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Ponderomotive shearing for spectral interferometry of extreme-ultraviolet pulses

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We propose a novel method for completely characterizing ultrashort pulses at extreme-ultraviolet (XUV) wavelengths by adapting the technique of spectral phase interferometry for direct electric-field reconstruction to this spectral region. Two-electron wave packets are coherently produced by photoionizing atoms with two time-delayed replicas of the XUV pulse. For one of the XUV pulses, photoionization occurs in the presence of a strong infrared pulse that ponderomotively shifts the binding energy, thereby providing the spectral shear needed for reconstruction of the spectral phase of the XUV pulse. © 2003 Optical Society of America

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Ultrashort pulses in the extreme-ultraviolet (XUV) spectral region can be obtained by high-order harmonic generation and will soon also be produced by free-electron lasers. For ultrashort XUV pulses to be a useful tool for spectroscopy, a complete characterization of their time-dependent frequency¹⁻⁴ is needed. Although such time-frequency characterization of visible and infrared (IR) pulses is standard in many laboratories,⁵⁻⁷ its extension to the XUV region is nontrivial, mainly because of the difficulty of introducing nonlinear processes in this spectral region.^{8,9} There have been successful attempts to make autocorrelation and frequency-resolved optical gating measurements of harmonics,¹⁰ but these methods have so far been limited to low-order harmonics because of the nonlinearity required. For higher-order harmonics, cross-correlation techniques have been used, in which the XUV pulse is mixed with an IR probe pulse.^{11,12} However, the temporal resolution of these techniques is limited by the duration and (or) the period of the IR probe. In the case of attosecond pulses it is possible to use directly the electric field of the IR probe in order to improve the time resolution of the cross correlation.^{13,14}

In this Letter we propose a novel method for the complete characterization of an ultrashort XUV pulse by use of spectral phase interferometry for direct electric-field reconstruction^{6,7} (SPIDER) adapted for the XUV regime. The XUV SPIDER is based on the ionization of a gas by two XUV pulse replicas separated in time. The electron wave packets thus generated will carry all the information about the spectral phase of the XUV pulses. In an electron spectrometer the wave packets will then interfere, creating an interferogram.¹⁵ To retrieve the phase, one has to introduce a spectral shear between the two wave packets. For IR pulses this is accomplished by sum-frequency mixing of two replicas with a stretched pulse.^{6,7} This procedure cannot easily be extended to the XUV regime. Our proposal is, instead, to have a strong IR pulse interacting

with the gas at the same time as the second XUV pulse, thus ponderomotively shifting the ionization potential of the gas and thereby introducing a shear. The IR pulse has to be long enough to shift the photoelectron spectrum without changing its shape but short enough to overlap only one of the XUV pulses. More fundamentally, the XUV pulse cannot be shorter than the period of the IR field if the ponderomotive shift is to be well defined. A similar concept, proposed by Quéré *et al.*,¹⁶ uses the electric field of the IR pulse instead of the envelope to characterize pulses of duration shorter than 400 as. Our proposal, which applies to longer pulses and can therefore be used to characterize single harmonics, is complementary and aims at providing a practical method that can easily be applied by use of current technology.

In a normal SPIDER, two pulses, identical except for a shift in time by τ and a shift in energy by Ω (the shear), are sent into a spectrometer, and their spectral interference is recorded. The relative phase can be determined by measurement of the interference of a consecutive pair of frequencies ω , $\omega + \Omega$. Spectral intensity $A(\omega)$ of the pair of pulses will then be given by

$$A(\omega) = |\tilde{\epsilon}(\omega) + \tilde{\epsilon}(\omega + \Omega)\exp(-i\omega\tau)|^2, \quad (1)$$

where $\tilde{\epsilon}$ represents the spectral amplitude of one pulse. Knowledge of the spectral phase and amplitude within a set of frequency components spaced by $\Omega < 2\pi/T$, where T is the duration of the pulse, will uniquely determine the temporal structure of the pulse. To make an XUV SPIDER we generate two electron wave packets through ionization of a gas, using two XUV pulses separated by τ . One of the wave packets is shifted in energy by the simultaneous interaction of a strong IR pulse. Time-varying intensity $I_{\text{IR}}(t)$ of the IR pulse makes the ground state and thereby ionization potential $I_p(t)$ time dependent, such that $I_p(t) = I_{p0} + U_p(t)$, where I_{p0} is the unperturbed ionization potential and

$U_p(t) = e^2 E_{\text{IR}}(t)^2 / 4m\omega_{\text{IR}}^2 = \alpha I_{\text{IR}}(t)$, where e and m are the charge and the mass of the electron, E_{IR} is the electric-field strength, and ω_{IR} is the carrier frequency. Assuming a structureless ionization continuum for the excitation wavelengths, the photoelectron spectrum generated by an XUV photon in the presence of a strong IR field will be proportional to¹¹

$$A(\omega) \propto \left| \int_{-\infty}^{\infty} dt \exp\left[-i\left(\omega_{\text{XUV}} - \frac{I_{p0}}{\hbar} - \omega\right)t\right] \times \epsilon_{\text{XUV}}(t) \exp[i\Phi_{\text{IR}}(t)] \right|^2, \quad (2)$$

where

$$\Phi_{\text{IR}}(t) = (\alpha/\hbar) \int_{-\infty}^t I_{\text{IR}}(t') dt', \quad (3)$$

ϵ_{XUV} is the complex XUV field envelope, and ω_{XUV} is the XUV photon carrier frequency. The XUV photon frequency and the unperturbed ionization potential will result only in a constant, and for the XUV SPIDER unimportant, energy shift of the electron spectrum. Hereafter, these terms will therefore be left out of the equations, resulting in a spectrum centered around zero energy when no IR field is present.

To make a model of the XUV SPIDER we add a second XUV pulse. This pulse is a replica of the first but shifted in time by τ . The IR pulse is also shifted in time to affect the second but not the first electron wave packet. The physical principle behind the XUV SPIDER is shown in Fig. 1. The resultant photoelectron spectrum with all three pulses present is given by

$$A(\omega) \propto \left| \int_{-\infty}^{\infty} dt \exp(i\omega t) [\epsilon_{\text{XUV}}(t) + \epsilon_{\text{XUV}}(t + \tau)] \times \exp[i\Phi_{\text{IR}}(t + \tau)] \right|^2, \quad (4)$$

where τ is the separation between the two XUV pulses. Relation (4) can be rewritten as

$$A(\omega) \propto |\tilde{\epsilon}_1(\omega) + \tilde{\epsilon}_2(\omega) \exp(-i\omega\tau)|^2, \quad (5)$$

where $\tilde{\epsilon}_1(\omega)$ is the Fourier transform of $\epsilon_{\text{XUV}}(t)$ and $\tilde{\epsilon}_2(\omega)$ is that of $\epsilon_{\text{XUV}}(t) \exp[i\Phi_{\text{IR}}(t)]$. If the IR probe pulse is longer than the XUV pulses, the ponderomotive shift will be approximately constant across the entire second XUV pulse, such that $\Phi_{\text{IR}}(t) \approx \Omega t$. This gives $\tilde{\epsilon}_2(\omega) \approx \tilde{\epsilon}_1(\omega + \Omega)$, which turns relation (5) into Eq. (1). It is therefore important to make the probe pulse duration long that a constant shift can be introduced throughout the second pulse without affecting the first pulse.

In a numerical test of the method described, a Gaussian spectrum, with a bandwidth corresponding to a transform-limited pulse duration of 5 fs, was constructed with a complicated spectral phase. Using this spectrum, we created two temporally shifted pulses in the time domain. The temporal structure was non-Gaussian, with a pulse duration of approximately 7.5 fs. To recover the phase accurately requires the interferogram to contain approximately

20 fringes.¹⁷ With the pulses used in the simulation, a separation of 80 fs will give a sufficient number of fringes, spaced apart from one another by 50 meV. The spectrometer's resolution needs to be of the order of 50 meV, which is achievable by most electron spectrometers. With 80 fs between the XUV pulses, a 30-fs probe pulse can easily be delayed to overlap only one of the XUV pulses. The pulse configuration is illustrated in Fig. 2(a). The full electron spectrum is calculated from expression (4), and the resultant interferogram is included in Fig. 2(b) together with the sheared replicas. Using the same SPIDER algorithm as for visible or IR pulses, we reconstructed the phase, which is shown in Fig. 3 in combination with the input phase and spectrum of the XUV pulse.

We tested the method with a variety of phases and found it to be accurate as long as the XUV pulses were sufficiently short compared with the IR pulse. To measure longer XUV pulses we need a longer IR pulse to recover the phase accurately. In this case the delay between the XUV pulses has to be increased, which puts higher demands on the resolution of the electron spectrometer. The shortest measurable pulse duration is limited to approximately one IR cycle because it is necessary to have a well-defined ponderomotive shift but also because the sidebands generated will start to overlap the wings of the interferogram.

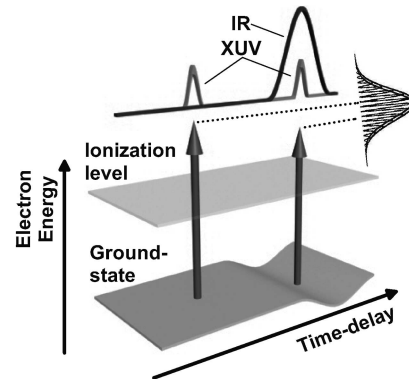


Fig. 1. Schematic of the XUV SPIDER. Two time-delayed XUV pulses generate two electron wave packets by ionizing a gas. The ground state is time dependent in the presence of a strong IR pulse, thus shifting the second electron wave packet in energy, introducing a spectral shear.

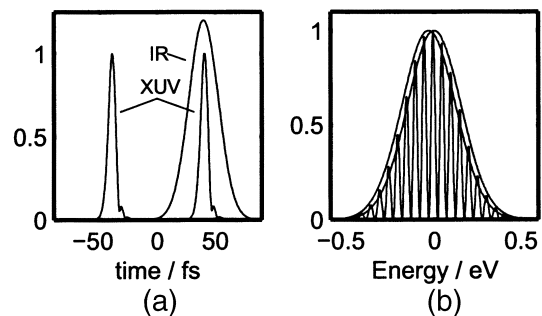


Fig. 2. (a) Pulses of the XUV SPIDER technique: two XUV pulses separated by 80 fs and a 30-fs IR pulse overlapping the second XUV pulse only. (b) Resultant XUV pulses' spectra and XUV SPIDER interferogram.

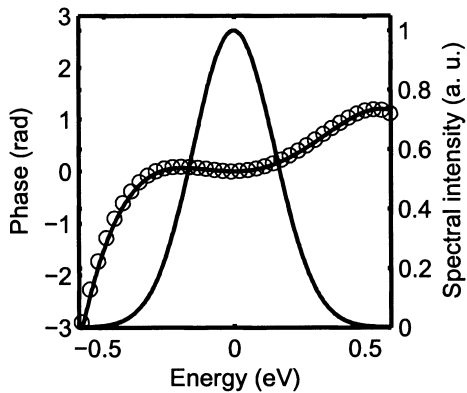


Fig. 3. Input spectrum and phase (solid curve) of the XUV pulse and the reconstructed phase (circles).

The generation of two XUV replicas constitutes the main difficulty in the experimental implementation of the XUV SPIDER. There are two solutions to this problem: The more intuitive alternative is to generate a single XUV pulse and then split it into two replicas; however, lack of good optics in the XUV spectral region makes this goal hard to achieve. For the second alternative it is assumed that the XUV pulses are produced through high-harmonic generation; the procedure consists of splitting the driving laser pulse into two replicas before the generation. This alternative is the more promising one because of its simpler design. A drawback is that the generation of the first pulse might affect the generation of the second, yielding two XUV pulses with different spectral phases. A separation of 80 fs between the driving laser pulses will enable two identical XUV pulses to be generated, provided that the intensity is not too high (i.e., is less than the saturation intensity).¹⁹

In conclusion, we have presented a novel method for the complete characterization of ultrashort pulses in the XUV spectral region. The validity of this method was tested by numerical simulations for XUV pulses available in many laboratories today. Detailed knowledge of the XUV spectral phase will improve the usefulness of such pulses and offer new insight into different XUV pulse-generation schemes.

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