Application of single-beam homodyne
SPIDER for the control of
complex spectral phase profiles

Peter Schöen and Sophie Brasselet*
Institut Fresnel, MOSAIC, CNRS, Aix-Marseille Université, École Centrale Marseille,
Domaine Universitaire St Jérôme, 13397 Marseille, France
*Corresponding author: sophie.brasselet@fresnel.fr

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We investigate the range of application of single-beam homodyne spectral phase interferometry for direct electric field reconstruction (SPIDER) for multiphoton microscopy. Simulations and experimental studies performed on model spectral phase profiles show that the phase reconstruction technique used in this method makes the phase retrieval quality highly sensitive to the complexity of the profile. In addition, we show that the use of iterative processes is likely to deteriorate the phase retrieval quality, especially for strongly varying phase profiles. These effects are illustrated and quantified for sinusoidal and quadratic spectral phase profiles. © 2011 Optical Society of America

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Nonlinear microscopy has considerably progressed recently, thanks to the increased control of the time and spatial profiles quality of ultrashort pulses at the focal spot of high NA objectives. A reliable tailoring of the phase and polarization spectral profiles of ultrashort pulses in condensed molecular media can lead to enhanced selection of multiphoton fluorescent signals [1,2] and bring new opportunities in the readout of nonlinear coherent anti-Stokes Raman [3], second-harmonic generation (SHG) [4,5], and third-harmonic generation [6,7] signals. A prerequisite for the application of these methods is the knowledge of the spectral phase of the optical pulse at the focal spot of the microscope objective, which needs primarily to be corrected for instrumental phase distortions.

Among the methods developed to access the spectral phase profiles of broadband laser pulses, frequency-resolved optical gating (FROG) [8] and SPIDER [9] have emerged as standard techniques, including possible variations to adapt to the collinear geometry imposed by microscopy imaging [10,11]. However, both require the implementation of interferometric setups, while FROG demands, in addition, large acquisition times. Multiphoton intrapulse interference phase scan [12] has been proposed to avoid these issues. Recently a single-beam SPIDER method has been shown to be an interesting alternative for fast analysis, in which two narrow polarization jumps close to one another (of which one is additionally phase-shifted with respect to the other) are shaped into a broadband pulse [13]. From the intrapulse interferences produced by this configuration the spectral phase can be obtained. A subsequent development, inspired by the homodyne optical technique for SPIDER (HOT-SPIDER) [14], has been introduced based on single pulse phase shaping, where only a narrow phase jump is inserted into the pulse without shaping its amplitude or its polarization [15]. In this scheme, a series of measurements is performed where a phase jump of magnitude \( \phi_{\text{pr}} \) is located either at the frequency \( \omega_{\text{pr}} \) or at a spectral position close by \( (\omega_{\text{pr}} + \delta \omega) \), the generated SHG signals being recorded on a spectrometer. They can be combined via

\[
\tan^{-1} \left( \frac{S^{(2)}(\omega, \phi_{\text{pr}} = \pi/2) - S^{(2)}(\omega, \phi_{\text{pr}} = -\pi/2)}{S^{(2)}(\omega, \phi_{\text{pr}} = 0) - S^{(2)}(\omega, \phi_{\text{pr}} = \pi)} \right) - \tan^{-1} \left( \frac{S^{(1)}(\omega, \phi_{\text{pr}} = \pi/2) - S^{(1)}(\omega, \phi_{\text{pr}} = -\pi/2)}{S^{(1)}(\omega, \phi_{\text{pr}} = 0) - S^{(1)}(\omega, \phi_{\text{pr}} = \pi)} \right) \equiv \theta(\omega - \omega_{\text{pr}})
\]

(1)

to calculate the construction phase \( \theta(\omega - \omega_{\text{pr}}) \), where \( S^{(1)}(\omega, \phi_{\text{pr}}) \) and \( S^{(2)}(\omega, \phi_{\text{pr}}) \) are the spectrally resolved SHG intensities produced by a pulse with a phase step of \( \phi_{\text{pr}} \) at respective frequencies \( \omega_{\text{pr}} \) and \( \omega_{\text{pr}} + \delta \omega \). Based on this operation the original laser phase \( \phi(\omega) \) is retrieved by

\[
\phi(\omega_0 + n\delta \omega) = \phi(\omega_0) + \sum_{k=1}^{n} \theta(\omega_0 + k\delta \omega),
\]

(2)

where \( \omega_0 \) is the lowest accessible frequency in the spectral range of the pulse, and \( \delta \omega \) defines the spectral resolution. This treatment has been shown to lead to similar results as for other SPIDER variants [15]. Sung et al. have shown that the method is successful in correcting for phase distortions present in a microscopy setup using an iterative process, although this is not required in principle by the technique [15]. In this work, we develop simulations and experiments to explore the range of application of this method for the retrieval of complex spectral phase profiles, and show the effect of iterative optimization on the reliability of the achieved phase reconstruction.

To quantify the phase reconstruction in single-beam homodyne SPIDER simulations, we define the phase retrieval quality for a number of different target phase profiles \( \phi_0(\omega) \), using a least-squares analysis between \( \phi_0(\omega) \) and the calculated retrieved phase \( \phi_r(\omega) \) as

\[
\sigma = \int_{\Omega} (\phi_0(\omega) - \phi_r(\omega))^2 d\omega,
\]

(3)

where \( \Omega \) is the spectral support of the laser pulse. We mirror experimental conditions by choosing the spectral...
position of the phase steps at 778 nm and 782 nm with a width of 1.2 nm each. The phase reconstruction uses Eqs. (1) and (2), including a systematic compensation for the superimposed linear phase dependency that can occur in this calculation. Figure 1 depicts simulation results for several phase profiles containing quadratic spectral dependencies (as often encountered in nonlinear microscopy), as well as sinusoidal profiles, which are widely used in coherent control for nonlinear microscopy [1,5] and represent high-order phase distortions. \( \sigma \) values below 100 can be considered to correspond to a good estimate of the spectral phase, while values far above that threshold indicate strong deviations between \( \phi_0 \) and \( \phi_f \). It can be seen that quadratic phases are reliably measured correctly, even for large quadratic phase coefficients [Fig. 1(a)]. For sinusoidal phases on the other hand, the retrieval quality is strongly dependent on the sinus amplitude. Once a value of about 0.35\( \pi \) is surpassed the method fails, while it has no difficulties for lower amplitudes [Fig. 1(b)]. However, the dependence on the period of the sinus is less striking [Fig. 1(b) inset]. Apart from a few isolated cases where the retrieval is not successful, the method is correct as long as the period is larger than the resolution limit \( \delta \omega \). Interestingly, for a combination of quadratic and sinus phases the tolerance of the modulation amplitude is much larger than in the pure sinus case [Fig. 1(c)].

Single-beam homodyne SPIDER was implemented experimentally using a 640 LC array dual spatial light modulator (SLM) in a \( 4f \) mirror-based setup as described in [5], using the SHG signal reflected from a KTP (KT\( \text{OPO}_4 \)) thick crystal interface. A target spectral phase profile is encoded in the SLM and measured by single-beam homodyne SPIDER using an interactive process to progressively correct for various phase distortions from the whole beam path. In each iteration cycle, the measured phase is subtracted from the SLM encoding, and the target phase is added again. In the end the target phase profile should be established while all distortions have been eliminated. Figure 2 shows the results for several sinusoidal phase profiles with different amplitudes \( (x_1) \) and periods \( (x_2) \), obtained after four iterations. In accordance with Fig. 1(b), a good convergence is observed between \( \phi_0 \) and \( \phi_f \) for small sine amplitudes of 0.3\( \pi \) over almost the whole laser spectrum. When the sine amplitudes grow, the phase retrieval quality clearly decreases.

Contrary to what would be intuitively thought, increasing the number of iterations does not improve the reliability of the reconstruction for complex phase profiles. Simulations shown in Fig. 3 show that this issue is moreover not purely experimental but rather intrinsically related to the method. Figures 3(a) and 3(b) depict respectively the measured and simulated phase after each iteration for a sinusoidal phase profile in the critical region where the method begins to fail. The simulation includes a Gaussian noise in the SHG spectra of similar magnitude as in experiments (a Poisson noise statistics leads to very similar conclusions). In the measurement, the phase is almost established after each of the first two

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**Fig. 1.** Phase retrieval simulations. \( x \) denotes the variable parameter in each series. (a) quadratic phase \( \phi_0 = x \omega (\omega - \omega_0)^2 \); (b) sinus with varying amplitude \( \phi_0 = x \sin(\pi \omega \omega) \); inset, sinus with varying period \( \phi_0 = \frac{x}{2} \sin(\pi \omega \omega) \); (c) quadratic phase and sinus with varying amplitude \( \phi_0 = 300 fs^4 (\omega - \omega_0)^2 + x \sin(\pi \omega \omega) \); inset, zoom of the graph. Left column, phase profile with a phase step of \( \sigma \) at \( \omega_0 + \delta \omega \), and laser spectrum (dashed curve). Right column, \( \sigma \) dependence on \( x \) for the corresponding series.

**Fig. 2.** Experimentally measured phases using single-beam homodyne SPIDER. Target phase profiles (solid curve), retrieved phases after the fourth iteration (dashed curve), laser spectrum (dotted curve). Sinusoidal phase profiles \( \phi_0 = x_1 \pi \sin(\omega \omega_0 (\omega - \omega_0)) \) (where \( \Delta \omega \) corresponds to an interval of \( \Delta \lambda = 50 nm \) around \( \lambda_0 = \frac{14}{3} \omega \) = 800 nm) are implemented for several amplitudes \( (x_1) \) and periods \( \Delta \lambda (x_2) \). (a) \( (x_1, x_2) = (0.3, 1.5) \); (b) \( (x_1, x_2) = (0.3, 2) \); (c) \( (x_1, x_2) = (0.3, 3) \); (d) \( (x_1, x_2) = (0.45, 1.5) \); (e) \( (x_1, x_2) = (0.5, 2) \); (f) \( (x_1, x_2) = (0.6, 1.5) \).
iterations, but deteriorates after the third one. A similar situation arises in the simulation.

Obviously increasing the number of iterations is not a solution to a better phase profile retrieval. It can even have a detrimental effect. This is illustrated in Figs. 3(c) and 3(d) for a sinusoidal phase where the method works correctly at the first iteration. Strong deviations from the target phase can be observed at the extrema of the sinusoidal profile after the fourth iteration. These deviations can be explained by the discretization of the operations performed in the method, imposed by the limited spectral resolution $\Delta \omega$. Because the retrieved phase cannot adapt to quickly varying phase slopes, such as at the extrema of a sinusoidal profile, the difference between $\phi_i$ and $\phi_0$ is then carried into the next iteration and increases successively. Therefore, except for cases where the phase changes are of low enough amplitude, the number of iterations should be kept small whatever the phase profile shape. Similar results are obtained for third-order phase distortions, which are frequently encountered in nonlinear microscopy and coherent control [2]. At last, the ratio performed in Eq. (1) makes this technique also sensitive to noise. Even in a low noise situation the quality of the phase retrieval can be affected, while without such noise the phase profile is measured correctly [Fig. 1(b)].

In summary, we have demonstrated that while single-beam homodyne SPIDER can be used to correct for phase distortions in many cases, special care has to be undertaken regarding the spectral shape of target phase profiles. It can in particular be applied to the measurement of quadratic phases spectral dependencies; however, it encounters difficulties when confronted with large phase changes at smaller frequency scales (here represented by a sinusoidal profile).

References