Single-beam homodyne SPIDER for multiphoton microscopy

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We report a new version of spectral phase interferometry for direct electric field reconstruction (SPIDER) requiring only a single phase-shaped laser beam. A narrowband probe pulse is selected out of a broadband ultrafast laser pulse by a phase pulse-shaping technique and mixed with the original broadband pulse to generate a second-harmonic generation (SHG) signal. Using another SHG signal solely generated by the broadband pulse as a local oscillator, the spectral phase of the broadband laser pulse can be analytically retrieved by a combination of double-quadrature spectral interferometry and homodyne optical technique for SPIDER (HOT SPIDER). An arbitrary spectral phase at the sample position of a microscope can be compensated with a precision of 0.05 rad over the FWHM of the laser spectrum. It is readily applicable to a nonlinear microscopy technique with a phase-controlled broadband laser pulse. © 2008 Optical Society of America

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Recently, there has been growing interest in nonlinear optical microscopy techniques utilizing phase-controlled ultrafast laser pulses [1–4]. Numerous nonlinear microscopy concepts have emerged with unprecedented capabilities, such as scanningless vertical sectioning [1], selective excitation [2], and chemical selectivity [3,4]. To apply these concepts to optical microscopy, it is critical to characterize and compensate for the spectral phase distortion of an ultrafast laser pulse at the sample position of a microscope [2,4]. In high resolution optical microscopy, a high numerical aperture (NA) microscope objective is required and introduces not only the group delay dispersion [(GDD), the second-order dispersion] but also significant high-order dispersions [2,4]. In general, the entire optical dispersion, not only the GDD, of the laser pulse should be compensated if one wishes to exploit the full potential of a broadband laser pulse. For example, Xi *et al.* demonstrated that the maximal two photon fluorescence signal by a sub-10 fs laser can be obtained only when the entire optical dispersion is compensated [2].

The use of a high NA objective lens also requires the pulse characterization technique to have either a collinear or a single-beam configuration owing to the large converging angle of the focused laser beam. There exist a few such pulse characterization techniques: Multiphoton intrapulse interference phase scan (MIIPS) [5,6] and some versions of spectral phase interferometry for direct electric field reconstruction (SPIDER) techniques utilizing a pulse shaper [7–9]. Since all of these techniques already use a pulse shaper, an arbitrary spectral phase can be compensated *in situ* once the spectral phase is characterized. The MIIPS technique developed by the Dantus group uses spectral interference between intrapulses inside a phase-controlled broadband pulse to retrieve the spectral phase [5]. It is an iterative method and takes a rather long measurement time (approximately minutes). Very recently, a noniterative version of MIIPS was also demonstrated [6]. Monmayrant *et al.* demonstrated a time-domain version of homodyne optical technique for SPIDER (HOT SPIDER) with an acousto-optic pulse shaper (Dazzler) [7]. von Vacano *et al.* also presented a shaper-assisted collinear SPIDER [8]. Both of the above SPIDER techniques generate a time-delayed replica of the target pulse by a phase and amplitude pulse shaper and apply SPIDER methods [10] to retrieve the spectral phase of the target pulse. Recently, Chen and Lim reported a single-beam SPIDER method [9] that selects two narrowband probe pulses by a phase and polarization pulse shaper to generate the spectral shear interferogram with the remaining broadband pulse and retrieves the spectral phase of the original pulse. However, all of the above SPIDER techniques require controlling either amplitude/phase or polarization/phase of the laser pulse and are not directly applicable to the experiments with a phase-only shaped pulse.

In this Letter, we report a new version of the single-beam SPIDER technique that requires a phase-only pulse shaping. It shares the same concept with the previously reported frequency-domain HOT SPIDER [11], but all the necessary pulses are selected inside a single broadband laser pulse by a phase-only pulse shaper. In the previous HOT SPIDER [11], a second-harmonic generation (SHG) signal from the mixing of a broadband target pulse and a long probe pulse interferes with a separately generated local oscillator. The spectral phase of the target laser pulse is obtained by Fourier transform spectral interferometry between the SHG signals and the local oscillator. In the current single-beam homodyne SPIDER method, we used a double quadrature spectral interferometry [12] with a local oscillator generated by the same pulse and retrieved the spectral phase via the HOT SPIDER algorithm [11].

The phase of a broadband laser pulse from a Ti:sapphire oscillator (80 MHz repetition rate, KM

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Labs) is controlled by an all reflective 4*f* pulse shaper with a 640 pixel liquid crystal spatial light modulator (SLM) [9]. The spectral resolution of our pulse shaper is 0.4 nm, limited by the size of a single SLM pixel at the Fourier plane. Figure 1(a) shows the spectrum of our laser pulse and a phase mask used in this experiment. The phase-shaped laser pulse was focused to a 10 μm thick type I β-barium borate crystal with a 1.2 NA microscope objective (Olympus). The SHG signals were collected with a 0.4 NA objective (Olympus), filtered by a BG 28 filter, and measured with a minispectrometer (USB4000, Ocean Optics). A pulse energy of 0.4 nJ, measured before the focusing objective, was used in this Letter. With the applied phase of ϕ_{pr} at the probe frequency of ω_{pr}, the original pulse can be considered as two separated pulses: A narrowband probe pulse at ω_{pr} with a phase of ϕ_{pr} [$E_{\mathrm{pr}}(\omega)$ in Fig. 1(b)] and a broadband pulse with a hole at ω_{pr} [$E_h(\omega)$ in Fig. 1(b)], i.e.,

$$E^0(\omega) = E_h(\omega) + E_{\mathrm{pr}}(\omega),$$

where we define $E_h(\omega) \equiv E^0(\omega) - E^0(\omega_{\mathrm{pr}})\delta(\omega-\omega_{\mathrm{pr}})$ and $E_{\mathrm{pr}}(\omega) \equiv E^0(\omega_{\mathrm{pr}})\delta(\omega-\omega_{\mathrm{pr}})$. $\delta(\omega)$ is the delta function, and we assume the probe pulse has a very narrow bandwidth. Note that $E_h(\omega)$ does not depend on ϕ_{pr}. Then the total SHG electric field, $E_{\mathrm{SHG}}(\omega)$ becomes

$$\begin{aligned}
E_{\mathrm{SHG}}(\omega) &\propto \int \mathrm{d}\Omega E^0(\omega-\Omega)E^0(\Omega) \\
&= \int \mathrm{d}\Omega[E_h(\omega-\Omega) + E_{\mathrm{pr}}(\omega-\Omega)] \\
&\quad \times [E_h(\Omega) + E_{\mathrm{pr}}(\Omega)] \\
&= \int \mathrm{d}\Omega[E_h(\omega-\Omega) + E^0(\omega_{\mathrm{pr}})\delta(\omega-\Omega-\omega_{\mathrm{pr}})] \\
&\quad \times [E_h(\Omega) + E^0(\omega_{\mathrm{pr}})\delta(\Omega-\omega_{\mathrm{pr}})] \\
&= \int E_h(\omega-\Omega)E_h(\Omega)\mathrm{d}\Omega + 2E^0(\omega_{\mathrm{pr}})E_h(\omega-\omega_{\mathrm{pr}}) \\
&\quad + (E^0(\omega_{\mathrm{pr}}))^2\delta(\omega-2\omega_{\mathrm{pr}}) \equiv E^{\mathrm{LO}}_{\mathrm{SHG}}(\omega) \\
&\quad + E^{(1)}_{\mathrm{SHG}}(\omega) + (E^0(\omega_{\mathrm{pr}}))^2\delta(\omega-2\omega_{\mathrm{pr}}),
\end{aligned}$$

where we define $E^{\mathrm{LO}}_{\mathrm{SHG}}(\omega) \equiv \int \mathrm{d}\Omega E_h(\omega-\Omega)E_h(\Omega)$ and $E^{(1)}_{\mathrm{SHG}}(\omega) \equiv 2E^0(\omega_{\mathrm{pr}})E_h(\omega-\omega_{\mathrm{pr}})$. Note that $E^{(1)}_{\mathrm{SHG}}(\omega)$ has the same spectral phase as $E_h(\omega)$ with a controllable phase offset (ϕ_{pr}) owing to the narrow bandwidth of the probe pulse. Figure 1(b) shows the first two components in the above equation. $E^{(1)}_{\mathrm{SHG}}(\omega)$ in Fig. 1(b) is the SHG signal from the mixing of the narrowband probe pulse ($E_{\mathrm{pr}}(\omega)$) and the broadband laser pulses with a hole ($E_h(\omega)$). $E^{\mathrm{LO}}_{\mathrm{SHG}}(\omega)$ in Fig. 1(b) is the SHG from the mixing of two $E_h(\omega)$'s and is independent of ϕ_{pr}. Since the third term ($(E^0(\omega_{\mathrm{pr}}))^2\delta(\omega-2\omega_{\mathrm{pr}})$) affects only the measured SHG spectrum at $\omega=2\omega_{\mathrm{pr}}$, we simply skip this single SHG frequency ($2\omega_{\mathrm{pr}}$) in the following phase retrieval process.

The detected SHG signal $S^{(1)}(\omega)$ at $\omega \neq 2\omega_{\mathrm{pr}}$ becomes

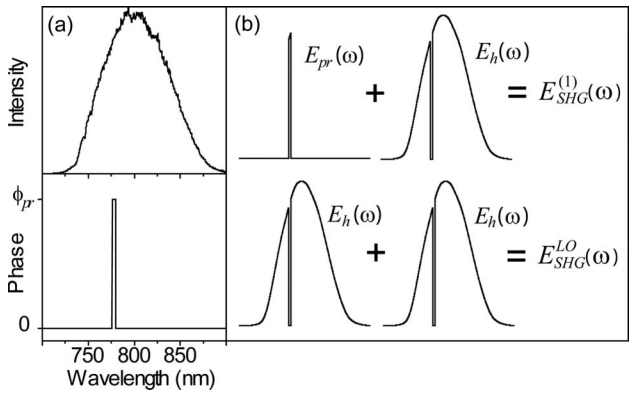

Fig. 1. (a) (Top) spectrum of the laser and (bottom) phase mask used in the experiment. (b) Two SHG components from the laser pulse with the phase mask in (a).

$$\begin{aligned}
S^{(1)}(\omega) &= |E^{\mathrm{LO}}_{\mathrm{SHG}}(\omega) + E^{(1)}_{\mathrm{SHG}}(\omega)|^2 \\
&= |E^{\mathrm{LO}}_{\mathrm{SHG}}(\omega)|^2 + |E^{(1)}_{\mathrm{SHG}}(\omega)|^2 + 2|E^{\mathrm{LO}}_{\mathrm{SHG}}(\omega) \\
&\quad \times E^{(1)}_{\mathrm{SHG}}(\omega)|\cos(\phi^{\mathrm{LO}}(\omega) - \phi_{\mathrm{pr}} - \phi(\omega-\omega_{\mathrm{pr}})),
\end{aligned}$$

where $\phi(\omega)$ and $\phi^{\mathrm{LO}}(\omega)$ are the spectral phases of $E_h(\omega)$ and $E^{\mathrm{LO}}_{\mathrm{SHG}}(\omega)$, respectively. ϕ_{pr} is the phase we apply to select the probe pulse. We can extract $\phi^{\mathrm{LO}}(\omega) - \phi(\omega-\omega_{\mathrm{pr}})$ from quadrature measurements [9,12] with four different probe phases ϕ_{pr} by

$$\begin{aligned}
&\phi^{\mathrm{LO}}(\omega) - \phi(\omega-\omega_{\mathrm{pr}}) \\
&\quad = \tan^{-1}\left[\frac{S^{(1)}(\omega,\phi_{\mathrm{pr}}=\pi/2) - S^{(1)}(\omega,\phi_{\mathrm{pr}}=-\pi/2)}{S^{(1)}(\omega,\phi_{\mathrm{pr}}=0) - S^{(1)}(\omega,\phi_{\mathrm{pr}}=\pi)}\right],
\end{aligned} \tag{1}$$

where $S^{(1)}(\omega,\phi_{\mathrm{pr}})$ is the measured SHG signal with a probe phase of ϕ_{pr} [13]. If we choose the probe at a slightly shifted frequency region (at $\omega_{\mathrm{pr}}+\delta\omega$ instead of ω_{pr}), the resulting spectral phase becomes

$$\begin{aligned}
&\phi^{\mathrm{LO}}(\omega) - \phi(\omega-\omega_{\mathrm{pr}}-\delta\omega) \\
&\quad = \tan^{-1}\left[\frac{S^{(2)}(\omega,\phi_{\mathrm{pr}}=\pi/2) - S^{(2)}(\omega,\phi_{\mathrm{pr}}=-\pi/2)}{S^{(2)}(\omega,\phi_{\mathrm{pr}}=0) - S^{(2)}(\omega,\phi_{\mathrm{pr}}=\pi)}\right],
\end{aligned} \tag{2}$$

where $S^{(2)}(\omega,\phi_{\mathrm{pr}})$ is the SHG signal with an applied phase mask of ϕ_{pr} at $\omega_{\mathrm{pr}}+\delta\omega$. Note that $\phi^{\mathrm{LO}}(\omega)$ in Eqs. (1) and (2) are almost identical, since the probe bandwidth is much narrower than the original bandwidth of the broadband laser pulse. By subtracting Eq. (1) from Eq. (2), we obtain

$$\begin{aligned}
&\phi(\omega-\omega_{\mathrm{pr}}) - \phi(\omega-\omega_{\mathrm{pr}}-\delta\omega) \\
&\quad = \tan^{-1}\left[\frac{S^{(2)}(\omega,\phi_{\mathrm{pr}}=\pi/2) - S^{(2)}(\omega,\phi_{\mathrm{pr}}=-\pi/2)}{S^{(2)}(\omega,\phi_{\mathrm{pr}}=0) - S^{(2)}(\omega,\phi_{\mathrm{pr}}=\pi)}\right] \\
&\qquad - \tan^{-1}\left[\frac{S^{(1)}(\omega,\phi_{\mathrm{pr}}=\pi/2) - S^{(1)}(\omega,\phi_{\mathrm{pr}}=-\pi/2)}{S^{(1)}(\omega,\phi_{\mathrm{pr}}=0) - S^{(1)}(\omega,\phi_{\mathrm{pr}}=\pi)}\right] \\
&\quad \equiv \theta(\omega-\omega_{\mathrm{pr}}).
\end{aligned} \tag{3}$$

From the measured $\theta(\omega)$ one can obtain the spectral phase of the original laser pulse, $\phi(\omega)$, by

$$\phi(\omega_0 + n\,\delta\omega) = \phi(\omega_0) + \sum_{k=1}^{n} \theta(\omega_0 + k\,\delta\omega), \qquad (4)$$

where n is positive integer. Note that $\theta(\omega)$ is equivalent to the spectral shear phase in the conventional SPIDER technique [10].

Figure 2(a) shows the four SHG spectra with $\phi_{\mathrm{pr}} = 0$, $\pi/2$, π, and $-\pi/2$. These traces were taken with a 10 ms exposure time and averaged 20 times. The probe pulse has a bandwidth of 1.6 nm (equivalent to four SLM pixel widths) at 778 nm. One can see that the spectral interference fringes change with ϕ_{pr} in Fig. 2(a). After measuring another set of SHG signals with the probe at 783 nm, we retrieve the spectral phase of the original laser pulse ($\phi(\omega)$) with Eqs. (3) and (4) [thin curve in Fig. 2(b)]. The spectral resolution of our SPIDER method is limited by the frequency difference between the two probe pulses (5 nm in the current demonstration). Note that we partially compressed the laser pulse by moving the grating in the pulse shaper, and it caused the unusual spectral phase in Fig. 2(b) [4]. After applying the compensating phase mask, the spectral phase becomes flat within 0.5 rad, which verifies the validity of our method. We also verify the result with the previously demonstrated single-beam SPIDER and MIIPS methods (data not shown). Although no iteration is required in principle, a few iterations help to compress the pulse further, and we speculate that it is probably due to the imperfect SLM calibration. The thick curve in Fig. 2(b) shows the measured spectral phase after three iterations. The standard deviation of the retrieved phase over the FWHM of the laser spectrum is less than ±0.05 rad, which is the current error limit of our technique. We took 20 spectral phase measurements to calculate the error bars in Fig. 2(b). Possible error sources can be imperfect SLM calibration, laser fluctuation during the measurements, and a transverse field effect of the SLM. The finite bandwidth of the probe also makes the local oscillators in the $S^{(1)}$ and $S^{(2)}$ measurements slightly different and may cause an additional error. The current method retrieves the spectral phase from relatively small variations in the SHG spectral intensity. However, one can increase the signal-to-noise ratio of the method by taking averages of multiple SHG spectra as long as the laser is stable. Here we demonstrate the pulse compression within ±0.05 rad over the FWHM of the laser spectrum under the experimental condition of a 1.6 nm probe bandwidth, 10 ms exposure time, and 20 spectral averages.

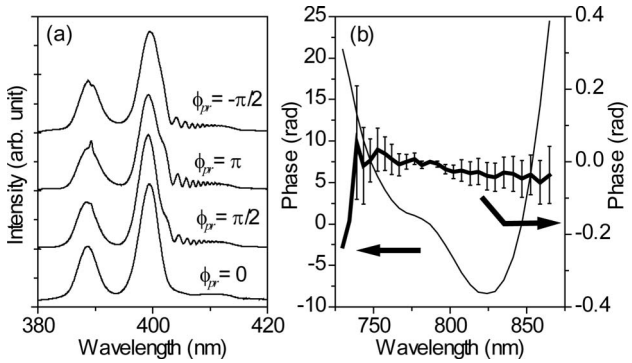

Fig. 2. (a) Measured SHG spectra with the probe phase (ϕ_{pr}) of 0, $\pi/2$, π, and $-\pi/2$. The probe wavelength is 778 nm. Each trace is vertically displaced for clarity. (b) Spectral phase of the laser pulse at the microscope sample position before (thin curve) and after (thick curve with error bars) the pulse compression. Note the difference in the vertical scales.

In summary, we demonstrated a new version of homodyne SPIDER requiring only a single phase-controlled laser beam. It can characterize and compress an arbitrary spectral phase at the sample position of a microscope with great precision. It has a very simple experimental setup, is relatively fast (it takes only eight SHG measurements, ~4 s in the current experiment), and is easily applicable to a nonlinear microscopy technique based on a phase-controlled pulse.

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References and Notes

1. D. Oron, E. Tal, and Y. Silberberg, Opt. Express **13**, 1468 (2005).
2. P. Xi, Y. Andegeko, L. R. Weisel, V. V. Lozovoy, and M. Dantus, Opt. Commun. **281**, 1841 (2008).
3. N. Dudovich, D. Oron, and Y. Silberberg, Nature **418**, 512 (2002).
4. B.-C. Chen and S.-H. Lim, J. Phys. Chem. B **112**, 3653 (2008).
5. B. W. Xu, J. M. Gunn, J. M. Dela Cruz, V. V. Lozovoy, and M. Dantus, J. Opt. Soc. Am. B **23**, 750 (2006).
6. V. V. Lozovoy, B. Xu, Y. Coello, and M. Dantus, Opt. Express **16**, 592597 (2008).
7. A. Monmayrant, M. Joffre, T. Oksenhendler, R. Herzog, D. Kaplan, and P. Tournois, Opt. Lett. **28**, 278 (2003).
8. B. von Vacano, T. Buckup, and M. Motzkus, Opt. Lett. **31**, 1154 (2006).
9. B.-C. Chen and S.-H. Lim, Opt. Lett. **32**, 2411 (2007).
10. C. Iaconis and I. A. Walmsley, Opt. Lett. **23**, 792 (1998).
11. C. Dorrer, P. Londero, and I. A. Walmsley, Opt. Lett. **26**, 1510 (2001).
12. L. Lepetit, G. Chériaux, and M. Joffre, J. Opt. Soc. Am. B **12**, 2467 (1995).
13. Here we use arctan for clarity of the discussion. In the experiment, we transformed the numerator and denominator inside the arctan term in Eqs. (2) and (3) into the polar coordinate to retrieve the phase.