Simultaneous measurement of two ultrashort near-ultraviolet pulses produced by multiplate continuum using dual self-diffraction dispersion-scan

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We present a new method based on self-diffraction dispersion-scan (SD d-scan) that enables the simultaneous measurement of two distinct ultrashort laser pulses in a region where they spatially and temporally overlap. This situation can arise when sampling and focusing two different spatial portions of a single inhomogeneous beam onto a medium. We demonstrate this new dual SD d-scan method by simultaneously characterizing two intense broadband ultraviolet pulses at 400 nm, with durations in the 10 fs range, originating from two different spatial portions of a beam produced by a multiplate continuum (MPC).

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Many experiments involving ultrashort laser pulses often require more than one beam. An important example are time-resolved pump-probe experiments, where in some cases both the pump and the probe pulses must be temporally characterized. Ideally, pulse characterization should be done on target and with the same interaction geometry used in the experiments, i.e., without changing the focusing conditions and the angles between the two beams and the sample, since these will also determine the actual temporal resolution. A different yet related situation can occur when apertures or d-shaped mirrors are used to spatially split a single beam into two (or more) beams, as commonly done in transient grating and degenerate four-wave mixing experiments. If the beam is inhomogeneous, its spectrum and spectral phase are spatially dependent, and two locally homogeneous but spatially separated regions within the beam profile will usually correspond to two different pulses. Several techniques have been developed that enable complete spatiotemporal characterization of ultrashort light pulses (see, e.g., [1–5]) but these are considerably more complex and difficult to implement than non-spatially-resolved methods.

In the two situations presented above, the ability to simultaneously characterize two different pulses in the most straightforward manner possible is highly desirable. Here we propose a method for the in-situ characterization of two overlapping ultrashort pulses using a new variant of self-diffraction dispersion-scan (SD d-scan) [6] and demonstrate its performance using two portions of an inhomogeneous ultraviolet (UV) beam produced by multiplate continuum (MPC).

The MPC technique has been recently proposed [7] for high-energy supercontinuum generation and pulse post-compression in solids while avoiding filamentation, by using a set of thin solid plates adequately spaced within the Rayleigh range of the focused input light beam. It has been demonstrated in a variety of spectral ranges, using a Ti:Sapphire amplifier [7], its second-harmonic at 400 nm [8], a tuneable near-infrared optical parametric amplifier (OPA) [9] and an Yb:KGW laser at 1025 nm [10]. MPC is also a suitable process to generate high-repetition rate carrier-envelope-phase stable pulses [11] and pump and seed pulses for ultra-broadband tuneable OPAs [12]. The approach is also scalable, since higher supercontinuum pulse energies can in principle be generated by distributing higher energy input pulses over a larger area in the plates. This fact is extremely interesting, but at the same time spatial inhomogeneities and distortions in the larger cross-section supercontinuum can arise [8, 9] due to subtle irregularities in the plates and in the input beam itself, or by incipient filamentation. The latter is more likely to occur when using high-energy photons in the ultraviolet region, or in general because self-focusing pulses undergo spatio-temporal transformations, which may lead to inhomogeneities across the beam [9]. Given that two different portions of such supercontinuum beams can be employed for pump-probe experiments, or for characterizing the pulses themselves via self-diffraction-based techniques (particularly important in the case of UV pulses [13, 14]), this further reinforces the need of simple and robust temporal diagnostics capable of characterizing two broadband pulses within spatially inhomogeneous beams. While many current pulse characterization methods rely on the condition that the pulse replicas participating in the required nonlinear optical effect are identical [8, 15, 16], which we will call the degenerate case, there are exceptions to this case, and several methods exist that rely on nonlinear optical effects produced by more than one input light field, such as XFROG [17], blind [18] and double-blind [19] FROG, and interferometric imaging of self-diffraction [20].

The d-scan technique [21] is a more recent approach for the temporal characterization and compression of femtosecond laser pulses that involves measuring the spectrum of a nonlinear process as a function of dispersion applied to the pulses, e.g., by using a pulse compressor, and retrieving the pulse from this measurement with an optimization algorithm. D-scan was first demonstrated with second-harmonic generation (SHG) [21, 22], which has enabled measuring pulses down to 1.04-cycles in duration [23], and has been extended to other nonlinearities, including third-harmonic generation (THG) [24], cross-polarized wave generation (XPW) [25] and self-diffraction (SD) [6]. The relatively high energy pulses generated by MPC allow implementing SD as the nonlinear process in the temporal diagnostic, which enables measuring pulses over the full transparency range of the nonlinear medium, typically from the infrared to the UV. Let us consider an arbitrary pulse in the frequency domain,

$$\tilde{E}(\omega) = \left|\tilde{E}(\omega)\right| e^{-i\phi(\omega)}, \tag{1}$$

with $\tilde{E}$ the electric field of the pulse and $\phi(\omega)$ its spectral phase. The measured d-scan trace, S_{meas}, is written as the product of a spectral response function, $R(\omega)$, and an ideal trace, S_{ideal} [22]

$$S_{\text{meas}} = R(\omega) \times S_{\text{ideal}} \equiv R(\omega) \left| \int_{-\infty}^{+\infty} E_{\text{NL}} e^{-i\omega t} \mathrm{d}t \right|^2, \tag{2}$$

where E_{NL} is the dispersion-dependent nonlinear signal in the time domain, which for SD d-scan implemented with a chirped mirror and glass wedge compressor is given by [6]

$$E_{\text{NL}}(t, l) = E^2(t, l) E^*(t, l), \tag{3}$$

where $E(t, l)$ is the time domain pulse after the compressor, i.e.,

$$E(t, l) \propto \int_{-\infty}^{+\infty} \tilde{E}(\Omega) e^{-ik(\Omega)l} e^{i\Omega t} \mathrm{d}\Omega, \tag{4}$$

with l the relative thickness of glass crossed by the pulse and $k(\Omega)$ the corresponding frequency-dependent phase per unit length. Applying the d-scan algorithm to the measured d-scan trace and linear spectrum enables retrieving both the electric field of the pulses and the response function $R(\omega)$ [6, 21, 22].

In the present nondegenerate SD case (see Fig. 1) we must consider the pulses from two portions of the beam to be different, and so the two generated SD pulses will also be different, as given by

$$E_{NL1}(t) = E_1^2(t) E_2^*(t) \tag{5}$$

and

$$E_{NL2}(t) = E_2^2(t) E_1^*(t). \tag{6}$$

Clearly, Eqs. (5, 6) are not invariant when exchanging E_1 with E_2, as in the degenerate case. Therefore, one equation or the other should be used in the retrieval, depending on which of the two generated SD signals is being measured. As we will show below, our proposed method enables simultaneous retrieval of the fields of the two pulses from a single d-scan measurement of just one of the SD signals. It can be easily shown that in order for two fields to generate the same dual SD d-scan trace as another pair of fields (with identical spectral intensities), their phases must differ from the latter by a constant value and/or by the same linear spectral phase. This contrasts with blind FROG, where different quadratic spectral phases can lead to the same trace even if the spectral intensities are preserved [26].

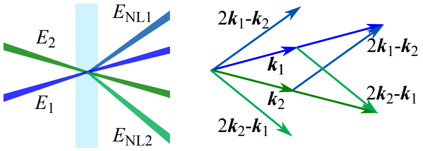

Fig. 1. Beam interaction geometry and wavevector diagram for nondegenerate self-diffraction.

The setups for both the MPC generation and the d-scan are depicted in Figure 2. A beam from a 1 kHz Ti:Sapphire amplifier (Femtopower Compact Pro CE-phase), centered at 800 nm and with a diameter of roughly 20 mm, undergoes a size reduction by passing through a telescope (×1/4). The beam then crosses a 200 μm-thick type-I BBO crystal, generating a SHG beam at 400 nm. The SHG beam was separated from the fundamental using a dichroic mirror and focused by a lens (f = 1000 mm). Five 100 μm-thick fused silica slides were placed at Brewster's angle along the Rayleigh range of the beam, with an average spacing of 4 cm, to maximize the spectral broadening. No further broadening was observed with additional slides. At the exit of the MPC, the beam is steered into the d-scan setup, comprising a pair of dispersion compensation mirrors (CM82, Ultrafast Innovations, GmbH) with a nominal group delay dispersion of -50fs^2 per bounce at 400 nm, where it experiences a total of 24 bounces, followed by a pair of fused silica wedges (one of them mounted on a translation stage to adjust the amount of positive dispersion). A mask with two circular holes is placed around the central part of the beam (see inset in Fig. 2). Due to the inhomogeneity of the original beam, the two beams that emerge after the mask have different spectra and can be considered as two distinct pulses. These two beams are then focused by a concave mirror (f = 200 mm) onto a 30 μm-thick fused silica slide and generate a dispersion-dependent nondegenerate SD signal that is collected into a fiber-coupled spectrometer.

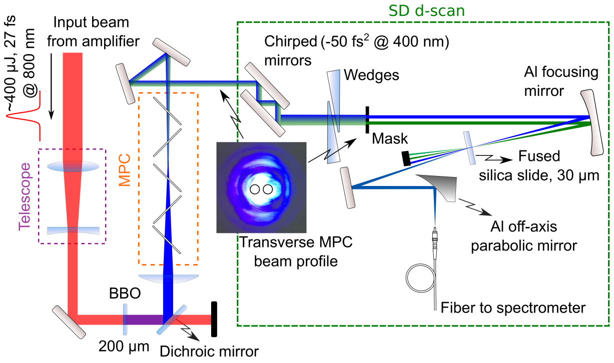

Fig. 2. Experimental setup for ultraviolet MPC pulse compression and dual SD d-scan (see text for more details).

The SHG power after the MPC slides was 60 mW for an input power of 72 mW, corresponding to an efficiency of 83 % similar to the one reported in [8]. After the mask, the total power in both beams was 1 mW. The diameter of the two circular apertures (1.2 mm) was chosen not only to ensure that the output power after the mask was small enough to avoid additional selfphase modulation in the fused silica slide, but also to obtain sufficiently homogeneous beams after each hole. An attempt at using smaller diameter (0.8 mm) holes proved unsuccessful in generating an observable self-diffraction signal.

To test the robustness of the method, we first performed numerical simulations using Eqs. (5, 6) and the measured spectra of pulses E_1 and E_2 to generate the d-scan traces of the two SD signals, E_{NL1} and E_{NL2}, assuming two different simulated spectral phases for each pulse (spline interpolations over 12 random points), followed by numerical retrieval of the two pulses using only signal E_{NL1} (Fig. 3) or only signal E_{NL2} (Fig. 4). We see that in both cases the retrieved spectral phases and temporal profiles of the two pulses are visually very similar to the simulated ones, with d-scan errors of ≈ 0.01 for 210×384 (insertion×frequency) sized traces. The corresponding standard deviations (obtained from 10 independent retrievals) are also shown (multiplied by a factor of 5 for visibility). This result supports our assumption that it is possible to retrieve two pulses with different known spectra and different unknown phases from the measurement of just one of the generated SD beams using the d-scan technique.

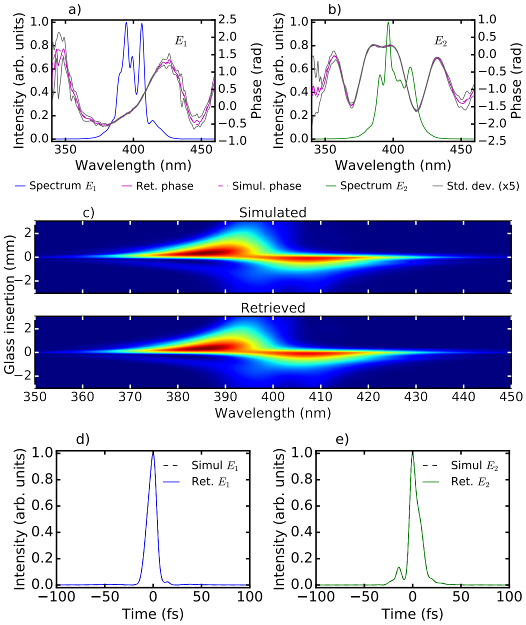

Fig. 3. Retrieval of two different pulses with unknown simulated phases by dual SD d-scan using signal E_{NL1}. (a, b) Measured spectra, simulated and retrieved phases of pulses E_1 and E_2. (c) Simulated and retrieved SD d-scan traces. (d, e) Simulated and retrieved temporal intensity profiles.

We now approach the problem of retrieving the actual phase of both pulses, E_1 and E_2, by measuring and retrieving the d-scan trace of one of the SD signals, namely E_{NL1}. To better illustrate the differences between standard SD d-scan and the new dual SD d-scan, we start by assuming that the two pulses are the same (A) - not a valid assumption in our case - and then proceed to the dual SD d-scan method (B).

A. Assuming an homogeneous beam (degenerate case)

In this case, we assume that the two fields E_1 and E_2 are identical. Figure 5 shows the corresponding d-scan analysis for signal E_{NL1}, using the linear spectrum of E_1 in the retrieval. In this case, the algorithm struggled to reproduce the measured trace and the retrieval had a large error (0.032). Even though the retrieved pulse duration (16.5±0.1 fs) has a relatively small uncertainty, this single parameter cannot reflect the overall larger uncertainty in the spectral phase compared to the simulations in Fig. 3. Similar results were obtained when using the linear spectrum of E_2. This further reinforces the notion that assuming identical pulses does not hold well for beams such as the one produced by MPC in the UV region, so this approach is not recommended in applications such as coherent control, where the phase of the field must be precisely known.

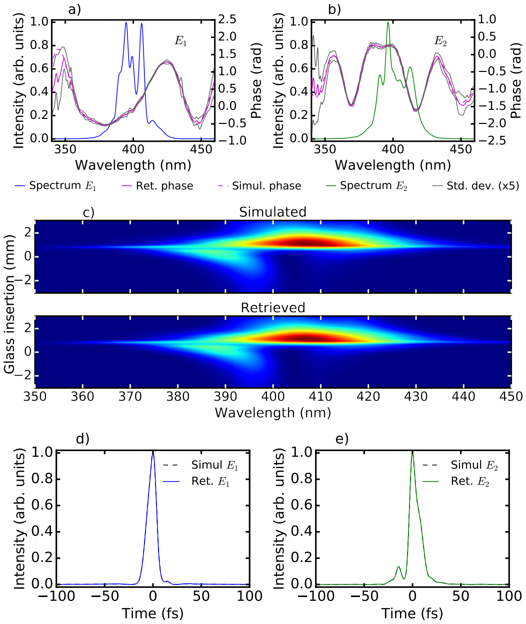

Fig. 4. Same as Figure 3, but using signal E_{NL2}.

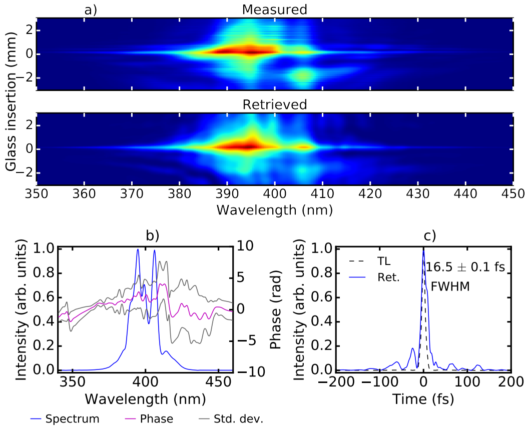

Fig. 5. D-scan analysis of signal E_{NL1}, assuming a degenerate case and the linear spectrum of pulse E_1. (a) Measured and retrieved d-scan traces. (b) Measured spectrum of pulse E_1 and retrieved spectral phase (standard deviation obtained from 20 independent retrievals). (c) Retrieved temporal intensity profile and transform-limited (TL) pulse (shown for reference).

B. Dual SD d-scan (nondegenerate case)

We now consider the case of two distinct fields, E_1 and E_2 (spectra and spectral phases bearing no relation between the pulses), exiting the mask to generate the self-diffraction signal. The corresponding analysis, assuming the nondegenerate signal in Eq. 5, is given in Fig. 6. In this case, the retrieved trace is visually more similar to the measurement (same as in Fig. 5), with a correspondingly lower d-scan error of 0.019. We obtain minimum (compressed) pulse durations of 8.5 ± 0.1 fs for pulse E_1 and 12.5 ± 1.7 fs for pulse E_2, which occur for different glass insertions, namely $z = 0.12$ mm (E_1) and $z = -0.79$ mm (E_2).

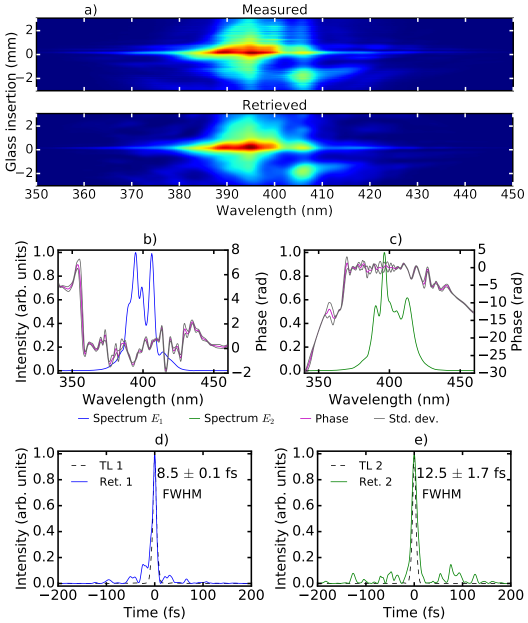

Fig. 6. Dual SD d-scan analysis of signal E_{NL1}. (a) Measured and retrieved d-scan traces. (b, c) Measured spectra of pulses E_1 and E_2 and retrieved spectral phases (standard deviations obtained from 20 independent retrievals). (d, e) Retrieved temporal intensity profiles of pulses E_1 and E_2 and corresponding transform-limited (TL) pulses.

In conclusion, we have proposed and demonstrated an implementation of SD d-scan, called dual SD d-scan, which allows simultaneous temporal characterization of two different pulses overlapping in a nonlinear medium. This method assumes that the SD process is produced by two distinct fields and is thus nondegenerate in nature. Dual SD d-scan enables measuring, e.g., pulses from different portions of a spatio-spectrally inhomogeneous beam, or overlapping pump and probe pulses in-situ, provided that the angle between the pulses is not so large as to prevent SD due to phase mismatch. The viability of dual SD d-scan was tested numerically and the diagnostic was then applied to two portions of a beam generated by MPC at 400 nm. The two pulses had similar intensities, but this method can in principle measure both a weak and a strong pulse (as used in most pump-probe experiments), since one of the SD signals will have a linear dependence on the weak pulse field. Since MPC is a promising technique for achieving large spectral broadening in solid media, and is scalable in energy by using larger area beams, we expect this dual SD d-scan diagnostic to be very useful for MPC with very high intensity lasers. The present dual SD d-scan technique and setup, as well as standard SD d-scan [6], can in principle be extended to deep-ultraviolet few-cycle pulses, since generating a spectrally resolvable SD signal is clearly possible [13, 14] and there are commercially available broadband chirped mirrors in this spectral range.

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