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Measuring ultrafast pulses in the near-ultraviolet using spectral phase interferometry for direct electric field reconstruction

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Abstract. A novel version of spectral phase interferometry for direct electric field reconstruction (SPIDER) based on parametric downconversion is demonstrated. This process is used to completely characterize low-energy, ultrashort optical pulses in the near-ultraviolet region of the spectrum.

Recent progress in the characterization of ultrashort optical pulses has led to the development of rapid and robust techniques for measuring pulses in the near-infrared [1–7] as well as in the ultraviolet regions of the spectrum [8–11]. All of these methods necessarily rely on nonlinear optical processes, since it is the only means available at present to implement a sufficiently rapid linear filter with non-stationary response [12]. In the near-IR it is possible to use sum-frequency generation (based on a $\chi^{(2)}$ response) as the nonlinear process [13]. It is not currently feasible to use this nonlinear process in the blue spectral region (fundamental wavelengths shorter than about 300 nm), however, since the most common nonlinear crystals are not transparent in this wavelength range, nor is it practical to phase match the upconversion process. For this reason, self-referencing techniques for measuring blue pulses have made use of higher-order nonlinear processes, which require higher pulse energies or significant signal averaging to produce a useable signal. In this paper we demonstrate a new technique for the complete characterization of ultrashort pulses at wavelengths in the blue spectral region, potentially as short as 170 nm. The method is based on a modification of spectral phase interferometry for direct electric field reconstruction (SPIDER) that makes use of $\chi^{(2)}$ a nonlinearity to implement the nonlinear time gate by means of difference frequency generation [14].

The same nonlinear process has been employed recently for characterizing blue pulses using a known reference pulse in the infrared. The method, called difference

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frequency generation cross-correlation frequency resolved optical gating (DFG XFROG) [10], is a spectrographic technique in which the spectrum of the cross-correlation signal between an unknown blue pulse and a previously characterized infrared reference pulse is recorded. Knowledge of the reference pulse field (obtained using FROG) is required for the field of the blue pulse to be reconstructed from the measured spectrogram by means of iterative deconvolution.

A straightforward modification of SPIDER enables complete characterization of blue ultrashort pulses by downconversion: we therefore label the method DC-SPIDER. The fundamental concepts involved are the same as those previously reported for SPIDER. In both versions spectral shearing is implemented by mixing two replicas of the input pulse with a strongly chirped pulse in a nonlinear crystal. Each member of the replica pair is mixed with a different frequency of the chirped pulse, because each time slice of this pulse corresponds to a different wavelength. In this case the shear is proportional to the delay between the two replicas. The frequency shifted and sheared pair is then sent into a spectrometer and the resulting spectral interferogram is recorded on a one-dimensional detector array.

In DC-SPIDER, an *uncharacterized* chirped pulse in the infrared acts as a signal wave, and the unknown blue pulse acts as the pump for the downconversion process. The idler wave is then a frequency-shifted replica of the signal wave. The method is entirely self-referencing as far as the blue pulse is concerned, and requires no knowledge of the spectral phase of the reference pulse. The inversion algorithm is identical to that used for SPIDER [2,15]. The interferogram is of the form

$$\begin{aligned} D(\omega_{BLUE} - \omega_0) = {} & |E(\omega_{BLUE} - \omega_0)|^2 + |E(\omega_{BLUE} - \omega_0 - \Omega)|^2 \\ & + |E](\omega_{BLUE} - \omega_0)||E(\omega_{BLUE} - \omega_0 - \Omega)| \\ & \times \mathrm{e}^{i\{\varphi(\omega_{BLUE}) - \varphi(\omega_{BLUE} - \Omega) - (\omega_{BLUE} - \omega_0)\tau\}} + c.c. \end{aligned} \tag{1}$$

where ω_{BLUE} is a blue frequency of interest, ω_0 is the frequency which seeds the downconversion for the leading replica pulse, Ω is the spectral shear, and τ is the temporal delay between the two replicas. Using standard processing algorithms [5] one can extract the argument of the cosine term and remove the known calibration phase $(\omega_{BLUE} - \omega_0)\tau$. The remaining phase, $\varphi(\omega_{BLUE}) - \varphi(\omega_{BLUE} - \Omega)$, is the relative phase between frequencies separated by the spectral shear. Provided the shear is chosen to satisfy certain sampling criteria, this is sufficient information to reconstruct the temporal field completely [2]. The data inversion is direct, as in traditional spectral interferometry.

As a demonstration of this method we used a test pulse generated by frequency-doubling the output of our chirped-pulse amplified laser system (CPA). The output of the CPA was a train of 400 μJ pulses at 1 kHz repetition rate, whose duration was measured, using both SPIDER and a scanning autocorrelator, to be 56 fs.

The DC-SPIDER apparatus is similar to that of regular SPIDER and is illustrated in figure 1. The laser pulse was first sent to a 50/50 beamsplitter, from which one of the output pulses was attenuated and then frequency-doubled in a type I BBO crystal. This produced blue pulses with energy of about 70 nJ. The blue pulse was then incident on a 250-μm-thick etalon which generated a pair of weak (3 nJ) blue pulses. The beam transmitted through the beamsplitter, at the

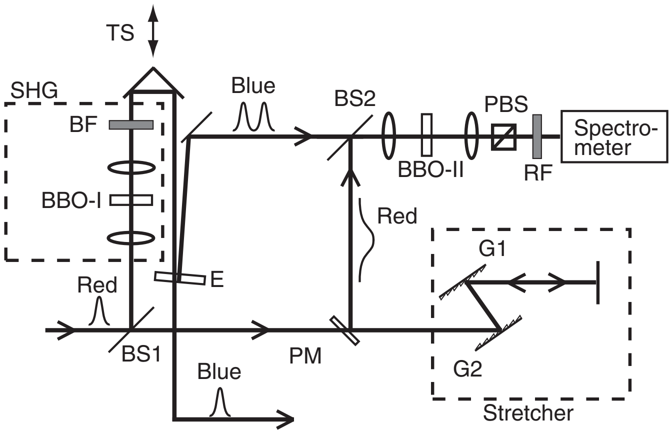

Figure 1. Experimental setup for DC-SPIDER. A red (825 nm) pulse is split into a pulse that is then frequency-doubled (the unknown pulse to be measured) and a second pulse to be chirped, in the stretcher. A replica pair is generated from the unknown blue pulse, and the pair is then overlapped with the chirped pulse on a second beamsplitter. The chirped pulse consequently seeds spectrally sheared downconversion for the blue replicas. BF = Blue Filter, RF = Red Filter, G1 = Grating 1, G2 = Grating 2, E = Etalon, PBS = Polarizing Beamsplitter.

fundamental frequency, passed through a grating pair with a second-order dispersion coefficient of -4.84×10^5 fs^2. This stretched the pulse to about 20 ps, so that it was highly chirped. The blue and fundamental beams were then combined colinearly on a broadband beamsplitter and directed to a 250-μm-thick type II BBO crystal. The blue pulses acted as a pump for the downconversion process, and the chirped red pulse seeded the signal beam. This resulted in the generation of idler pulse pairs polarized orthogonally to the pump beam.

The spectrometer used to measure the downconverted pulse-pair spectrum was an ISA Model HR320139838 whose focal plane was imaged onto a 1D Hamamatsu S3903-512Q CCD array. The array was read into and analysed by a PC computer using LabView 4.0 software. The acquisition and analysis operated at 10 Hz, so each recorded interferogram was actually the sum of 100 interferograms generated from individual pulse pairs. A calibration interferogram in the blue was measured simultaneously with the DC-SPIDER interferogram in the red by making use of the second-order grating reflection in the spectrometer [4].

We note that this apparatus is flexible enough to operate as a standard SPIDER with only minor modification. In particular, if the second beamsplitter is broadband the apparatus can run in upconversion or downconversion mode, simply by rotating the type II crystal through 90 degrees. This dual-mode capability exemplifies the simplicity and robustness of DC-SPIDER. Furthermore, since the upconversion process is much more efficient than downconversion, it is possible to generate both upconversion and downconversion signals simultaneously by rotating the crystal slightly off from the phase-matched downconversion orientation. One could then measure both red and blue signals simultaneously on a single array and extract both spectral phases simultaneously.

The results of the measurement are shown in figures 2 and 3. The input pulse field was measured using the standard SPIDER method. The measured spectral

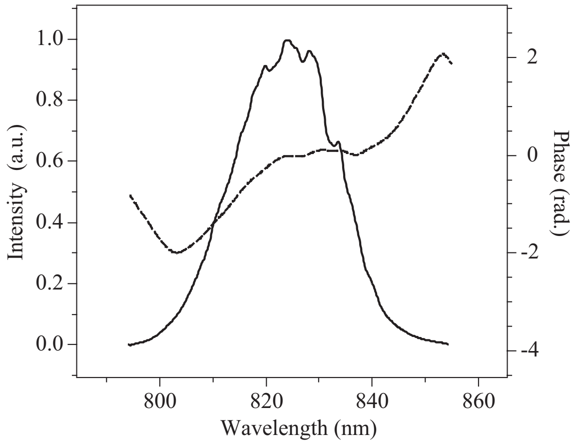

Figure 2. Spectral intensity (——) and spectral phase (– – –) of the near-infrared pulse before frequency doubling, as measured by SPIDER. Note the cubic phase structure over the region of significant spectral amplitude.

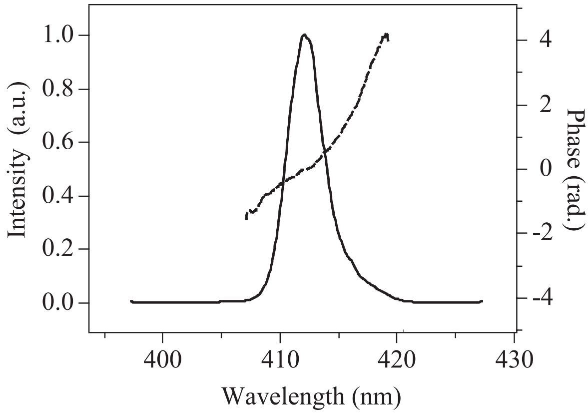

Figure 3. Spectral intensity (——) and spectral phase (– – –) of the input second-harmonic pulse, as measured by DC-SPIDER. Note the less structured spectrum expected in second-harmonic generation with low conversion efficiency, and the preserved shape of the spectral phase of the fundamental.

amplitude and phase of the input pulse field are shown in figure 2, and those of the blue pulse in figure 3. These pulses were generated when an infrared pulse of energy 1.2 μJ was focused to a spot of 55 μm on the second-harmonic crystal. The conversion efficiency was 17%. Note the strong modulation of the spectral phase, and the relatively smooth intensity spectrum.

Some measure of the fidelity of the blue pulse reconstruction can be obtained by a comparison of the parameters of the measured input and output pulses. The power bandwidth of the input infrared pulse before upconversion is approximately 23 nm FWHM, or 10.2 THz. The largely cubic spectral phase profile ranges over 4 radians, across 22.6 THz of bandwidth (beyond this there is not enough signal to reconstruct the phase precisely). This results in an temporal pulse duration of 56 fs (RMS). The signal to noise ratio for these measurements was 50:1, which implies negligible error for phase and temporal reconstruction [15].

The bandwidth of the blue second-harmonic radiation is approximately 4 nm FWHM, or 7.2 THz. The measured phase profile of the blue pulse clearly has a component that is linear and one that is cubic in frequency. The linear component represents an arbitrary pulse delay that has no physical meaning. The remaining cubic spectral phase ranges over 2.4 radians, across 18 THz of bandwidth. The temporal duration of the blue pulse is therefore approximately 140 fs (RMS). The signal to noise ratio of the measured interferogram was 10:1, which gives about 2% accuracy in the temporal reconstruction when the energy spectrum signal to noise is high (also 50:1) [12]. It should be noted that the window function of the SPIDER algorithm significantly filters the noise [16].

The bandwidth and duration of the blue pulse are consistent with both the dispersion of the frequency-doubling crystal used in the experiments, and the fact that the input pulse has a cubic chirp. This implies a bandwidth that is approximately the same as that of the input pulse, providing the spectral shapes are similar. However, because of the distortions arising from the interplay of various nonlinear effects, such as the nonlinear refractive index of the crystal, cascaded nonlinearities and blue-induced infrared absorption, we do not expect such simple calculations to give a reliable estimate for the output pulse duration and bandwidth.

Several scientists have already presented spectra from ultrafast SHG processes which they were unable to explain with their models. For example, Furbach *et al.* directly upconverted a high-power Ti:Sapph oscillator in a bulk doubling crystal [17]. Their model, based on standard plane-wave interactions, could not explain the rapid structure of their spectrum or its bandwidth. This was attributed to the tight focus of the beam into the doubling crystal. Gallman *et al.* noticed similar fluctuations in their upconverted spectrum when doubling a Ti:Sapph oscillator in a QPM structure [18].

We are currently developing a comprehensive theoretical model of pulse propagation to better understand the sensitivity of the process to small angular phase-matching, space-time coupling, competing nonlinear processes and dispersion.

The DC-SPIDER technique provides a new means for experimental exploration of the propagation of ultrashort pulses in nonlinear media. In particular, it is now possible to extend the pioneering studies of continuum generation [19, 20] into a new spectral regime where dispersive properties are quite different from those in the IR, and where second-order nonlinearities, such as the generation of second-harmonic radiation, play a central role. Experiments using higher-order nonlinearities are also possible.

In summary, we have developed a new version of SPIDER that enabled us to fully characterize femtosecond optical pulses in the blue region of the spectrum while remaining self-referencing. The phase retrieval algorithm for this technique is direct, non-iterative, and capable of operating at 20 Hz with standard lab equipment and modest computational power. Furthermore, the setup is simple and robust. We demonstrated this method by measuring the amplitude and phase of blue pulses obtained by frequency doubling the output of a CPA.

The sensitivity of the apparatus can be significantly improved by generating the pulse pair with a Michelson–Morley interferometer rather than an etalon. This would increase the blue pair intensity entering the SPIDER apparatus by a factor of five, at the expense of half of the output blue power. In addition, the use of an appropriate dichroic beamsplitter in the apparatus (BS2) would increase the blue power at the downconversion crystal by a factor of 1.5, and the infrared by a factor of four. This would improve the SNR of the interferogram by a factor of about thirty. With these improvements our data could be reproduced with an input pulse energy of 1.5 nJ. In fact, by reducing the SNR and using longer detector integration times (thus performing reconstructions at a lower repetition rate), reliable measurements of the pulse shape could be made at energies as low as 100 pJ. It is important to note that although the above experiment measured a blue signal accompanied by its fundamental, this is not a necessary requirement. The chirped pump pulse can be generated independently of the signal pulse since no previously known phase relationship, other than their relative delay, is required between the two. We expect that limitations on the brevity of pulses that can be measured using this method are similar to those for upconversion SPIDER [2, 4, 14, 15] since the crystal's downconversion bandwidth is very large.

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