Direct measurement of spectral phase for ultrashort laser pulses

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Abstract: We present an intuitive pulse characterization method that provides an accurate and direct measurement of the spectral phase of ultrashort laser pulses. The method requires the successive imposition of a set of quadratic spectral phase functions on the pulses while recording the corresponding nonlinear spectra. The second-derivative of the unknown spectral phase can be directly visualized and extracted from the experimental 2D contour plot, without any inversion algorithm or mathematical manipulation.

OCIS codes: (320.0320) Ultrafast optics; (320.7100) Ultrafast measurements

References and links

1. K. Naganuma, K. Mogi, and H. Yamada, "General-Method for Ultrashort Light-Pulse Chirp Measurement," IEEE J. Quantum Elect. **25,** 1225-1233 (1989).
2. R. Trebino, and D. J. Kane, "Using Phase Retrieval to Measure the Intensity and Phase of Ultrashort Pulses - Frequency-Resolved Optical Gating," J. Opt. Soc. Am. A **10,** 1101-1111 (1993).
3. C. Iaconis, and I. A. Walmsley, "Spectral phase interferometry for direct electric-field reconstruction of ultrashort optical pulses," Opt. Lett. **23,** 792-794 (1998).
4. D. Yelin, D. Meshulach, and Y. Silberberg, "Adaptive femtosecond pulse compression," Opt. Lett. **22,** 1793-1795 (1997).
5. T. Baumert, T. Brixner, V. Seyfried, M. Strehle, and G. Gerber, "Femtosecond pulse shaping by an evolutionary algorithm with feedback," Appl. Phys. B-Lasers O. **65,** 779-782 (1997).
6. K. A. Walowicz, I. Pastirk, V. V. Lozovoy, and M. Dantus, "Multiphoton intrapulse interference. 1. Control of multiphoton processes in condensed phases," J. Phys. Chem. A **106,** 9369-9373 (2002).
7. V. V. Lozovoy, I. Pastirk, K. A. Walowicz, and M. Dantus, "Multiphoton intrapulse interference. II. Control of two- and three-photon laser induced fluorescence with shaped pulses," J. Chem. Phys. **118,** 3187-3196 (2003).
8. V. V. Lozovoy, I. Pastirk, and M. Dantus, "Multiphoton intrapulse interference. IV. Ultrashort laser pulse spectral phase characterization and compensation," Opt. Lett. **29,** 775-777 (2004).
9. B. W. Xu, J. M. Gunn, J. M. Dela Cruz, V. V. Lozovoy, and M. Dantus, "Quantitative investigation of the multiphoton intrapulse interference phase scan method for simultaneous phase measurement and compensation of femtosecond laser pulses," J. Opt. Soc. Am. B **23,** 750-759 (2006).
10. The MIIPS technology is protected by U.S. patent No. 7,105,811; and other patents pending.
11. B. W. Xu, Y. Coello, V. V. Lozovoy, D. A. Harris, and M. Dantus, "Pulse shaping of octave spanning femtosecond laser pulses," Opt. Express **14,** 10939-10944 (2006).
12. Y. Coello, B. W. Xu, T. L. Miller, V. V. Lozovoy, and M. Dantus, "Group-velocity dispersion measurements of water, seawater, and ocular components using multiphoton intrapulse interference phase scan (MIIPS)," Appl. Opt. 46, 8394 (2007).

1. Introduction

Pulse characterization and compression are of critical importance in ultrashort laser science and technology. Among the available spectral phase characterization techniques, the early development of Naganuma *et al*. [1], and the development of FROG [2] and SPIDER [3] represent milestones in the field. There are also optimization algorithm approaches for pulse

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compression that do not require phase measurement [4], and that are able to characterize the phase after pulse compression, provided a calibrated pulse shaper is used [5]. Ideally, a spectral phase measurement should be simple, direct and insensitive to noise. Here, we demonstrate such a method.

It has long been recognized that nonlinear optical (NLO) processes are sensitive to the second derivative of the phase. In keeping with the above requirements, we consider the direct measurement of the second derivative of an unknown phase $\phi''(\omega)$. We start by envisioning $\phi''(\omega)$ plotted as a function of frequency as shown by the red line in Fig. 1a. The goal can be restated as mapping the unknown function in the two dimensional space. This can be simply achieved by introducing a grid of reference functions $f''(\omega)$, and finding for what values of ω they intersect the unknown $\phi''(\omega)$. For each intersection, identified by a green circle in Fig. 1a, the equation $\phi''(\omega)=f''(\omega)$ is satisfied. Given that each of the functions $f''(\omega)$ are known, the function $\phi''(\omega)$ can be obtained directly.

Practically speaking, the simplest reference functions are horizontal lines, which correspond to different amounts of linear chirp. When the introduced chirp locally compensates the unknown distortion, the equation

$$\phi''(\omega)-f''(\omega)=0 \tag{1}$$

is satisfied. It is at that frequency that any NLO process, for example second harmonic generation (SHG), reaches its maximum possible intensity (Fig. 1b). This condition allows us to identify the position of every intersection. Experimentally, a NLO spectrum, for example second-harmonic generation (SHG), is acquired for each horizontal line on the grid (Fig. 1b). The spectra are used to construct a two-dimensional contour map, shown in Fig. 1c, in which the height corresponds to the SHG intensity. When a line is drawn through the maxima in the contour map, the unknown $\phi''(\omega)$ is directly obtained.

The procedure described in Fig. 1 is, to our knowledge, the simplest and most direct method for measuring the phase of an ultrashort pulse, and can be used to measure the chromatic dispersion introduced by passive optics, adaptive pulse shapers or by nonlinear optical effects. The method is based on the fundamental concept of multiphoton intrapulse interference [6, 7], which explains why NLO processes are maximized when Eq. 1 is satisfied. Our group has been using the multiphoton intrapulse interference phase scan (MIIPS) method, which typically uses a sinusoidal function for $f(\omega)$ because it offers some advantages [8-10]. Here we highlight the choice of a reference quadratic phase function to obtain $\phi''(\omega)$ directly. We include measurements from a sub-5fs laser system obtained performing a single chirp scan.

2. Experimental

We used an ultrabroad-bandwidth femtosecond Ti:Al_2O_3 laser oscillator with a double chirped mirror pair, whose spectrum spans 620-1050 nm (Fig. 3) and that has been used to generate a SHG spectrum spanning almost 200 nm [11]. The pulse shaper used to introduce the spectral phase $-f(\omega)$ was a folded all-reflective grating-based system containing a 150-lines-per-mm grating, a 762-mm–focal-length spherical mirror, and a 640-pixel dual-mask spatial light modulator (SLM-640, CRi Inc.). After the shaper, the pulses were focused onto a 20-μm type-I KDP crystal, and the SHG signal was separated from the fundamental before it was directed to a spectrometer (QE65000, Ocean Optics Inc.). The setup has been extensively described in an earlier publication [11].

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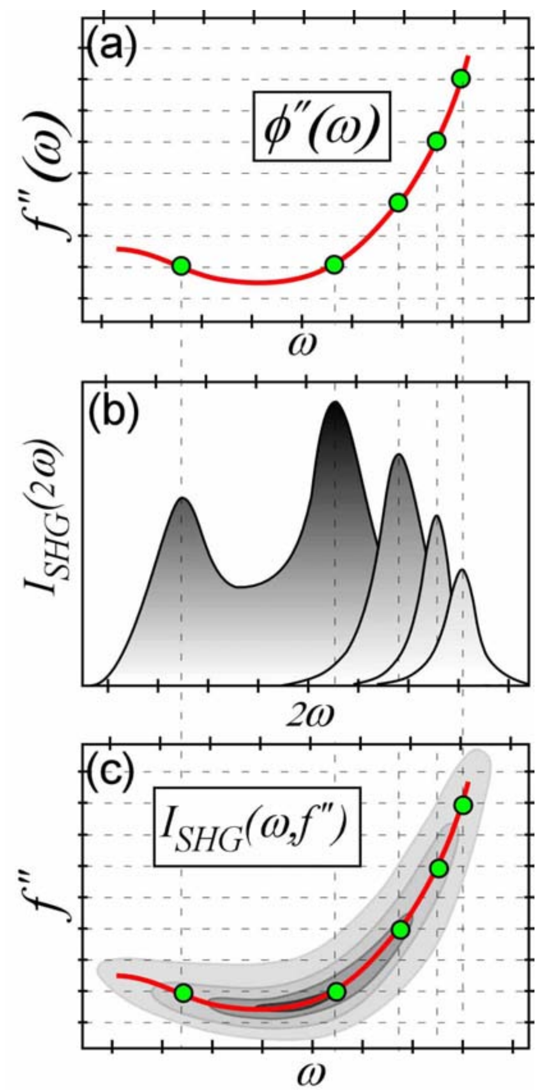

Fig. 1. Principle of the method. (a) The unknown $\phi''(\omega)$ function is probed using a set of reference linear chirps represented by the horizontal grid. (b) The maximum SHG intensity for every frequency indicates that the corresponding reference chirp value compensates the unknown function at the position of the maximum. (c) A two-dimensional contour plot mapping the intensity of the SHG as a function of chirp and frequency directly reveals the unknown $\phi''(\omega)$.

3. Results

Transform-limited pulses were obtained by measuring and compensating the spectral phase of the system using the sinusoidal MIIPS method [8, 9]. A cubic spectral phase function defined as $\phi(\omega)$=500fs^3 $(\omega$-$\omega_0)^3$, which corresponds to a linear $\phi''(\omega)$, was then introduced to the pulses and measured with the method described in this paper. The experimental trace is shown in Fig. 2. The red dashed line indicates the spectral maxima, which directly correspond to the measured $\phi''(\omega)$. Note that both the accuracy and precision of the measurement are ~1-2 fs^2, and results were obtained from a single chirp scan with grid-step of 5 fs^2.

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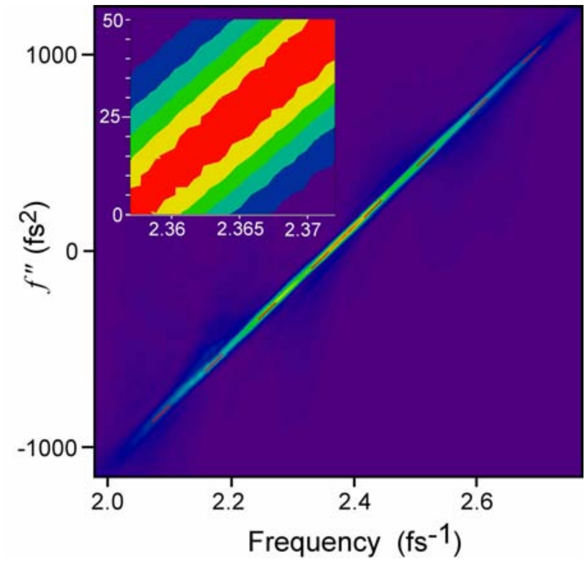

Fig. 2. Experimental measurement of a cubic phase obtained by a single chirp scan. The figure is a contour plot of the SHG spectra measured at each value of applied chirp. The feature revealed by the spectral maxima corresponds to the second derivative of the cubic phase introduced. As expected, the second derivative is linear with frequency. The inset shows a magnified portion of the trace.

Once $\phi''(\omega)$ is obtained, double integration can be used to calculate $\phi(\omega)$. Figure 3 shows the measured and the introduced functions, together with the spectrum of the laser. Note the excellent accuracy of the results, which were obtained from a single chirp scan. The data shown in Fig. 3 measured the third-order dispersion (TOD) with 0.5% accuracy.

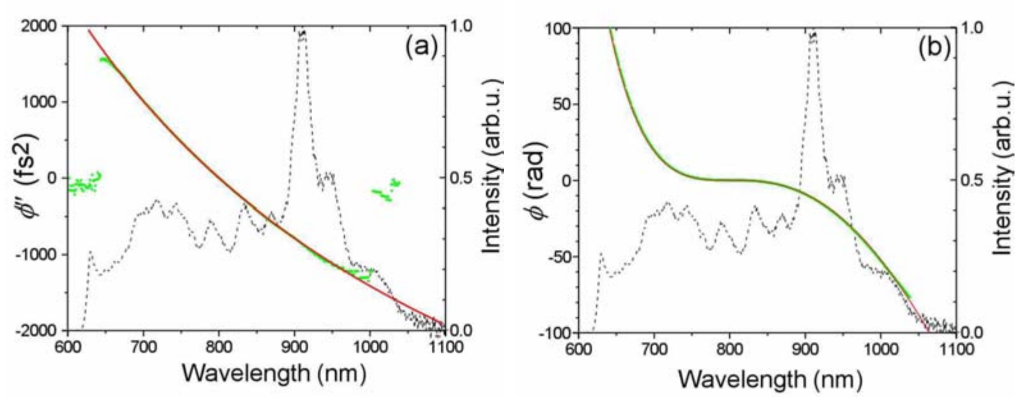

Fig. 3. Spectral phase measurement. The introduced (red) and measured (green) phase functions agree without adjusting parameter. (a) Shows the second derivative of the spectral phases. (b) Shows the spectral phases. The spectrum of the pulses is also shown (dashed).

The method presented is able to measure arbitrarily complex spectral phases, as has been shown for MIIPS [8, 9]. To demonstrate this ability, a sinusoidal spectral phase function defined by $\phi(\omega)=5\pi sin[7\text{fs}\,(\omega-\omega_0)]$, was introduced using the pulse shaper and then measured by the method described here (see Fig. 4). As evident from the screen shot shown in Fig. 4a, the second derivative of the introduced phase is obtained from a chirp scan. The measured phase (green) is very close to the phase introduced by the pulse shaper (red), as shown in Fig. 4b. To improve the quality of this method, an iterative measurement-compensation routine can be used [8, 9].

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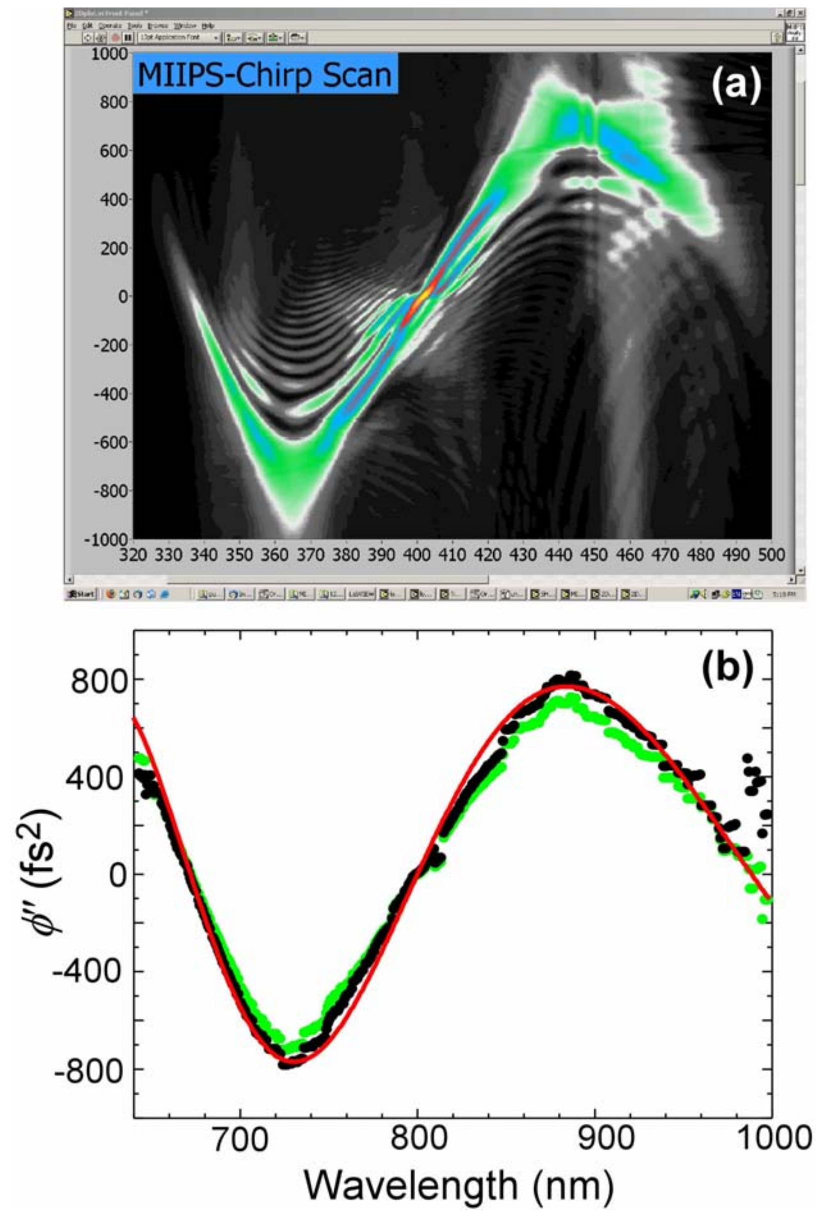

Fig. 4. Sinusoidal spectral phase measurement. (a) Shows the experimental trace. The measured second-derivative of the phase can be directly visualized from the feature corresponding to the spectral maxima. (b) Shows the measured second-derivative after a chirp scan (green) and after one measurement-compensation iteration (black). The red curve corresponds to the introduced sinusoidal function.

4. Discussion

This new MIIPS implementation does not necessarily require the use of an adaptive pulse shaper. Given that different amounts of chirp can be applied using passive optics, the method can be conveniently implemented using these devices. A more versatile option is the use of an adaptive pulse shaper, as shown in this work. In this case, compression can be thoroughly accomplished by applying $-\phi(\omega)$ to null the measured phase distortions.

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In addition to linear chirp, other reference functions can be employed when using an adaptive pulse shaper. Even though the simplicity of the measurement resulting from using a constant $f''(\omega)$ has been highlighted in the present study, there are as many variations of the method as reference functions one can implement. The use of a sinusoidal $f''(\omega)$ and its accuracy have been demonstrated [8, 9, 12]. Other options include cubic reference phases, which correspond to a diagonal grid. We have implemented this approach with great success on arbitrarily complex distortions. These results will be published in a more detailed communication.

The MIIPS chirp scan implementation demonstrated in this paper is especially suitable for sub-50fs pulses. For a measurable distortion $\Delta\phi''$, the corresponding change $\Delta I_{SHG}(\Delta\phi'')=I_{SHG}(\Delta\phi''=0)\times\beta^2/2(\Delta\phi''/\tau_0^2)^2$ needs to be bigger than the noise N, where τ_0 is the time duration of the pulses. For a Gaussian pulse, we obtain $I_{SHG}(\Delta\phi''=0)\times\beta^2/2(\Delta\phi''/\tau_0^2)^2\approx N$, where $\beta=4ln2$. Typically, the noise of the SHG signal is about a few percent. Therefore, the precision of the ϕ'' measurements is about $0.1\tau_0^2$. For the laser system used in this study $\tau_0\approx5$fs and we calculate a 2.5fs^2 precision, which is in excellent agreement with our experimental results.

In conclusion, a new MIIPS implementation based on a simple chirp scan was presented. The corresponding experimental trace directly yields the second derivative of the unknown spectral phase, without any mathematical treatment.

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