Appl. Phys. B 91, 283–286 (2008)
DOI: 10.1007/s00340-008-2978-6

Applied Physics B
Lasers and Optics

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Complete characterization of weak ultra-short near-UV pulses by spectral interferometry

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Received: 13 November 2007 /
Revised version: 28 January 2008
Published online: 27 March 2008 •

ABSTRACT We present a method for a complete characterization of a femtosecond ultraviolet pulse when a fundamental near-infrared beam is also available. Our approach relies on generation of the second harmonic from the pre-characterized fundamental, which serves as a reference against which an unknown pulse is measured using spectral interferometry (SI). The characterization apparatus is a modified second harmonic frequency resolved optical gating setup which additionally allows for taking SI spectra. The presented method is linear in the unknown field, simple and sensitive. We checked its accuracy using test pulses generated in a thick nonlinear crystal, demonstrating the ability to measure the phase in a broad spectral range, down to 0.1% peak spectral intensity as well as retrieving π leaps in the spectral phase.

PACS 42.65.Ky; 42.65.Re

1 Introduction

Techniques allowing for a complete characterization of the instantaneous intensity and frequency of a femtosecond pulse were first introduced in the 1990s. The FROG (frequency-resolved optical gating) [1], SPIDER (spectral interferometry for direct e-field reconstruction) [2] and sonographic techniques [3] are the most popular. These techniques are usually based on sum-frequency generation of an unknown near-infrared (NIR) pulse with a modulated copy of itself. The resultant ultraviolet (UV) radiation is registered as a function of modulation. Finally, the pulse envelope and phase are retrieved from acquired data. It is crucial for those methods to filter out the fundamental components from the sum-frequency signal. Fortunately, it can be easily accomplished by inserting a colored glass or spatial filter. Moreover, the UV signal is measurable with inexpensive silicon-based photodetectors.

Frequently, one faces the need to characterize the second harmonic (SH) of nanojoule pulses of Ti:sapphire lasers. This is the case in ultra-fast spectroscopy [4, 5], micromachining [6] or downconversion-based photon-pair sources [7–9]. Directly generalized FROG and SPIDER techniques suitable for characterization of the UV pulses are based on upconverting or downconverting the unknown second-harmonic pulse using the fundamental beam [10, 11]. The resultant radiation, which carries information about the unknown pulse, is difficult to detect. In the case of using upconversion the detected signal is centered around 266 nm where the silicon detectors are inefficient. On the other hand using downconversion produces weak pulses at the fundamental wavelength that are easily overwhelmed by the stray background. Moreover, the nonlinear up- or downconversion process requires substantial intensity of the measured beam. The latter requirement has recently been diminished by an application of an optical parametric amplification based conversion process [12]; however, these new approaches are complicated in use. Whereas for sub-μJ pulses other nonlinear processes can be used to characterize the pulses as in self-diffraction FROG [13] and zero additional phase SPIDER [14], the method described below could be useful also in those cases.

In this paper we present a method for a complete characterization of SH pulses when the fundamental beam is also available. This is a typical experimental situation, which can be exploited to avoid nonlinear conversion of a SH beam. It is assumed that the fundamental pulses are characterized prior to the measurement, for example using the well-established second harmonic generation frequency-resolved optical gating (SHG FROG) technique [1]. The scheme of our method is presented in Fig. 1. The idea of measurement is analogous to TADPOLE [15], but extended to near-UV pulses. A portion of the fundamental beam is converted into a reference second harmonic. Thanks to application of a very thin crystal in this step the complex spectral field of the reference pulse can be precisely calculated. Then, the reference pulse and the unknown pulse are brought to interfere on a slit of a spectrometer and spectral fringes are registered. The phase of the unknown pulse is retrieved from this signal using the Fourier filtering technique [16]. We demonstrate this method for a pulse generated in a 1-mm-thick beta barium borate (BBO) crystal oriented for type I phase matching. We were able to reconstruct the phase in a spectral range where the pulse intensity is above 0.1% maximum, including the π phase leaps.

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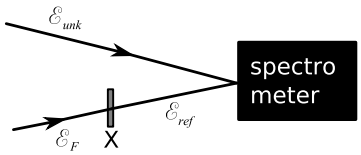

FIGURE 1 Pictorial diagram of the measurement scheme. $\mathcal{E}_{\mathrm{unk}}$: unknown pulse, $\mathcal{E}_{\mathrm{F}}$: fundamental pulse, $\mathcal{E}_{\mathrm{ref}}$: reference second harmonic, X: thin nonlinear crystal

2 Method

The first step in our measurement method is registering a FROG trace for the fundamental beam [1]. Next, we run the standard retrieval procedure using FROG software (Femtosoft Technologies) and obtain complete information on the electric field of the fundamental pulses. In particular, we learn about their complex spectral field $\mathcal{E}_{\mathrm{F}}(\omega)$:

$$\mathcal{E}_{\mathrm{F}}(\omega) = \sqrt{I_{\mathrm{F}}(\omega)}\mathrm{e}^{\mathrm{i}\phi_{\mathrm{F}}(\omega)}\,, \tag{1}$$

where $I_{\mathrm{F}}(\omega)$ is the spectral intensity and $\phi_{\mathrm{F}}(\omega)$ is the spectral phase. Note that $I_{\mathrm{F}}(\omega)$ can be measured directly with a spectrometer. This gives us a possibility to check the consistency of the data retrieved from FROG. Next, we double the known fundamental pulses in a BBO crystal. We choose a thin crystal because of negligible group-velocity mismatch between NIR and UV bands and large conversion bandwidth.

We take advantage of this fact to calculate precisely the output second harmonic and use it as a reference. Its complex spectral field $\mathcal{E}_{\mathrm{ref}}(\omega)$ is given by the usual convolution formula

$$\mathcal{E}_{\mathrm{ref}}(\omega) \propto \int \mathrm{d}\omega' \mathcal{E}_{\mathrm{F}}(\omega')\mathcal{E}_{\mathrm{F}}(\omega-\omega')\,. \tag{2}$$

In particular, we learn about the spectral phase of the reference pulses $\phi_{\mathrm{ref}}(\omega)$. The above formula is a reflection of the fact that the second-harmonic field in time is the fundamental field squared: $\mathcal{E}_{\mathrm{ref}}(t) = \mathcal{E}_{\mathrm{F}}^2(t)$. Note that FROG retrieves the reference pulse including the phase acquired during propagation through the FROG setup. Also note that even if the crystal narrows down the bandwidth of the reference pulse, it contributes a negligible phase to it.

We exploit the spectral interferometry method in order to retrieve the phase difference between the unknown second-harmonic pulse $\mathcal{E}_{\mathrm{unk}}(\omega)$ and the reference pulse $\mathcal{E}_{\mathrm{ref}}(\omega)$. This is accomplished by directing them with a relative delay τ into the spectrometer slit, where they interfere. Hence, we measure the interference spectrum $I_{\mathrm{SI}}(\omega)$ of the form

$$\begin{aligned} I_{\mathrm{SI}}(\omega) &= \left|\mathcal{E}_{\mathrm{ref}}(\omega)\mathrm{e}^{-\mathrm{i}\omega\tau} + \mathcal{E}_{\mathrm{unk}}(\omega)\right|^2 \\ &= I_{\mathrm{ref}}(\omega) + I_{\mathrm{unk}}(\omega) + 2\sqrt{I_{\mathrm{ref}}(\omega)I_{\mathrm{unk}}(\omega)} \\ &\quad \times \cos[\phi_{\mathrm{ref}}(\omega) - \phi_{\mathrm{unk}}(\omega) - \omega\tau]\,. \end{aligned} \tag{3}$$

An exemplary interference spectrum $I_{\mathrm{SI}}(\omega)$ is shown in Fig. 2. The key information about the unknown phase $\phi_{\mathrm{unk}}(\omega)$ is contained in the $\cos[\phi_{\mathrm{ref}}(\omega) - \phi_{\mathrm{unk}}(\omega) - \omega\tau]$ term. First, we retrieve $\phi_{\mathrm{ref}}(\omega) - \phi_{\mathrm{unk}}(\omega)$ using the Fourier filtering technique [16]. The interference spectrum can be separated into three distinct elements: the sum of spectral intensities $I_{\mathrm{ref}}(\omega) + I_{\mathrm{unk}}(\omega)$, which is slowly varying with ω, and the

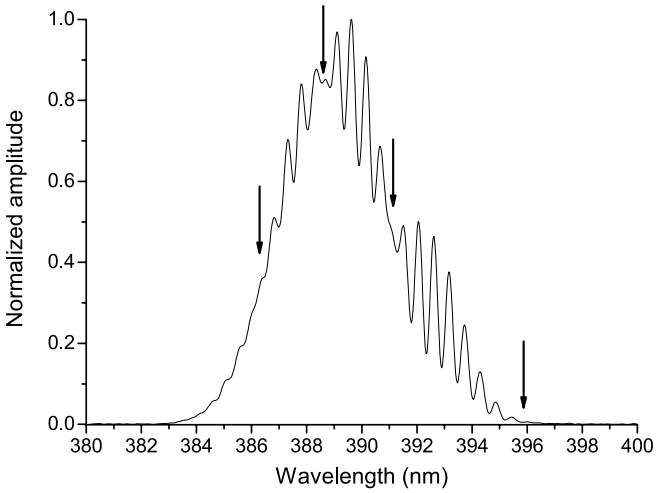

FIGURE 2 Interference fringes measured by the spectrometer used in the method. For a detailed description of the setup see Sect. 3. *Arrows* mark points of π spectral phase leaps

fringes described by the last term in (3), which consist of positive and negative frequency parts varying as $\mathrm{e}^{+\mathrm{i}\omega\tau}$ and $\mathrm{e}^{-\mathrm{i}\omega\tau}$. We calculate the Fourier transform of the interference spectrum, which splits those parts in the Fourier domain, provided that the delay τ is large enough. A typical result is shown in Fig. 3. Next, the Fourier transform is multiplied by a supergauss filter function $F(\tilde{t})$:

$$F(\tilde{t}) = \exp\left[-\left(\frac{\tilde{t}-\tau}{\Delta t}\right)^8\right], \tag{4}$$

where $\tilde{t}$ denotes time in the Fourier domain and Δt is the filter width. $F(\tilde{t})$ is non-zero near the $\tilde{t} = \tau$ point and zero elsewhere. Thus, the product of the filter function $F(\tilde{t})$ and the interference spectrum in the Fourier domain $\tilde{I}_{\mathrm{SI}}(\tilde{t})$ contains only the term originating from the part of $I_{\mathrm{SI}}(\omega)$ which varies as $\mathrm{e}^{+\mathrm{i}\omega\tau}$. It is transformed back into the frequency domain,

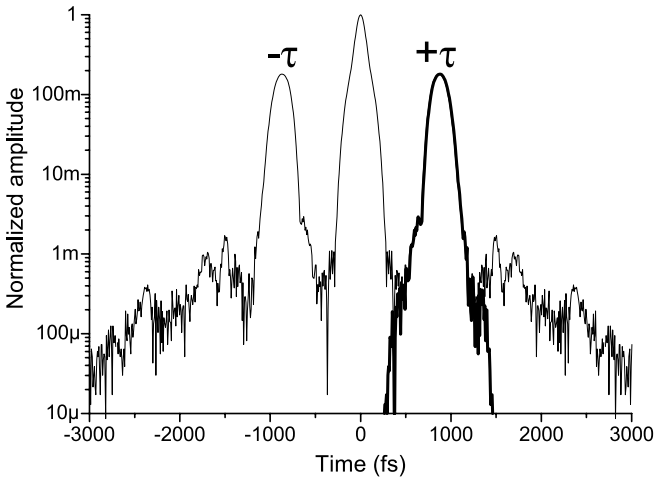

FIGURE 3 The Fourier transform of the interference fringe pattern (*thin line*) and the filtered signal (*thick line*) obtained using the supergauss filter from (4) with filter width $\Delta t = 500$ fs and temporal delay $\tau = 1000$ fs. Note the logarithmic scale on the vertical axis

yielding

$$I_{\mathrm{SI}}^{(+)}(\omega) = \sqrt{I_{\mathrm{ref}}(\omega) I_{\mathrm{unk}}(\omega)} \times \exp[\mathrm{i}(\phi_{\mathrm{ref}}(\omega) - \phi_{\mathrm{unk}}(\omega) + \omega\tau)] \,. \quad (5)$$

The argument of $I_{\mathrm{SI}}^{(+)}(\omega)$ is the phase difference $\phi_{\mathrm{unk}}(\omega) - \phi_{\mathrm{ref}}(\omega)$ and the linear term $\omega\tau$. Hence, the unknown phase is given by the following formula:

$$\phi_{\mathrm{unk}} = -\arg\left(I_{\mathrm{SI}}^{(+)}(\omega)\right) + \phi_{\mathrm{ref}} + \omega\tau \,. \quad (6)$$

Finally, the spectral intensity of the unknown pulse $I_{\mathrm{unk}}(\omega)$ is measured with a spectrometer and the spectral envelope can be reconstructed, which concludes the characterization:

$$\mathcal{E}_{\mathrm{unk}}(\omega) = \sqrt{I_{\mathrm{unk}}(\omega)}\mathrm{e}^{\mathrm{i}\phi_{\mathrm{unk}}(\omega)} \,. \quad (7)$$

The pattern of the fringes obtained in the experiment depends on the sign of τ. From the convention used in (3), it follows that if the reference beam path is delayed then positive τ is to be inserted in the reconstruction algorithm.

3 Measurement setup

Our measuring apparatus is depicted in Fig. 4. We used a mode-locked Ti:sapphire oscillator of 200-mW power and 80-MHz repetition rate. A 780-nm centered beam is split by a beam splitter BS1 into two paths. One part goes directly to the FROG-SI measurement apparatus, while the other is sent to the test pulse preparation arm.

In the first step we measured the SHG FROG trace of the fundamental beam. This measurement requires two replicas of the fundamental beam, which are produced by the beam splitter BS2. One replica reflected off BS2 undergoes an adjustable delay by bouncing off the corner-cube mirror CCM, while the second, transmitted through BS2, is directed with help of the flipping mirror FM. Both thus-produced pulses are focused on a 0.05-mm-thick BBO type I crystal X where the second-harmonic generation occurs and three beams emerge. Two of them, the upper and lower beams, are blocked by a diaphragm D. The middle beam is filtered out of a scattered fundamental beam by a blue filter BF and focused with a lens L2 on the spectrometer slit. We retrieved the fundamental pulse with a FROG error [1] of 0.0012 and we compared the retrieved spectrum with the fundamental beam spectrum measured directly by the spectrometer. The shape of the pulse was approximately Gaussian of spectral FWHM 17.5 nm and temporal FWHM 75 fs.

The second step is to measure the interference of the reference pulse and the unknown UV pulse generated by an experiment. For this purpose we switched the position of the flipping mirror to direct the generated UV pulse to the spectrometer. This pulse is assumed to pass unaffected through the crystal X, while the NIR beam reflected off the corner-cube mirror CCM generates the second harmonic of the form given by (2). These two UV pulses are let through by removing the diaphragm D and made to interfere on the slit of the spectrometer.

Note that the crystal suitable for FROG must be thin enough to guarantee the validity of (2). Therefore, we can use the same crystal for reference-pulse generation as for SHG FROG measurement.

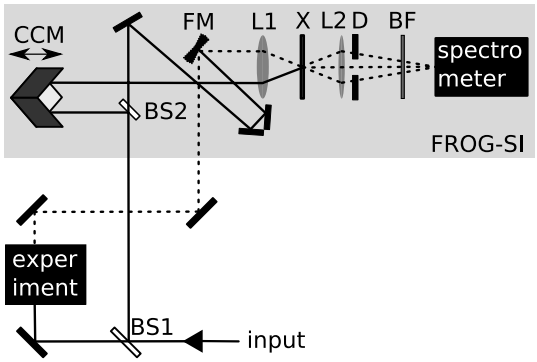

FIGURE 4 Experimental setup. BS1, BS2: beam splitters; FM, CCM: mirrors; L1, L2: lenses; X: SHG crystal; D: diaphragm; BF: blue filter

The spectral width of the calculated reference pulse was 5.6 nm FWHM, being in good agreement with the direct measurement that indicated 5.8 nm FWHM with the central frequency shifted by 0.3 nm to the blue.

4 Results

We verified the accuracy of our method using test pulses generated in a 1-mm-thick BBO crystal, filtered out of the remaining NIR light and attenuated with neutral density filters. The crystal and filters are represented as an 'experiment' in Fig. 4. The test pulse is directed to the FROG-SI apparatus. When producing the test pulses, we did not focus the fundamental beam in the crystal to achieve a well-defined phase-matching angle. This simplified the calculation of the test-pulse envelope and made the spectral phase features more pronounced.

In Fig. 5 we plot a comparison of a theoretically calculated pulse and one retrieved experimentally. The results displayed in Fig. 5a were obtained for a thick crystal oriented for phase matching at the wavelength of 778 nm, which is equal to the central wavelength of the incident fundamental beam. Figure 5b shows the result of a similar measurement with the crystal tilted by 63 mrad, which results in phase matching at 785 nm. Note the fidelity of π phase leaps at wavelengths where the spectral intensity reaches zero. The leaps can be noticed in raw interference fringes in Fig. 2. The inversion procedure obviously fails at wavelengths where the unknown pulse intensity is below the noise level and the fringes disappear. From Fig. 5 we infer that the threshold spectral intensity above which phase reconstruction is possible is about 0.1% of the maximum. Note that our method is capable of retrieving phase in the case when the spectrum of the unknown pulse is non-zero in two or more separated regions.

We calculated the pulse intensity and phase at the output face of the crystal producing test pulses using the formula for the second harmonic $\mathcal{E}_{\mathrm{SH}}(\omega)$ generated in a thick nonlinear medium in the case when pulse distortion can be neglected [17]:

$$\mathcal{E}_{\mathrm{SH}}(\omega) \propto \mathrm{sinc}\left(\frac{\Delta\beta L}{2}(\omega - 2\omega_{\mathrm{pm}})\right) \times \int \mathrm{d}\omega' A_1(\omega') A_2(\omega - \omega') \,, \quad (8)$$

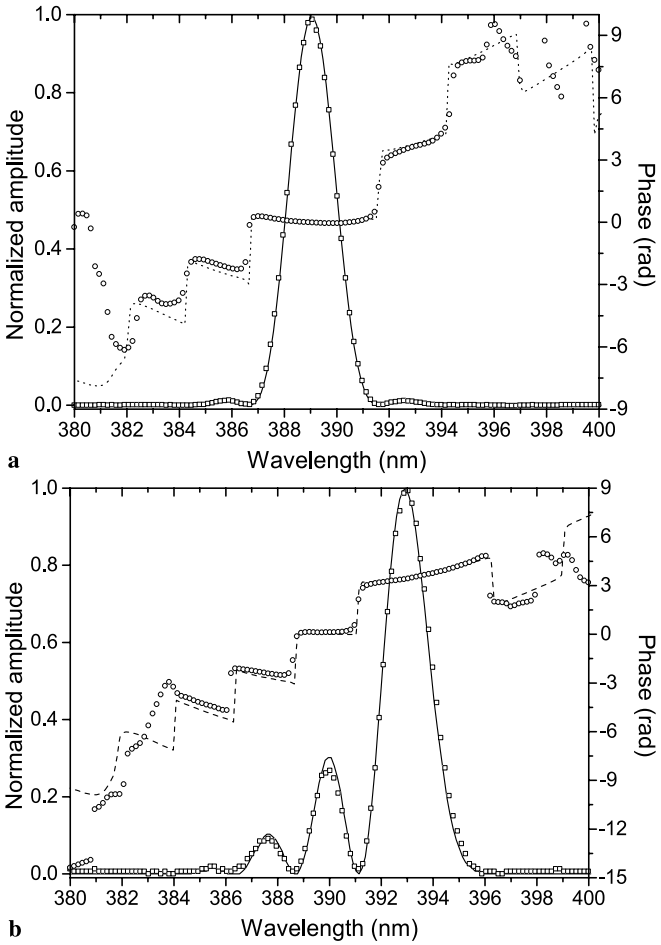

FIGURE 5 Spectral intensity (*solid line* – simulated, *rectangles* – retrieved experimentally) and phase (*dashed line* – simulated, *circles* – retrieved) obtained for a thick crystal oriented for phase matching at 778 nm (**a**) and with the crystal tilted by 63 mrad, which results in phase matching at 785 nm (**b**)

where L is the crystal thickness, ω_{pm} is the frequency of perfect phase matching and $\Delta\beta$ is the difference of the reciprocals of group velocities of fundamental and second-harmonic waves.

The necessary presence of neutral and colored glass filters in the test-pulse path influences it by contributing an additional phase $\Delta\phi_{\mathrm{filter}}(\omega)$. We describe it by Taylor expansion around the pulse central frequency ω_0:

$$\Delta\phi_{\mathrm{filter}}(\omega) = \beta_{0f} + \beta_{1f}(\omega-\omega_0) + \frac{\beta_{2f}}{2}(\omega-\omega_0)^2 + \ldots \qquad (9)$$

The constant and linear phase terms β_{0f} and β_{1f} correspond to pulse retardation. The only relevant and appreciable contribution is the quadratic dispersion $\beta_{2f}/2(\omega-\omega_0)^2$.

For measuring β_{2f}, we replaced the 1-mm BBO crystal with one that was 0.1-mm thick, which generated more broadband test pulses. Next, we acquired a reference spectral interferogram. Then, we inserted additional filters, identical to those to be characterized, in the test path and registered the interference again. The difference of the spectral phases retrieved in those two cases equals $\Delta\phi_{\mathrm{filter}}(\omega)$. By fitting the latter by a square polynomial we computed β_{2f}. Using these data we could subtract the influence of the filters from the measured test-pulse phase, obtaining the result shown in Fig. 5.

The agreement of measurement results with theoretical models certifies the validity of the presented method.

5 Summary

We have demonstrated a novel method for a complete characterization of the near-UV second harmonic of ultra-short pulses. It is applicable in situations when a nanojoule intense NIR beam is available. Additionally, the unknown pulse spectrum must lie within the spectral range of the second harmonic generated in a very thin crystal.

The main advantage of the presented method is a simple setup. It is a hybrid of standard SHG FROG with a spectral interferometer and there is no need to employ additional spectrometers or crystals. The pulse shape is inferred from a UV spectrum, which is easily registered. The algorithm used for phase retrieval is simple and non-iterative. In addition, our method is linear in the unknown pulse field.

The interference acquisition is single shot, without a need to scan a range of time delays τ. Additional averaging over various τ is possible with compensation of a linear phase.

Finally, our method allowed us to characterize a rather small phase contributed to the UV pulses by glass filters. Measurement of such a small phase would be very difficult to achieve using FROG.

ACKNOWLEDGEMENTS We acknowledge insightful discussions with C. Radzewicz, Ł. Kornaszewski and P. Wasylczyk. This work has been supported by the Polish budget funds for scientific research projects in years 2006–2008 and the European Commission under the Integrated Project Qubit Applications (QAP) funded by the IST directorate as Contract No. 015848. It has been carried out in the National Laboratory for Atomic, Molecular, and Optical Physics in Toruń, Poland.

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