κ is the difference in the mean transverse wave vectors of the driving pulses, given by $\kappa = kX/L$, where X is the lateral separation between the driving pulses in the

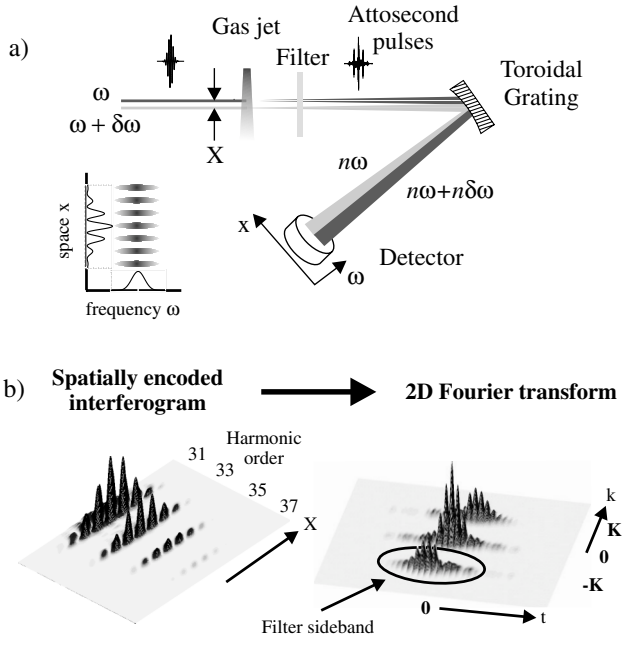

FIG. 3. (a) Schematic arrangement for SEA-SPIDER characterization of attosecond pulses produced by high harmonic generation. (b) Diagram showing spatial interference fringes for SEA-SPIDER, conversion to the pseudotime/k-space domain and sideband filtering.

gas jet, L is the distance from the sources to the detector, and k is the mean wave vector of the XUV spectrum. The separation of the driving pulses is constrained by the spatial resolution of the spectrometer, the distance to the detector, and the wavelength of the XUV radiation. It needs to be large enough that the two pulses do not overlap spatially but close enough that the gas jet density is not significantly different in the two generation regions. As an example, we take a detection system with a pixel size of 15 μm, and require there to be 6 pixels per spatial fringe, i.e., the fringe spacing is 90 μm. For a wavelength of 26 nm and a detector-source distance of 50 cm this gives a driving pulse separation of $\sim$145 μm. This is perfectly feasible, as is demonstrated in [24], where stable spatial interference fringes are observed between harmonics generated by driving pulses which have a focal diameter of $<$50 μm and are separated by 150 μm. As the SEA-SPIDER signal is spatially resolved it is possible to extract the temporal shape of the XUV pulses at each transverse coordinate across the beam. The reconstruction of harmonic phase at a single location in the beam from simulated SEA-SPIDER data is identical to that shown in Fig. 2 for XUV-SPIDER.

The spectral correlation term is extracted from the SEA-SPIDER interferogram using the same algorithms as in conventional SPIDER, except that the Fourier transform is taken with respect to the spatial axis, so that the interferometric component is extracted in the k_x domain. After the relevant interference term is selected, an inverse Fourier transform is taken, and the resulting function resolved with respect to the XUV beam transverse spatial coordinate x. Then the extracted phase is $\varphi(x, \omega) - \varphi(x, \omega + \Omega) + \kappa x$. This is the same as the spectral phase difference returned in conventional SPIDER. The calibration of the κx term can be performed by setting the shear between the two driving pulses to zero. This guarantees that the shear Ω is also zero, which returns the carrier phase term to be subtracted from the argument of the filtered and transformed sideband. This may also be used as a test of the accuracy and precision of the reconstruction, as any deviation from a linear phase term when Ω is zero represents a systematic error.

We have shown that both SEA-SPIDER and XUV-SPIDER enable the characterization of trains of attosecond x-ray pulses created by high-order harmonic generation (i.e., several separate harmonics). They will also be able to characterize individual attosecond pulses produced from a high harmonic continuum, which will indeed be simpler as fewer fringes will be needed in total for the phase reconstruction across a continuous spectrum than across separate harmonics. There are three important advantages to the SEA-SPIDER configuration. First, it avoids the problem of generating two harmonic pulses in the same region of the atomic gas; second, the detector spatial resolution required is a less stringent condition than that of the spectral resolution required for XUV-SPIDER and, third, the reconstruction returns the complete electric field of the pulse train at every point in the beam. Moreover, as with XUV-SPIDER, it is suitable for single-shot operation, thus raising the possibility of having a shot-to-shot diagnostic tool for attosecond pulses.

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