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Discrete dispersion scanning as a simple method for broadband femtosecond pulse characterization

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Abstract: A simple and easy to implement technique for femtosecond pulse characterization is proposed and experimentally verified. It is based on the introduction of a known amount of dispersion (by controlling the number of passes through dispersive material) and subsequent recording of the spectral positions of second harmonic peaks obtained in a non-linear crystal. Such dependence allows for direct retrieval of the pulse spectral phase. The presented pulse characterization method is beneficial especially for broadband pulses, where the second harmonic spectrum exceeds the detection bandwidth of a single spectrometer.

OCIS codes: (320.7100) Ultrafast measurements; (320.7110) Ultrafast nonlinear optics; (320.5520) Pulse compression; (320.7090) Ultrafast lasers.

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1. Introduction

With the continuous pursuit to reliably generate femtosecond pulses with high peak powers, proper dispersion management plays a critical role. Pulse duration and its spectral phase characterization are key in achieving that goal. The two most established techniques for femtosecond pulse characterization are Frequency Resolved Optical Gating - FROG [1,2] and Spectral Phase Interferometry for Direct Electric-field Reconstruction – SPIDER [3]. Both techniques are matured and well verified, but still new, simpler, robust and more sensitive methods are desirable. One class of such femtosecond characterization techniques relies on the scanning of a known test phase and simultaneously recording the Second Harmonic (SH) spectrum. Representatives of that approach are MIIPS [4] and D-scan [5]. The first one employs a femtosecond pulse shaper, for example a spatial light modulator in a 4f setup [6], which from one hand allows to arbitrarily shape the test phase, but form the other, increases the price and complexity of the setup. The D-Scan technique points toward simplicity, where dispersion scanning is realized with just two glass wedges. Although that technique proved its noise resistance and insensitivity to narrowing of the conversion bandwidth, it still has some limitations. Especially, it is not easy to measure pulses which are chirped quite significantly. In that case the thickness of the wedges starts to be impractical.

In this paper we propose a simple and robust method of pulse characterization which is also based on dispersive scanning. In this realization continuous phase scanning is replaced by a single piece of dispersive material, and just the number of passes of the beam through it is varied – which can be considered as a discreet realization of the MIIPS technique. Moreover, instead of recording the full SH spectrum for each amount of the introduced dispersion, only the position of the peak of the second harmonic is registered. Such information suffices to retrieve the spectral phase of the measured pulse. This simple approach is beneficial for the characterization of broadband and significantly chirped pulses, typical for picosecond Optical Parametric Chirped Pulse Amplifiers (OPCPA) systems before compression. Since the only observable is the position of the SH peak, this method is especially useful to characterize broadband mid-IR pulses (1500-3000 nm), where SH spectra, necessary for other common characterization techniques, could not be recorded with just a single spectrometer.

2. Theory and simulation

Although the unprecedented increase in operational speed of electronic detectors allowed to directly trace pulses in the picosecond range, they are still too slow to trace the temporal waveform on femtosecond time scales. The most advanced streak cameras in the VIS-NIR range allow to achieve 200 fs temporal resolution, still for the mid-IR range are limited to tens of picoseconds. It could be shown that full characterization of ultrashort pulses requires a

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nonlinear process [7], with the exception of reference techniques, where a temporally synchronized reference pulse with known spectral phase is accessible [8]. The most easily accessible nonlinearity associated with femtosecond pulses is Second Harmonic Generation – SHG. In the undepleted regime and for perfect phase matching, the SH spectrum can be described by the following expression [9]:

$$I^{SH}(2\omega) = C\left|\int_{-\infty}^{+\infty}\left|E(\omega-\Omega)\right|\left|E(\omega+\Omega)\right|\exp\left[i(\phi(\omega-\Omega)+\phi(\omega+\Omega))\right]d\Omega\right|^2 \qquad (1)$$

Analysis of this equation shows that the SH spectrum does not only depend on the spectral amplitude of the fundamental pulse $|E(\omega)|$, but particularly the spectral phase $\phi(\omega)$ affects the final shape and conversion efficiency. One interesting observation is that for any phase which is antisymmetric around a certain frequency ω_0 the intensity of the SH at the doubled frequency $2\omega_0$ gives the same amplitude as the Fourier limited pulse [10], i.e. $I^{SH}(2\omega)|_{\omega 0} = I^{SH/FL}(2\omega)\ |_{\omega 0}$. At the same time, the intensity of the SH for other frequencies decreases rapidly, leading to formation of a pronounced peak in the SH spectrum.

Strictly speaking Eq. (1) shows that the process typically termed SHG is indeed the phase sensitive addition of sum frequency mixing (SFM) of various spectral components. For simplicity we still refer to it as SHG. It is often overlooked that any non-trivial spectral phase will lead to a group delay of the various spectral components. In the temporal picture only specific combinations of spectral slices will be able to interact in the SFM process and contribute to the SHG spectrum [11].

To obtain deeper insight into the relation between SH spectral shape and fundamental phase, the spectral phase $\phi(\omega)$ in Eq. (1) can be expanded into a Taylor series (up to second order) around a central frequency ω. The first order coefficient cancels, leaving only the second order term:

$$I^{SH}(2\omega) = \left|\int_{-\infty}^{+\infty}\left|E(\omega-\Omega)\right|\left|E(\omega+\Omega)\right|\exp\left[i\frac{\partial^2\phi(\omega)}{\partial\omega^2}\Omega^2\right]d\Omega\right|^2 \qquad (2)$$

due to the absolute value of the spectral amplitudes, both contributions from amplitudes are positives. Thus, the whole expression reaches a maximum only if the imaginary part fulfils the condition: $d^2\phi/\partial\omega^2 = 0$. This second derivative of the spectral phase is called Group Delay Dispersion (GDD). As a consequence the GDD has to vanish for the SH signal to be maximal. If a known amount of dispersion $GDD_{test}(\omega)$ is added to the pulses and the resulting position ω_{peak} of the SH peak is registered, we can solve the following equation: $GDD_{in}(\omega_{peak}) + GDD_{test}(\omega_{peak}) = 0$, allowing to retrieve the initial GDD_{in} at ω_{peak}. Repeating this step for different amounts of GDD_{test} allows retrieving the full spectral dependence of the initial GDD_{in} of the fundamental pulse. Subsequent double integration of the determined GDD_{in} function over the frequency ω gives the initial spectral phase $\phi_{in}(\omega)$.

This interplay between the fundamental phase and the SH spectrum is a key element of such characterization techniques as D-Scan, MIIPS, as well as the one presented here. In MIIPS the test phases (typically sinusoidal) are continuously scanned across the spectrum by a dedicated pulse shaper while the positions of the SH maxima are being recorded. It was also proposed that the test phase could be introduced by a simple linear device such as a stretcher-compressor or a pair of prisms [12–14], enabling full characterization of the femtosecond pulses. It is worth mentioning that the observable dependence of the position of the SHG peak for different amounts of the introduced GDD is only possible if there is a contribution of odd phase terms (like TOD). In case of pure second order phase there would be no observable change in the SH peak position, but only its amplitude would change, while for fourth order phase a double SH peak structure should be expected. Such limitation could be avoided with dispersion scanning techniques like MIIPS if more general test functions are used [15]. Nevertheless, in most of the typical cases (material dispersion or prism compressor) there is already a sufficient contribution of the third order phase to observe shifts of the SH peaks.

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The above derived condition for the position of the SH peak was obtained by expanding the spectral phase only to the second order. It is worth mentioning, that this condition is valid only if there is just one frequency fulfilling GDD(ω) = 0. Otherwise, the SH spectrum becomes more complicated, and it is no longer possible to assign single locations of the SH peak. However, the latter is not typically expected for the case of chirp originating from propagation through media, where GDD varies monotonically (away from absorption bands). For the pulses which are close to the Fourier limit, more advanced phase characterization techniques should be used. On the other hand, these are difficult to implement for significantly chirped pulses – the situation where the present method is readily applicable.

According to the more exact Eq. (1), the final position of the SH peak (due to integration over the whole frequency range) is not only defined by the GDD zero crossing point, but is also affected by the higher order terms. Utilizing the fact that the spectral amplitude of the fundamental pulses $|E(\omega)|$ can be easily measured, the position of the SH peak can be exactly calculated using Eq. (1) for any guess spectral phase $\phi_{in}(\omega)$. Such an approach allows to reproduce the position of the SH peaks more precisely, accounting for such effects as skewness of the fundamental spectrum or contribution from higher order terms of the spectral phase. For this reason the proposed pulse characterization method relies on Eq. (1) instead of GDD zeroing, for more accurate reproduction of the location of the SH peaks.

The idea of the proposed characterization method is based on the measurement of the position of the SH peak for different test phases. In our realization the test phase $\phi_{test}(\omega)$ is introduced by a slab of material with known dispersive properties. Changing the number of passes *n* of the beam through the material affects the spectral phase of the pulses accordingly: $\phi_n(\omega) = n\phi_{test}(\omega) + \phi_{in}(\omega)$, which in turn reshapes the spectrum of the SH. As a result the position of the SH peak vs. total thickness of the dispersive material can be obtained. The shift of the peak of the SH spectrum with varying linear chirp has already been reported some time ago [11] but was not used for pulse characterization. A similar set of points can be calculated using Eq. (1) assuming some initial spectral phase $\phi_{in}(\omega)$. When choosing a polynomial as the functional form for $\phi_{in}(\omega)$, the calculated SH peak positions can be fitted to the measured ones, with the coefficients of the polynomial as the fit parameters.

To verify this approach we numerically simulated the reliability of our phase retrieval algorithm. Femtosecond pulses with a spectral FWHM of 350 nm centred around 1800 nm were assumed. The Taylor coefficients of the initial spectral phase were set to 1500 fs^2 and 6000 fs^3 accordingly. To further test the approach against experimental noise, the spectral intensity was intentionally modulated by 20% noise (see Fig. 1(a), grey line). In this simulation discreet dispersion scanning was obtained by varying the number of passes from 1 to 9 through a sapphire plate with the thickness of 5 mm. The introduced test phase was calculated using the Sellmeier equation of sapphire [16]. For each configuration the SH spectrum was calculated according to Eq. (1), and was additionally modulated with noise on the same level as before. The position of the SH was then determined using a peak finding algorithm. The procedure was repeated for each number of passes through the dispersive material. An example of the SH spectrum (with added noise) corresponding to three passes through a sapphire plate is shown as an orange line on Fig. 1(a). The result of the discreet dispersion scanning is depicted on Fig. 1(b) together with a fit to the data (assuming second and third order spectral coefficients).

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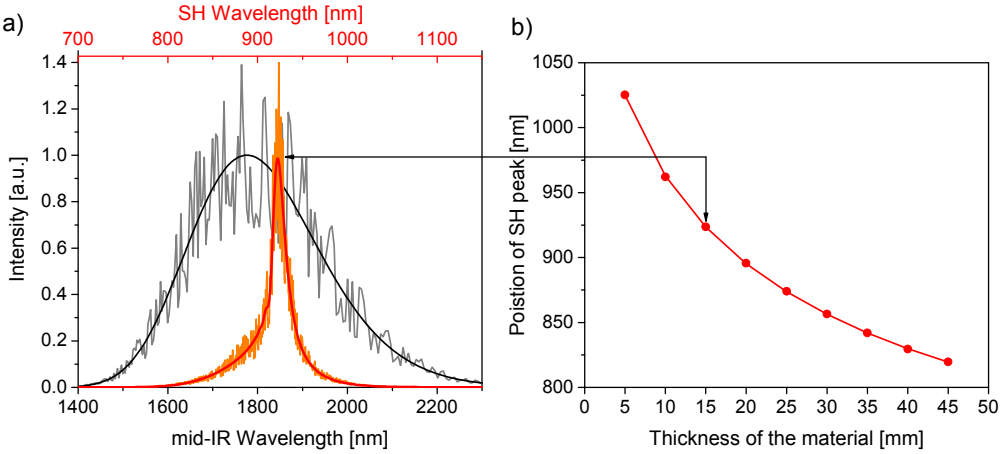

Fig. 1. Simulation results of the phase retrieval from SH peak positions: a) black line and red line – fundamental and SH spectrum, corresponding to 3 passes through sapphire plate (normalized to 1). Grey and orange – the same spectra but calculated with 20% amplitude noise. b) set of positions of SH peaks for different amount of material (red dots) together with the fitted values (red line).

In the next step, the minimization algorithm was used to find such an input phase $\phi_{in}(\omega)$, that would best reproduce the previously obtained locations of the SH peaks. As a result Taylor coefficients of the retrieved phase are 1498 ± 6 fs^2 and 5965 ± 75 fs^3 – which equal to the assumed ones within the numerical accuracy given by the added noise. This simulation proves that despite the high amount of the additional noise (for both fundamental and SH spectra), the initial phase can still be accurately retrieved. Demonstrated high immunity to experimental noise is especially beneficial for systems with low pulse energy or low repetition rate. However care has to be taken when the fundamental spectrum is far from Gaussian shape (with double or few peaks). Then the simple peak location algorithm may not be sufficient to properly identify the position of the SH peak and more advanced localization methods should be used.

3. Experiment

For the experimental verification of the usability of the presented pulse characterization technique, we used a broadband mid-IR source in a similar configuration to the one described in [17,18]. First, broadband pulses around 680 nm were generated using an OPCPA setup, pumped by the SH of an Innoslab pump laser [19] running at 100 kHz repetition rate. The mid-IR pulses were then generated in a Difference Frequency Generation (DFG) process, by mixing the visible pulses with the fundamental 1030 nm pulses.

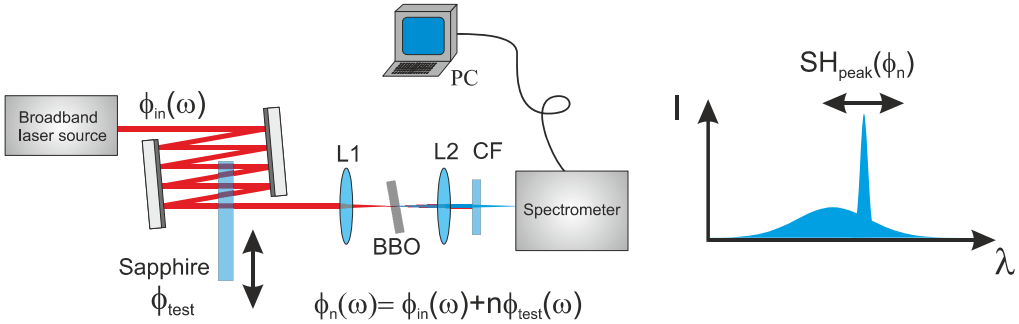

Fig. 2. Experimental setup; L1,L2 – focusing lenses, BBO – nonlinear crystal, CF – short pass glass color filter.

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As a result broadband infrared pulses spanning the range of 1500 – 2400 nm were generated. It is worth mentioning, that since the DFG was driven by the pulses with the same carrier frequency offset, the generated mid-IR pulses possess a passively stable carrier envelope phase [17,18,20,21].

The mid-IR pulses were then sent onto a pair of mirrors in a zig-zag pattern, so that the number of passes through the dispersive material can be easily controlled, see Fig. 2. Since the spectrum of the characterized pulses spans up to 2400 nm (see Fig. 3(a)), sapphire as dispersive material was chosen, having high transmission in that spectral range. For more precise measurements, we used sapphire plates with two thicknesses: 5 and 3 mm (with a beam propagating along the ordinary axis). In theory only one thinner plate should be sufficient, but it would result in a higher number of passes for the same amount of dispersion, and thus higher losses.

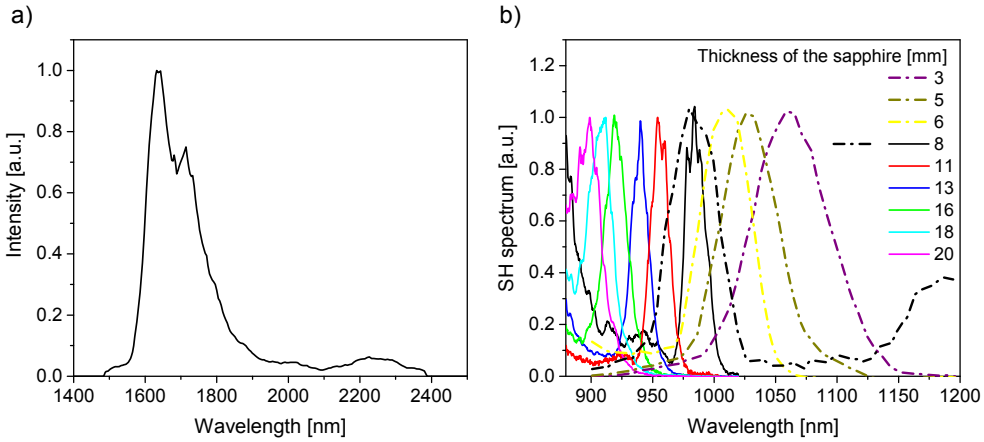

Fig. 3. a) Spectrum of the mid-IR pulses and b) second harmonic spectra for different amounts of dispersive material. Solid and dash-dotted lines represent the spectra measured with silicon and InGaAs spectrometers, respectively.

The SH beam was generated by focusing the mid-IR beam in a 200 μm BBO crystal. The fundamental beam was then rejected by the color filter, while the SH beam was fiber coupled to the spectrometer. Due to the broad bandwidth of the fundamental spectrum and its central wavelength, its second harmonic would cover the range of 750-1200 nm. This spectral range is somewhat difficult to register using just a single commercial spectrometer, because the sensitivity of regular silicon based spectrometers rapidly drops for wavelengths longer than 1000 nm, while commercial InGaAs spectrometers are not sensitive for wavelength shorter than 950 nm at present. With the availability of commercial InGaAs arrays sensitive down to 500 nm this should soon improve. In any case the spectrometers should be carefully calibrated against a black body source of known temperature [22]. The other common characterizations techniques based on second harmonic generation (such as SH-FROG or SPIDER) require precise measurement of the whole SH spectrum. In principle, the combination of two different spectrometers could be used (silicon and IR InGaAs), under the condition that both spectra are carefully stitched together. But due to the high dynamic range of the SH spectrum and different noise characteristic of the two spectrometers, such an approach is prone to significant systematic errors. Due to the fact, that our technique relies just on determining the locations of the SH peaks, even two spectrometers with different semiconductor materials can be applied to cover a broad spectral range. In our case, for wavelengths shorter than 970 nm, a silicon based spectrometer was used, while for longer wavelengths an InGaAs spectrometer was employed.

The effective material thickness was varied in the range from 3 to 20 mm. The resulting spectra for different amounts of sapphire material, recorded with the silicon (solid lines) and InGaAs spectrometers (dash-dotted lines) are shown in Fig. 3(b). It can be noticed, that as the

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amount of the dispersive material is increased, the position of the SH peak shifts towards shorter wavelengths. Taking into account, that in this spectral range GDD introduced by the sapphire plate is negative, it can be already concluded, that the measured pulses have positive GDD with a significant contribution of positive third order phase. The obtained positions of the SH peaks vs. the amount of introduced dispersive material are shown in Fig. 4.

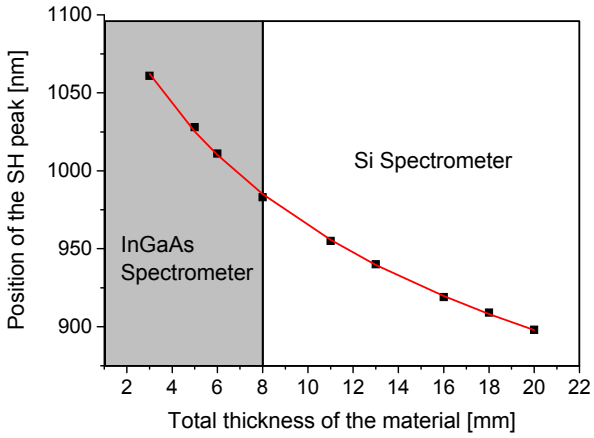

Fig. 4. Position of the SH peaks for different amount of the dispersive material. Longer wavelength points were measured with an InGaAs spectrometer and the ones for shorter wavelengths with a Si based spectrometer. The red line is a fit (GDD and TOD) to the experimental points.

In the next step the fitting procedure was used to find such a spectral phase of the input pulses that would closest reproduce the measured data points. As a result a phase with second and third order Taylor coefficients was sufficient to obtain a good fit to the experimental data (see red line on Fig. 4.) with an RMS value of 0.94 nm. Although the positions of the SH peaks do not completely cover the spectrum of the measured pulses (due to high asymmetry of the fundamental spectrum intensity), fit quality and dispersion properties of the characterized laser setup justify extrapolating the spectral phase for the full bandwidth of the pulses. The Taylor coefficients of the retrieved input spectral phase equal: 1532 ± 68 fs^2 and 6456 ± 650 fs^3. Those coefficients could later be used to design a chirped mirror compressor or as a control signal for a programmable pulse shaper, enabling compression of the infrared pulses close to the Fourier limit.

In case of our laser system, the measured pulses were positively chirped i.e. GDD>0, and subsequent GDD zeroing was possible by changing the number of passes of the beam through sapphire plates, which introduced a negative dispersion. In general, if the initial dispersion of the measured pulses had the same sign as the dispersion of the material, GDD zeroing would not be possible. In such case discrete dispersion scan can be achieved by replacing the mirrors with chirped mirrors, and varying the number of reflections. The other possibility is to pre-chirp the pulses with known phase (by compressor, chirped mirrors or grisms) and then performing a discrete scan as described previously.

4. Conclusions

The proposed characterization method, based on a discrete dispersion scanning with subsequent SH peak localization, represents a simple and useful method of femtosecond pulse characterization. Due to its simplicity, with a setup based just on a single piece of dispersive material and SHG crystal, it can be easily constructed and used as a quick and handy characterization technique. Its resistance on experimental noise and its high sensitivity allow characterizing femtosecond pulses with low input powers. It is especially useful for measuring pulses with wavelengths in the mid-IR range, where either second order techniques

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require cumbersome stitching of the spectra registered with different spectrometers or higher nonlinearity techniques (like THG-FROG) are necessary.

The presented discrete dispersion scanning method is mainly applicable to the characterization of pulses with smooth, monotonic dependence of GDD. This type of the dispersion, however, is common for most laser systems (like OPCPA), where ultrashort pulses experience material dispersion associated with their propagation through optical and dispersive elements.

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