Spectral-temporal Measurement Capabilities of Third-order Correlators

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Abstract: We present a method extending scanning third-order correlator temporal pulse evolution measurement capabilities of high power short pulse lasers to spectral sensitivity within the spectral range exploited by typical chirped pulse amplification systems. Modelling of the spectral response achieved by angle tuning of the third harmonic generating crystal is applied and experimentally validated. Exemplary measurements of spectrally resolved pulse contrast of a Petawatt laser frontend illustrate the importance of full bandwidth coverage for the interpretation of relativistic laser target interaction in particular for the case of solid targets.

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1. Introduction

Scanning third-order correlators [1–4] are state of the art reference devices for pulse contrast characterization of high-power high-intensity lasers [5–8] driving relativistic laser-plasma research [9]. In particular for the controlled interaction with solid targets knowledge of this temporal intensity distribution, easily covering a dynamic range of 12 orders of magnitude, is of crucial importance. Laser plasma ion acceleration [10–15] and its various applications [9, 16, 17] represent a prominent example in this field. The dynamic range covered by scanning devices is unprecedented as compared to single-shot techniques featuring complementary advantages [18]. Contrast of a laser pulse is defined as the instantaneous intensity ratio at given delays in the temporal pulse distribution relative to the peak intensity. This ratio is dominated by different effects at different time scales and is in its complexity an ongoing topic of research [7, 8, 10, 19–21]. At short times around the main pulse (<10 ps) the so-called coherent contrast resulting from imperfections of the main pulse compression is dominating. For intermediate delays up to several tens of picoseconds coherent pulse distortions, broad pedestal as well as distinct pulses, generated e.g. by scattering or phase noise in stretcher systems and non-linear coupling during propagation, are the main origin of contrast degradation [22–24]. At larger delays, on the order of the duration of the stretched pulse and beyond, incoherent amplified spontaneous emission and pre-pulse replica from the main pulse are the prevalent cause of contrast degradation, where both can extend to many nanoseconds, related to path length in amplifiers. Commercially available third-order correlators are employed to measure the signal level of all of these features. The laser pulse is sampled in time by delaying the fundamental laser pulse relative to a frequency doubled reference pulse, derived from the fundamental. The third-harmonic signal is generated by sum-frequency generation of the two. The absolute level of the third-harmonic signal is derived from calibrated
neutral density filters used for attenuation of the input beam in combination with the signal level of the detector for the third-harmonic, namely a secondary electron multiplier (SEM). The temporal pulse evolution of the original pulse is recovered by recording the third-harmonic signal together with the delay between fundamental and second-harmonic light. The nature of the signal generation process in TOCs via second-harmonic and third-harmonic generation allows only a characterization of the contrast for a limited bandwidth of the initial laser pulse. Thus the measured pulse contrast is only representative for this spectral bandwidth.

In this paper a model of the sum-frequency process of the third-harmonic signal generation is presented. This model supports the extension of the application range of third-order correlators to spectrally resolved contrast measurements through controlled angle-tuning of the THG crystal with the potential of revealing previously non-detected contrast degrading spectral features. The validity of this model is demonstrated by the representation of measured third-harmonic spectra for different wavelength settings and by measuring the spectral-temporal distribution of a sample pulse stretched by material dispersion. Finally, the frequency resolved characterization of the contrast of a high-intensity high peak-power laser pulse is discussed, indicating a significant difference of the measured temporal contrast for different spectral components.

2. Theory

Modelling the sum-frequency generation in a nonlinear optical crystal requires energy and momentum conservation. Energy conservation is expressed as

\[ \omega_{TH} = \omega_F + \omega_{SH}, \]  

where \( \omega_F \) and \( \omega_{SH} \) refer to the fundamental and second-harmonic optical radial frequencies, while \( \omega_{TH} \) is the resulting third-harmonic radial frequency [25].

Momentum conservation is achieved by phase matching in nonlinear optical crystals. In case of type I phase matching in a negative uniaxial nonlinear optical crystal like barium borate (BBO) the fundamental and the second-harmonic are propagating in ordinary polarization, while the pulse is generated in extra-ordinary polarization experiences a refractive index depending not only on the wavelength (as for ordinary polarization), but also on the phasematching angle \( \theta \) relative to the optical axis of the crystal. Momentum conservation is defined by \( k_{TH}(\theta) = k_F + k_{SH} \), with \( k_{TH} \), \( k_F \) and \( k_{SH} \) representing the wavevectors of the third-harmonic, the fundamental and the second-harmonic respectively. Note that for convenience and as relative angles involved are small we use the scalar field notation. The wavevector \( k \) is then defined as \( k = \omega \cdot n / c = 2 \cdot \pi \cdot n / \lambda \), with \( n \) representing the wavelength depending refractive index, \( c \) the speed of light in vacuum and \( \lambda \) the wavelength.

\( k_{TH}(\theta) \) is propagating with extra-ordinary polarization. This allows for fulfilling the conditions for momentum conservation for a set of single wavelengths by tuning \( \theta \). If the combination of wavelength is not matching, which typically applies in broad bandwidth pulses and setting \( \theta \) for a specific wavelength combination, the equation must be expanded by \( \Delta k \) representing the wavevector mismatch:

\[ \Delta k(\theta) = k_{TH}(\theta) - (k_F + k_{SH}). \]

The wavevector mismatch is leading to a reduction of conversion efficiency \( \eta \) by \( \eta \sim \text{sinc}^2(0.5 \cdot L \cdot \Delta k(\theta)) \) with \( L \) representing the interaction length in the specific non-linear crystal [25].

The conversion efficiency further depends on the shape of the spectral intensity distribution of fundamental and second harmonic pulses generating the third harmonic by \( \eta \sim I(\omega_F) \cdot I(\omega_{SH}). \)

In our case the fundamental spectrum is a super-Gaussian. The spectral shape of the second-harmonic can be expressed in a similar manner as the third-harmonic generation with a sinc function shape and a bandwidth depending on SHG crystal type and crystal length [25, 26].
The combination of both efficiency considerations leads to an overall dependency of

\[ \eta(\omega_F, \omega_{SH}, \theta) \sim I(\omega_F) \cdot I(\omega_{SH}) \cdot \text{sinc}^2\left(\frac{L \cdot \Delta k(\theta)}{2}\right). \] (3)

Here several common assumptions have been made. A stationary approach for the sum-frequency

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Fig. 1. Synthesis of the THG conversion efficiency \( \eta \) (a) resulting from the combination of fundamental (super-Gaussian) and second-harmonic (sinc function) spectral intensity distributions. The fundamental super-Gaussian spectrum with order 2.5 is centered at 800 nm with a FWHM (full width at half maximum) bandwidth of 42 nm. The sinc function of the second harmonic pulse is centered at 400 nm with a FWHM bandwidth of 5.6 nm. (b) Conversion efficiency in an exemplary BBO crystal with an interaction length for the simulation of 0.5 mm and internal angle of 44.3°, the optimal angle for generation of third-harmonic output at 266.7 nm, resulting from the wavevector mismatch. (c) Combination of both efficiencies results in the spectral efficiency map. The point of highest efficiency is marked by a ★. Iso-wavelength lines for third-harmonic wavelengths following energy conservation are shown as dash-dotted lines. (d) Generated third-harmonic spectrum. All plots are normalized. The colorbar is valid for all plots.
length of 100 fs for the second-harmonic probe pulse and oo-e phase matching in BBO, the
characteristic length is larger than 300 μm. As we will see later this is easily fulfilled by the third-
harmonic crystal thickness. The spatial walk-off under the same conditions amounts to less than
20 μm. This is negligible compared to typical beamsizes in the millimeter range. Furthermore
non-depletion of the fundamental and second-harmonic can be assumed, as well as that no
absorption takes place. Now, for a specific angle θ a spectral efficiency map η of third-harmonic
generation, synthesized from the combination of fundamental and second-harmonic spectra,
shown in figure 1(a), and the efficiency given by the wavevector mismatch, shown in figure 1(b),
can be generated [28]. The resulting spectral efficiency map is shown in figure 1(c). For each
combination of wavelength a third-harmonic generation efficiency is calculated according to
equation 3. Equation 1 is represented by iso-wavelength lines for the third-harmonic light, as
shown as dash-dotted lines in figure 1(c). For retrieving the third-harmonic spectrum shown
in figure 1(d) the intensity values along the iso-wavelength lines are summed up. The spectral
efficiency map shown in figure 1(c) can be evaluated for different angles θ as shown in figure
2. Tuning the angle leads to a change of the position of the efficiency map governed by the
wavevector mismatch and thus a change of the third-harmonic spectrum. As long as the peak
efficiency lies within the plateau of the spectrum of the fundamental pulse it is as well close to the
central wavelength of the second-harmonic. When tuning further in angle the peak of efficiency
is not only moving along the fundamental wavelength axis, but also along the second-harmonic
axis, illustrated in figure 2 (b) and (d).
The combination of fundamental and second-harmonic spectra is generating a two-dimensional
spectral intensity profile (see figure 1a). The phase matching then slices out a slant cut. As
long as the slant cut is dominantly within the plateau of the spectral intensity profile the shift
along the second-harmonic axis is minor. In contrast, when this slant cut is touching the slopes
of the spectral intensity profile the peak of highest efficiency is shifted in both spectral ranges
(c.f. transition between figs. 2 (a) to (b) and (d) to (e)). Tuning θ further to the edge of the
spectral intensity profile the side wings, originating from the second-harmonic spectral sinc shape,
become strongly visible, as depicted in figure 2 (a) and (e). Thus the peak of efficiency is even
stronger shifted along the second-harmonic spectral axis, and spectral parts of the fundamental
closer to the central wavelength are used for third-harmonic generation leading to a different
combination range for third-harmonic signal generation.
The spectral bandwidth of the third-harmonic signal is also affected by tuning of angle θ. While
within the plateau the bandwidth stays nearly the same and is dominated by the wavevector
mismatch. At the tuning edges the bandwidth significantly broadens and modulates due to the
prominent mixing with the side-bands of the second-harmonic sinc. Also, when tuning away
from the second-harmonic central wavelength the effective usage of all spectral components of
the second-harmonic is not given anymore, resulting in a less efficient third-harmonic signal
generation. This effect can be studied in comparison to the original fundamental spectrum. For
the given parameters this attenuation effect is on a bandwidth of 50 nm less than a factor of 1.5.
These findings illustrate that the spectral details of fundamental and second-harmonic pulses as
well as the phase matching conditions must be considered for spectral-temporal measurements
with third-order correlators. Due to the wavevector mismatch and the reduced bandwidth of the
second-harmonic, the bandwidth of the generated third-harmonic is limited in comparison to
the original fundamental spectrum. This shows the necessity for tuning the spectral position
of the given measurement window by tuning the angle θ. Eventually, the full spectral contrast
representation of broad bandwidth laser pulses can in principle be retrieved with existing devices
from a series of reduced bandwidth measurements.
Fig. 2. Effect of tuning the angle $\theta$ on the spectral efficiency map. For each plot the relative angle to $44.30^\circ$, representing the optimal angle for the combination of 800 nm and 400 nm, is given. All plots are individually normalized and the color scaling of figure 1 is used. A reduced number of iso-wavelength lines of the third-harmonic are shown as well for 260 nm, 266.67 nm and 273.33 nm respectively. Note that for the relative angle change of $\pm 1.6^\circ$ ((a) and (e)) the first side maximum of the sinc function becomes dominant. For total signal level comparison the integrated signal values relative to the case of optimal angle (c) are given as $S_{rel}$.

### 3. Experimental Evaluation

A systematic experimental investigation of the consequences and opportunities resulting from bandwidth restrictions in third order correlators was performed at the CPA1 probe beam installation of the Draco Petawatt laser system [29, 30] shown in figure 3. The aim is to test the model for its ability to represent the third-harmonic spectrum and as a second step to characterize a chirped post-pulse as test case for the potential to measure and retrieve spectral-temporal structures. The main part of the laser pulses amplified in the CPA1 stage is compressed and sent to an XPW stage for contrast cleaning. The XPW pulses are then seeding the CPA2 stage and its subsequent main amplifiers.

As part of the CPA1 output is permanently made available for development purposes the present
systematic study was performed there. In order to generate a spectral-temporal pulse structure applied to validate the method the pulse was first sent into a Mach-Zehnder interferometer enabling the generation of pulse pairs with defined delay. By adding an SF6 glass block representing additional material dispersion into one of the interferometer arms a delayed and stretched post-pulse was generated in a well-defined way. After post-pulse generation both pulses are recombined and propagate co-linearly into an in-air optical compressor tuned to compensate the total dispersion accumulated by the main pulse in the stretcher and the amplifier system of CPA1. The residual dispersion of the post-pulse is then only the dispersion gained by the SF6 material. As a consequence of the temporal stretching of the post-pulse it will exhibit a residual chirp, characterized by different delays of its different spectral components. These delays can later be measured to test the model against the introduced chirp.

After compression the generated temporal two-pulse-structure was sent into a third-order correlator, namely a Sequoia from Amplitude [31]. The crystal used for third-harmonic generation is a BBO crystal type I with a cut of 44.3° internal phase matching angle and a thickness of 200 \( \mu m \pm 20 \mu m \) [32]. The crystal is mounted angle-tuneable in a mirror holder, nominally optimized for the combination of fundamental light of 800 nm and second-harmonic light of 400 nm central wavelength, generating 266 nm light. For spectral measurements of the fundamental, second-harmonic and third-harmonic signal inside the Sequoia we used an Avantes AvaSpec spectrometer (AvaSpec-3648-USB2, 181-1100 nm, 300 l/mm, 0.25 nm/px). The spectral characteristics of the fundamental and second-harmonic signal were measured, as shown in figure 4 together with their idealization by a super-Gaussian and a sinc function respectively. The idealizations of the spectra are used for further comparison of modeling and measurements. The fundamental spectrum is characterized by a FWHM of 50 nm and a super-Gaussian order of 2.5 centered at 793.8 nm. The sinc function is centered at 398.8 nm and shows a FWHM of 5.1 nm.

As first test of the model, expected and measured third-harmonic spectra are compared at zero delay with highest third-harmonic yield to demonstrate the capability to reconstruct the central wavelength. For this measurement the Mach-Zehnder interferometer arm containing the SF6 glass block was blocked. The third-harmonic crystal was tuned in angle and third-harmonic spectra were recorded. The sum-frequency conditions were then simulated and compared to the measured spectra, as shown in figure 5. For optimal matching of the simulation to the measured spectra the crystal thickness has been found to be 180 \( \mu m \), which is in good agreement with the specified thickness. The internal tuning angle of the third-harmonic crystal could not be measured in the actual device with absolute precision due to space constrains and given mount design. Nevertheless, this is of minor importance since the phase matching conditions are contained in the individual THG spectra, thus the peak wavelength of fundamental and second-harmonic and the crystal tuning angle can be retrieved from the model.

In the next step to validate the concept a series of temporal contrast measurements of a chirped post-pulse is analyzed. Within the series angle tuning of the THG crystal provides for spectral sensitivity and thus enables the spectral-temporal characterization of the post-pulse with dedicated chirp as shown in figure 6. The peak wavelength of the third-harmonic signal was thereby tuned from 261.9 nm to 269.4 nm. The model is applied to retrieve the central wavelength of the fundamental for individual contrast measurements at different angle tuning. Plotting the central wavelength of the narrow band windows versus the delay of the peak corresponding to the post-pulse in the contrast curve reveals the expected trace of its dominantly linear stretching. The material dispersion of 194 nm SF6 glass caused a group velocity dispersion (GVD) of 38.6 \( \cdot 10^3 \) fs\(^2\) [33]. The red line in figure 6 represents a group delay with a group delay dispersion of 36 \( \cdot 10^3 \) fs\(^2\) (a systematic error of 6.7 %). In contrast to the matching result derived from the presented model, the simple application of a fixed peak wavelength of the second-harmonic would lead to a non-linear slope of the group delay. This is due to the spectral part of the second-harmonic used for the sum-frequency process not being constant, as can be seen in figure
Fig. 3. Experimental setup implementation at Draco: (a) a part of the laser pulses amplified in the CPA1 stage of Draco is used for the experiment. (b) the experimental setup consists of a Mach-Zehnder interferometer (c) used for post-pulse generation. The post-pulses are additionally stretched by material dispersion in a SF6 block double-pass (with 194 mm total path length). The interferometer is followed by a grating compressor (d) compensating the stretching applied in the CPA1 stage of the main pulse. For measuring the temporal evolution of the main and residual stretching of the post-pulse a third-order correlator is used (e). A spectrometer (f) was applied on the fundamental (red), the second-harmonic (blue) and the third-harmonic (purple) beam. For the sake of completeness the full Draco laser chain is depicted. The XPW is followed by a second complete CPA system with two individual power amplifier / compressor systems.

Fig. 4. Spectra of the (a) fundamental and (b) second-harmonic pulses (black) and the idealized spectra used for further modeling (red).

2. The usage of a constant SHG in combination with equation 1 would lead to an S-shape of the chirp measurement, representing a non-existing TOD.
Fig. 5. Third-harmonic measured (black) and modelled (red) spectra for different tuning angles. The relative angle to 44.3° is given for each plot. Note, that the given angles are retrieved via the simulation.

A special situation occurs at the blue wing of the spectrum. The first side wing of the second-harmonic sinc is becoming dominant in signal intensity generation. This is leading to a change of the wavelength combination in the mixing process and to double peaking in the contrast trace visible in figure 6 at 770nm and for delays of 89.5 ps and 94.5 ps. This detail is already visible at neighbouring contrast traces and described by and thus confirming the model (see figure 2).

The dominance of the effect on the blue wing of the spectrum originates from the blue shifted fundamental and SHG compared to the model presented above. In summary, this correct analysis of the known chirp of an artificially generated post-pulse confirms the validity of the presented model linking the spectra of fundamental, second, and third harmonic spectra. Angle tuning of the THG crystal of the scanning TOC can thus be reliably exploited to measure spectrally resolved contrast curves of high power laser systems.

4. Spectrally resolved contrast measurement of the Draco frontend

The spectral-temporal tuning capabilities of scanning third-order correlators can be applied to perform a comprehensive characterization of the temporal contrast of high-power ultra short pulse laser systems, where standard diagnostic protocols do not cover the full spectral range of the
Fig. 6. Direct measurement of the spectral-temporal evolution (chirp) of the artificial post-pulse generated by the Mach-Zehner interferometer in combination with an SF6 glass block in the post-pulse arm. The time is given on the y-axis, while the fundamental central wavelength derived via the model. The black dots represent the peaks of the individual measurements in time vs. the retrieved central wavelength, while the individual normalized intensity is encoded in the color scale. The non-equidistant spectral position results from the individual angle tuning. For comparison the retrieved THG crystal tuning angles relative to 44.3° are given for each spectral slice. The width of the color scaled traces is not representative for bandwidth considerations or as error bar, but helps guiding the eye. The spectral width of the fundamental of each measurement can be retrieved by the model and is in the order of 10nm. For comparison the linear fit for the accumulated GVD of the SF6 glass block is given as red dashed line. The temporal position of the post-pulse relative to the main pulse was chosen to be well after the main pulse and to experience no other dominant pulse structures.

amplified pulse. A first measurement exploiting this additional tuning capability was performed at the CPA2 stage of the Draco frontend at 1.5 J pulse energy level, exhibiting a spectrum, red shifted due to gain saturation in the complex amplifier chain to a central wavelength of 805 nm with a FWHM bandwidth of 55.5 nm. In-air pulse compression was employed analogous to the scheme introduced with figure 3 yet bypassing the Mach-Zehnder interferometer. Apart from applying angle tuning of the THG crystal, the Sequoia was operated in standard configuration. Exemplary results are shown in figure 7 depicting spectrally resolved temporal contrast curves, where the center wavelength of individual traces corresponds to the tuning angle of the THG crystal and the spectral bandwidth of the fundamental is of the order of 10 nm. All measurements presented were performed under the same conditions, reflected in the same noise level of about $10^{-10}$ relative to the highest signal intensity at 814.8 nm and zero delay. They exhibit significant differences in pre- as well as in post-pulse contrast behaviour of up to two orders of magnitude.
on few tens of picoseconds time scales, in reference to just a factor of two in peak level for the central measurements (see inset in figure 7b).

Fig. 7. Spectrally resolved contrast measurement of the CPA2 stage of the Draco laser, displayed (a) as false color coded traces for each wavelength setting. All measurements are normalized to the highest overall peak intensity at 814.8 nm. For comparison the retrieved THG crystal tuning angles relative to 44.3° are given for each spectral slice. Sample line-outs at the center and at the edges of the spectral range are presented in (b). Both plots share the same dynamic scaling expressed by the identical colorbar (a) and y-axis (b) scaling.

5. Discussion and Conclusion

The presented model for sum-frequency generation for third-order generation was successfully applied to measurements performed with a standard third-order correlator, namely a Sequoia from Amplitude. The third-harmonic generation was investigated with spectral resolution and spectrally resolved contrast traces were recorded with the usual multi-shot scanning techniques. In a first demonstration the linear chirp, introduced by an SF6 glass block to an artificially generated post-pulse, was found to nearly match the expected GVD for SF6 within an error of 6.7%. This small mismatch is consistent with the expected accuracy of the spectral and temporal measurements. Thus, the method enables novel insight into spectro-temporal pre- and post-pulses features, in particular when chirped.

The second exemplary measurement of the spectrally resolved pulse contrast of the Draco frontend, amplified to a pulse energy level of 1.5 J, underlines the need for such extended metrology capabilities. It has to be noted that, at present, the origin of the striking deviation
in pulse contrast observed here between the blue and red side of the spectrum of the amplified pulse is not understood. It requires further systematic investigation of the amplification process as well as of components and techniques used in the high power laser chain and in the metrology devices. Given the high relevance for any controlled laser interaction with solid targets and their quantitative representation in computer simulations, such investigations are essential for the validation of established measurement protocols as well as drive laser performance, the latter in particular performed at full energy of the system and on target. If the observation presented in figure 7 can be traced back to the laser itself, it may hold promise for general improvements in overall temporal pulse contrast.

One way of in principle improving measurement quality could be by enhancing the bandwidth of the second-harmonic reference and reducing the THG crystal thickness. This would lead to the inclusion of more spectral components of the fundamental pulse in one measurement and thus to a more complete contrast information in a single diagnostic scan application. Yet, a compromise has likely to be found between increased bandwidth and reduced signal strength and thus dynamic range. A complementary approach could involve a reduction of spectral bandwidth of the SHG reference, enabling sharper spectral resolution in the individual contrast traces or spectral response calibration of third-order correlators. As a side effect broadening the SHG reference might lead to a violation of the quasi-static approach of the model introduced and used in this work. The influence of different spectral shapes on the measurement should be investigated as well. Therefore, further systematic studies of the potential and drawbacks of these measures have to be undertaken.

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