Gold-SPIDER: spectral phase interferometry for direct electric field reconstruction utilizing sum-frequency generation from a gold surface

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We report on a version of spectral phase interferometry for direct electric field reconstruction (SPIDER) that uses no nonlinear crystals, instead relying on the harmonic signal from a gold mirror. Surface harmonic generation holds the promise of being able to upconvert extremely broad bandwidths over a large tuning range, thus providing access to both extremely short pulses and wavelengths outside of traditional methods. In this proof of principle demonstration, SPIDER traces for chirped and transform-limited 55 fs pulses are presented.

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1. INTRODUCTION

Ultrashort pulse-measurement techniques have been an active area of research for the past several decades. The initial investigations by Bradley and New [1] on characterizing picosecond pulses emphasized the importance of being able to measure the temporal or spectral phase of light fields. Ultrashort laser pulses, with their short temporal duration and large spectral bandwidths, pose a unique problem. Techniques such as autocorrelation reveal the temporal intensity profile, and a spectrometer or scanning monochrometer reveals the spectral intensity profile. But recovering the full field information, amplitude and phase, requires more-sophisticated techniques. Approaches include interferometric autocorrelation (IAC) [2], frequency-resolved optical gating (FROG) [3], phase and intensity from spectrum and autocorrelation (PICASO) [4], dynamic sonograms [5], and spectral phase interferometry for direct electric-field reconstruction (SPIDER) [6]. The majority of these techniques rely on harmonic generation from a nonlinear crystal to synthesize time gates or temporal modulators suitable for femtosecond pulses. Nonlinear crystals, however, still provide some obstacles: They are relatively expensive, they require careful alignment, and their phase-matching bandwidth scales as the inverse of their length. This necessarily limits the wavelength tunability of these devices and the ability to measure short pulses of ever-increasing bandwidth. Some pulse-measurement techniques already sidestep the use of nonlinear crystals. For instance, autocorrelators [7] and sonogram-based devices [8] now routinely use two-photon detectors, and versions of SPIDER in the time domain that use pulse shaping and a two-photon photodiode have been presented [9]. The two-photon absorption method provides a broader response [10] than nonlinear crystals but is still somewhat limited in that the bandgap of the material must be bigger than one-photon energy but smaller than the energy of two photons ($\hbar\omega < E_{\mathrm{gap}} < 2\hbar\omega$). Recently, surface harmonic generation from metallic surfaces has gained interest in the ultrafast community, because it is inexpensive and simple to align, and has an extremely broad upconversion bandwidth [11–13]. It is also possible to perform second-harmonic generation (SHG) and third-harmonic generation (THG) from dielectric mirrors, where THG often dominates SHG [14]. In this paper we wish to test whether surface harmonic generation will provide the necessary nonlinear interaction to produce the requisite spectral shear for SPIDER to function. As a testbed we present Gold-SPIDER, a variation of classic SPIDER that instead uses the harmonic generation from a gold mirror to perform the sum-frequency upconversion.

2. SURFACE SECOND-HARMONIC GENERATION

If a medium possesses inversion symmetry, the second-order nonlinear coefficient vanishes. These materials cannot generate second-harmonic light. However, at an interface, the inversion symmetry is necessarily broken, leading to a nonzero nonlinearity. Incident light at frequency ω can generate light at frequency 2ω. Likewise, two incident beams at frequency ω_1 and ω_2 can generate sum-frequency light at $\omega_1+\omega_2$. SHG at interfaces has a long history. The first description of this phenomenon was by Bloembergen and Pershan in 1962 [15]. Since then, several seminal papers have appeared, and a good discussion of the physical picture and history is presented by Shen [16]. For many researchers, surface SHG has provided a means to study the properties of the interface and in particular to study the adsorption of molecular layers onto substrates, permitting submonolayer molecular sensitivity [17]. Surface harmonic generation is possible from dielectrics [18] and metals [19], and since the harmonic generation relies on the first few atomic layers, there is essentially no phase-matching requirement, thus permitting SHG over an enormous range of frequencies, from IR to UV, limited only by the plasma frequency which is approximately 10 eV (120 nm) for gold.

Surface harmonic generation has been utilized as a tool for ultrafast laser diagnostics. Autocorrelators based on the second and third harmonic signal from metal surfaces have been realized with near-IR ultrafast pulses [13,20,21]. UV pulses of 248 nm have been autocorrelated using surface SHG from silicon [11]. Second harmonic has also been generated from interfaces such as Si/SiO_2 [22]. Sum-frequency generation for crosscorrelation of NIR and UV pulses has been demonstrated on GaAs and Pd [12]. The THG on a glass surface was employed to record interferometric autocorrelation traces [23] and carry out pulse characterization by third-order FROG [24].

The details of the surface harmonic generation spectral response may depend on the material and surface, and therefore it is important to test the suitability of this interaction for SPIDER. The choice of gold is one of convenience, since its surface harmonic response covers our wavelength range of interest. Other metals such as silver would likely be suitable as well [18].

3. SPIDER

SPIDER is a technique for characterizing the field of ultrashort laser pulses, both the amplitude and phase. It is a self-referencing implementation of spectral shearing interferometry, a technique whereby two replica pulses, separated slightly in central frequency by Ω, the spectral shear, interfere in a spectrometer. As was originally pointed out by Zubov and Kuznetsova [25] and later by Wong and Walmsley [26], the resulting interferogram recorded by the spectrometer contains information about the spectral phase of the original pulse. In the original SPIDER [6], the incident pulse is split into three pulses, a short-pulse pair separated by τ and a highly stretched pulse. The requisite spectral shear is obtained by frequency mixing the two time-delayed short replica pulses with this highly chirped pulse in a nonlinear optical crystal. Since the fundamental short pulses overlap with a quasi-monochromatic temporal slice of the chirped pulse, the upconverted pulse maintains a true representation of the spectral phase of the original fundamental pulse. And since the two time-delayed fundamental replica pulses overlap with different frequency slices in the chirped pulse, the upconverted pulses have a spectral shear between them, governed by the pulse-pair temporal separation and the amount of chirp in the stretched pulse. The upconverted pulses then enter the spectrometer, where they generate a SPIDER interferogram, or Spidergram.

The Spidergram is then processed in a computer program that uncovers the original spectral phase. The process works as follows. The interferogram is Fourier transformed to the pseudo-time-domain, and three peaks are found, at $t=0$ and at $t=\pm\tau$. The peak at $t=+\tau$ is filtered from the rest and inverse Fourier transformed back to the frequency domain. The argument of this complex quantity is then retrieved and is integrated to recover the spectral phase. The process is direct and noniterative and requires knowledge of the spectral shear, obtained in a calibration step. The spectral phase, when combined with a spectral amplitude (the square root of the spectral intensity), yields the complete pulse information.

4. EXPERIMENT

The laser system used in our experiment is a home-built chirped pulse amplification (CPA) system. The oscillator consists of a Ti:sapphire double-prism folded cavity pumped by a frequency-doubled $Nd:YVO_4$ (Spectra-Physics Millennia). The CPA regenerative amplifier is pumped by a frequency-doubled Nd:YLF laser (Coherent Evolution). The system parameters are as follows: 200 μJ pulse energy at 2 kHz, 55 fs pulse duration at center wavelength 804 nm. The Gold-SPIDER apparatus is shown in Fig. 1. The laser pulse enters the setup and is immediately separated into three pulses. The reflection from an etalon of thickness 160 μm gives two pulses separated by $\tau=1.544$ ps. The stretched pulse is generated in a grating pair compressor arrangement that introduces a group-delay dispersion (GDD) of 1.8×10^5 fs^2. Note that the steps required to calibrate Gold-SPIDER are identical to conventional SPIDER. A pickoff mirror steers this stretched pulse to a turning mirror that sends this beam parallel to the pulse pair. The two beams reflect from a silver spherical mirror (20 cm radius of curvature) and focus onto a flat gold mirror, where they produce sum-frequency upconversion. The sum-frequency beam that carries the SPIDER signal bisects the two reflected fundamental beams is filtered by a pinhole and a blue pass filter (not shown) and imaged into the spectrometer (Ocean Optics USB2000).

Several technical points warrant clarification. The first is polarization. It was found by Bloembergen *et al.* [18] and later verified [20,21] that the surface harmonic generation requires the electric field to be polarized in the plane of incidence. Thus in our setup the incident beams are all *p* polarized. Furthermore, surface SHG is optimized at an incident angle of approximately 65 deg [18]; thus we used this incident angle in our setup. Alignment of the setup is fairly straightforward, since the upconverted beam is guaranteed by momentum conservation to bisect the reflected fundamental beams. Removing the blue filters and aligning both fundamental beams into the spectrometer via the focusing lens ensures that the upconverted beam will also focus into the spectrometer. Once an upconverted signal is detected with this technique, fine tweaks may be performed. Finally, it should be

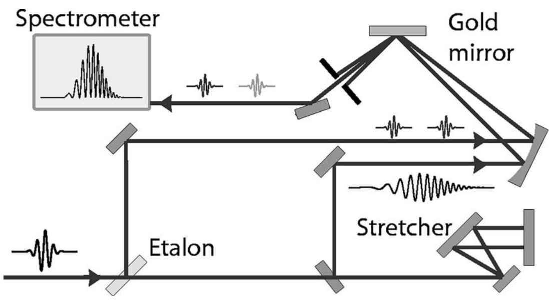

Fig. 1. Schematic of the experimental setup.

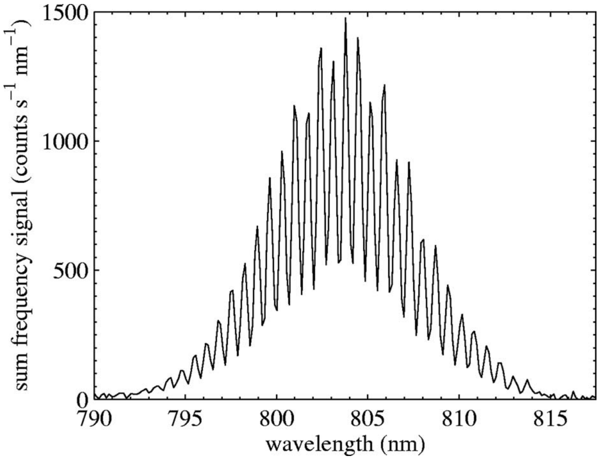

Fig. 2. Sample Spidergram for a near-transform-limited 55 fs pulse recorded with an integration time of 5 s.

noted that with our setup the harmonic signal is reasonably strong. In previous work with ultrafast oscillators, the harmonic signal was collected with a photomultiplier tube; yet with our amplified beam we were able to use the spectrometer's integrating CCD array. In fact, with good alignment we could see a Spidergram signal with a CCD integration time of only 300 ms. This is quite useful for real-time tweaking. For our reconstructions, however, we used longer integration times (3–20 s) to improve the signal-to-noise ratio.

5. RESULTS

A Gold-SPIDER interferogram is shown in Fig. 2. This was recorded with an integration time of 5 s. To record a transform-limited pulse we used a separate SHG crystal and optimized the CPA's compressor grating separation to give maximum SHG output. We then redirected this beam into our Gold-SPIDER apparatus and recovered the pulse profile shown in Fig. 3. This shows the spectrum (dashed curve) and spectral phase (solid curve). Characteristic of a broad upconversion bandwidth, the spectral phase is well defined into the wings of the pulse, where the spectral intensity drops to 1% of its peak. The temporal intensity profile is shown in the inset, which confirms that the pulse is close to the transform limit.

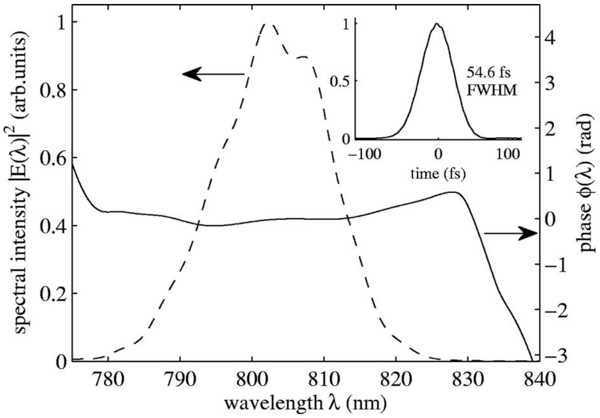

Fig. 3. Retrieved pulse at $\lambda_0 = 803$ nm, FWHM = 18.5 nm. The inset shows the recontructed temporal intensity distribution. The FWHM is 54.6 fs, which is 2% over the FTL of 53.6 fs.

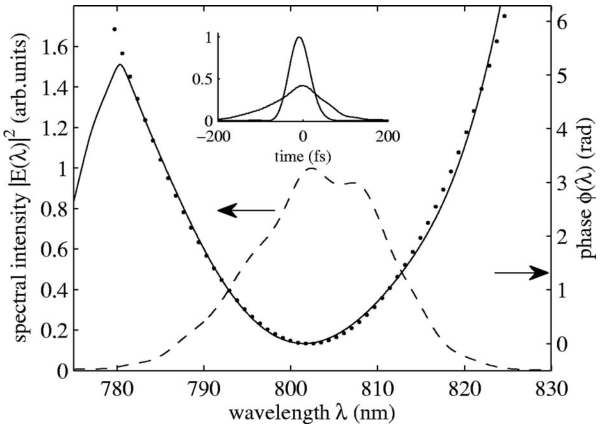

Fig. 4. Phase difference of two CPA compressor settings retrieved with Gold-SPIDER. The measured phase, i.e., the difference phase from two consecutive SPIDER measurements, (solid curve) agrees well with the calculated compressor phase (dots). The inset shows the near FTL and the slightly stretched pulse (125.5 fs) temporal profiles.

In order to quantify the accuracy of our Gold-SPIDER we introduced a known chirp into the pulse and compared it to a theoretical calculation. Maintaining the same alignment of the beam into the Gold-SPIDER apparatus, we introduced a GDD of 5500 fs^2 into our input pulse by slightly changing the grating separation in the CPA's compressor. This chirped pulse was measured and reconstructed to yield a highly quadratic spectral phase. We then subtracted the phase reconstructed for the near-transform-limited pulse. The results are shown in Fig. 4. The dashed curve in Fig. 4 is the spectrum of the pulse; the solid curve is the spectral phase difference between the measured chirped pulse and the near-transform-limited pulse of Fig. 3. In this way we can make a direct comparison to our theoretical calculation of the spectral phase induced by the CPA's compressor tuning. This is shown as dots in Fig. 4. The inset shows the temporal intensity profile of both the optimally compressed and chirped pulse with duration of 125.5 fs. Our results confirm that the nonlinear response of a gold surface is suitable for generating the shear necessary for SPIDER.

6. CONCLUSION

This proof-of-principle Gold-SPIDER has been presented as a variant of classic SPIDER utilizing surface harmonic generation from a gold surface and was found to reliably reconstruct ultrashort pulses from our Ti:sapphire laser. Since it contains no nonlinear crystals, there are several advantages to Gold-SPIDER over classic SPIDER: it is lower cost, since metallic mirrors are readily available; it is easy to align, since there are no phase-matching requirements; and it is extremely broadband, meaning that a single apparatus should be capable of measuring sub-10 fs pulses over many different wavelength ranges (UV to IR). In fact, there is a unique opportunity to measure near single-cycle octave-spanning pulses with this technique. The surface harmonic generation will easily cover this range, and SPIDER is ideally suited to reject the residual fundamental light, first, by using the angular separation of the beams to spatially filter the desired signal; and second, by taking advantage of SPIDER's frequency-shifted signal. Since the upconverted frequencies are produced by the frequency sum of the fundamental pulse to be measured and a quasi-cw component of an ancilla pulse (which may or may not originate from the fundamental pulse), there is freedom in choosing this quasi-cw component, hence shifting the resultant upconverted frequencies. Indeed, the upconverted signal may be tailored such that there is no spectral overlap with the original fundamental pulse.

Variants of classic SPIDER should also be possible. For instance, downconversion SPIDER was used to measure UV pulses [27]. Since a gold surface is capable of nonlinear interaction down to 120 nm, there is potential for extending this technique to ultrashort extreme UV pulses, possibly employing difference-frequency generation with the original femtosecond NIR pulse. Also, the spatially encoded arrangement for SPIDER (SEA-SPIDER) [28] could benefit from the broad upconversion bandwidth of metallic surfaces. In fact, many other SPIDER incarnations could use the surface nonlinearity of metals to generate the spectral shear necessary for complete pulse reconstruction.

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