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High-dynamic-range pulse-front steepening of amplified femtosecond pulses by third-order dispersion

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Abstract. Pulses of 25-fs duration from a Ti:sapphire multipass amplifier system have been temporally characterized over a high dynamic range with autocorrelation based on third harmonic generation. The investigated high-order-dispersion-controlled kHz system produces millijoule-energy 25-fs pulses with a symmetric intensity envelope which decreases by five orders of magnitude at a delay of 300 fs. The low-intensity wings of the pulse result from residual dispersion of fifth and higher order. Further steepening of the leading edge of these high-contrast femtosecond pulses over a dynamic range of five orders of magnitude has been achieved by introducing appropriate amount of third-order dispersion with specially designed chirped multilayer mirrors.

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Laser–matter interactions at intensity levels exceeding $10^{15}\,\mathrm{W/cm^2}$ call for intense ultrashort pulses with a nearly exponential intensity envelope over several orders of magnitude, particularly on the leading edge of the pulse. The high intensity contrast is required to prevent the formation of a plasma in an uncontrolled manner before the peak of the pulse impinges on the target [1]. Even though oscillators can now produce high-contrast femtosecond pulses [2–4], chirped-pulse amplifiers (CPA) [5] tend to generate high-energy laser pulses with a pedestal-like background extending over a timescale orders of magnitude longer than the FWHM (full width at half maximum of the intensity envelope) duration of the pulse. This temporally extended background co-propagating with the intense amplified pulse is a consequence of the fact that usually only the lowest-order contribution to the frequency-dependent group delay of the system

$$T_g(\omega) = \frac{\mathrm{d}\phi}{\mathrm{d}\omega} = \phi'(\omega_0) + \phi''(\omega_0)(\omega-\omega_0) + \frac{1}{2}\phi'''(\omega_0)(\omega-\omega_0)^2 + \frac{1}{6}\phi''''(\omega_0)(\omega-\omega_0)^3 + \dots\,, \tag{1}$$

namely $\phi''(\omega_0)$, commonly referred to as group delay dispersion (GDD), can be precisely compensated, whereas residual higher-order contributions, such as third-order dispersion (TOD) $\phi'''(\omega_0)$, tend to distort the temporal shape of the amplified recompressed pulse. Here $\phi(\omega)$ is the overall phase delay introduced by the system and ω_0 is the center frequency of the amplified femtosecond pulses.

Several devices including Pockels cells [6], saturable absorbers [7], and specifically designed pulse cleaners [8] have been used to improve the contrast of high-energy subpicosecond pulses generated by CPA systems. More recently, improved high-order dispersion control resulted in the generation of 32-fs, 25-TW pulses with an intensity contrast of better than 10^{-5} [9]. In this paper, we demonstrate that *precise control of third-order dispersion allows steepening of the leading edge of high-contrast* 25-fs *femtosecond pulses over a high dynamic range*. Intense pulses with a steep leading edge may benefit several applications in high-field physics. A key to controlling the leading edge of 20-fs-scale pulses over many orders of magnitude is a precise dispersion control up to and including fourth order, i.e. controlling $\phi''(\omega_0)$, $\phi'''(\omega_0)$, and $\phi''''(\omega_0)$, in the system. In this communication we demonstrate, as a result of this capability, the generation of 1.5-mJ, 25-fs, kHz-repetition-rate laser pulses, whose intensity increases by five orders of magnitude within a time interval as short as 300 fs and a reduction of this "rise time" to less than 200 fs by nearly loss-free reflection of the pulses off chirped dielectric mirrors exhibiting nonzero TOD. Introducing a well-controlled amount of broadband TOD can steepen the temporal pedestal caused by spectral phase errors in the wings of the amplified pulse spectrum.

The Ti:sapphire (Ti:S) oscillator–amplifier system characterized in this work is shown in Fig. 1. It is an improved version of the system described in detail in [10]. The previously demonstrated 8-pass confocal Ti:S amplifier has been modified to accommodate a 9th pass with a slightly expanded beam for efficient gain saturation across the entire pumped volume. This improved amplifier seeded with a MDC sub-10 fs Ti:S oscillator (FemtoSource Pro, FemtoLasers GmbH) is capable of boosting the pulse energy up to 2 mJ and routinely generates pulses of around 1.5 mJ on a day-to-day

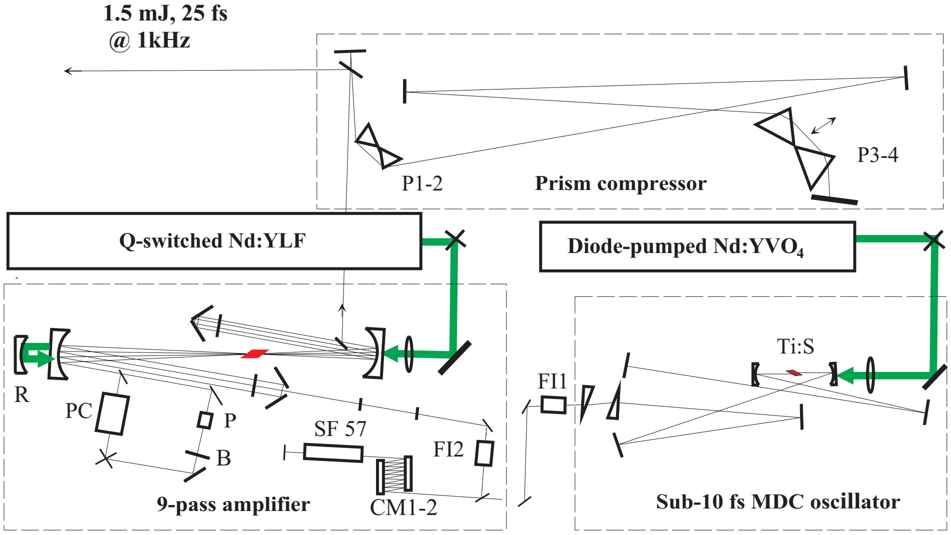

Fig. 1. Schematic of the laser system. FI1, FI2, Faraday isolators; CM1-2, chirped mirrors for cubic and quartic phase control; PC, Pockels cell; B, Berek's (polarization) controller; P, Polarizer; R, reflector for the residual pump beam transmitted through the amplifier crystal; P1–4, Brewster-angled fused silica prisms

basis when pumped with ≈ 12-mJ pulses from a cw-lamp-pumped Q-switched intracavity-frequency-doubled Nd:YLF laser (Model 621D, Thomson CSF Laser) at a 1 kHz repetition rate. Pulse stretching before amplification is accomplished by a comparatively small amount of dispersion introduced by a 10-cm-long block of heavy flintglass (SF 57, Schott) and other system components (e.g. Faraday isolators, Pockels cell, and polarizers) to a duration of ≈ 20 ps, allowing compression by a high-throughput (> 90%) dispersive delay line consisting of Brewster-angled fused silica prisms [10]. Pulses at the 1-kHz repetition rate of the pump laser are selected after the full 80 MHz train delivered by the oscillator has been passed four times through the amplifier. As a consequence, the Pockels cell also suppresses amplified spontaneous emission (ASE) emerging from the preamplification process, resulting in an ASE-to-amplified-pulse energy contrast of better than 2×10^{-3} [10]. Broadband phase control up to fourth order is accomplished with special chirped mirrors exhibiting positive third-order and fourth-order dispersion [10]. The group delay error can be kept below 5 fs over the wavelength range 750–850 nm with these mirrors.

For a complete, amplitude and chirp, characterization of the output over a limited dynamic range ($\approx 10^2$), we have used a frequency-resolved optical gating (FROG) [11] based on noncollinear second-harmonic generation (SHG) in a 25-μm BBO crystal. The adverse effects of temporal smearing due to the finite beam size in the noncollinear configuration [12] and the finite phase matching bandwidth have been found to be negligible for pulse durations ≥ 20 fs owing to a small beam crossing angle and to the thin nonlinear crystal, respectively. Figure 2 depicts the spectral and temporal intensities as well as phases of the amplified pulses as retrieved from the measured SHG-FROG trace. Figure 2a also shows the measured spectrum (dotted line), which compares well with the retrieved one. The nearly constant spectral phase over a substantial fraction of the laser spectrum (Fig. 2a) indicates the excellent high-order dispersion control in the system. The small temporal wings (Fig. 2b) originate from the rapid variation of the spectral phase in the wings of the spectrum. This off-center spectral phase error is assumed, in turn, to be a consequence of self-phase modulation (SPM) in the amplifier. In fact, the B-integral has been estimated to be of the order of 1 radian. Kane et al. [13] have shown that the effect of SPM acting on a strongly chirped pulse is to impart a *frequency-dependent* phase and, consequently, can be dispersively compensated. As a matter of fact, our dispersion control up to fourth order is capable of compensating the

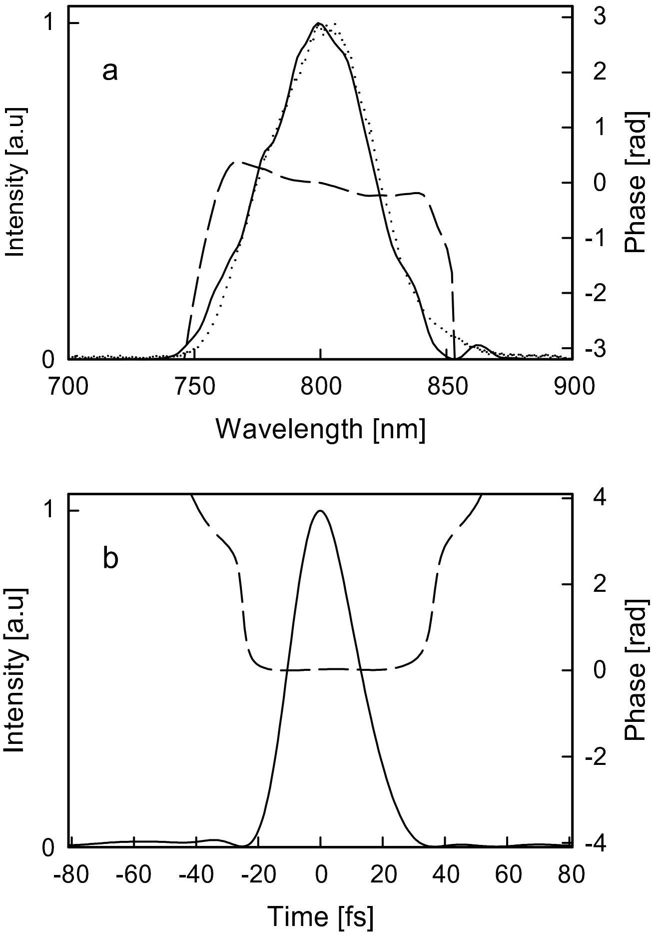

Fig. 2. Spectral (**a**) and temporal (**b**) intensity (*full lines*) and phase (*dashed lines*) as retrieved from SHG-FROG measurements. The *dotted line* depicts the measured spectrum of the amplified pulses

SPM-induced spectral phase over a substantial fraction of the spectrum. However, compensation of the SPM-induced phase in the wings would require even higher-orders in the Taylor expansion of the phase, which cannot be controlled in our present system.

For high-dynamic-range measurements another, somewhat modified, noncollinear autocorrelator has been constructed. Both the crossing angle and the spot size of the beams impinging on the nonlinear medium have been increased in order to reduce undesirable stray light contributions to the correlation signal and to enhance the correlation signal (limited by damage to the nonlinear medium), respectively. The two beams have been gently focused (f-number ≈ 100) onto the back surface of a 0.25-mm-thick microscope cover glass plate and generate surface-enhanced third-harmonic radiation [14–16] (see Fig. 3). The time-averaged third-harmonic signal yields the third-order autocorrelation function $G^{(3)}(\tau) = \int_{-\infty}^{\infty} I^2(t) I(t-\tau)\,\mathrm{d}t$, which, in contrast to $G^{(2)}(\tau)$, reveals possible asymmetries in the pulse shape.

To further suppress stray-light contributions to the signal, i.e. to improve the signal-to-noise ratio, a sum-frequency lock-in technique has been employed. A dual-frequency wheel chops the two beams at frequencies of f_1 and f_2, respectively. The third harmonic signal detected by the photomultiplier is then selectively amplified within a narrow frequency interval around $f_1 + f_2$. This technique [3, 17] results in an efficient suppression of (scattered) third-harmonic signal produced by the individual beams on their own, because these contributions are modulated at either f_1 or f_2. Using $\approx 1\%$ of the amplified pulse energy (i.e. an energy of $\approx 15\,\mu\text{J}$), this system provides a dynamic range of $\approx 10^5$. The delay τ can be varied by a retroreflector (RR in Fig. 3) mounted on a stepping-motor-controlled translational stage. Scattered fundamental laser light is prevented from entering the photomultiplier by multiple reflections off dichroic mirrors exhibiting a high reflectivity at the third harmonic only.

The third-harmonic autocorrelation trace of the laser output is shown by the full line in Fig. 4a. The symmetric, nearly exponential intensity envelope over a dynamic range of five orders of magnitude provides evidence for the powerful high-order dispersion control in the multipass amplifier. The intensity rises by five orders of magnitude within 300 fs on the leading edge and falls within 300 fs by the same factor on the trailing edge. In many high-field interactions, a high contrast and short "switching" time is important only on the leading edge, prompting the question of whether this can be improved by controlling only the first few terms in the Taylor expansion of the frequency-dependent phase delay of the system. In what follows, we demonstrate that the leading edge of nearly bandwidth-limited amplified femtosecond pulses imposed by a phase error of fifth or higher order can be made significantly steeper over an intensity range of (at least) five orders of magnitude by adding an appropriate amount of *positive* TOD to the phase delay of the system.

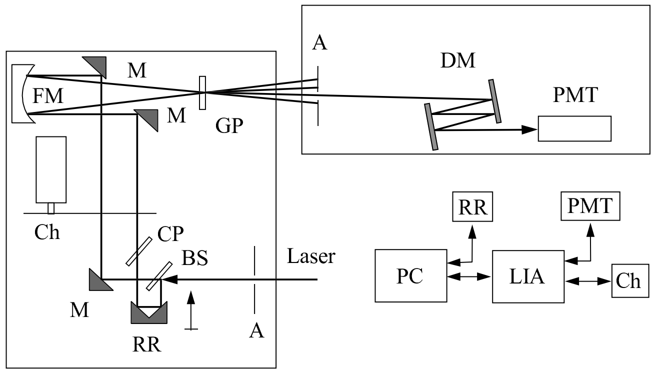

Fig. 3. Schematic of the high-dynamic-range third-harmonic-generation autocorrelator. A, aperture; BS, beam splitter; RR, retroreflector; M, mirror; CP, compensation plate; Ch, chopper; FM, focusing mirror; GP, glass plate; DM, dichroic mirror; PMT, photomultiplier tube; LIA, lock-in amplifier; PC, personal computer

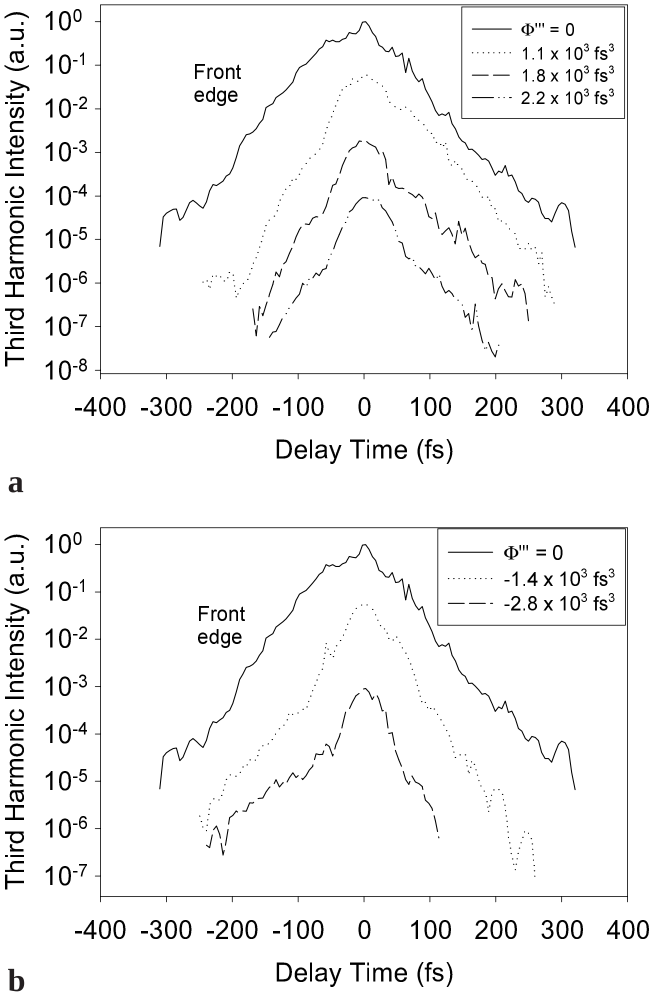

Fig. 4a,b. Third-harmonic autocorrelation traces of the 25-fs pulses delivered by the amplifier (*full lines*) and reflected several times off **a** positive-TOD and **b** negative-TOD chirped mirrors

TOD has been imposed on the 25-fs pulses by multiple bounces off a pair of chirped multilayer mirrors, with a high reflectivity over a bandwidth that well exceeds the pulse spectral width. The chirped mirrors introduce a TOD of $\approx 160\,\text{fs}^3$ (per bounce) that is approximately constant over the wavelength range 740–840 nm. Figure 4a depicts the third-harmonic autocorrelation traces obtained for different numbers of reflections off the TOD mirrors. The dotted and dashed lines in Fig. 4a indicate that in the range of 7–11 reflections, i.e. for a positive TOD in the range of approximately 1100–1800 fs^3, the front edge of our 25-fs pulses can be steepened over five orders of magnitude without significant change in the pulse duration (< 2 fs), resulting in a rise time of 200 fs over this dynamic range. This improvement comes at the expense of an enhanced temporal extension of the pulse tail. Figure 4b reveals that, as anticipated, the effect can be reversed by applying TOD of opposite sign (introduced, once again, with chirped mirrors having $\phi'''_{\mathrm{cm}} = -280\,\mathrm{fs}^3$).

The physical origin of this phenomenon is straightforward and can be understood with the assistance of Fig. 5, which shows the correlation traces calculated for a 25-fs, sech^2-shaped pulse in the absence of a frequency-dependent spectral phase $\phi(\omega)$ (thin full line) and for $\phi(\omega) = (1/6)\phi'''(\omega-\omega_0)^3$ with $\phi''' = 1100\,\mathrm{fs}^3$ (thin dashed line) and the corresponding measured traces without additional TOD (thick full line) and with a positive TOD of $\phi''' = 1100\,\mathrm{fs}^3$ imposed on the amplified pulses (thick dashed line). Whereas the applied TOD does not change the pulse envelope appreciably at intensities more than an order of magnitude below the peak intensity, experimentally we observe a steepening of the front edge.

This paradox can be resolved as follows. Dispersion in the amplifier cannot be compensated to all orders, resulting in a significant broadening of the pulse tails at low intensity levels, as revealed by Fig. 5. The residual (uncompensated) high-order dispersion (fifth and higher orders in our case) imposes the largest phase errors on spectral components far from the center of the pulse spectrum (ω_0), implying that these off-center frequency components provide the dominant contribution to the broadening of the low-intensity pulse tails.

Positive TOD imposes a group delay increasing quadratically with $(\omega-\omega_0)$, i.e. towards the spectral tails of the pulse. As a consequence, off-center frequency components, which carry most of the energy at low intensity levels far off the temporal pulse center according to the above considerations, are rearranged and shifted to the trailing edge of the pulse due to positive TOD, resulting in a steepening of the front edge over a high dynamic range. In other words, elimination of the off-center spectral components from the pulse front allows the actual pulse intensity envelope to follow more closely the analytic one on the leading edge of the pulse.

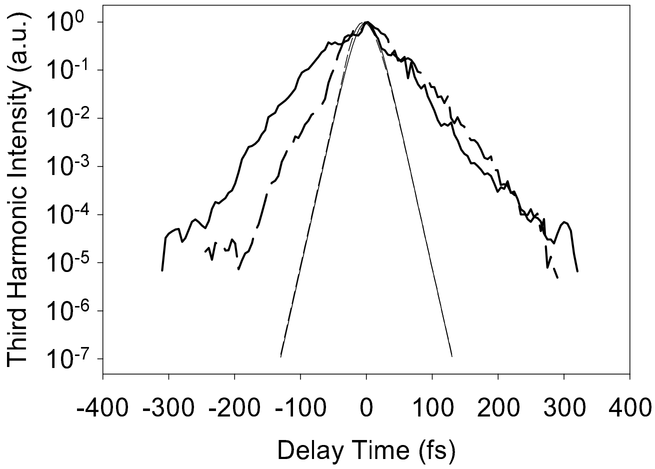

Fig. 5. Third-harmonic autocorrelation traces of a 25-fs sech^2-shaped pulse with no spectral phase (*thin full line*) with a TOD of $\phi''' = 1100\,\mathrm{fs}^2$ imposed (*thin dashed line*) and the corresponding measured traces (*thick full* and *dashed lines*, respectively)

In conclusion, we have investigated 1.5-mJ, 25-fs pulses from a Ti:sapphire multipass amplifier system using a high-dynamic-range third-harmonic autocorrelator. Owing to a precise high-order dispersion control up to and including fourth-order dispersion, the system delivers high-quality pulses with a symmetric intensity envelope over a range of five orders of magnitude with rise and fall times of 300 fs. These rise and fall times are limited by uncompensated dispersion of fifth and higher order. Adding an appropriate amount of *positive* third-order dispersion has been shown to shorten the rise time to 200 fs. Because dispersion in CPA systems can never be compensated to all orders, this simple manipulation is expected to be generally applicable for improving prepulse contrast in advanced femtosecond CPA systems with efficient high-order dispersion control and ASE suppression.

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