Few-cycle Pulses Amplification For Attosecond Science Applications Modeling And Experiments

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FEW-CYCLE PULSES AMPLIFICATION
FOR ATTOSECOND SCIENCE
APPLICATIONS: MODELING AND EXPERIMENTS

by

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ABSTRACT

The emergence of mode-locked oscillators providing pulses with durations as short as a few electric-field cycles in the near infra-red has paved the way toward electric-field sensitive physics experiments. In addition, the control of the relative phase between the carrier and the pulse envelope, developed in the early 2000’s and rewarded by a Nobel price in 2005, now provides unprecedented control over the pulse behaviour. The amplification of such pulses to the millijoule level has been an on-going task in a few world-class laboratories and has triggered the dawn of attoscience, the science of events happening on an attosecond timescale.

This work describes the theoretical aspects, modeling and experimental implementation of HERACLES, the Laser Plasma Laboratory optical parametric chirped pulse amplifier (OPCPA) designed to deliver amplified carrier-envelope phase stabilized 8-fs pulses with energy beyond 1 mJ at repetition rates up to 10 kHz at 800 nm central wavelength. The design of the hybrid fiber/solid-state amplifier line delivering 85-ps pulses with energy up to 10 mJ at repetition rates in the multi-kHz regime tailored for pumping the optical parametric amplifier stages is presented. The novel stretcher/compressor design of HERACLES, suitable for handling optical pulses with spectra exceeding 300 nm of bandwidth with unprecedented flexibility, is fully modeled and also presented in the frame of this thesis. Finally, a 3D model of the multi-stage non-collinear optical parametric amplifier is also reported.

The current and foreseen overall performances of HERACLES are presented. This facility is designed to enable attosecond physics experiments, high-harmonic generation and physics of plasma studies.
To my family...
# TABLE OF CONTENTS

LIST OF FIGURES ........................................................................................................................................ viii
LIST OF TABLES .......................................................................................................................................... xvi
LIST OF ABBREVIATIONS ................................................................................................................................ xviii
1 INTRODUCTION ...................................................................................................................................... 1
  1.1 Attoscience, a new field of physics ................................................................................................. 1
  1.2 Motivation of the thesis .................................................................................................................... 2
  1.3 Focus and objectives of the thesis ................................................................................................... 3
  1.4 Thesis outline .................................................................................................................................. 4
2 FROM CPA TO OPCPA: THE PLACE OF HERACLES IN 30 YEARS OF LASER DEVELOPMENT ...................................................................................................................................................... 6
  2.1 Chirped-Pulse Amplification: amplification to high peak powers .................................................. 6
     2.1.1 Providing positive and negative chirp: prerequisite to CPA techniques ................................ 6
     2.1.2 Invention of CPA and consequences ......................................................................................... 9
     2.1.3 The limits of CPA technologies ............................................................................................... 12
  2.2 Optical Parametric Chirped-Pulse Amplification ........................................................................... 14
  2.3 The place of HERACLES, OPCPA system at LPL ........................................................................ 15
3 OPTICAL PARAMETRIC AMPLIFIER PUMP BEAM GENERATION: MODEL AND SIMULATIONS ........................................................................................................................................................................ 17
  3.1 Requirements on HERACLES pump beam parameters ............................................................... 17
  3.2 Multi-stage architecture of the HERACLES pump beam amplifier ............................................. 19
  3.3 Physical properties of Neodymum doped laser media .................................................................. 20
  3.4 Regenerative amplifier modeling and parametric study ............................................................... 21
     3.4.1 Principle of operation ............................................................................................................... 22
     3.4.2 Regenerative amplifier model ................................................................................................ 23
     3.4.3 Parametric investigation of regenerative amplifier performances ......................................... 24
     3.4.4 Cavity design and stability ....................................................................................................... 39
     3.4.5 Intra-cavity volume Bragg grating ............................................................................................ 42
  3.5 Post-amplifier modeling and parametric study .............................................................................. 47
     3.5.1 Post-amplifier model ............................................................................................................... 47
     3.5.2 Parametric investigation of post-amplifier performances ....................................................... 47
  3.6 Multi-pass booster amplifier ........................................................................................................... 50
3.6.1 Post-amplifier model .................................................................50
3.6.2 Parametric investigation of booster amplifier performances ...50
3.6.3 Depolarization effects .............................................................54
3.6.4 Multi-pass booster amplifier layout model .............................57
3.7 Conclusion and recommendation for HERACLES amplifier chain 59

4 ULTRA-BROADBAND PULSE STRETCHING AND COMPRESSING: MODELING AND IMPLEMENTATION .................................................................61
4.1 Limitations of traditional stretcher/compressor assemblies 61
4.2 Strategy for efficient pulse compression.................................62
4.3 Traditional pulse chirping techniques: model ........................ 63
4.3.1 Spectral phase: definition and convention ........................ 64
4.3.2 Material dispersion and angular dispersion ....................... 66
4.3.3 Brewster prism pair ..............................................................70
4.3.4 Treacy grating pair ............................................................76
4.4 Solutions for efficient stretching and compressing of ultra-broadband pulses: model and prospects ........................................................80
4.4.1 Grism pairs semi-analytical model ......................................83
4.4.2 Acousto-optic programmable dispersion filter ....................94
4.4.3 Chirped mirrors .................................................................97
4.5 The HERACLES stretcher compressor assembly .................104
4.5.1 Hybrid grism-grating stretcher ..........................................104
4.5.2 Bulk glass pulse compression ............................................113

5 NON-COLLINEAR OPTICAL PARAMETRIC AMPLIFICATION: THEORY AND SIMULATIONS .................................................................123
5.1 Theory of optical parametric amplification ............................123
5.1.1 Nonlinear polarization and introduction of coupled wave-equations ....124
5.1.2 Analytical solutions of the coupled wave equations ............127
5.2 Broadband phase-matching..................................................134
5.2.1 Broadband phase-matching: conceptual approach ............134
5.2.2 Broadband phase-matching: mathematical description ......137
5.3 Phase evolution of the optical parametric process ................140
5.4 Numerical modeling of a broadband non-collinear optical parametric amplifier 142
5.4.1 Broadband phase-matching and non-collinear geometry .......143
5.4.2 Introduction of the model ..................................................148
LIST OF FIGURES

Figure 1: Layout of the HERACLES facility. ................................................................. 4
Figure 2: Layout of a grating compressor as proposed by Treacy ............................... 7
Figure 3: a. Layout and principle of operation of the grating pair assembly providing positive dispersion as proposed by Martinez [45] ............................................................................................................. 8
Figure 4: Comparison of the volume gain media required to store a given amount of energy (to scale), saturation fluence and excited state lifetime of a dye and several solid-state gain media [55].......................................................................................................................... 9
Figure 5: Principle of the chirped-pulse amplification technique proposed by Mourou et al. ......10
Figure 6: Evolution of optical pulses peak power from the invention of lasers to today [55].......12
Figure 7: Layout of the HERACLES hybrid amplifier chain featuring fiber and solid-state amplifiers stages. ............................................................................................................ 19
Figure 8: Operation principles of a regenerative amplifier in the time domain. A period of low intra-cavity losses during which the seeded pulses are amplified is followed by a period of high intra-cavity losses in which lasing is prevented while inversion in the crystal is re-established [158]. ......................................................................................................................... 22
Figure 9: Output energy performances and number of round trips required to reach saturation for a regenerative amplifier featuring either Nd:YAG, Nd:YLF or Nd:YVO4 as a gain medium for intra-cavity losses of 40% (a and b), 20% (c and d) or 10% (e and f). Calculations are performed for a 2 mm diameter rod at 1 kHz repetition rate with 5 pJ seed energy. .................................................. 27
Figure 10: Output energy performances and number of round trips required to reach saturation for a regenerative amplifier featuring either Nd:YAG, Nd:YLF or Nd:YVO4 as a gain medium for rod diameters of 2 mm (a and b), 3 mm (c and d) or 4 mm (e and f). Calculations are performed for at 1 kHz repetition rate with 5 pJ seed energy with 40% intra-cavity losses. ..........30
Figure 11: Output energy performances and number of round trips required to reach saturation for a regenerative amplifier featuring either Nd:YAG, Nd:YLF or Nd:YVO4 as a gain medium for seed energies of 5 pJ (a and b), 50 pJ (c and d) or 500 pJ (e and f). Calculations are performed for a 2 mm diameter rod at 1 kHz repetition rate with 40% intra-cavity losses. ............33
Figure 12: Output energy as a function of number of round trip of ejection for a Nd:YAG-based regenerative amplifier operated at 1 kHz (a), 2 kHz (b), 5 kHz (c) and 10 kHz (d). .........................35
Figure 13: Output energy as a function of number of round trip of ejection for a Nd:YVO4-based regenerative amplifier operated at 1 kHz (a), 2 kHz (b), 5 kHz (c) and 10 kHz (d). .........................36
Figure 14: Phase diagram showing the gain remaining after pulse ejection as a function of output energy for Nd:YVO4 regenerative amplifier operated at repetition rate below 5 kHz (a) and at 10 kHz (b).................................................................37
Figure 15: Layout of the oscillator as modeled by LASCAD – the rod provides the cavity stability via thermal lensing.

Figure 16: Mode volume diameter as a function of position in the cavity of a Nd:YVO₄-based regenerative amplifier for an 85 cm long cavity (a) and a 95 cm long cavity (b).

Figure 17: Schematic view of a volume Bragg grating illustrating constructive interferences at the Bragg angle.

Figure 18: Simulated influence of ASE on the output spectrum of a Nd:YVO₄ regenerative amplifier with after 1 pass in the cavity (a and b) and 11 passes in the cavity (c and d).

Figure 19: Simulated output spectra from a YVO₄ regenerative amplifier without intra-cavity VBG (a) an equipped with a VBG (inset) and corresponding output pulse durations (b).

Figure 20: Output energy performances as a function of the number of passes and the pump power for a multi-pass amplifier featuring either Nd:YVO₄ or Nd:YAG as a gain medium for rod diameter of 5 mm (a and b), 7 mm (c and d) and 10 mm (e and f). Calculations are performed for a 12.5 cm long rod at 1 kHz repetition rate with 2 mJ seed energy.

Figure 21: Output energy performances as a function of the number of passes and pump power for a multi-pass amplifier featuring either Nd:YVO₄ or Nd:YAG as a gain medium for seed energy of 1 mJ (a and b), 2 mJ (c and d) and 3 mJ (e and f). Calculations are performed for a 7 mm diameter, 12.5 cm long rod at 1 kHz repetition rate.

Figure 22: Computed refractive index profile for a 150 W thermal load on a 10 mm diameter YAG rod. The index of refraction variation $\Delta n$ is computed with respect to the index of refraction at the edge of the rod.

Figure 23: Computed spatial output profile resulting from the interaction of a 12.5 cm long YAG rod and a polarizer under (a) 10 W heat load and resulting $\Delta n = 5.5 \times 10^{-6}$, (b) 50 W heat load and resulting $\Delta n = 2.7 \times 10^{-5}$, (c) 100 W heat load and resulting $\Delta n = 5.5 \times 10^{-5}$ and (d) 150 W heat load and resulting $\Delta n = 8.1 \times 10^{-5}$.

Figure 24: Computed depolarization induced losses based on [177] for a 12.5 cm long, 10 mm diameter YAG rod.

Figure 25: Layout of the multi-pass booster amplifier with sketched mode volume.

Figure 26: Beam size along multi-pass amplifier cavity for (a) a 130 cm long cavity at 200 W and 300 W pump power, (b) a 90 cm long cavity at 300 and 450 W pump power.

Figure 27: Low loss stretching and compressing strategy used on HERACLES relying on down-chirp in the stretcher and up-chirp in the compressor.

Figure 28: Schematic representation of the convention used for computing the spectral phase. $\delta$ represents the optical path difference between the k-vector $k_1$ and $k_2$ form which the spectral phase is inferred.
Figure 29: Prism pair assembly with apex angle $\alpha$, incident angle $\theta_i$, diffracted angle $\theta_d$ and optical path experienced by the center wavelength $d$ ..........................................................70

Figure 30: a. GDD and TOD for a center wavelength $\lambda_0 = 850$ nm for a fused silica prism pair operated at Brewster’s angle as a function of apex to apex separation; b. GDD and TOD for a center wavelength $\lambda_0 = 850$ nm for an SF10 prism pair operated at Brewster’s angle as a function of apex to apex separations ..........................................................75

Figure 31: a. Variation of the GDD and TOD for a center wavelength $\lambda_0 = 850$ nm as a function of depth of insertion in the second prism of the pair for a fused silica prism pair; b. Variation of the GDD and TOD for a center wavelength $\lambda_0 = 850$ nm as a function of depth of insertion in the second prism of the pair for an SF10 prism pair ..........................................................76

Figure 32: Treacy grating pair assembly with incident angle $\theta_i$, diffracted angle $\theta_d$ and optical path experienced by the center wavelength $d$ ...........................................................................77

Figure 33: a. GDD and TOD/GDD ratio as a function of groove density for a grating separation of 20 cm and an incident angle $\theta_i = 20^\circ$; b. GDD and TOD/GDD ratio as a function of grating to grating distance for a 300 l/mm groove density and an incident angle $\theta_i = 20^\circ$; c. GDD and TOD/GDD ratio as a function of incident angle for a 300 l/mm groove density and a grating separation of 20 cm ..........................................................79

Figure 34: a. Effects of 100 fs$^2$ residual GDD (blue curve) on a transform-limited pulse (black curve) featuring 100 nm FWHM spectral bandwidth; b. Effects of 100 fs$^3$ residual TOD (blue curve) on a transform-limited pulse (black curve) featuring 100 nm FWHM spectral bandwidth; c. Effects of 100 fs$^2$ residual GDD (blue curve) on a transform-limited pulse (black curve) featuring 200 nm FWHM spectral bandwidth; b. Effects of 100 fs$^3$ residual TOD (blue curve) on a transform-limited pulse (black curve) featuring 200 nm FWHM spectral bandwidth ..........81

Figure 35: Layout of the modeled grism pair with associated notations used in the derivation ....84

Figure 36: Phase-matching diagram of the interaction between a tailored acoustic wave and an optical wave in an acousto-optic programmable dispersion filter ...............................................................................96

Figure 37: Principle of operation of an acousto-optic programmable dispersion filter: wavelengths making up the input chirped pulse are switched from the fast-axis to the slow-axis of the birefringent crystal allowing the introduction of an arbitrary spectral phase upon the input pulse to cancel out the initial chirp [198] ........................................................................................................96

Figure 38: Representation of a chirped mirror showing the depth-dependent reflectivity of the mirror [205] .................................................................................................................................97

Figure 39. Evolution of the GDD in a Gires Tournois Interferometer [157] .........................100

Figure 40. GTI effect due to front reflection between air and mirror [205] ............................101

Figure 41. Effects of the air mirror interface on the GD and GDD over broad bandwidth ......102

Figure 42. Comparison between a chirped mirror and a double chirped mirror [208] ..........103
Figure 43: a. Group delay (set to zero at the center wavelength of 850 nm) produced by a 1.5 cm effective BBO crystal (blue line) and a 4.5 cm TeO\textsubscript{2} crystal (black line) normalized to the center wavelength $\lambda_0 = 850$ nm; b. GDD produced by a 1.5 cm effective BBO crystal (blue line) and a 4.5 cm TeO\textsubscript{2} crystal (black line); c. TOD by a 1.5 cm effective BBO crystal (blue line) and a 4.5 cm TeO\textsubscript{2} crystal (black line); d. FOD produced by a 1.5 cm effective BBO crystal (blue line) and a 4.5 cm TeO\textsubscript{2} crystal (black line).

Figure 44: a. GDD and TOD behavior for a grating and grism pair adjusted such that the total GDD provided by both pairs equal -40000 fs\textsuperscript{2} for a range of grism separation from 0.5 to 6 cm; b. TOD/GDD ratio of a grating and grism pair adjusted such that the total GDD provided by both pairs equal -40000 fs\textsuperscript{2} for a range of grism separation from 0.5 to 6 cm.

Figure 45: a. Group delay (set to zero at the center wavelength of 850 nm) and GDD provided by an SF11 grism pair featuring 300 l/mm gratings and a 5.4 cm grism separation; b. TOD and FOD provided by an SF11 grism pair featuring 300 l/mm gratings and a 5.4 cm grism separation.

Figure 46: a. Group delay (normalized to the center wavelength) and GDD provided by a 300 l/mm grating pair and a 17.8 cm grating separation; b. TOD and FOD provided by a 300 l/mm grating pair and a 17.8 cm grating separation.

Figure 47: Evolution of the TOD/GDD ratio over a 300 nm spectral band ranging from 700 to 1000 nm for a grating pair, a grism and grism/grating pair combination showing the superior behavior of the combination.

Figure 48: Quasi-Gaussian temporal profile of the stretched pulse after experiencing propagation through the grism pair, grating pair, AOPDF and OPA stages.

Figure 49: Residual group delay, set to zero at the center wavelength of 850 nm, after propagation through the stretcher and compressor assemblies.

Figure 50: a. Residual group delay of the modeled HERACLES stretcher/compressor assembly; b. residual GDD of the modeled HERACLES stretcher/compressor assembly; c. residual TOD of the modeled HERACLES stretcher/compressor assembly; d. residual FOD of the modeled HERACLES stretcher/compressor assembly.

Figure 51: a. Evolution of the pulse duration upon propagation in the SF57 glass compressor; b. evolution of the B-integral value upon propagation in the SF57 glass compressor for a 1 cm diameter beam (black line) and a 5 cm diameter beam (blue line).

Figure 52: Output energy as a function of nonlinear medium length ($z$) for a gain $g = 2000$, a signal seed energy of 20 pJ, an idler seed level of 1 fJ and a pump energy of 500 $\mu$J represented on a logarithmic scale.

Figure 53: Schematic of the temporal walk-off between an idler and signal wave during a collinear nonlinear interaction.

Figure 54: Schematic of the non-collinear geometry allowing compensation of the temporal walk-off between idler and signal wave during nonlinear interaction.
Figure 55: Effects of beam size correlated with non collinear geometry on the spatial overlap and consequently on the parametric gain ................................................................. 136

Figure 56: Schematic of the arrangement used in a non-collinear optical parametric amplifiers. The notation used for the derivation of the phase-matching condition are gathered on the schematic .................................................................................................................. 137

Figure 57: Evolution of the ordinary and extraordinary refractive index of BBO as a function of wavelength [215] ......................................................................................................................... 144

Figure 58: Alternative non-collinear configurations featuring the signal wave-vector either between the pump wave-vector and the optical axis (TPM configuration) or the idler wave-vector between the pump wave-vector and the optical axis (PCWC configuration) ...................... 145

Figure 59: Simulations of the effects of phase-matching angle tuning (a, $\theta = 23.83^\circ$, c, $\theta = 23.86^\circ$, e, $\theta = 23.89^\circ$) and non-collinear angle (b, $\alpha = 2.36^\circ$, d, $\alpha = 2.39^\circ$, f, $\alpha = 2.42^\circ$) tuning on the phase mismatch and gain spectral profile. ................................................................. 147

Figure 60: Schematic representation of the strategy employed to solve the partial differential equations ruling the optical parametric amplification process in space, time and spectral domains ........................................................................................................................................................................ 148

Figure 61: Comparison of the spectral gain profile for the case when the signal wave-vector is between the pump wave-vector and the optical axis (TPM configuration – black line) and the case when the idler wave-vector is between the pump wave-vector and the optical axis (PVWC configuration – blue line). ........................................................................................................................................................................ 150

Figure 62: Simulation of the pump and signal beam propagating through the nonlinear crystal for (a,c) $\alpha = -2.36^\circ$, (b,d) $\alpha = +2.36^\circ$ showing a better overlap between the pump and signal in PVWC (right column) configuration than TPM configuration (left column).................................................................................................................. 151

Figure 63: Effects of time delay between the pump and signal pulses on the spectral gain profile for -100 ps (a), -50 ps (c), 0 ps (e), 50 ps (g) and 100 ps (i) delays and effects of time delay between the pump and signal pulses on the amplified spectrum (dark line) for -100 ps (b), -50 ps (d), 0 ps (f), 50 ps (h) and 100 ps (j) delays compared to the seeded spectrum (grey area). .......... 153

Figure 64: a. Fraction of the seeded bandwidth being amplified as a function of ratio between the pump pulse and signal pulse duration; b. corresponding time domain FWHM pulse duration for the extreme cases of $\tau_s/\tau_p = 0.2$ and $\tau_s/\tau_p = 1.1$ ........................................................................................................................................ 155

Figure 65: Pump depletion for 20 pJ (red line), 200 pJ (green line), 2 nJ (black line) and 20 nJ seed energy ......................................................................................................................................................... 156

Figure 66: a. Effects of nonlinear crystal length on spectral gain profile and bandwidth; b. effects of nonlinear crystal length on parametric gain shown on a logarithmic scale (blue curve) and linear curve (black curve). The linear curve has been normalized to the maximum gain. .......... 157

Figure 67: a. Amplified spectrum under two-color pumping (532 and 355 nm) conditions (blue line) compared to seed spectrum (gray shaded region); b. comparison of minimum achievable
pulse duration between single-color pumping at 532 nm (black curve) and two-color pumping at 532 nm and 355 nm (blue curve). ................................................................. 159

Figure 68: Layout of the sub 6 fs Ti:Sapphire oscillator ................................................................. 162

Figure 69: Measured output spectrum of the Ti:Sapphire oscillator on a log scale ............... 163

Figure 70: Measured interferometric autocorrelation trace showing the 6 fs pulse produced by the Octavius oscillator ............................................................................................................................ 164

Figure 71: Measured output profile at the output of the Octavius oscillator ............................... 165

Figure 72: Left column – evolution of temperature and power as a function of time for 3.5W pump power, $T_{\text{crystal}} = 14 \degree \text{C}$ and $T_{\text{base plate}} = 25\degree \text{C}$ in non-mode locked regime, right column – evolution of temperature and power as a function of time for 3.5W pump power, $T_{\text{crystal}} = 14 \degree \text{C}$ and $T_{\text{Base Plate}} = 25\degree \text{C}$ in mode locked regime .................................................. 168

Figure 73: Left column – evolution of temperature and power as a function of time for 3.5W pump power, $T_{\text{crystal}} = 14 \degree \text{C}$ and $T_{\text{base plate}} = 25\degree \text{C}$ in non-mode locked regime, right column – evolution of temperature and power as a function of time for 5.5W pump power, $T_{\text{crystal}} = 14 \degree \text{C}$ and $T_{\text{Base Plate}} = 25\degree \text{C}$ in non-mode locked regime .............................................................................. 169

Figure 74: Layout of the f-2f interferometer used to stabilize the carrier-envelope phase of the Octavius oscillator ............................................................................................................................ 172

Figure 75: Measured error signal between the beat signal and the quarter of the reference signal with feedback loop enabled (left) and disabled (right) ................................................................................................................................. 173

Figure 76 : Influence of the seed energy level on the output signal to ASE ratio for high gain amplifier [218]. ........................................................................................................................................... 174

Figure 77: Cross-section of the polarization maintaining Yb fiber used in the regenerative amplifier. The 5 $\mu$m core appears between the two large stress rods ensuring horizontally polarized output. ............................................................................................................................... 176

Figure 78: Layout of the fiber pre-amplifier showing the two 15 m long identical stages pumped by 750 mW single emitter diodes and separated by an 8 nm wide spectral filter. ...................... 177

Figure 79: a. Average output power provided by the first (blue triangles) and second (black squares) stages of the fiber pre-amplifier; b. output spectra measured for a pump power of the first stage 0 W (black dotted line) – 45 mW (red long dashed line) – 110 mW (blue dashed-dotted line) – 300 mW and beyond (pink solid line). .................................................................................................................... 178

Figure 80: Autocorrelation trace of the output pulses of the fiber pre-amplifier. ...................... 178

Figure 81: Layout of the HERACLES regenerative amplifier seed injection and regenerative amplifier. ................................................................................................................................. 180

Figure 82: a. Gain medium of the HERACLES regenerative amplifier; (b) image of the fluorescence distribution obtained at full power from the pump module. .............................................. 180
Figure 83: Transmissivity of the VBG used in the HERACLES regenerative amplifier (source: specification sheet from Optigate Corp.)

Figure 84: Characteristics output energy versus input power (a) and corresponding number of round trips required to reach saturation at 1 kHz repetition rate (black squares), 2 kHz (red circles), 5 kHz (blue upward triangles) and 10 kHz (pink downward triangles).

Figure 85: a. Characteristics output energy versus input pump power with an intra-cavity VBG (black squares) or a R = 40% output coupler (red circles) for a modified regenerative amplifier design enhancing the regeneration of ASE; b. output spectrum from the HERACLES regenerative amplifier operated with a VBG or a R = 40% output coupler (inset).

Figure 86: a. Autocorrelation trace of the seed pulse (red dashed line) and output pulse (black solid line) of the HERACLES regenerative amplifier; b. Energy in second harmonic as a function of fundamental energy (black squares) and corresponding SHG efficiency curve (blue triangle).

Figure 87: Beam profile at the output of the HERACLES regenerative amplifier at 1 kHz (upper left hand corner), 2 kHz (upper right hand corner), 5 kHz (lower left hand corner) and 10 kHz (lower right hand corner).

Figure 88: a. Layout of the single pass amplifier; b. image of the end of the Nd:YVO4 rod showing the seed pulse centered onto the rod.

Figure 89: Characteristics output energy versus pump power at 1 kHz (a), 2 kHz (c) and 5 kHz (e) and output energy in the second harmonic as a function of amplifier pump power at 1 kHz (b), 2 kHz (d) and 5 kHz (f) for various IR seed energies.

Figure 90: a. Measurement of the pointing stability over 1000 shots on a CCD camera; b. beam profile in the far field measured by relay imaging the end of the amplifier rod.

Figure 91: a. Picture of one of the grisms of the HERACLES stretcher; b. Picture of the compact layout of both the grating pair stretcher and grism pair stretcher. This layout enabled stretching ratio of up to 10000.

Figure 92: Measured throughput of the grism stretcher with 7 cm separation distance (blue line) compared to the input spectrum (black line).

Figure 93: a,b. Measured autocorellation of a pulse stretch in grism/grating pair assembly with a grism separation of 4 cm and grating separation of 14.5 cm (blue line) and corresponding computed autocorellation trace (black line), and corresponding measured spectrum (blue line) and computed spectral phase c,d. Measured autocorellation of a pulse stretch in grism/grating pair assembly with a grism separation of 6 cm and grating separation of 14.5 cm (blue line) and corresponding computed autocorellation trace (black line), and corresponding measured spectrum (blue line) and computed spectral phase Optical parametric amplifier performances; e,f. Measured autocorellation of a pulse stretch in grism/grating pair assembly with a grism separation of 7 cm and grating separation of 14.5 cm (blue line) and corresponding computed autocorellation trace (black line), and corresponding measured spectrum (blue line) and
computed spectral phase Optical parametric amplifier performances; .......... \textbf{Error! Bookmark not defined.}

Figure 94: Representation of the spectrum as a frequency comb and highlighting the role of $f_{CEO}$
## LIST OF TABLES

Table 1: Limitations in pulse duration and maximum extractable energy of CPA system [43]....13
Table 2: Typical physical parameters for Nd:YAG, Nd:YLF and Nd:YVO4 laser materials......20
Table 3: Summary of pump power and number of round trip required to reach the damage threshold of the rod for Nd:YAG, Nd:YVO4 and Nd:YLF for intra-cavity losses of 10%, 20% and 40%.................................................................28
Table 4: Comparison of the power distribution in continuous wave and Q-switched mode for the regenerative amplifier cavity. ........................................................................................................41
Table 5: Output energy as a function of rod diameter for a single and double pass amplifier featuring either Nd:YVO4 or Nd:YAG as a gain medium. Computations assumed 500 µJ seed pulses and 180 W pump power ......................................................48
Table 6: Output energy as a function of seed energy for a single and double pass amplifier featuring either Nd:YVO4 or Nd:YAG as a gain medium. Computations assumed a 2 mm rod and 180 W pump power. ........................................................................................................49
Table 7: computations of GDD, TOD and FOD for fused silica grism pairs with an apex angle $\alpha = 23.2^\circ$ and featuring 300 l/mm, 400 l/mm and 500 l/mm gratings and adjusted to provide 35000 fs$^2$ of GDD...........................................................................................................................................90
Table 8: computations of GDD, TOD and FOD for a fused silica grism pairs featuring 300 l/mm gratings with apex angles of $\alpha = 23.2, 23.5$ and 23.8° and adjusted to provide 35000 fs$^2$ of GDD .................................................................................................................................................91
Table 9: computations of GDD, TOD and FOD for grism pairs featuring 300 l/mm, made of fused silica, SF11 and SF58 glass and adjusted to provide 35000 fs$^2$ of GDD .................................................................92
Table 10: computations of GDD, TOD and FOD for an SF11 grism pairs featuring 300 l/mm gratings and adjusted to provide 35000 fs$^2$ of GDD assuming alignment error of the input beam ....................................................................................................................93
Table 11: computations of GDD, TOD and FOD for an SF11 grism pairs featuring 300 l/mm gratings and adjusted to provide 35000 fs$^2$ of GDD assuming slight misalignment of the parallelism of the grisms .................................................................94
Table 12: Total amount of dispersion provided by an SF11 grism pair featuring 300 l/mm gratings separated by 5 cm, a grating pair featuring 300 l/mm gratings separated by 13.8 cm, a 4.5 cm long TeO2 piece modeling an AOPDF and a 1.5 cm BBO crystal modeling three OPA stages. ................................................................................................................................................114
Table 13: Total amount of dispersion provided by an SF11 grism pair featuring 300 l/mm gratings separated by 5 cm, a grating pair featuring 300 l/mm gratings separated by 17.8 cm, a
4.5 cm long TeO$_2$ piece modeling an AOPDF and a 1.5 cm BBO crystal modeling three OPA stages. ................................................................................................................................................ 115

Table 14: Dispersive properties of fused silica, SF11 and SF59 glass............................................. 117

Table 15: Computed required glass length to compensate the GDD introduced by a 5 cm grism pair, a 17.8 cm grating pair, a 4.5 cm TeO$_2$ crystal and a 1.5 cm effective BBO crystal assuming a fused silica, SF11 or SF59 glass compressor. Residual TOD and FOD upon matching GDD are also computed ................................................................................................................................... 117

Table 16: Residual GDD, TOD and FOD when a 21.2 cm long SF57 compressor is used to compensate the dispersion induced by a grism pair with 5 cm separation, a grating pair with 17.8 cm separation, a 4.5 cm long TeO$_2$ AOPDF and an effective 1.5 cm long BBO crystal.........118
<table>
<thead>
<tr>
<th>Acronym</th>
<th>Abbreviation</th>
</tr>
</thead>
<tbody>
<tr>
<td>AR</td>
<td>Anti-reflection</td>
</tr>
<tr>
<td>ASE</td>
<td>Amplified Spontaneous Emission</td>
</tr>
<tr>
<td>BBO</td>
<td>Beta Baryum Oxyde</td>
</tr>
<tr>
<td>CEP</td>
<td>Carrier-Envelope Phase</td>
</tr>
<tr>
<td>CPA</td>
<td>Chirped-Pulse Amplification</td>
</tr>
<tr>
<td>FWHM</td>
<td>Full Width at Half Maximum</td>
</tr>
<tr>
<td>FOD</td>
<td>Fourth Order Dispersion</td>
</tr>
<tr>
<td>GD</td>
<td>Group Delay</td>
</tr>
<tr>
<td>GDD</td>
<td>Group Delay Dispersion</td>
</tr>
<tr>
<td>HERACLES</td>
<td>High Energy Repetition Rate Adjustable Carrier Locked to Envelope System</td>
</tr>
<tr>
<td>HHG</td>
<td>High-Harmonic Generation</td>
</tr>
<tr>
<td>HR</td>
<td>High Reflector</td>
</tr>
<tr>
<td>LPL</td>
<td>Laser Plasma Laboratory</td>
</tr>
<tr>
<td>Nd</td>
<td>Neodymium</td>
</tr>
<tr>
<td>NOPCPA</td>
<td>Non-collinear Optical Parametric Chirped Pulse Amplifier</td>
</tr>
<tr>
<td>OPA</td>
<td>Optical Parametric Amplifier</td>
</tr>
<tr>
<td>OPCPA</td>
<td>Optical Parametric Chirped-Pulse Amplifier</td>
</tr>
<tr>
<td>PTR</td>
<td>Photo-Thermo Refractive – a type of holographic glass</td>
</tr>
<tr>
<td>SFG</td>
<td>Sum-Frequency Generation</td>
</tr>
<tr>
<td>SHG</td>
<td>Second-Harmonic Generation</td>
</tr>
<tr>
<td>SPM</td>
<td>Self Phase Modulation</td>
</tr>
<tr>
<td>TEM</td>
<td>Transverse Electro-Magnetic</td>
</tr>
<tr>
<td>TFP</td>
<td>Thin Film Polarizer</td>
</tr>
<tr>
<td>Ti</td>
<td>Titanium</td>
</tr>
<tr>
<td>TOD</td>
<td>Third Order Dispersion</td>
</tr>
<tr>
<td>VBG</td>
<td>Volume Bragg Grating</td>
</tr>
<tr>
<td>YAG</td>
<td>Yttrium Aluminium Garnet</td>
</tr>
<tr>
<td>YLF</td>
<td>Yttrium Lithium Fluoride</td>
</tr>
<tr>
<td>YVO₄</td>
<td>Yttrium Vanadate</td>
</tr>
<tr>
<td>Yb</td>
<td>Ytterbium</td>
</tr>
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</table>
1 INTRODUCTION

At the time of its invention, 50 years ago, the laser was called “a solution looking for a problem”. Fifty years later lasers have enabled breakthroughs in science that were unthinkable at the time. Further developments are continuously being made to push the limits of laser systems and enable more exciting fundamental and applied physics research. The present work follows this dynamics consisting of the development of a novel, state of the art laser light source designed to enable physics experiments at the attosecond timescale.

1.1 Attoscience, a new field of physics

Investigating physical processes that occur on a sub-femtosecond timescale can only be performed if an optical pulse with duration shorter than the event under consideration is available. Such optical pulses have recently been made available via the high-harmonic generation process described by Corkum in 1993 [1]. In this paper, Corkum proposed a semi-classical three-step model that provides a simple explanation to the work performed a few years earlier by Mainfray et al. [2-4] where odd high-harmonics of the fundamental driving wavelength were observed when a high peak power optical pulse was being used to ionize a rare gas.

In the three step model, Corkum suggests that upon ionization under a linearly polarized optical electric field, an electron is freed and accelerated before recombining with its parent ion and releasing its excess kinetic energy in the form of a photon with an energy higher than that of photon absorbed in the initial ionization. This process occurs for every cycle of the electric field
under the pulse envelope as long as the field is strong enough to ionize the gas and results in photons with higher energy for electrons recombining after a few round trips around their parent ions. This swinging of the electron around its ion results in the odd high-harmonics observed by Mainfray et al. Such optical combs are of interest to some research but are impractical since they do not allow the generation of single isolated attosecond pulses but rather, a train of attosecond bursts [5].

The main breakthrough occurred in 2006 when Sansone et al. implemented a polarization gate that reduced the duration of the driving NIR pulses from a few tens of cycles to quasi-single cycle [6-8]. The reduction of the number of cycles on the generation of high-harmonics resulted in a single-cycle ionization process, therefore suppressing the electron swinging motion in the high-harmonic generation process yielding the emission of a broad, coherent EUV spectrum. This broad spectrum translated in the temporal domain to the generation of an isolated attosecond pulse synchronized with the driving field. The technique was further refined in 2008 by Goulielmakis et al.[9] by directly generating and amplifying optical pulses with few-cycle duration. This improvement allowed removing the optical gate and therefore use the full energy contained in the pulse.

1.2 Motivation of the thesis

The construction of the High Energy Repetition-rate Adjustable Carrier-Locked to Envelope System (HERACLES) at the Laser Plasma Laboratory (LPL) enables the group to take part in attosecond research and follows the group’s philosophy of building one-of-a-kind laser systems to investigate physical processes in regimes that are left unexplored by commercially available systems.
Gaining access to the few-cycle range with sufficient energy to generate plasmas, filaments or optical damages in solids offers a significant broadening of the current research programs on-going within LPL. In addition to broadening the scope of current research projects, HERACLES is designed to enable both high-harmonic generation and single isolated attosecond pulses production.

1.3 Focus and objectives of the thesis

The focus of this thesis is on the design, modeling and building of HERACLES. The output parameters were chosen to enable as wide of range of experiments as possible.

- The pulse duration was set to ~8 fs to enable the generation of isolated attosecond pulses directly from the laser system without resorting to optical gating techniques. The pulses can still be gated to reach the true single-cycle regime but the toll on available energy is limited thanks to the initial short duration of the pulses.

- The system is designed to operate in a wide range of repetition rates ranging from single-shot to 10 kHz with minor adjustments to the system required to switch from one regime to the other. This tunability enables a wide range of experiments. The choice of kilohertz repetition rate operation is dictated by the need of large photon flux in EUV when using HERACLES for high-harmonic generation.

- The energy is set to a ~0.5-2 mJ, depending on the repetition rate of operation. This energy is limited by the energy that can be stored in the pump pulse of the parametric amplifier. Nevertheless, HERACLES is designed such that, if further developments in
solid-state laser technology enable energy scaling of the pump beam, the output energy of the facility could be scaled up as well.

The design can be separated in several stages as shown in Figure 1. A front-end carrier-envelope phase stabilized, octave-spanning Ti:Sapphire oscillator is used to seed both the optical parametric amplifier stages after passing through a custom-made stretcher and an amplifier chain designed to provide the pump beam for the optical parametric amplifiers. The three parametric amplifier stages are pumped with increasing pump energy to overcome saturation and noise related issues. Finally the amplified broadband pulses are recompressed.

Figure 1: Layout of the HERACLES facility.

1.4 Thesis outline

In order to motivate the work performed in this thesis an overview of ultrafast technology over the past 30 years showing the move toward OPCPAs for the amplification of few-cycle pulses is presented in chapter 2. The rest of the thesis follows the organization of the laser itself, each chapter being devoted to the modeling of a building block of the system as highlighted by the dashed lines in Figure 1, experimental results being gathered in a separate chapter. In chapter
a simple and general approach is introduced that allows modeling of the entire pump beam generation amplifier line. Each stage of this amplifier line is simulated in a parametric and comparative study providing an in-depth laser design analysis covering a wide range of experimental parameters. Chapter 4 is devoted to the modeling of the stretcher/compressor assembly of HERACLES. An analysis of the traditional techniques is proposed to introduce, in two simple cases, the numerical techniques implemented when modeling dispersive lines. The analysis is then carried to model the more advanced grism/Treacy design introduced in HERACLES. Simulation results are presented in a parametric study and experimental design parameters are deduced from this analysis. Chapter 5 provides the theoretical background of optical parametric amplification and a classical analytic solution to the equations ruling the process. The limitations of these analytical solutions are presented and a numerical model is proposed that offers temporal, spatial and spectral description as a superior alternative to analytical approaches to model the HERACLES optical parametric amplifier stages. Simulations are performed to address major experimental difficulties. Finally, experimental results showing the performances of HERACLES are gathered in chapter 6. The thesis is concluded by a discussion of the future evolutions of HERACLES and the capabilities offered by this new laser facility.
2.1 Chirped-Pulse Amplification: amplification to high peak powers

2.1.1 Providing positive and negative chirp: prerequisite to CPA techniques

A few years after the invention of the laser by Schawlow and Townes [10] and the first demonstration of laser action by Maiman [11, 12], pulses as short as a few picoseconds were within reach from mode-locked He-Ne, Ruby and Nd:Glass lasers [13-22]. It was found that the pulses generated from these systems were not transform-limited [23, 24] and such pulse broadening was attributed to frequency chirping, or temporal spreading of the frequency components of the optical pulse. Schemes were proposed to compensate the spectral chirp and ensure that all wavelengths within the pulse were localized at the same point in time. Schemes inspired by microwave technology relying on Doppler shifting [25], schemes involving frequency dependent Bragg diffraction [26, 27] and techniques featuring Gires-Tournois interferometers were successfully implemented [28-30]. Notice that the technique relying on Bragg diffraction is nowadays employed in chirped fiber Bragg gratings [31, 32] and chirped volume Bragg gratings [33] while the Gires-Tournois compressor that has been implemented in mirror coatings as an alternative to chirped mirrors.

The introduction by Treacy of a grating based compressor [34-37] surpassed all previous designs and is one of the preferred embodiments to provide negative chirp to an optical pulse. The design of a Treacy compressor is shown in Figure 2.
Figure 2: Layout of a grating compressor as proposed by Treacy

The assembly consists of a pair of identical gratings facing each other parallel. Diffraction of the incident pulse from the first grating creates a path to the second grating with a frequency-dependent optical path length. Upon diffraction on the second grating the angular spreading provided by the first grating is cancelled out. A retro-reflector mirror is typically placed after the second grating at a tilt angle, sending the pulse for a second path into the setup. An analytical description of these assemblies is provided in section 4.3.4. These compressors were first implemented in the early 1980’s for compression of self-phase modulated (SPM) pulses to durations shorter than 30 fs [38-41]. The energy of these pulses was limited by the amount of energy that could be passed into the single mode fibers used to broaden the spectrum via SPM.

In 1984, Martinez proposed a prism pair compressor operating similarly to the grating pair proposed by Treacy 15 years earlier but relying on refraction rather than diffraction [42]. Several general analysis of negative chirp provided by angular dispersion were provided by Martinez et al., applied to the case of prism pairs and grating pairs, investigating the effects of beam size and divergence [42-44].
The development of pulse compressors providing negative chirp in the visible and near infrared spectral region was directly driven by the quest for high peak power via pulse shortening. Providing positive chirp was of little interest to this field and it was only in 1984 that an assembly providing positive chirp was proposed by Martinez [43] and fully described in 1987 [45]. These assemblies were designed to compensate for the negative chirp accumulated by optical pulses in the 1.3 to 1.6 µm spectral region when propagating in telecommunication optical fibers. The layout of the assembly, commonly referred to as Martinez stretcher, is shown in Figure 3.

![Figure 3: a. Layout and principle of operation of the grating pair assembly providing positive dispersion as proposed by Martinez [45]](image)

The difference between the Treacy compressor and the Martinez stretcher is the presence of a 4f system between the two gratings in the Martinez assembly. Positive chirp is obtained by placing the gratings within the focal distance of the two lenses. The development of both Treacy compressor and Martinez stretcher provided unprecedented control over the temporal and spectral behavior of optical pulses and led to the invention of the chirped-pulse amplification technique.
2.1.2 Invention of CPA and consequences

In the early 1980s the peak power limits set by damages of optical surfaces and detrimental nonlinear effects had been reached in the nanosecond [46], picosecond [47] and femtosecond regime [41]. In order to produce pulses with peak power greater than \(~1 \text{ GW cm}^{-2}\), the clear aperture of the gain media had to be increased, resulting in increased costs and added complexity. Liquid or gas gain media offer high damage thresholds and low nonlinearities [48-51]; however these media have low saturation fluences. Solid-state gain media, though suffering from lower damage threshold compared to liquids or gases, remained theoretically the ideal gain media for amplification of optical pulses due to high saturation fluence and high energy storage capability in a reduced volume [52-54]. Figure 4 compares the saturation fluence of a dye amplifying medium with that of a few solid-state gain media [55].

Figure 4: Comparison of the volume gain media required to store a given amount of energy (to scale), saturation fluence and excited state lifetime of a dye and several solid-state gain media [55]

Figure 4 also illustrates the size of a gain medium required to store the same amount of energy in a dye gain medium versus five solid-state gain media. Dye amplifier can handle optical pulses with high peak power but their poor energy storage capability makes them unable to
amplify to high energies while solid-state lasers can amplify pulses to high energy but cannot handle the resulting peak power.

This dilemma was solved in 1985 by Strickland and Mourou who amplified short pulses to unprecedented levels of energy by using the chirped-pulse amplification technique [56, 57]. The principle of the technique is shown in Figure 5 [55].

![Figure 5: Principle of the chirped-pulse amplification technique proposed by Mourou et al.](image)

A short pulse generated by an oscillator is temporally stretched via spectral chirp then amplified and recompressed by cancelling out the spectral phase induced by the stretcher. The first demonstration of CPA produced pulses with 2 ps duration and millijoule level energy at 1.06 µm wavelength [56, 57].

The early implementations of CPA relied on Nd:Glass as a gain medium and used fiber-based stretchers to both spread the pulse and broaden its spectrum via SPM [58]. These Nd:Glass systems enabled the generation of pulses with several hundred millijoules energy in picosecond duration, the pulse duration being limited by the inherent mismatch between the fiber stretcher and the grating compressor [59]. Engineering of the Martinez stretcher used in conjunction with a fiber stretcher and a Treacy compressor allowed spectral shaping of the pulses leading to a record pulse duration of 750 fs with 2.4 J output energy from an Nd:Glass amplified system [60]. Energies up to 20 J in 2.4 ps were later reported for an Nd:Glass CPA system [61].
The invention of CPA was coincident with new developments in laser materials. In the mid 1980s, the development of rare-earth (Er, Yb, Tm) doped materials [62-64], the development of chromium doped materials [65-74], and the development of Ti:Sapphire [75-80] led to the generation of pulses with unprecedented spectral width spanning several tens of nanometers – and corresponding pulse duration in the sub-100 fs regime, generated directly from oscillators. The availability of gain media with broad emission bandwidth allowed removing fiber stretchers since SPM was not longer needed and shorter pulse duration could be achieved by using gratings pair for both stretching and compressing.

The CPA techniques originally developed for Nd:Glass [81] were successively adapted for use with Alexandrite [82, 83], Ti:Sapphire [78, 84-86] and Cr:LiSAF [86-91] resulting in optical pulses with peak powers beyond the TeraWatt level. Since its invention in 1985, the CPA technique has been further refined [92-95] and expanded to other architectures such as fiber-based systems or thin-disk systems [96-100]. Figure 6 summarizes the evolution of achieved peak power over the years.
The availability of pulses with short duration and high energy has opened the way for a wide range of applications spanning from pump probe experiments [101-103], material processing [104, 105], plasma physics [106] and all the way to the possibility of laser-induced nuclear fusion [107, 108].

2.1.3 Limits of CPA technologies

After over 25 years of improvements, none of the theoretical limits of CPA have been approached, neither in term of achievable pulse duration nor in terms of extracted energy. Mourou suggests taking as a measure of minimum achievable pulse duration $\tau$ the spectral bandwidth ($\Delta\lambda$) of the gain medium and as a measure of maximum extractable energy the saturation fluence and gathers this data in Table 1 [55].
Table 1: Limitations in pulse duration and maximum extractable energy of CPA system [55]

<table>
<thead>
<tr>
<th>Material</th>
<th>Saturation fluence</th>
<th>$\Delta\lambda$ (nm)</th>
<th>$\tau$ (fs)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nd:Glass phosphate</td>
<td>5</td>
<td>22</td>
<td>80</td>
</tr>
<tr>
<td>Nd:Glass silicate</td>
<td>10</td>
<td>28</td>
<td>60</td>
</tr>
<tr>
<td>Ti:Sapphire</td>
<td>0.8</td>
<td>200</td>
<td>5</td>
</tr>
<tr>
<td>Alexandrite</td>
<td>28</td>
<td>100</td>
<td>10</td>
</tr>
<tr>
<td>Cr:LiSAF</td>
<td>8</td>
<td>50</td>
<td>15</td>
</tr>
<tr>
<td>Yb:Silica</td>
<td>40</td>
<td>200</td>
<td>8</td>
</tr>
</tbody>
</table>

The introduction of chirped mirrors and pairs of double-chirped mirrors (see section 4.4.3 for a description of chirped mirrors) in Ti:Sapphire have enabled the generation of output spectra as wide as an octave directly from the oscillator [109, 110]. Similar developments in Cr:LiSAF-based oscillators have led to pulse energies as high as 100 nJ directly from the oscillator and pulses as short as 12 fs [111, 112]. Nevertheless, maintaining such short pulse durations in CPA amplifier systems is a goal currently out of reach due to spectral narrowing, inherent to stimulated emission-based processes. Schemes have been proposed to address this issue [113, 114] but cannot be scaled to systems designed to reach the saturation fluence of systems featuring large aperture amplifiers. Spectral narrowing is currently the main limitation in preventing peak power scaling in CPA systems.
2.2 Optical Parametric Chirped-Pulse Amplification

Optical parametric amplification has been described experimentally [115, 116] and theoretically [117-121] since the early days of lasers. It has been extensively used in the 1990s, since the invention of CPA techniques, to produce optical pulses in the nanosecond [122], picosecond [123] and femtosecond regimes [124] at wavelengths ranging from 200 nm to 18 µm [125]. In the late 1990s it was found that implementing optical parametric amplification in non-collinear geometries [126-132] increased phase-matchable bandwidths and could permit amplification of pulses with duration shorter than 20 fs. Phase-matching over spectral bandwidth exceeding $\Delta \lambda = 300$ nm at 800 nm center wavelength and beyond $\Delta \lambda = 500$ nm at 2 µm center wavelength has been reported [133, 134], therefore exceeding the capabilities of CPA systems in terms of minimum amplified pulse duration. Combining the concept of CPA with optical parametric amplification allows suppression of gain narrowing limitations, reducing detrimental effects of nonlinear phase acquired in amplifiers and greatly simplifies thermal management of the amplifiers. These advantages have enabled the emergence of ultrafast systems operating in the sub-10 fs regime producing high energy pulses [135, 136], at high repetition rate [137] at various wavelength [133, 138, 139] employing various architectures [140]. The emergence of these systems in the mid-2000s has triggered the dawn of attoscience [5, 7, 141-145] and enabled probing atoms and molecules at the electronic time scale [8, 146, 147] opening up new extremely active fields of research.
2.3 The place of HERACLES, OPCPA system at LPL

The High Energy Repetition-rate Adjustable Carrier-Locked-to-Envelope System (HERACLES) is the Laser Plasma Laboratory OPCPA facility. It is designed to enable a broad range of experiments ranging from high harmonic generation (HHG), attosecond science, plasma studies, filamentation, spectroscopy and material processing. The facility is designed to be relatively hands-off and yet flexible to enable such variety of experiments. The HERACLES was designed as part of this thesis in the fall of 2007 at a time when only a few OPCPA systems had been reported worldwide. Several novelties were considered in the original design, including cryogenically-cooled amplifiers and thin-disk amplifiers to provide energetic pump pulses for the optical parametric amplifiers. These techniques have not been utilized in the HERACLES design but have been implemented – or are about to be – in OPCPA facilities [148, 149].

The modular design of HERACLES makes the facility readily upgradable. To illustrate the modularity of the design and its upgradability, a beam line operating at 1 Hz repetition rate and high energy, non-part of the original design, has recently been added to the original kHz beam line while the system was being developed. Similar developments are expected in the future to expand the capabilities of the system, in particular the addition of a beam line delivering few-cycle, carrier-envelope phase stabilized pulses in the 2 \( \mu \text{m} \) spectral region.

The development of HERACLES has also led to a number of innovations and achievements such as the invention of a novel stretcher design enabling unprecedented flexibility and control for stretching pulses with spectra spanning over 300 nm of bandwidth [150, 151]. Volume Bragg gratings (VBG) have been implemented for the first time in a picosecond regenerative amplifier [152] in the frame of this thesis. The assembly of HERACLES also led the current record of continuous wave power on a single-longitudinal mode from a Nd:YVO\(_4\) laser
[153] and record energy in the Q-switch regime from the same oscillator [154]. Finally, the delivery of 1 J, 20 ns pulses featuring spectra as narrow as a ~20 pm at 1 Hz repetition rate [155] has been performed as a test bed for the future upgrade of the LPL Terawatt facility.
3 OPTICAL PARAMETRIC AMPLIFIER PUMP BEAM GENERATION: MODEL AND SIMULATIONS

This chapter provides a model of the amplifier line generating the pump beam of the HERACLES optical parametric amplifier. The energy, repetition rate, beam profile and stability requirements on the pump beam are first presented. The multi-stage architecture implemented on HERACLES permitting it to reach the required output parameters is presented. Subsequently, each stage of the amplifier line is modeled via comparative and parametric studies.

3.1 Requirements on HERACLES pump beam parameters

The strong single pass gain of optical parametric amplifiers, as high as $10^6$, results in a strong imprint of the pump beam properties onto the output beam and will be discussed in chapter 5. Thus, one of the challenges of an OPCPA design lies in producing a pump beam with excellent optical properties. General requirements placed on OPCPA pump beams and specifics of the HERACLES are discussed below [138].

- **Pulses with 85 ps duration are required.** The constraints on pulse duration are dictated by the need of an optimized temporal overlap between the pump and signal pulses in the optical parametric amplifiers. This pulse duration is chosen as a trade-off between the minimum stretching factor of the signal pulse – ensuring accurate recompression – and the maximum pump pulse duration – limiting the peak power in the amplifier stages.

- **Synchronization between pump and signal beam must be jitter free.** With the pump pulse duration being set to 85 ps, its synchronization with the signal pulse must be achieved with picosecond accuracy. The HERACLES design relies on optical
synchronization: the pump pulse and signal pulse are both derived from a master Ti:Sapphire oscillator ensuring consistency of any jitter and allowing sub-picosecond accuracy synchronization.

- **Pump pulses with 15 mJ of energy at 1064 nm are required.** The energy transfer between the pump pulse at the fundamental wavelength and the final output of an optical parametric amplifier is typically ~10%. In order to ensure that the output of HERACLES reaches energies beyond 1 mJ, the pump pulses are required to reach energies beyond 10 mJ at 1064 nm.

- **Repetition rates in the kilohertz regime.** HERACLES is designed to provide a large of photon flux. This is achieved by providing both high energy and high repetition rate.

- **Excellent beam profile is required.** Providing an excellent beam profile is a major constraint for successful optical parametric amplification. A poor beam quality of the pump results in limited efficiency in the optical parametric amplification process and transfer of the spatial defects to the signal beam.

- **The footprint of the amplifier needs to be limited.** HERACLES is designed to fit on a 3 m long and 1.5 m wide single optical table. The choice of building the laser system on a single table is dictated by the need to limit vibrations and the potential of isolating the whole system from vibrations by floating the optical table. It was therefore necessary that each stage be designed with as reduced a footprint as possible.
3.2 Multi-stage architecture of the HERACLES pump beam amplifier

The pump beam of the HERACLES optical parametric amplifier is generated in a hybrid amplifier line that gathers advantages of fiber and solid-state technologies. The amplifier chain operates at a center wavelength of 1064 nm, enabling the use of mature GaAs diode pumping, common gain media such as Nd:YAG and Nd:YVO₄ and well-established optical components such as volume Bragg gratings. Figure 7 shows a schematic of the amplifier line.

![Figure 7: Layout of the HERACLES hybrid amplifier chain featuring fiber and solid-state amplifiers stages](image)

The seed for the amplifier line is derived from the master Ti:Sapphire oscillator. A section of the ultra-broadband spectrum centered at 1064 nm is seeded into a two stage fiber pre-amplifier. The output of the pre-amplifier is then seeded into a Nd:YVO₄-based regenerative amplifier to reach ~700 µJ of energy (section 3.4). The pulses, spectrally and temporally tailored in the regenerative amplifier via an intra-cavity volume Bragg grating are sent into a Nd:YVO₄-based single pass amplifier that brings the output energy to 2 mJ (section 3.5). Finally a Nd:YAG-based amplifier boosts the energy up to 15 mJ (section 3.6).
3.3 Physical properties of Neodymium doped laser media

The operating wavelength of the HERACLES pump beam generation line is set to 1064 nm. Therefore, the choice of gain media is limited to Nd doped materials. Nd:YLF is included in the study, despite its center wavelength of 1053 nm, because optical components such as mirrors, coatings, isolators, pump diodes available for 1064 nm wavelength are readily extended to 1053 nm. The physical properties of each crystal are gathered in Table 2.

Table 2: Typical physical parameters for Nd:YAG, Nd:YLF and Nd:YVO4 laser materials.

<table>
<thead>
<tr>
<th>Materials</th>
<th>Nd:YAG</th>
<th>Nd:YLF</th>
<th>Nd:YVO4</th>
</tr>
</thead>
<tbody>
<tr>
<td>Emission wavelength (nm)</td>
<td>1064</td>
<td>1053</td>
<td>1064</td>
</tr>
<tr>
<td>Emission cross-section, $\sigma$ ($m^2$)</td>
<td>2.8x10^{-23}</td>
<td>1.2x10^{-23}</td>
<td>1.14x10^{-22}</td>
</tr>
<tr>
<td>Peak absorption cross-section ($m^2$)</td>
<td>7.7x10^{-24}</td>
<td>17x10^{-24}</td>
<td>60x10^{-24}</td>
</tr>
<tr>
<td>Excited state lifetime, $\tau$ (s)</td>
<td>230x10^{-6}</td>
<td>480x10^{-6}</td>
<td>90x10^{-6}</td>
</tr>
<tr>
<td>$\frac{\partial n}{\partial T}$ (K^{-1})</td>
<td>7.3x10^{-6}</td>
<td>-4.3x10^{-6}</td>
<td>3x10^{-6}</td>
</tr>
<tr>
<td>Thermal conductivity $\kappa$ (W.m^{-1}.K^{-1})</td>
<td>13</td>
<td>6</td>
<td>5.1</td>
</tr>
<tr>
<td>Saturation fluence $F_{sat}$ (J.m^{-2})</td>
<td>6.6x10^{3}</td>
<td>1.57x10^{4}</td>
<td>1.63x10^{3}</td>
</tr>
</tbody>
</table>

Nd:YLF and Nd:YAG exhibit small emission cross-section compared to Nd:YVO4 resulting in large saturation fluence $F_{sat}$ ($F_{sat} = h\nu/\sigma$) and reduced small signal gain $g_0$ ($g_0 = \sigma n$), where $n$ is the frequency at the signal wavelength. These materials exhibit a long excited state lifetime, consequently they are suitable for storing energy and providing a large amount of energy by multi-passing through the crystals. Nd:YVO4 offers a large emission cross-section, yielding a large small signal gain $g_0$. The large absorption cross-section of Nd:YVO4, makes it a
good candidate to operate at high repetition rate since population inversion can be restored rapidly. Nd:YAG and Nd:YLF on the other hand are suitable for power amplifiers where a large energy storage capability is required.

It is also necessary to consider the thermal properties of each of these materials to evaluate the magnitude of thermal lensing and depolarization. Thermal lensing is a major parameter to consider when designing a resonator while depolarization needs to be considered for energy extraction efficiency and beam quality purposes. A simple expression for the strength of the thermal lens (in diopters) is given by [156]:

\[
 f^{-1} = \frac{\partial n/\partial T}{2\kappa A} P_{\text{Heat}} 
\]

where \( A \) is the cross-section of the rod, \( \partial n/\partial T \) is the thermo-optic coefficient, \( \kappa \) is the thermal conductivity of the crystal and \( P_{\text{Heat}} \) is the dissipated heat [157]. The low thermal conductivity and low thermo-optic coefficient of YVO\(_4\) compared to YAG makes it a crystal much more prone to strong thermal lensing. YLF’s negative \( \partial n/\partial T \) produces a negative thermal lens. Unlike Nd:YAG, Nd:YVO\(_4\) and Nd:YLF are birefringent materials, having an emission cross-section larger on one crystal axis than the other, making them insensitive to depolarization, a typical side effect of heavily pumped gain media.

3.4 Regenerative amplifier modeling and parametric study

This section introduces the models developed for simulating the performance of the regenerative amplifier implemented in the HERACLES pump beam generation line. First, a comparative study between several Nd doped gain media is presented along with a parametric
study covering the main design parameters of the gain media. Then, the beneficial effects of spectral narrowing via volume Bragg gratings on ASE limitation and pulse tailoring in a picosecond regenerative amplifier are modeled. Finally, investigations of cavity designs are presented to provide a stable and efficient amplifier.

3.4.1 Principle of operation

The operation principles of a regenerative amplifier present similarities with a Q-switched laser. One period \( T \) consists of a low-Q state dominated by intra-cavity losses and a high-Q state where amplification takes place (Figure 8) [158].

![Figure 8: Operation principles of a regenerative amplifier in the time domain. A period of low intra-cavity losses during which the seeded pulses are amplified is followed by a period of high intra-cavity losses in which lasing is prevented while inversion in the crystal is re-established [158].](image)

In a typical regenerative amplifier operating at kilohertz repetition rates, the duration of the amplification period \( (T_{\text{high}}) \) is a few hundreds of nanoseconds while the duration of the pumping period with low-Q factor \( (T_{\text{low}}) \) is in the microsecond range (Figure 8).
3.4.2 Regenerative amplifier model

The high-Q and low-Q states of the regenerative amplifier are modeled separately since different physical processes are involved. The high-Q state is modeled by two coupled partial differential equations involving the density of photons \( \phi \) present in the cavity and the density of excited electrons (or population inversion) \( n \) in the gain medium.

\[
\frac{\partial n}{\partial t} = -nc\sigma \phi \\
\frac{\partial \phi}{\partial t} = nc\sigma \phi,
\]

where \( n \) is the population inversion per unit volume, \( c \) is the speed of light, \( \sigma \) is the emission cross-section and \( \phi \) is the photon flux in the cavity. From Eqn. (2) the inversion \( n \) is reduced proportionally to both the amount of photons \( \phi \) in the cavity and the amount of inversion achieved at the moment of interaction between the excited electrons and the photons in the pulse. Conversely from Eqn. (3), the photon flux \( \phi \) increases proportionally to the number of photons already present in the cavity for a given inversion \( n \). As the inversion \( n \) is reduced, the rate of increase in number of photons is reduced, leading to saturation.

The low-Q state is modeled by a single partial differential equation describing the evolution of the inversion over time (4).

\[
\frac{\partial n}{\partial t} = \frac{P_{\text{power}}}{E_{\text{Photon}} V} - \frac{n}{\tau},
\]

where \( P_{\text{power}} \) is the pump power in Watts, \( E_{\text{Photon}} \) is the energy of a pump photon in Joules, \( V \) is the volume of the rod in m\(^3\) and \( \tau \) is the excited state lifetime (in seconds). The evolution of the
inversion over time in the high-Q state results from the balance between the pumping rate, 
\[
\frac{P_{\text{Power}}}{E_{\text{Photon}} V},
\]
and the losses via stimulated emission, \(\frac{n}{\tau}\).

3.4.3 Parametric investigation of regenerative amplifier performance

3.4.3.1 Regenerative amplifier output requirements

A number of practical constraints on pulse duration, minimum extracted energy, repetition rate, beam profile and footprint are placed on the HERACLES regenerative amplifier. **The duration of the pulses** at the output of the regenerative amplifier should be \(\sim 85\) ps. The pulse duration of the pump pulse needs to match that of the signal pulse in the parametric amplifier. The duration of the pump pulse therefore results from a tradeoff between minimizing the stretching factor (to ensure successful compression) of the signal pulses and minimizing the peak power of pump pulse in the amplifier chain. This compromise resulted in duration of 85 ps being chosen for the HERACLES system. **The minimum energy that needs to be extracted** in the regenerative amplifier is governed by the amplification stages following the regenerative amplifier. In order to reach the final target energy of 15 mJ from the booster amplifier (3.2), a minimum of 500 \(\mu\)J needs to be extracted from the regenerative amplifier. **The repetition rate** of the regenerative amplifier controls the repetition rate of the entire system. The HERACLES system is designed to operate at repetition rates as high as 10 kHz, it is therefore necessary for the regenerative amplifier to successfully operate up to at least 10 kHz. **The constraints on beam profile and footprint** have been discussed in previous sections (3.1 and 6.2.1 respectively) and are identical for the regenerative amplifier.
The constraints on pulse duration, output energy and repetition rate can be addressed by the model previously described, these three properties being parameters of the model. Several design parameters have been investigated to meet the needs of the HERACLES regenerative amplifier:

- crystal material
- crystal size
- pump power
- seed energy level

The parametric study assumes continuous diode pumping as an energy source. This is dictated by the current limitations of diodes when switched at kilohertz repetition rates.

### 3.4.3.2 Influence of intra-cavity losses

The effect of intra-cavity and output coupling losses on both the output energy and the number of round trips required to reach saturation is investigated. The losses can arise from diffraction, clipping of the beam, depolarization and the value of output coupling. In the case of the HERACLES regenerative amplifier, large output coupling losses need to be considered due to the presence of the intra-cavity VBG ($R = 70\%$ over a 50 pm spectral line centered at 1064 nm). The influence of losses has been investigated for Nd:YAG, Nd:YLF and Nd:YVO$_4$ laser gain media. For this simulation (Figure 9), a 4 cm long, 2 mm diameter rod was chosen. The continuous pump power was increased from 40 W to 200 W and it was assumed that all the pump light was absorbed in the gain medium. The repetition rate of operation was 1 kHz and the seed energy was set to 5 pJ.
The output energy and number of round trips required to reach saturation are plotted versus pump power for Nd:YAG, Nd:YLF and Nd:YVO₄ crystals for cumulated intra-cavity and output coupling losses per round trip of 10% (Figure 9 a and b), 20% (Figure 9 c and d) and 40% (Figure 9 e and f). Maximum output energies of 15 mJ, 8.5 mJ and 3.5 mJ can be extracted from a cavity with 10% round trip losses featuring respectively Nd:YLF, Nd:YAG and Nd:YVO₄ as a gain medium. Comparatively, energies up 10 mJ, 6 mJ and 3 mJ are expected from an amplifier cavity with 40% round trip losses. Quadrupling the cavity losses results in an output energy loss of ~30% for Nd:YLF, ~25% for Nd:YAG and 15% for Nd:YVO₄. Due to their reduced small signal gain, Nd:YLF and Nd:YAG are more sensitive to additional cavity losses.

At low pump powers, the effects of the reduced single-pass gain in Nd:YLF and Nd:YAG leads to a dramatic increase in number of round trips needed for saturation to be reached. At a pump power of 40 W with cavity losses set to 40%, saturating a Nd:YLF or a Nd:YAG rod requires up to 250 and 110 round trips respectively while saturating a Nd:YVO₄ rod requires only 40 round trips. At high pump power, the differences between these three materials are less, but Nd:YVO₄ remains superior for rapid amplification due to its higher single pass gain.
Figure 9: Calculate output energy performances and number of round trips required to reach saturation for a regenerative amplifier featuring either Nd:YAG, Nd:YLF or Nd:YVO₄ as a gain medium for intra-cavity losses of 40% (a and b), 20% (c and d) or 10% (e and f). Calculations are performed for a 2 mm diameter rod at 1 kHz repetition rate with 5 pJ seed energy.
The higher energy extraction capabilities of Nd:YLF and Nd:YAG are practically limited by damage threshold considerations in the picosecond regime. The dielectric coating of optical components displays a damage threshold, in the picosecond regime, of ~10 GW.cm\(^{-2}\) and we define a maximum tolerable intensity that equals 10% of the damage threshold. This reduced damage threshold is used in the design process to prevent unexpected experimental failures caused by operation in the vicinity of the damage threshold. Table 3 summarizes the required pump power and number of round trips needed to reach the design-tolerated damage threshold for the three Nd-doped materials for 40%, 20% and 10% cavity losses.

<table>
<thead>
<tr>
<th></th>
<th>10% losses</th>
<th>20% losses</th>
<th>40% losses</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pump power (W)</td>
<td>Nd:YAG</td>
<td>Nd:YLF</td>
<td>Nd:YVO(_4)</td>
</tr>
<tr>
<td>Number of round trips</td>
<td>40</td>
<td>30</td>
<td>70</td>
</tr>
<tr>
<td></td>
<td>43</td>
<td>90</td>
<td>14</td>
</tr>
<tr>
<td>Pump power (W)</td>
<td>Nd:YAG</td>
<td>Nd:YLF</td>
<td>Nd:YVO(_4)</td>
</tr>
<tr>
<td>Number of round trips</td>
<td>50</td>
<td>45</td>
<td>80</td>
</tr>
<tr>
<td></td>
<td>39</td>
<td>70</td>
<td>13</td>
</tr>
<tr>
<td>Pump power (W)</td>
<td>Nd:YAG</td>
<td>Nd:YLF</td>
<td>Nd:YVO(_4)</td>
</tr>
<tr>
<td>Number of round trips</td>
<td>80</td>
<td>70</td>
<td>100</td>
</tr>
<tr>
<td></td>
<td>28</td>
<td>52</td>
<td>11</td>
</tr>
</tbody>
</table>

The output energy can be reduced to match the design-tolerated damage threshold by reducing the pump power. Pump powers as low as 30 W, 40 W and 70 W are required to reach that threshold for Nd:YLF, Nd:YAG and Nd:YVO\(_4\) amplifiers respectively. These required pump powers are doubled when the cavity losses are quadrupled. The pump power required to reach the design-tolerated damage threshold in a Nd:YVO\(_4\) amplifier is systematically 30 to 40 W higher than that required for Nd:YAG and Nd:YLF amplifiers (Table 3), making such designs slightly more demanding on pump diode requirements. Nevertheless, this increase in required pump power is balanced by a reduced number of round trips required to reach the damage threshold (about 80% and 60% less round trips required for a Nd:YVO\(_4\) amplifier compared to a
Nd:YLF and a Nd:YAG amplifier respectively). A smaller number of round trips is desirable, both to ensure a constant voltage on the Pockels cell and to limit the accumulation of nonlinear spatial phase that alters the beam profile.

3.4.3.3 Influence of crystal diameter

Reducing the intensity on the crystal to prevent damage can be achieved by reducing the pump power or by increasing the rod diameter. An increase in rod diameter results in a quadratic decrease of the peak intensity but also results in a decrease in inversion per unit volume for a given pump power. The density of excited electrons is reduced in a quadratic fashion as the rod diameter is increased, resulting in a quadratic decrease in small signal gain. The effects of increasing the crystal diameter on output energy and number of round trips required to reach saturation have been modeled for 4 cm long rods of Nd:YLF, Nd:YAG and Nd:YLF in a cavity with 40% round trip cavity losses, 5 pJ seed energy and operating at 1 kHz repetition rate (Figure 10).
Figure 10: Calculate output energy performances and number of round trips required to reach saturation for a regenerative amplifier featuring either Nd:YAG, Nd:YLF or Nd:YVO₄ as a gain medium for rod diameters of 2 mm (a and b), 3 mm (c and d) or 4 mm (e and f). Calculations are performed for at 1 kHz repetition rate with 5 pJ seed energy with 40% intra-cavity losses.
When the rod diameter for Nd:YLF, Nd:YAG and Nd:YVO₄ is doubled the output energy decreases by 90%, 85% and 30%, respectively. Such a decrease in output energy is attributed to the quadratic reduction of the available inversion $n$ and the increase in saturation energy, therefore requiring higher pump power to reach the saturation energy of the material. Simultaneously, the energy required to reach the design-tolerated damage threshold increases from 1 mJ to 2.4 mJ and 4.2 mJ for rods with 2 mm, 3 mm and 4 mm diameter (defined for a beam filling 75% of the rod section). Table 4 summarizes the pump power and number of round trips required to reach the damage threshold.

Table 4: Summary of pump power and number of round trip required to reach the damage threshold of the rod for Nd:YAG, Nd:YVO₄ and Nd:YLF for rod diameters of 2 mm, 3 mm and 4 mm

<table>
<thead>
<tr>
<th></th>
<th>Nd:YAG</th>
<th>Nd:YLF</th>
<th>Nd:YVO₄</th>
</tr>
</thead>
<tbody>
<tr>
<td>2 mm</td>
<td>Pump power (W)</td>
<td>75</td>
<td>75</td>
</tr>
<tr>
<td></td>
<td>Number of round trips</td>
<td>50</td>
<td>35</td>
</tr>
<tr>
<td>3 mm</td>
<td>Pump power (W)</td>
<td>180</td>
<td>165</td>
</tr>
<tr>
<td></td>
<td>Number of round trips</td>
<td>35</td>
<td>55</td>
</tr>
<tr>
<td>4 mm</td>
<td>Pump power (W)</td>
<td>&gt;200</td>
<td>&gt;200</td>
</tr>
<tr>
<td></td>
<td>Number of round trips</td>
<td>&gt;110</td>
<td>&gt;70</td>
</tr>
</tbody>
</table>

The effects of increasing the rod diameter are also dramatic on the number of round trips required to achieve saturation as shown in Table 4. Nd:YVO₄ is a good candidate: with its large small signal gain, the output energy is mildly affected by the increase in rod diameter and a 3 mm rod pumped with 200 W allows operation at the design-tolerated damage threshold with only 13 round trips in the cavity or operation at 70 W with a 2 mm rod allows generation of 1 mJ of output energy, twice the minimum amount required to meet the specifications for HERACLES.
3.4.3.4 **Influence of seed energy**

The influence of the seed energy is relevant in the case of HERACLES because of the presence of the intra-cavity VBG. The pulses initially seeded into the regenerative amplifier cavity have energies up to 2 nJ and a spectral bandwidth as wide as 20 nm. The reflectivity bandwidth of the VBG is limited to 50 pm (see 3.4.5 for discussion on the VBG), resulting in dramatic energy losses in the first round trip. The amount of seed energy within the reflectivity bandwidth of the VBG is estimated to be in the pJ level of energy. It is therefore necessary to investigate the effects of seed energy level on the amplifier performances. The ASE is not taken into account in the model, this approximation being valid thanks to the presence of the VBG; only the low amount of ASE matching the reflectivity bandwidth of the VBG is oscillating in the cavity, making the ASE level a negligible quantity. A more detailed analysis of the VBG effect on the ASE is provided in 3.4.5. Computations of the output energy and number of round trip required to achieve saturation have been performed for Nd:YLF, Nd:YAG and Nd:YVO₄ rods, 4 cm long, 2 mm in diameter assuming 40% round trip cavity losses and a repetition rate of 1 kHz (Figure 11).
Figure 11: Calculated output energy performances and number of round trips required to reach saturation for a regenerative amplifier featuring either Nd:YAG, Nd:YLF or Nd:YVO₄ as a gain medium for seed energies of 5 pJ (a and b), 50 pJ (c and d) or 500 pJ (e and f). Calculations are performed for a 2 mm diameter rod at 1 kHz repetition rate with 40% intra-cavity losses.
Adjusting the seed energy over two orders of magnitude does not affect the output energy. This result is expected since neither the saturation energy or the small single gain depend on the seed energy. On the other hand, the number of round trips required to reach saturation increases as the seed energy decreases. A decrease in seed energy of an order of magnitude results in an increase of ~10-20% (depending on pump power level) on the number of round trips required to reach saturation. This increase is not significant for Nd:YVO_4 but can have severe consequences for Nd:YAG and Nd:YLF at moderate pump powers, making Nd:YVO_4 a much more flexible material.

3.4.3.5 Influence of repetition rate scaling

The scaling of the repetition rate under continuous pumping leads to a reduction of the output energy. As the duration of the high-Q period is reduced, less time is given to pump the crystal back to its initial inversion resulting in reduced gain and potentially uneven gain from pulse to pulse. Such fluctuations of the gain can result in bifurcation and chaotic behavior both of which should be avoided in a high energy amplifier chain to prevent damage. This section shows the limitations of Nd:YAG and Nd:YLF for operation at high repetition rates and investigates the behavior of Nd:YVO_4 in the multi-kHz repetition rate regime.

The limitations of low gain media for operation at high repetition rates are illustrated in Figure 12 for the case of Nd:YAG. The model previously described was modified such that a pulse train is generated at a different ejection time. Experimentally, this would correspond to setting the ejection trigger signal to several successive timings and observing the pulse train. The pulse energy of each pulse train was then compiled into a matrix and plotted against the number
of round trips in the cavity. The phase diagrams (right column) are obtained by calculating the gain remaining in the cavity after the pulse is ejected.

Figure 12: Calculated output energy as a function of number of round trip of ejection for a Nd:YAG-based regenerative amplifier operated at 1 kHz (a), 2 kHz (b), 5 kHz (c) and 10 kHz (d)

Simulations were performed with a 4 cm long crystal, 2 mm in diameter. The seed energy was 5 pJ and the cavity losses set to 40% per round trip. For repetition rates beyond 2 kHz, instabilities start appearing in the output energy, eventually causing a clear splitting of output energy, resulting in period doubling at 5 kHz for ejection times of 20 to 30 round trips. If the repetition rate is further increased, chaotic behavior starts occurring. Nevertheless, the
regenerative amplifier can be operated at high repetition rates in the regions of period doubling (round trip 20 to 30 at 5 kHz and round trip 40 to 50 at 10 kHz). In these regions the output energy is stable and the energy in the missed pulses is comparable to the seed energy *id est* negligible compared to period doubled pulses. The case of Nd:YLF is similar to that of Nd:YAG, bifurcations and chaotic behavior being even more pronounced due to smaller gain.

Since the small signal gain of Nd:YVO$_4$ exceeds that of Nd:YAG and Nd:YLF, the detrimental effects of repetition rate scaling are expected to be less dramatic (Figure 13).
Energies as high as 1 mJ can be extracted at up to 5 kHz repetition rate without instabilities occurring. At 10 kHz, after a region of period doubling (from ejection round trip 10 to 15) and the beginning of a chaotic region (from ejection round trip 15 to 18), stable output can be obtained with up to ~450 µJ.

Delaying the extraction of the pulse results in reduced output energy but also contributes to stabilizing the dynamics of the amplifier. The overall dynamics of the amplifier can be better understood by investigating the amount of gain remaining in the rod after ejection of the pulse. The plotted phase diagrams represent the evolution of the gain as a function of extracted energy for different repetition rates provides a view of the entire dynamics of the amplifier. After each pulse is extracted numerically from the amplifier scheme, the remaining inversion is multiplied by the emission cross-section to provide the value of the small signal gain after the extraction of the pulse.

Figure 14: Calculated phase diagram showing the gain remaining after pulse ejection as a function of output energy for Nd:YVO₄ regenerative amplifier operated at repetition rate below 5 kHz (a) and at 10 kHz (b)
Figure 14 shows the phase diagram of a regenerative amplifier featuring a Nd:YVO₄ operated at repetition rates below 5 kHz (a) and at 10 kHz and beyond. Examination of the bifurcated phase diagram can be performed in three steps.

- When the pulse remains in the cavity less than 10 round trips, the regenerative amplifier is operated essentially in the small signal regime, no significant depletion is achieved and each pulse experiences the same gain when seeded into the amplifier.

- In the bifurcation region, the pumping rate is too low to replenish the inversion fully leading to a significant difference of available gain for each seeded pulse. Since the pulses are kept in the cavity for a set amount of time, the pulses that experience a low gain will saturate the gain medium less than the pulses which experience a high gain. Simultaneously, as the gain medium is less saturated by the low energy pulses, the inversion is easily restored, yielding another high gain cycle.

- The final regime is when the pulse is ejected after the bifurcation regime. This case is opposite to the initial regime: with the gain completely saturated and essentially no gain remaining, the pulse is ejected. The gain leftover after each pulse is therefore consistent from pulse to pulse. Since the pumping rate is also constant, the inversion seen by each seeded pulse is constant leading to a stable output pulse train. The energy of each pulse is reduced because of the low initial gain available to the seeded pulses.

It was numerically found and experimentally proven by that increasing the amount of seed energy allows suppression of the bifurcation phenomena [159, 160]. Fast pump modulations also enable suppression of chaotic regimes [161].
3.4.4 Cavity design and stability

The design of the cavity needs to ensure maximum extraction of the energy and preserve the excellent beam profile provided by the fiber pre-amplifier while preventing damage to the optical surfaces. Ensuring an excellent beam profile is typically achieved in end pumped systems by focusing the pump beam down to a small waist in the crystal, therefore providing a soft aperture in the cavity filtering the higher-order transverse modes. This allows for maximum energy extraction and provides excellent beam profiles assuming that the profile of the pump beam is excellent.

In the case of the HERACLES regenerative amplifier, focusing the beam to a small waist in the crystal is prohibited by the high peak power inherent to the short pulse duration of the seed pulses. The opposite strategy is therefore adopted: a large and uniform pumped region is provided by a side-pumped geometry. The cavity must therefore be designed to prevent the excitation of higher order transverse modes. Lastly, the beam size needs to be maximized in the crystal to ensure maximum energy extraction.

The modeling of the resonator was performed using LASCAD, a commercially available software package relying on the mathematical analysis detailed in [162]. LASCAD is based on a finite element analysis that can predict the mode size at any point in the cavity, taking into account the thermal lensing in the crystals and predicting the output beam profile. The findings of LASCAD have been confirmed experimentally as part of this thesis.

Several cavity designs have been investigated numerically and experimentally, however in consideration of space and relevance, only the finally implemented design is described here. The design consists of a simple flat-flat resonator with the gain medium placed in the middle of the cavity. In the numerical investigations, we set the minimum cavity length to 85 cm
corresponding to a round trip time of ~6 ns, the limit allowed by the switching time of the Pockels cell.

Investigation of intra-cavity beam size as a function of cavity length and pump power have been performed. A 4 cm long Nd:YVO₄ rod, 2 mm in diameter with 0.3% doping concentration was chosen for the simulation. The simple layout of the cavity under investigation is shown in Figure 15.

![Figure 15: Layout of the oscillator as modeled by LASCAD – the rod provides the cavity stability via thermal lensing.](image)

The main source of mode size modification arises from adjusting the pump power because of thermal lensing. The increase in beam diameter with pump power in the rod is accompanied with a decrease of the beam diameter on the end mirrors of the cavity (position \( z = 0 \) cm and \( z = 90 \) cm or \( z = 100 \) cm). The simultaneous increase of beam diameter in the rod and reduction of beam size at the cavity ends evolves in a quadratic fashion with pump power, making catastrophic damage on the end mirror more likely when increasing the pump power; the simultaneous reduction of beam size on the end mirror correlated with the increase in extracted energy can avalanche and damage dielectric coatings.
Figure 16: Calculated mode volume diameter as a function of position in the cavity of a Nd:YVO₄-based regenerative amplifier for an 85 cm long cavity (a) and a 95 cm long cavity (b).

The main effects of increasing the cavity length are visible for the highest pump power. A longer cavity enables a larger beam diameter at the rod, even allowing for the mode diameter to exceed the rod diameter (Figure 16).

The output profile is provided by LASCAD as a fraction of the total output energy. The beam profile is consistent for all previously described configurations. The profile has been computed for both continuous wave and Q-switch regime and the results are gathered in Table 5.

Table 5: Comparison of the power distribution in continuous wave and Q-switched mode for the regenerative amplifier cavity.

<table>
<thead>
<tr>
<th></th>
<th>Continuous wave</th>
<th>Q-switched</th>
</tr>
</thead>
<tbody>
<tr>
<td>Power in TEM₀₀ mode</td>
<td>16.3%</td>
<td>25.4%</td>
</tr>
<tr>
<td>Power in TEM₀₁ mode</td>
<td>27.1%</td>
<td>74.6%</td>
</tr>
<tr>
<td>Power in TEM₁₀ mode</td>
<td>2 %</td>
<td>0%</td>
</tr>
<tr>
<td>Power in TEM₂₀ mode</td>
<td>0 %</td>
<td>0%</td>
</tr>
<tr>
<td>Power in TEM₀₂ mode</td>
<td>54.6%</td>
<td>0%</td>
</tr>
</tbody>
</table>
The discrepancy between Q-switched and continuous wave operation results from the beam being shaped by the gain, the difference of gain dynamics in the two modes of operation potentially leading to different output beam profiles. The improvement in beam quality in the Q-switched mode has been observed experimentally. It was found experimentally that, when seeded with a Gaussian profile, the regenerative amplifier preserves the beam profile. Such conservation of beam profile is attributed to the relatively good behavior of the oscillator as a standalone Q-switched oscillator, to the quality of the seed pulses and to the reduced number of passes through the cavity.

3.4.5 Intra-cavity volume Bragg grating

Volume Bragg gratings are holographic grating recorded in photo-thermo refractive (PTR) glass. An interference pattern generated by two UV beams crossed into the PTR glass is recorded yielding a succession of planes of alternate refractive indices. When a laser beam illuminates the stack, partial reflections occur at each interface between planes of high and low refractive indices.

![Figure 17: Schematic view of a volume Bragg grating illustrating constructive interferences at the Bragg angle.](image)
For an incident angle equal to the Bragg angle, the optical path difference experienced by all the partially reflected beams is such that the reflected beams interfere constructively producing an effective reflected beam (Figure 17). The Bragg condition is expressed by:

\[ 2nd \cos \theta = m \lambda, \]  

(5)

where \( n \) is the refractive index of the PTR glass, \( d \) is the distance between planes of different indices of refraction, \( \theta \) is the angle of incidence of the beam with respect to the normal of the stack, \( m \) is the diffraction order and \( \lambda \) the wavelength of the incident beam. In the case of a VBG designed to be operated at normal incidence (\( \theta = 0^\circ \)) and in the first diffracted order (\( m = 1 \)), the equation simplifies to:

\[ \lambda_0 = 2nd, \]  

(6)

where \( \lambda_0 \) is the resonant wavelength where the VBG is designed to operate. The Bragg condition can be written, in the case of a VBG designed for a center wavelength \( \lambda_0 \) operated at normal incidence and in the first diffracted order:

\[ \lambda = \lambda_0 \cos \theta. \]  

(7)

The spectral selectivity of the Bragg condition increases as the depth of the stack increases. The derivation of this phenomenon is beyond the scope of this discussion. VBGs with spectral reflectivity linewidths ranging from 1 nm to 50 pm FWHM have been produced and have been implemented in both solid-state, fiber and diode lasers to achieve dramatic spectral narrowing, wavelength stabilization [152, 153, 163-176] and spectral beam combining. The properties of the PTR glass are similar to that of fused silica (aside from absorption values), therefore peak powers in the GW.cm\(^2\) can be handled and transparency beyond 90% from 350
nm to 2700 nm is achieved. The peak reflectivity of the VBG can be adjusted in the hologram development process and can reach up to 99.9%.

One of the end-mirrors of the regenerative amplifier of HERACLES is replaced by a VBG and fulfills two roles:

- Enhancing the signal to ASE ratio by spectrally filtering out of band ASE
- Tailoring the pulse duration via spectral narrowing

A simple qualitative model has been developed to illustrate the behavior of the VBG with respect to the ASE in the regenerative amplifier cavity. The seed spectrum is modeled by a 20 nm wide Gaussian function normalized such that its integral is equal to the number of photon in the experimental seed pulse. The ASE is modeled by a 1.5 nm wide Gaussian (Figure 18.a) containing an adjustable amount of energy and the gain is modeled by a square function also 1.5 nm wide. The choice of a square function for the gain prevents numerical gain narrowing effects in the amplifier, making the VBG cause gain narrowing. The gain is initially set to $g_0 = 8$ and follows a geometrical series evolution $g_0/r$ after each round trip to model the saturation of the gain. The factor $r$ is adjusted such that the output energy, computed by integrating the output spectrum is within the range of values predicted by the accurate model presented in 3.4.2. The reflectivity linewidth of the VBG is set to 50 pm and its reflectivity to 70%.

The amount of ASE photons is challenging to estimate and is therefore simply compared to the amount of seeded photons. Computations have been performed with an amount of ASE photons 10 times larger than the seeded photons, equal to the seeded photons and 10 times smaller than the seeded photons.
Figure 18: Simulated influence of ASE on the output spectrum of a Nd:YVO₄ regenerative amplifier with after 1 pass in the cavity (a and b) and 11 passes in the cavity (c and d).

The VBG strongly contributes to the reduction of the ASE. After only one round trip in the cavity, the presence of the VBG enables the reduction of the background signal (amplified ASE and signal out of the reflectivity bandwidth) down to 60 dB below the signal level while narrowing the spectrum down to ~70 pm. After 11 round trips, the level of the background has become negligible compared to the signal ensuring a clean output from the regenerative amplifier. The only ASE that can affect the cleanliness of the output pulse train, is the ASE contained in the 50 pm band corresponding to the reflectivity linewidth of the VBG. It was
numerically found that after 11 passes on the VBG, the FWHM spectral width had shrunk to ~30 pm. Such behavior has been reported experimentally by Okishev et al. [174].

The VBG also controls the pulse duration via spectral narrowing. Experimentally, the seed pulses have a 20 nm-broad spectrum and are delivered by a 30 m long fiber pre-amplifier generating a large amount of spectral phase via material dispersion and self-phase modulation. The seed pulse duration has been measured to be ~20 ps, confirming the presence of spectral chirp. Dramatic spectral narrowing allows for both suppression of most of the complex spectral phase and lengthening of the pulse.

Figure 19: Simulated output spectra from a YVO₄ regenerative amplifier without intra-cavity VBG (a) an equipped with a VBG (inset) and corresponding output pulse durations (b).

In Figure 19.a, a 20 nm wide spectrum was numerically chirped until the duration of the corresponding pulse reaches 20 ps, the experimentally measured duration of the seed pulses provided by the fiber pre-amplifier. For simplification, only second order chirp was considered. The width of the spectrum was then changed to 30 pm to simulate the effect of the VBG, while maintaining the spectral phase. Figure 19.b shows the resulting temporal profile of the VBG narrowed pulse. The calculated pulse duration with the VBG is ~85 ps. The pulse profile was
also computed for a VBG narrowed spectrum with no spectral phase; the predicted pulse
duration is within 2% of the predicted pulse duration with the large spectral phase, proving that
the pulse is essentially transform limited after experiencing the spectral narrowing in the VBG.

3.5 Post-amplifier modeling and parametric study

3.5.1 Post-amplifier model

Single and double-pass configurations have been modeled to investigate the performances of the post-amplifier. The code used in the previous section was modified to suit such a geometry: the high-Q period is limited to one pass or two passes and the intra-cavity losses are set to zero while the low-Q period is still governed by Eqn. (4).

Based on the results of section 3.4.3, Nd:YLF is not a candidate as a gain medium for the regenerative amplifier which sets the output wavelength of the chain to 1064 nm and leaves only the choice between Nd:YVO₄ and Nd:YAG for all subsequent amplifiers. Therefore an investigation similar to that carried out in 3.4.3 of Nd:YVO₄ and Nd:YAG is reported in this section.

3.5.2 Parametric investigation of post-amplifier performances

3.5.2.1 Post amplifier output requirements

A single pass amplifier needs efficient extraction of the stored energy and therefore a large small signal gain. It is also preferable to have the saturation energy larger than the product of the seed energy and the single pass gain to achieve efficient energy extraction. The post-
amplifier implemented in the pump beam generation line of HERACLES is required to provide pulses with \( \sim 2 \text{ mJ of energy} \) and \( 85 \text{ ps} \) duration at \( 1-10 \text{ kHz} \) repetition rate.

3.5.2.2 Influence of crystal diameter

An increase in rod volume results in a decrease of the density of excited electrons \( n \) which reduces the small signal gain \( g_0 = n\sigma \), where \( \sigma \) is the emission cross-section of the gain medium. An increase of rod diameter is often necessary to reduce the peak intensity on the rod surfaces. A simulation of the influence of the rod diameter has been performed for a 4 cm long Nd:YVO\(_4\) rod and 4 cm long Nd:YAG rod, side pumped with a seed energy of 500 \( \mu \text{J} \) at 1 kHz repetition rate with 180 W pump power (Table 6).

Table 6: Output energy as a function of rod diameter for a single and double pass amplifier featuring either Nd:YVO\(_4\) or Nd:YAG as a gain medium. Computations assumed 500 \( \mu \text{J} \) seed pulses and 180 W pump power.

<table>
<thead>
<tr>
<th>Rod diameter</th>
<th>Nd:YVO(_4)</th>
<th>Nd:YAG</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Single pass</td>
<td>Double pass</td>
</tr>
<tr>
<td>2 mm</td>
<td>2.4 mJ</td>
<td>4.5 mJ</td>
</tr>
<tr>
<td>3 mm</td>
<td>1.23 mJ</td>
<td>2.43 mJ</td>
</tr>
<tr>
<td>4 mm</td>
<td>856 ( \mu \text{J} )</td>
<td>1.38 mJ</td>
</tr>
</tbody>
</table>

The saturation energy of the YVO\(_4\) rod is \( \sim 5 \text{ mJ} \) while that of the YAG rod is \( \sim 12 \text{ mJ} \), therefore saturation in the YVO\(_4\) rod is achievable in a double-pass configuration. Nevertheless, assuming that the rod is 75\% filled by the seed beam, a 4.5 mJ energy beam on the end facet of the rod corresponds to a peak power of \( \sim 4 \text{ GW.cm}^2 \) for an 85 ps pulse, closely approaching the damage threshold of the dielectric coating. The performance of the YAG rods are close to that of
the small diameter YVO₄ but rapidly decay as the rod diameter is increased. Therefore, a single pass through a small diameter YVO₄ rod appears to be the most valuable choice.

3.5.2.3 Influence of seed energy

For a single pass amplifier, the amount of output energy is proportional to the seed energy as long as the amplifier is not strongly saturated. Saturation ensures complete extraction of the gain from the amplifier and stable output energy but limits the efficiency of the extraction. A simulation of the influence of the seed energy on the output energy has been performed for a 4 cm long Nd:YVO₄ rod and a 4 cm long Nd:YAG with 2 mm diameter, side-pumped with 180 W of continuous power at 1 kHz repetition rate (Table 7).

Table 7: Output energy as a function of seed energy for a single and double pass amplifier featuring either Nd:YVO₄ or Nd:YAG as a gain medium. Computations assumed a 2 mm rod and 180 W pump power.

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>100 µJ</td>
<td>757 µJ</td>
<td>2.81 mJ</td>
<td>400 µJ</td>
<td>1.47 mJ</td>
</tr>
<tr>
<td>500 µJ</td>
<td>2.4 mJ</td>
<td>4.5 mJ</td>
<td>1.84 mJ</td>
<td>5.12 mJ</td>
</tr>
<tr>
<td>1 mJ</td>
<td>3.6 mJ</td>
<td>5.1 mJ</td>
<td>3.4 mJ</td>
<td>7.66 mJ</td>
</tr>
</tbody>
</table>

The amount of available seed energy is the main deciding factor for the gain medium and the amplifier geometry (single or double pass). From Table 7, if the seed energy is high and a double or multi-pass configuration can be implemented in a rod 2 or 3 mm rod, the best energy output is obtained from an Nd:YAG amplifier. On the other hand, if only a limited amount of seed energy is available, which is the case for the HERACLES post amplifier, Nd:YVO₄ is a
better choice since the amplifier is operated in the small signal gain regime and Nd:YVO₄ has a small signal gain $\sim \sigma_{\text{YVO}_4}/\sigma_{\text{YAG}} = 4$ times larger than that of Nd:YAG. If a double-pass geometry can be implemented with a limited (~100 $\mu$J) seed energy, a 2 mm diameter rod allows output energies approaching 1/10 of the damage threshold, an experimentally achievable target. Finally, for repetition rate scaling purposes it is preferable to not fully saturate the amplifier because complete depletion of the gain leads to uneven gain between pulses at high repetition rates as shown in 3.4.3.5.

3.6 Multi-pass booster amplifier

3.6.1 Post-amplifier model

A model similar to that used in the previous section is used to investigate the performance of a booster amplifier. The booster amplifier design should allow amplification of pulses with 1 to 3 mJ to the 10 to 20 mJ energy level at multi-kHz repetition rates in 85 ps. Similarly to previous discussions, only Nd:YAG and Nd:YVO₄ crystals are being investigated due to the poor performance of Nd:YLF as a regenerative amplifier in the case of HERACLES.

3.6.2 Parametric investigation of booster amplifier performance

3.6.2.1 Influence of crystal diameter and pump power

The effects of gain reduction attributed to the increase in rod diameter is investigated for the case of 12.5 cm long Nd:YVO₄ and Nd:YAG rods for continuous side-pumping at power levels of 500 W, 750 W and 1 kW. The frequency of the seed pulses is assumed to be 1 kHz and the seed energy 2 mJ. Results of the simulation are shown in Figure 20.
Figure 20: Calculate output energy performances as a function of the number of passes and the pump power for a multi-pass amplifier featuring either Nd:YVO₄ or Nd:YAG as a gain medium for rod diameter of 5 mm (a and b), 7 mm (c and d) and 10 mm (e and f). Calculations are performed for a 12.5 cm long rod at 1 kHz repetition rate with 2 mJ seed energy.
The expected reduction in gain with increasing rod diameter is observed in the simulations for both gain media. The 5 mm diameter rod allows the saturation energy to be reached in both YVO₄ (~22 mJ) and YAG (~55 mJ) at all pump powers. Nevertheless, pulses with peak powers approaching 1 GW.cm⁻² propagating for an effective crystals length of \(N\times12.5\) cm (where \(N\) is the number of passes in the gain medium) are likely to accumulate a large amount of nonlinear phase, resulting in amongst other effects, a deteriorated beam profile. Reaching saturation is a desirable feature for the last amplifier of the chain since it provides output energy stability. For diameters greater than 5 mm, saturation cannot be reached in YAG and is almost reached in YVO₄. Therefore the experimental design will require using a 7 mm or 10 mm diameter rod and the introduction of a controlled amount of losses in the multi-pass amplifier, thereby limiting the extracted energy to ~20 mJ while maintaining a saturated output. Notice that YVO₄ crystals, though attractive for their saturation energy are currently available in limited sizes, the largest available size being ~3 mm x 4 cm, therefore limiting the practical use of YVO₄ in high energy amplifiers.

3.6.2.2 Influence of seed energy and pump power

The effects of variations in seed energy on the output energy have been modelled for the case of 12.5 cm long Nd:YVO₄ and Nd:YAG rods for continuous side-pumping at power levels of 500 W, 750 W and 1 kW. The frequency of the seed pulses is assumed to be 1 kHz in the simulation and the beam diameter was set to 7 mm. Results of the simulation are shown in Figure 21.
Figure 21: Calculated output energy performances as a function of the number of passes and pump power for a multi-pass amplifier featuring either Nd:YVO₄ or Nd:YAG as a gain medium for seed energy of 1 mJ (a and b), 2 mJ (c and d) and 3 mJ (e and f). Calculations are performed for a 7 mm diameter, 12.5 cm long rod at 1 kHz repetition rate.
Tripling the amount of seed energy in YAG results in doubling the output energy as long as the pump power is maintained below 1 kW. For pump powers as high as 1 kW, increasing the seed energy only results in better saturation of the gain medium. In the case of YVO₄, increasing the seed energy has little effect on the output energy and only affects the number of passes required to saturate the gain medium. In this regard, it is worth providing as high of a seed energy level as possible since it results in a reduced number of passes in the gain medium.

3.6.3 Depolarization effects

From the results of previous sections, it appears that Nd:YAG is the prime candidate as a gain medium for the booster amplifier of HERACLES because of the unavailability of Nd:YVO₄ in large sizes.

The output produced by Nd:YAG-based lasers is typically unpolarized due to the anisotropic nature of the crystal (see section 3.3). In the case of the HERACLES booster amplifier, polarization switching in and out of the amplifier needs to be considered as an option. Furthermore, the output of the booster amplifier stage must be polarized to ensure phase-matching in the OPA stages.

A YAG based system with an intra-cavity polarizer is subject to thermally induced depolarization effects. As the pump power is increased thermal stress birefringence results in the variation of the index of refraction across the gain medium. This birefringence can be described via a radial refractive index variation $n_r$ and a tangential component $n_\phi$. A calculated refractive index profile obtained under a 150 W thermal load using a derivation provided by Koechner [156] is shown in Figure 22.
Figure 22: Computed refractive index profile for a 150 W thermal load on a 10 mm diameter YAG rod. The index of refraction variation $\Delta n$ is computed with respect to the index of refraction at the edge of the rod.

At any point $P(r, \phi)$ in the section of the rod, an incident polarized radiation must therefore be decomposed along the two axis of birefringence. Since $\Delta n_\phi \neq \Delta nr$, a phase difference is introduced by propagating through the gain medium resulting in an elliptically polarized output except for regions where the induced axis are along the polarizer axis [156]. The effects of this depolarization are twofold: first some of the horizontally polarized light will couple into the orthogonal polarization and the light in the orthogonal polarization will be rejected by the polarizer. The second effect of depolarization is to strongly affect the beam profile due to the removal of the orthogonal polarization state. Simulations of spatial output profiles under several levels of heat load have been performed implementing the analysis provided by Koechner [156] into MATLAB for a 12.5 cm long, 10 mm diameter YAG rod. The obscured rings observed at the highest heat load are the regions where the retardation reaches a full wave.
The overall losses introduced by depolarization in an Nd:YAG cavity equipped with a polarizer have also been computed based on the formula provided by Karr [177], the derivation of which being beyond the scope of this discussion. The results obtained for a resonator provide a close approximation to the results expected in the case of a multi-pass amplifier. Results of the simulation applied to the case of a 12.5 cm long, 1 cm diameter YAG rod are shown in Figure 24.
Without any depolarization scheme, losses as high as 30% are expected, which significantly impairs the amplifying abilities of the gain medium. From Figure 20, considering the case of 10 mm diameter YAG rod, the single pass gain is $\sim 1.15$ and therefore less than the depolarization losses. In order to effectively obtain amplification, schemes need to be implemented to compensate for the phase lag introduced by the thermally induced birefringence.

3.6.4 Multi-pass booster amplifier layout model

Several schemes have been proposed to compensate for depolarization effects, involving either quarter-wave plates [178] or Faraday rotators [179, 180]. The principle of these techniques is to flip the polarization of the beam between each passes to add an equal amount of phase lag from the $n_\phi$ and $n_r$ birefringent axis onto the beam. Another technique, sharing the same principle but relying on two identical gain media is typically implemented when high average output power and high pump power are involved [181, 182]. The end of one rod is imaged onto the other via a $4f$ system, making the two rods effectively one rod with twice the length of each
individual rod. A half-wave plate is introduced within the 4f system, rotating the polarization of the incident beam such that the $n_r$ axis is imaged onto the $n_\phi$ axis and vice-versa. In this way, the same amount of phase accumulated on both axes will effectively cancel out the depolarization effects. Implementing this type of depolarization compensation scheme complicates the design of a multi-pass amplifier, in particular, since the depolarization is typically accompanied by thermal lensing.

In order to overcome these difficulties, LASCAD (see section 3.4.4) was used to model a suitable design. The design must be robust with respect to thermal lensing while compensating for thermally induced depolarization. The layout of a multi-pass booster amplifier implemented on HERACLES is shown in Figure 25.

![Figure 25: Layout of the multi-pass booster amplifier with sketched mode volume](image)

The beam enters off the thin-film polarizer (TFP) and is polarization switched into the cavity by the Pockels cell. Polarization switching is advantageous since it allows for the beam to travel parallel to the crystal longitudinal axis, as opposed to common multi-pass “butterfly-type” amplifiers. An output coupler is placed at the end of the cavity for both monitoring and controlling the maximum extracted energy to prevent damages to the components in the cavity.
Beam diameters filling the rod entirely can be achieved for the 130 cm long cavity. Nevertheless as the pump power is increased, the beam is increasingly focused on the end mirrors of the cavity. For a pump power of 450 W and a 130 cm long cavity (not shown on Figure 26.a), the mode volume at the end of the cavity is as small as 1 µm and exceeds 6 cm in the region between the YAG rods and the lenses, clearly showing that this cavity reaches the edge of its stability region for pump power of ~400 W. In order to overcome this issue, the cavity is shortened to 90 cm allowing a stable cavity for pump power of up to half a kilowatt (Figure 26.b) at the expense of a smaller beam diameter in the YAG rods. For pump power beyond 600 W this shorter cavity also became unstable setting a maximum operating pump power.

3.7 Conclusions and recommendations for HERACLES amplifier chain

The parametric and comparative simulations performed in this chapter give an extensive view of a number of aspects for the design of the HERACLES amplifier line. Several conclusions can be drawn from these simulations:
• The design of the regenerative amplifier should rely on a small diameter Nd:YVO$_4$ rod as it limits the number of passes in the amplifier, therefore limiting accumulation of nonlinear phase.

• The insertion of VBG in the regenerative amplifier enables efficient pulse tailoring and ASE reduction while limiting insertion losses.

• The cavity design of a regenerative amplifier featuring a side-pumped Nd:YVO$_4$ rod needs to be carefully handled to prevent damage to components and ensure efficient energy extraction. A simple flat-flat cavity with carefully mastered symmetry ensures high energy extraction and, within computed boundaries, prevents damages to components.

• A post amplifier, operated preferentially in a single-pass configuration relying on Nd:YVO$_4$ as a gain medium enables high gain and energy outputs up to 2 mJ for a 2 mm rod diameter. Nd:YVO$_4$ offers superior capabilities compared to Nd:YAG as a gain medium for this amplification stage.

• The final booster amplifier stage can be readily be saturated and provide energies in the 10-20 mJ range if Nd:YVO$_4$ is used as a gain medium. Due to the unavailability of Nd:YVO$_4$ in sizes large enough to prevent optical damages, it is necessary to resort to Nd:YAG as a gain medium which requires implementing depolarization compensation schemes. A careful design of a multi-pass amplifier is proposed that addresses both thermal lensing and depolarization issues while enabling energy extraction up to the desired energy range. Saturation can be reached by adding a limited amount of losses in the amplifier.
This chapter provides a theoretical background and models of traditional stretching and compressing techniques. These techniques – successful for the compressing and stretching of pulses with spectra up to ~100 nm spectral widths – are presented as an introduction to the more exotic techniques implemented for ultra-broadband pulses [183-186]. A model of the grism (a hybrid dispersion line based on both gratings and prisms) based HERACLES stretcher is developed in a semi-analytical approach and simulation results along with a parametric study are presented. Finally, the HERACLES compressor is discussed and critical parameters are described via simulation results.

4.1 Limitations of traditional stretcher/compressor assemblies

Traditional ways of chirping optical pulses rely either on Treacy compressors, Martinez stretchers (see section 2.1.1), or prism pairs operated at Brewster’s angle. The optical frequency-dependent path difference provided by a prism pair arises from refraction and is typically limited. Prism pairs - thanks to their low insertion loss – are mostly used to compensate for the low amount of dispersion inside mode-locked oscillators. The optical path difference provided by a grating pair arises from diffraction and therefore differentiates optical frequencies much more strongly than refraction. Grating pairs are typically used for stretching and compressing in CPA systems. This technique has proven to be robust, easy to implement and is common in most commercial chirped-pulse amplifier systems. It nevertheless presents several drawbacks:
• The diffraction efficiency of ruled gratings typically used in stretchers and compressors is at most 90% upon reflection and results in **high insertion losses**. The geometry of a grating stretcher or compressor requires four reflections on the grating surfaces, typically reducing the throughput of these assemblies to a maximum of ~70%.

• The use of gratings in the case of ultra-broadband pulse stretching requires great care in the choice of the grating due to the **non-uniform diffraction efficiency** of gratings that can result in amplitude modulations in the spectrum and alter the output pulse duration.

• Grating pairs have been implemented to stretch and compressor pulses as short as 20-25 fs. Beyond this limit, the third order dispersion effects start to be sizable. **Grating pair stretcher and compressor assemblies are enable to compensate for dispersion only up to the third order** unless extreme care is taken [92].

4.2 **Strategy for efficient pulse compression**

The 30% losses experienced in a grating stretcher or compressor are a major limitation to high-energy laser systems. While the loss induced by the stretcher can usually be compensated for, the 30% loss of the compressor is simply wasted power.

In typical CPA systems, the pulse is up-chirped in the stretcher (i.e. the pulse experiences normal dispersion) and down-chirped in the compressor (i.e. the pulse experiences anomalous dispersion). Reversing this strategy can offer an alternative design that limits the loss in the compressor by removing the need for a grating pair (Figure 27).
The pulse is initially down-chirped in a grating based stretcher and compressed by propagating through a large piece of glass designed to provide the same amount of spectral phase – but with opposite sign – as that of the grating pair. This strategy can successfully be applied in the near IR since most glasses provide normal dispersion in this spectral range. The choice of the glass composition and length allow tuning of the spectral phase provided by the compressor. Provided that the glass is anti-reflection coated and transparent at the wavelength of operation, the compressor is essentially lossless.

4.3 Traditional pulse chirping techniques: model

In this section, traditional pulse stretching and compressing techniques are discussed and analytical solutions for group delay and high order dispersion are presented. The conventions used for the derivations are introduced. Traditional approaches are presented as a prelude to the more complex assemblies developed later in this chapter that are suitable for ultra-broadband pulse stretching.
4.3.1 Spectral phase: definition and convention

The key parameter to be computed when developing a stretcher or compressor assembly is the spectral phase [187, 188]. The spectral phase can be simply introduced by mathematically expressing the spectrum of a pulse as:

\[ E(\omega) = E_0(\omega) e^{i\phi}, \]  

(8)

where \( E_0(\omega) \) is the spectral amplitude of the electric field and \( \phi(\omega) \) is the spectral phase. The modulus squared of the spectral amplitude \( E_0 \) is the spectral intensity measured experimentally by e.g. spectrometers. The spectral phase is typically expressed as a Taylor expansion around a central frequency \( \omega_0 \):

\[
\phi(\omega) = \phi(\omega_0) + \frac{d\phi}{d\omega} \bigg|_{\omega_0} (\omega - \omega_0) + \ldots \\
\frac{1}{2} \frac{d^2\phi}{d\omega^2} \bigg|_{\omega_0} (\omega - \omega_0)^2 + \frac{1}{6} \frac{d^3\phi}{d\omega^3} \bigg|_{\omega_0} (\omega - \omega_0)^3 + \frac{1}{24} \frac{d^4\phi}{d\omega^4} \bigg|_{\omega_0} (\omega - \omega_0)^4 + \ldots
\]

(9)

where \( \omega_0 \) is the central frequency. The first term of the expansion is the absolute phase of the spectrum at the center frequency \( \omega_0 \). The second term is proportional to \( \frac{d\phi}{d\omega} \bigg|_{\omega_0} \), the group delay (GD), or the time for the pulse envelope to propagate through the medium or apparatus under consideration. The third term is proportional to \( \frac{d^2\phi}{d\omega^2} \bigg|_{\omega_0} \), called the group delay dispersion (GDD) which relates to the frequency dependence of the delay experienced by each wavelength as they propagate through the medium or apparatus under consideration. Similarly the fourth and fifth terms of the expansion are proportional to, respectively, the third and fourth derivative of the spectral phase and are called third order dispersion (TOD) and fourth order dispersion (FOD).
Such an expansion makes apparent the influence of increased bandwidth on spectral phase: for a narrow spectral bandwidth, the higher order terms \((\omega - \omega_0)^3\) and \((\omega - \omega_0)^4\) terms are negligible compared to the first order dispersion (GDD). However, for pulses with octave-spanning spectra, higher order terms need to be considered. In the case of HERACLES, the first four orders are taken into consideration and compensated for.

The spectral phase induced by stretchers and compressors is the result of either refraction or diffraction as discussed in section 4.1. It is therefore necessary to develop a formalism that can handle all situations in order to compute the spectral phase introduced by stretcher and compressor assemblies. For this purpose the spectral phase is expressed as the dot product between a frequency dependent wave vector and an arbitrary vector \(\mathbf{R}\) that extends throughout the assembly being considered [188]:

\[
\phi(\omega) = \overline{k(\omega) \cdot \mathbf{R}} = |k(\omega)| |\mathbf{R}| \cos \theta, \\
\tag{10}
\]

where \(\theta\) is the angle between the wave vector \(k\) of the frequency \(\omega\) and the vector \(\mathbf{R}\). \(\mathbf{R}\) can be chosen arbitrarily as long as it extends throughout the entire system. The dot product ensures that all vectors will be projected onto the \(\mathbf{R}\) vector and the phase computed this way gives a relative measurement of the delay experienced by each frequency in the dispersive line. Figure 28 summarizes the vector-convention used for the calculation of the spectral phase. The symbol \(\delta\) represents the optical path difference experienced in an arbitrary dispersion line from which the spectral phase is computed.
The choice of \( R \) only results in a phase offset. Although this value does not aid in the design of stretcher-compressor assembly, it is convenient for computational purposes to choose \( R \) to be the optical path experienced by the center frequency \( \omega_0 \) as suggested by Kane [188]. In this way the \( \cos \theta \) and \( \sin \theta \) terms will always be one or zero at the center wavelength which simplifies the expressions. The spectral phase can then be expressed as:

\[
\phi(\omega) = \frac{d}{c} n(\omega) \omega \cos \theta,
\]

where \( d \) is the optical path length experienced by the center wavelength through the dispersive line and \( n(\omega) \) is the frequency dependent refractive index of the medium in which the pulse propagates.

### 4.3.2 Material dispersion and angular dispersion

Dispersion effects can arise from either angular dispersion (due to refraction or diffraction) or material dispersion (due to the frequency dependence of refractive index). Equation (11) is general enough to contain both type of dispersion. This can be made apparent by computing the group delay by taking the first and second derivatives of Eqn.(11) with respect to frequency:
\[
\frac{d\phi}{d\omega} = \frac{d}{c} \left[ \frac{\omega}{d\omega} \left( \frac{dn(\omega)}{d\omega} \cos \theta + n(\omega) \cos \theta - n(\omega) \omega \frac{d\theta}{d\omega} \sin \theta \right) \right] \\
= \frac{d}{c} \left[ \left( \frac{\omega}{d\omega} + n(\omega) \right) \cos \theta - n(\omega) \omega \frac{d\theta}{d\omega} \sin \theta \right] 
\]

and at the center frequency:

\[
\left. \frac{d\phi}{d\omega} \right|_{\omega_0} = \frac{d}{c} \left[ \frac{\omega}{d\omega} + n(\omega) \right] 
\]

The group delay at the center wavelength is only dependent on the length and properties of the material since all dependence on \( \theta \) disappears. Nevertheless, inspecting Eqn.(12) shows that any angular dispersion will be accounted for if the group delay is computed over the entire spectral range of interest. The GDD is computed by taking the second derivative of the spectral phase. After processing the algebra and gathering the terms in \( \cos \theta \) and \( \sin \theta \), one obtains:

\[
\frac{d^2\phi}{d\omega^2} = \frac{d}{c} \left[ \cos \theta \left( 2 \frac{dn(\omega)}{d\omega} + \omega \frac{d^2n(\omega)}{d\omega^2} - n(\omega) \omega \left( \frac{d\theta}{d\omega} \right)^2 \right) + \ldots \right] \\
\quad \sin \theta \left( -2 \omega \frac{dn(\omega)}{d\omega} \frac{d\theta}{d\omega} - 2n(\omega) \frac{d\theta}{d\omega} - n(\omega) \omega \omega \frac{d^2\theta}{d\omega^2} \right) \right], 
\]

which, at the center wavelength, simplifies to:

\[
\left. \frac{d^2\phi}{d\omega^2} \right|_{\omega_0} = \frac{d}{c} \left[ 2 \frac{dn(\omega)}{d\omega} + \omega \frac{d^2n(\omega)}{d\omega^2} - n(\omega) \omega \left( \frac{d\theta}{d\omega} \right)^2 \right]. 
\]

Since it provides more physical insight, Kane [188] suggests writing the expression of the GDD as follows:

\[
\left. \frac{d^2\phi}{d\omega^2} \right|_{\omega_0} = \frac{d}{c} \left[ \frac{\omega}{d\omega} \left( \frac{dn(\omega)}{d\omega} \right) \right] - \omega \left( \frac{d\theta}{d\omega} \right)^2. 
\]
The GDD can be separated in a contribution from the material properties and a contribution from angular dispersion. From this expression it can be inferred that the sign of any GDD generated via angular dispersion is negative. This general statement applies to any dispersive line as long as no imaging optics are inserted in the line; in the case of a Martinez stretcher, the 4f system placed between the gratings reverses the sign of \( d \) resulting in positive dispersion from a dispersive line relying on angular dispersion. The material dispersion on the other hand can vary, depending only on the variations in the Sellmeier equation of the material at the wavelength of interest. The expression for the TOD is calculated by computing the third derivative of the spectral phase. After simplification, the expression for the TOD is given by:

\[
\frac{d^3 \phi}{d\omega^3} = \frac{d}{c} \left[ \cos \theta \left( \frac{d^3 n(\omega)}{d\omega} + 3 \frac{d^2 n(\omega)}{d\omega} - 3 \frac{d \theta}{d\omega} \right)^2 \left[ \frac{d n(\omega)}{d\omega} + n(\omega) \frac{d}{d\omega} \frac{d^2 \theta}{d\omega} \right] \right] + \ldots \tag{17}
\]

and, at the center wavelength, collapses to:

\[
\left. \frac{d^3 \phi}{d\omega^3} \right|_{\omega_0} = \frac{d}{c} \left[ \frac{d^3 n(\omega)}{d\omega} + 3 \frac{d^2 n(\omega)}{d\omega} - 3 n(\omega) \left( \frac{d \theta}{d\omega} \right) \left( \frac{d n(\omega)}{d\omega} \right)^2 \left[ \frac{\omega}{n(\omega)} \frac{d n(\omega)}{d\omega} + 1 + \omega^2 \frac{d^2 \theta}{d\omega} \frac{d \theta}{d\omega} \right] \right] \tag{18}
\]

Neglecting the term in \( \frac{\omega}{n(\omega)} \frac{d n(\omega)}{d\omega} \) which is typically much smaller than one for all practical applications, Kane suggests to rewrite the TOD in a more insightful manner [188]:

68
The angular dispersion term, also in agreement with Kane, provides little physical insight. There are no mathematical constraints on the sign or the magnitude of the angular TOD which suggests that only the design of the dispersive line affects the TOD. It is therefore necessary to investigate thoroughly the link between TOD and experimental parameters in order to tune the TOD to the desired range.

In order to simplify the algebra when investigating the properties of a particular assembly, it is useful to express these relations as a function of wavelength rather than frequency. This is achieved by using the following set of conversion equations [187]:

\[
\frac{d\theta}{d\omega} = -\frac{2\pi c}{\omega^2} \frac{d\theta}{d\lambda}
\]

\[
\frac{d^2\theta}{d\omega^2} = \frac{\lambda^2}{(2\pi)^2} \left( \frac{\lambda^2}{\lambda^2} \frac{d^2\theta}{d\lambda^2} + 2\lambda \frac{d\theta}{d\lambda} \right)
\]

\[
\frac{d^3\theta}{d\omega^3} = -\frac{\lambda^3}{(2\pi)^3} \left( \frac{\lambda^3}{\lambda^3} \frac{d^3\theta}{d\lambda^3} + 6\lambda^2 \frac{d^2\theta}{d\lambda^2} + 6\lambda \frac{d\theta}{d\lambda} \right)
\]

and results in the following expressions for the GD, GDD and TOD:

\[
\frac{d\phi}{d\lambda} = \frac{d}{c} \left( n - \lambda \frac{dn}{d\lambda} \right)
\]

\[
\frac{d^2\phi}{d\lambda^2} = \frac{\lambda^3}{2\pi c^2} \frac{d^2n}{d\lambda^2} - \frac{\lambda^3 n}{2\pi c^2} \frac{d\theta}{d\lambda} \left( \frac{d\theta}{d\lambda} \right)^2
\]
\[
\frac{d^3 \phi}{d\lambda^3} = -\frac{\lambda^4}{4\pi^2 c^3} d \left[ 3 \frac{d^2 n}{d\lambda^2} + \lambda \frac{d^3 n}{d\lambda^3} \right] + \frac{3\lambda n}{2\pi^2 \lambda^2} d \frac{d\theta}{d\lambda} \left( \frac{d\theta}{d\lambda} \right)^2 \left[ 1 + \lambda \frac{d^2 \lambda}{d\theta} \right]
\] (23)

4.3.3 Brewster prism pair

In this section, a two-prism assembly is considered and arranged such that the input beam is incident at Brewster's angle on each prism. The formalism developed earlier is used to describe the behaviour of a Brewster prism pair, in the approximation of pulses with duration greater than \( \sim 20 \text{ fs} \) (for shorter pulses, TOD effects start becoming relevant) and allows computation of the GD, GDD and TOD at the center wavelength.

![Figure 29: Prism pair assembly with apex angle \( \alpha \), incident angle \( \theta_i \), diffracted angle \( \theta \) and optical path experienced by the center wavelength \( d \)](image)

The apex angle of the prism is \( \alpha \), the incidence angle \( \theta_i \) and the refracted angle \( \theta \). In order to simplify the analysis the general expressions Eqn.(21), Eqn.(22) and Eqn.(23) are split into their material and angular contribution as follows:

\[
GDD_n = \frac{\lambda^3}{2\pi^2 \lambda^2} d \frac{d^2 n}{d\lambda^2} \]
(24)

\[
GDD_\theta = -\frac{\lambda n}{2\pi^2 \lambda} d \left( \frac{d\theta}{d\lambda} \right)^2
\]
(25)

\[
TOD_n = -\frac{\lambda^4}{4\pi^2 c^3} d \left[ 3 \frac{d^2 n}{d\lambda^2} + \lambda \frac{d^3 n}{d\lambda^3} \right]
\]
(26)
where the subscript $\theta$ refers to the angular contribution and the subscript $n$ relates to the material contribution. The frequency dependent angular dependence $\theta(\omega)$ of both diffraction and refraction mechanisms is usually expressed as a $\sin\theta$ function: in the case of diffraction gratings, the grating equation is $\sin\theta = m\lambda/g - \sin\theta_i$ and in the case of refraction $\sin\theta = n_1/n_2\sin\theta_i$, where $\theta_i$ is the incident angle, $g$ is the groove density of the grating, $m$ the order of diffraction and $n_1$ and $n_2$ the refractive indices of the two media on each side of the refractive interface. The algebra can be simplified by modifying the expressions of GDD$_\theta$ and TOD$_\theta$ to account for this property. Assuming:

$$f(\lambda) = \sin\theta,$$  \hspace{1cm} (28)

is the function ruling the process involved in the dispersive line, the successive derivatives of $f(\lambda)$ can be expressed as:

$$f'(\lambda) = \cos\theta \frac{d\theta}{d\lambda}$$  \hspace{1cm} (29)

$$f''(\lambda) = \cos\theta \frac{d^2\theta}{d\lambda^2} - \sin\theta \left(\frac{d\theta}{d\lambda}\right)^2.$$  \hspace{1cm} (30)

The GDD and TOD can then be expressed as a function of $f(\lambda)$ [188]:

$$GDD_\theta = -\frac{\lambda^3}{2\pi^2} d\left(\frac{f'(\lambda)}{\cos\theta}\right)^2$$  \hspace{1cm} (31)

$$TOD_\theta = -3GDD_\theta \frac{\lambda}{2\pi} \left[1 + \lambda \frac{f''(\lambda)}{f'(\lambda)} + \lambda f''(\lambda) \frac{\sin\theta}{\cos^2\theta}\right].$$  \hspace{1cm} (32)
In the case of prism pairs, the beam incident on the first prism is typically incident near the apex; therefore, the dispersion experienced in the first prism is negligible. The deviation angle $\theta$, which sets the angular dispersion of the first prism can be calculated by applying Snell’s law at the point of impact on the prism and at its exit resulting in the following expression:

$$\sin \theta = \sin \alpha \sqrt{n(\omega)^2 - \sin^2 \theta_i} - \cos \alpha \sin \theta_i = f(\lambda)$$  \hspace{1cm} (33)

The two first derivatives of $f(\lambda)$ can be readily evaluated assuming Brewster incidence [187, 188], and the expression of the angular GDD for the air gap between the two prisms is:

$$GDD_{air} = -\frac{2\lambda^3}{\pi c^2} \left[ \frac{dn}{d\lambda} \right]^2_{\theta_o}$$  \hspace{1cm} (34)

where $d$ is the distance from prism to prism experienced by the central wavelength. The dispersion in the second prism is the sum of the material and angular dispersion. The angular dispersion is again governed by refraction, according to:

$$\sin \theta = n(\lambda) \sin \theta_2,$$  \hspace{1cm} (35)

where $\theta_2$ is the incidence angle of the center wavelength on the second prism. Again, this is set to Brewster’s angle to minimize losses, producing the following expression for the combined GDD [188]:

$$GDD_{prism2} = \frac{\lambda^3}{2\pi c^2} d_p \left[ \frac{d^2 n}{d\lambda^2} - n \left( \frac{dn}{d\lambda} \right)^2 \right]$$  \hspace{1cm} (36)

where $d_p$ is the propagation distance of the central wavelength in the prism. The first order derivative term in Eqn.(36) is typically two orders of magnitude smaller than the second order.
derivative term and therefore can be neglected in the rest of the derivation. The overall GDD is then given by the sum of GDD\textsubscript{prism2} and GDD\textsubscript{air}:

\[
GDD = \frac{\lambda^3}{2\pi c^2} \left[ d_p \frac{d^2 n}{d\lambda^2} - 4d \left( \frac{dn}{d\lambda} \right)^2 \right].
\] (37)

The second term in the expression of the GDD, corresponding to the angular dispersion, always provides negative dispersion. This term can be balanced by the first term in the bracket, corresponding to the material dispersion in the second prism, allowing the user to experimentally tune the amount of GDD provided by the prism pair by simply controlling the amount of insertion in the second prism. In general, the amount of insertion in the second prism is kept small such that the GDD remains negative.

With the GDD quantified, it is necessary to compute the TOD, as excessive TOD can result in temporal distortion of the pulse and the creation of pre-and post-pulses. The dispersion experienced by the optical pulse between the two prisms is governed by angular dispersion, therefore Eqn. \( TOD_{\theta} = -3GDD_{\theta} \frac{\lambda}{2\pi c} \left[ 1 + \frac{f''(\lambda)}{f'(\lambda)} + \lambda f''(\lambda) \frac{\sin \theta}{\cos^2 \theta} \right] \) (32) applies and the TOD acquired in air, TOD\textsubscript{air}, can be expressed as:

\[
TOD_{air} = -\frac{3\lambda}{2\pi c} GDD_{air} \left[ 1 + \frac{f''(\lambda)}{f'(\lambda)} + \lambda f''(\lambda) \frac{\sin \theta}{\cos^2 \theta} \right].
\] (38)

Expressing the derivatives of \( f(\lambda) \) in Eqn.(38) and simplifying yields the following:

\[
TOD_{air} = -\frac{3\lambda}{2\pi c} GDD_{air} \left[ 1 + \lambda \left( \frac{d^2 n}{dn^2} - \frac{dn}{dn} \left( \frac{d\lambda}{d\lambda} + 2n \frac{dn}{d\lambda} \right) \right) \right].
\] (39)
Eqn.(39) can be further simplified since the first term in the parenthesis is typically orders of magnitude larger than the two others [188], thus the expression for TOD\textsubscript{air} can be reduced to:

\[
TOD\textsubscript{air} = -\frac{3\lambda}{2\pi c} \frac{d^2 n}{dn} \left[ 1 + \lambda \frac{d\lambda^3}{d\lambda} \right].
\] (40)

The TOD after the second prism is limited to the material dispersion since the angular dispersion is negligible afterward. In the prism, the angular dispersion is minimal since all frequency-dependent optical paths are propagating almost collinearly. The TOD introduced by the material of the material of the second prism is given by:

\[
TOD\textsubscript{prism} = -\frac{\lambda^4}{4\pi^2 c^3} d_p \left[ 3 \frac{d^2 n}{d\lambda^2} + \lambda \frac{d^3 n}{d\lambda^3} \right].
\] (41)

The overall TOD for a prism pair operated at Brewster’s angle is therefore given by:

\[
TOD = -\frac{\lambda^4}{4\pi^2 c^3} d_p \left[ 3 \frac{d^2 n}{d\lambda^2} + \lambda \frac{d^3 n}{d\lambda^3} \right] - \frac{3\lambda}{2\pi c} GDD\textsubscript{air} \left[ 1 + \lambda \frac{d\lambda^3}{dn} \right].
\] (42)

For common glasses, the first term of the expression is positive while the second term is negative. The angular dispersion introduces negative TOD while the material dispersion of the second prism adds positive TOD, therefore permitting tuning of the TOD/GDD ratio. This valuable feature is discussed in more details in section (4.5).

The performance of two prism pairs (fused silica prisms and SF10 prisms) operated at Brewster’s angle have been numerically investigated to provide orders of magnitude
expectations for GDD and TOD of a dispersive line. The variations of GDD and TOD are linear with prism separation and with the insertion of the second prism as shown in Figure 30.

![Graph showing GDD and TOD](image)

Figure 30: a. Computed GDD and TOD for a center wavelength $\lambda_0 = 850$ nm for a fused silica prism pair operated at Brewster’s angle as a function of apex to apex separation; b. Computed GDD and TOD for a center wavelength $\lambda_0 = 850$ nm for an SF10 prism pair operated at Brewster’s angle as a function of apex to apex separations.

The SF10 prism pair induces significantly larger values of GDD and TOD than the fused silica pair, as well as a larger GDD/TOD ratio. This ratio is limited to 1.1 for a fused silica pair and reaches up to 2.5 for an SF10 pair. Fused silica pairs are typically used in Ti:Sapphire oscillators to compensate for intra-cavity dispersion since they introduce little third order dispersion. The model developed earlier shows that the TOD/GDD ratio can be adjusted by increasing the insertion of the second prism into the beam path. This feature has been investigated for the fused silica and SF10 prism pair by adding extra glass from the second prism (Figure 31).
This simulation was performed assuming a 20 cm separation between the prisms to minimize the effects of angular dispersion and make the stretcher sensitive to the material insertion. The TOD/GDD ratio can be tuned by ~20% in the case of fused silica while it can only be tuned by ~10% in the case of SF10. This lack of tuning in the case of SF10 – even for a short apex to apex distance – is attributed to the strong dominance of angular dispersion in such a dispersive line.

### 4.3.4 Treacy grating pair

A Treacy grating pair consists of one pair of identical gratings placed parallel to each other. The input pulse impinging on the first grating is diffracted into the first order. The diffracted beam from the first grating propagates to the second grating, where it is diffracted again in the first order but with opposite sign to that of the first grating. This now collimated beam is then retro-reflected into the apparatus with a vertical offset for the second identical length path and exits via a pick-off mirror placed close to the entrance of the setup.
The angular separation of the optical spectrum causes a path difference that generates the spectral phase required to stretch or compress the optical pulse. The sign of the spectral phase (or the frequency dependent optical path difference) can be reversed by inserting a 4f telescope between the two gratings (see section 2.1.1). A model similar to that proposed for the Brewster prism pair can be developed using the same mathematical tools [187]. All general formulae derived in section 4.3.1 can be applied here in the case of a grating compressor, the grating equation rules the diffraction experienced by the incoming pulses:

$$\sin \theta = m \frac{\lambda}{g} - \sin \theta_1 = f(\lambda),$$  \hspace{1cm} (43)

where $\theta_1$ is the incident angle of the beam onto the first grating, $m$ is the diffraction order, $g$ is the groove spacing of the grating and $\theta$ is the angle at which the wavelength $\lambda$ is diffracted. The dispersion experienced in a grating pair is exclusively angular which simplifies the algebra. Furthermore, the form of $f(\lambda)$ is such that its first and second derivative are readily expressed by:

$$f'(\lambda) = \frac{1}{g} \hspace{1cm} (44)$$

$$f''(\lambda) = 0. \hspace{1cm} (45)$$

Recalling Eqn.(31) and Eqn.(32), the GDD of a grating pair can be expressed by:
\[ GDD_{\text{grating}} = -\frac{\lambda^3}{2 \pi c^2} \frac{d}{g^2 \cos^2 \theta}. \] (46)

The TOD follows by using the equation for TOD\(_0\):

\[ TOD_{\text{grating}} = -\frac{3\lambda}{2\pi c} GDD_{\text{grating}} \left[ 1 + \frac{\lambda}{g} \frac{1}{\cos^2 \theta} \right]. \] (47)

The sign of \(GDD_{\text{grating}}\) is always negative, therefore a grating pair will always provide negative dispersion, consistent with the general formula of angular dispersion derived in section 4.3.2. The sign of \(TOD_{\text{grating}}\) requires more care to be determined [188]: for \(TOD_{\text{grating}}\) to be negative, the term in brackets needs to be negative as well which means:

\[ \frac{\lambda}{g} \frac{1}{\cos^2 \theta} < -1 \iff -1 + \frac{m\lambda}{g} \sin \theta + \sin^2 \theta < 0 \] (48)

Solving this polynomial by setting \(X = \sin \theta\) yields two real solutions [188]:

\[ X = \sin \theta = \frac{1}{2} \left( \frac{m\lambda}{g} \pm \sqrt{1 + m^2 \left( \frac{\lambda}{2g} \right)^2} \right) \] (49)

In order for these solutions to be valid and enable negative TOD from a grating pair, they need to satisfy the grating equation. Substituting into the grating equation yields:

\[ \sin \theta = \sin \theta_i \pm 2 \sqrt{1 + m^2 \left( \frac{\lambda}{2g} \right)^2} \] (50)

The term under the square root is always greater than 1, which results in complex angles for \(\theta\); thus a grating pair produces only positive third order dispersion for any incident angle, any grating groove density and any diffracted order. Similar to the case of the prism pair, a numerical simulation has been performed to investigate some features of a grating pair and provide orders of magnitude expectations for GDD, TOD and TOD/GDD ratio provided by such dispersive line.
The amount of GDD and TOD provided by a grating pair exceeds that provided by a prism pair by at least two orders of magnitude for a comparable footprint. The main adjustment available to the user when operating a grating pair is the grating separation distance: it is numerically confirmed that the evolution of both GDD and TOD is linear with grating separation and the TOD/GDD ratio is constant. Another parameter available experimentally, yet more challenging to adjust practically, is the angle of incidence on the first grating. Increasing the incidence angle on the first grating results in wider angular spreading of the optical frequencies.
and in a larger magnitude GDD. Even though the increase in GDD with angle is linear, the
TOD/GDD ratio decreases as the incidence angle is increased allowing tuning of the TOD/GDD
ratio. Finally, an increase of the grating groove density results in larger magnitude GDD and the
TOD/GDD ratio also increases in magnitude as the groove density is increased suggesting that
the TOD increases faster than the GDD.

4.4 Solutions for efficient stretching and compressing of ultra-broadband pulses: model and
prospects

The previous section describes techniques that are suitable for stretching and
compressing pulses with spectra as wide as ∼50 to 100 nm FWHM, or correspondingly pulses
with duration ≥ 20 fs. For such pulses, managing the GDD is sufficient to obtain pulses close to
the transform limit, higher dispersion orders are of minor importance since the term \((\omega-\omega_0)^n\) in
the Taylor expansion of the spectral phase (Eqn. (9)) are negligible. For spectra with bandwidth
exceeding ∼100 nm the effects of the higher order terms become sizable in the temporal domain
if they are not properly compensated for. The effects of residual spectral phase on pulses with
100 nm and 200 nm FWHM spectral bandwidth are compared and illustrated on Figure 34. All
computations are performed at a central wavelength of 850 nm.
A residual GDD of 100 fs$^2$ and a residual TOD of 100 fs$^3$ have been applied to a pulse with a FWHM spectral width of 100 nm and a pulse with FWHM spectral width of 200 nm. The 100 nm wide spectrum results in a transform-limited pulse of duration of ~20 fs. A residual 100 fs$^2$ of uncompensated GDD yields a 50% increase in pulse duration. The effects of TOD on this same pulse are negligible in terms of pulse duration, only a slight steepening of the trailing edge of the pulse can be observed. The spectrum with 200 nm of spectral width on the other hand,
results in a transform-limited pulse of 5 fs duration. A residual GDD of 100 fs\(^2\), results in an increase in pulse duration of 1200\%, an effect much more dramatic than that observed for a 20 fs pulse. The effects of TOD, negligible in the case of the 20 fs transform-limited pulse, results in the creation of pre-pulses in the case of an ultra-broadband pulse. The magnitude of the first pre-pulse reaches 10\% of the peak amplitude of the main pulse, an unacceptable feature for many high-energy applications. The number of pre-pulses and their magnitude increase with the amount of residual TOD. Simulations were performed to investigate the effects of residual fourth order dispersion: as expected, these effects are not visible in the case of the 20 fs pulse and are essentially negligible for the 5 fs pulse, up to values as large as \(10^5\) fs\(^4\).

Compensating the residual TOD and GDD is crucial to reaching high peak power with good temporal behavior. It is also necessary to have efficient compression to reach high output peak power, as mentioned in section 4.2. Grating pairs are ruled out when trying to achieve efficient pulse compression since they offer typically 70\% throughput. Prism pairs can be implemented as compressor but suffer from the low GDD provided by the arrangement as shown in 4.3.3. Schemes based on two triplets of prisms have been proposed and exhibit a factor of 9 increase in GDD compared to a simple pair of prism for the same apex to apex separation at the cost of an increased complexity [189]. Using the normal dispersion of a piece of glass to cancel out the GDD and TOD provided by the stretcher is an elegant approach for systems with energies up to a few tens of millijoules. At 800 nm center wavelength, most common glass exhibit positive GDD and positive TOD and the ratio between these two values is \(~0.7\) fs for all common glasses. There is therefore a need for a stretcher that can offer negative GDD and negative TOD and such that TOD/GDD = 0.7 fs. From the previous investigations of prism and grating pairs, neither of these dispersive lines enables such ratio to be readily achieved.
Several approaches have been proposed to resolve this issue: grism pair stretchers combining the flexibility of prism pairs and the stretching efficiency of gratings have been proposed and show potential at achieving negative GDD and negative TOD in large amounts [183, 185, 186]. Chirped mirrors have also been developed and provide GDD and TOD without additional loss and reduced footprint [190]. However, chirped mirrors provide relatively small amounts of GDD and TOD, and offer no freedom of adjustment since the user has no access to the GDD, the TOD and the TOD/GDD ratio. Their use is typically limited to fine-tuning of the total compression or compensation by windows placed in the beam. An additional device that has been developed is the acousto-optic programmable dispersion filter (AOPDF or Dazzler) that offers exceptional flexibility since it enables adaptive control of the GDD, TOD and FOD within a few $\text{fs}^2$, $\text{fs}^3$ and $\text{fs}^4$ respectively [191-195]. A typical Dazzler such as the one implemented in HERACLES can provide group delays up to ~1.5 ps over 250 nm of bandwidth at the expense of throughput. An AOPDF operated at its maximum capacities can have a throughput as low as 10%. The following sections describes in detail the grism pair and provides a simple approach to AOPDFs since both devices are implemented on HERACLES. Chirped mirrors are also mentioned since they are important components of the ultrafast toolbox.

4.4.1 Grism pair semi-analytical model

A simple model based on analytical ray-tracing is proposed to investigate the performances of a grism pair stretcher for down chirping ultra-broadband pulses. The effects of misalignment are quantitatively investigated, and the advantages of a grism pair stretcher over a traditional Martinez design are discussed for the case of ultra-broadband pulses. A simple
analytical ray-tracing model that can predict the performances of a grism stretcher, including the effects of experimental misalignment of the system is detailed.

The grism pair consists of a set of two identical prisms and two identical gratings. Each grating can be optically contacted to the hypotenuse of a prism or simply brought close and parallel to it. The design modeled in this paper is shown in Figure 35.

A Cartesian set of coordinates is chosen having its origin at the apex of the lower grism. The $x$-axis is along the interface between the lower grism and the gap between the two grisms. In these coordinates, the refracting and diffracting surfaces are treated mathematically as lines. The beams of different wavelength are also treated as lines and the coordinates of the intersection of each beam with each interface are calculated. The optical path can then be calculated for each wavelength between each surface and the resulting optical path of the system can be calculated, yielding an expression for the spectral phase introduced by the grism arrangement. The input beam on the lower grism has an arbitrary incident angle $\theta_i$. The refracted angle $\theta_r$ into the prism is obtained via Snell’s law, $\theta_r = \sin^{-1}(n(\omega) \sin(\theta_i))$, where $n(\omega)$ is the frequency dependent refractive index in the prism. By definition of the coordinate system, the equation of the input
interface is $y = 0$. The equation of the line representing the grating surface is given by $y = x(\tan(\alpha))$, where $\alpha$ is the apex angle of the prism. The coordinates of $A(x_A, y_A)$, the intersection between the input beam and the input surface, are simply $A(U, 0)$, where $U$ is the distance between the apex of the lower grism and the input beam location. The equation for the beam propagating from the input interface to the grating is given by:

$$y = x \tan\left(-\left(\frac{\pi}{2} - \theta_r(\omega)\right)\right) + U \tan\left(\frac{\pi}{2} - \theta_r(\omega)\right) = \frac{1}{\tan \theta_r(\omega)}(x - U).$$

(51)

The location $B(x_B, y_B)$ of the beam on the grating, as a function of frequency, is found by equating Eqn.(51) and the equation of the grating surface.

$$x_B(\omega) = \frac{U}{1 + \tan \alpha \tan \theta_r(\omega)}$$

$$y_B(\omega) = \frac{U \tan \alpha}{1 + \tan \alpha \tan \theta_r(\omega)}$$

(52)

Assuming that the grating is in contact with the prism, the incident angle on the grating is $\theta + \alpha$, the diffracted angle $\theta_d$ is found via the grating equation

$$\theta_d(\omega) = \arcsin\left(\sin(\theta_r(\omega) + \alpha) - \frac{2\pi}{\omega n(\omega) d}\right),$$

(53)

where $d$ is the groove density of the grating. The expression of the line $(BC)$ representing the beam propagating from the grating $B(x_B, y_B)$ to the interface prism/air gap $C(x_C, y_C)$ is expressed in an $(x', y')$ coordinate system rotated from the original $(x, y)$ coordinate by $\alpha$, such that $x'$ is along the axis of the grating surface and rotated back to the $(x, y)$ coordinate system via a usual rotation matrix. The equation of the line $(BC)$ is given by:
The coordinates \( C(x_C, y_C) \) of the intersection between the diffracted rays from the grating and the interface prism/air gap are obtained by equating Eqn.(54) to \( y = 0 \), the equation of the interface.

\[
x_C(\omega) = \frac{y_b(\omega)\sin(\alpha) - x_b(\omega)\cos(\alpha)}{\cos(\alpha) + \tan(\theta_d(\omega))\sin(\alpha)}  
\]

(55)

\( y_C = 0 \).

The refracted angle \( \theta_g \) through the interface is obtained using Snell’s law, \( \theta_g = \sin(n(\omega)\sin(\theta_g + \alpha)) \). The width \( G \) of the gap between the two grisms is conveniently defined as the normal distance between the interface prism/air gap of each prism, therefore the equation for the line representing the interface air gap/prism for the upper grism is simply given by \( y = G \).

The equation for the beam propagating between the two prisms is given by:

\[
y = \tan\left(\frac{\pi}{2} - \theta_g(\omega)\right)x - x_C \tan\left(\frac{\pi}{2} - \theta_g(\omega)\right).  
\]

(56)

The coordinates \( D(x_D, y_D) \) of the intersection between the refracted rays from the lower grism to the upper grism is obtained by equating Eqn.(56) to \( y = G \).

\[
x_D(\omega) = G \tan(\theta_g(\omega)) + x_C
\]

\[
y_D = G
\]

(57)

The refracted angle \( \theta_n \) through the interface can again be readily calculated using Snell’s law, \( \theta_n = \sin(\sin(\theta_g)/n(\omega)) \). The distance between the \( y \)-axis and the tip of the upper prism is \( x = S \) and the length of the hypotenuse of both prisms is \( H \). The equation of the line representing the surface of the grating is given by:
\[ y = -x \tan \alpha + G + \left( h + S \right) \tan \alpha. \]  
\[ (58) \]

The equation for the beam propagating from \( D(x_D, y_D) \) on the interface air gap/prism of the upper prism to the grating is given by:

\[ y = \frac{1}{\tan \theta_n(\omega)} \left( x - x_D(\omega) \right) + G. \]
\[ (59) \]

The wavelength dependent location \( E(x_E, y_E) \) of the beam on the grating can be obtained by equating Eqn.(58) and Eqn.(59):

\[ x_E(\omega) = \frac{(h + S) \tan \alpha + x_E \tan \left( \frac{\pi}{2} - \theta_n(\omega) \right)}{\tan \alpha + \tan \left( \frac{\pi}{2} - \theta_n(\omega) \right)} \]
\[ (60) \]

\[ y_E(\omega) = -x_E(\omega) \tan \alpha + G + (h + S) \tan \alpha. \]

The input angle on the second grating is \( \theta_{g2} = \alpha - \theta_m \) and the diffraction angle \( \theta_{d2} \) from the grating is obtained via the grating equation \( \theta_{d2} = \sin^{-1}(\sin \theta_{g2} - 2 \pi/(\omega n(\omega)d)) \). The equation of the line representing the beam propagating from \( E(x_E, y_E) \) to the prism/air interface is given by:

\[ y = -x \tan \left( \frac{\pi}{2} + \theta_{d2} + \alpha \right) + x_E \tan \left( \frac{\pi}{2} + \theta_{d2} + \alpha \right) + Y_E. \]
\[ (61) \]

The location of the intersection of the beam with the prism/air interface is given, as a function of frequency, by:

\[ x_{E'}(\omega) = \frac{y_E(\omega) - G + x_E(\omega) \tan \left( \frac{\pi}{2} + \theta_{d2}(\omega) + \alpha \right)}{\tan \left( \frac{\pi}{2} + \theta_{d2}(\omega) + \alpha \right)} \]
\[ (62) \]

\[ y_{E'} = G. \]
For practical designs it is desirable to propagate the beam to a retro-reflector and back into the grism sequence. In our model the retro-reflector is modelled by a line that can be translated along the y-axis by an amount $N$. In order to ensure that the retro-reflector is normal to the output beam, the line equation representing it is given by:

$$y = x \tan \theta_i + N. \quad (63)$$

The beam refracted at the prism/air interface emerges from the prism with an angle $\theta_{\text{out}} = \sin(n(\omega)\sin(\theta_d + \alpha))$. The equation of the beam propagating from the output of the prism to the mirror is then obtained:

$$y = -x \tan \left( \frac{\pi}{2} + \theta_{\text{out}} \right) + x_F \tan \left( \frac{\pi}{2} + \theta_{\text{out}} \right) + G. \quad (64)$$

The location of the different beam on the retro-reflector $H(x_H, y_H)$ can be expressed, as a function of frequency by:

$$x_H(\omega) = \frac{x_F(\omega) \tan \left( \frac{\pi}{2} + \theta_{\text{out}}(\omega) \right) + G - N}{\tan \theta_i(\omega) + \tan \left( \frac{\pi}{2} + \theta_{\text{out}}(\omega) \right)} \quad (65)$$

$$y_H(\omega) = x_H \tan \theta_i(\omega) + N.$$

From the knowledge of the location of every frequency dependent intersection between beams and interfaces, the distances travelled by each wavelength throughout the system can be calculated by $IJ = \sqrt{(x_i - x_j)^2 + (y_i - y_j)^2}$ where $I$ and $J$ are any points in the system. The overall spectral phase generated by the optical path difference can then be calculated by:

$$\phi_{\text{OPD}}(\omega) = \frac{\alpha}{c} (n(\omega)AB + n(\omega)BC + CD + n(\omega)DE + n(\omega)EF + FH) \quad (66)$$
In order to accurately describe the overall spectral phase, a phase shift needs to be added to the red wavelength upon reflection on the grating [34]. This can be readily achieved by calculating the extent of the beam, $K$, on the second grating, dividing this extent by the groove separation and multiplying by $2\pi$. This extra phase term is then added to Eqn.(66). The total spectral phase after a round trip through the system is then given by:

$$
\phi(\omega) = 2\left(\phi_{\text{OPD}} - 2\pi \frac{K}{d}\right)
$$

(67)

The group delay, GDD, TOD and FOD are obtained by taking consecutive numerical derivatives of $\phi(\omega)$. A set of simulations, representing a parametric study of the main design parameters of a grism pair, have been performed by implementing the previously derived equations in MATLAB including the material of the prisms, the apex angle of the prisms and the groove density of the gratings [151].

4.4.1.1 Influence of grating groove density

Experimental parameters related to the HERACLES design were chosen to perform the simulations: in order to stretch a 5 fs pulse with a central wavelength of 850 nm to 20 ps, an amount of GDD ~35000 fs$^2$ is needed. Therefore, for each groove density, the GDD was adjusted to that value by compensating the separation $G$ between the two grisms and the variations of the values of TOD and FOD were recorded. The effect of grating groove density is investigated by choosing a pair of fused silica gratings for with a grating groove density of 300, 400 and 500 l/mm.
Table 8: computations of GDD, TOD and FOD for fused silica grism pairs with an apex angle $\alpha = 23.2^\circ$ and featuring 300 l/mm, 400 l/mm and 500 l/mm gratings and adjusted to provide 35000 fs$^2$ of GDD

<table>
<thead>
<tr>
<th></th>
<th>300 l/mm</th>
<th>400 l/mm</th>
<th>500 l/mm</th>
</tr>
</thead>
<tbody>
<tr>
<td>G (cm)</td>
<td>6.2</td>
<td>2.95</td>
<td>1.42</td>
</tr>
<tr>
<td>GDD (fs$^3$)</td>
<td>-3.5x10$^4$</td>
<td>-3.5x10$^4$</td>
<td>-3.5x10$^4$</td>
</tr>
<tr>
<td>TOD (fs$^3$)</td>
<td>-8.9x10$^4$</td>
<td>-1x10$^5$</td>
<td>-1.1x10$^5$</td>
</tr>
<tr>
<td>FOD (fs$^4$)</td>
<td>4.5x10$^4$</td>
<td>2.8x10$^4$</td>
<td>2x10$^4$</td>
</tr>
</tbody>
</table>

All configurations provide negative GDD and negative TOD a feature necessary for successful lossless compression through common glass. The relatively large positive FOD presents a problem since it cannot readily be compensated for in a bulk glass compressor. The 400 l/mm and 500 l/mm stretcher designs exhibit TOD/GDD ratio significantly larger (2.85 and 3.14 respectively) than that of the 300 l/mm grism pair (2.54), making the latter more suitable for glass compression. Furthermore, the reduced footprint of the 400 l/mm and 500 l/mm designs, though apparently attractive, is impractical for laboratory implementations: an excessively reduced footprint results in beam clipping and makes it impossible for the pulse to exit the stretcher. The 300 l/mm option is therefore the most suitable for practical implementation. Nevertheless, such low groove density grating reduces diffraction efficiency for the near infrared pulse and results in reduced diffraction efficiency, as the diffraction is most efficient when the wavelength of the incoming light approaches the groove spacing (the optimum for 800 nm being ~1200 l/mm). Such losses can nonetheless be afforded since they can be compensated by the large gain provided in the optical parametric amplifiers.
4.4.1.2 Influence of apex angle

For this study, the prism material was chosen to be fused silica, and the grating groove density was set to 300 l/mm based on the results of the previous section. The apex angle was adjusted from 23.2° to 23.8°. The choice of 23.2° as a lower bound was dictated by the shape of the group delay curve which started increasing dramatically for wavelengths beyond 900 nm (see 4.5.1.3 for investigation of grism pair behavior away from the central wavelength).

Table 9: computations of GDD, TOD and FOD for a fused silica grism pairs featuring 300 l/mm gratings with apex angles of $\alpha = 23.2, 23.5$ and 23.8° and adjusted to provide 35000 fs$^2$ of GDD

<table>
<thead>
<tr>
<th></th>
<th>$\alpha=23.2^\circ$</th>
<th>$\alpha=23.5^\circ$</th>
<th>$\alpha=23.8^\circ$</th>
</tr>
</thead>
<tbody>
<tr>
<td>G(cm)</td>
<td>6.2</td>
<td>3.8</td>
<td>2.4</td>
</tr>
<tr>
<td>GDD (fs$^2$)</td>
<td>-3.5x10$^4$</td>
<td>-3.5x10$^4$</td>
<td>-3.5x10$^4$</td>
</tr>
<tr>
<td>TOD (fs$^3$)</td>
<td>-8.9x10$^4$</td>
<td>-7.4x10$^4$</td>
<td>-6.6x10$^4$</td>
</tr>
<tr>
<td>FOD (fs$^4$)</td>
<td>4.5x10$^4$</td>
<td>1.5x10$^4$</td>
<td>-1.35x10$^4$</td>
</tr>
</tbody>
</table>

Increasing the apex angle results in a decrease in TOD and a reduction of the TOD/GDD ratio, of 2.5, 2.1 and 1.88 for $\alpha = 23.2^\circ, 23.5^\circ$ and $23.8^\circ$ respectively. This reduction in TOD/GDD is accompanied by a drastic reduction in footprint making setups with apex angles larger than 23.2° impractical. Notice that only for large apex angle do the FOD starts to become negative in fused silica grisms.

4.4.1.3 Influence of material choice

Similar numerical parameters and similar approach are used for investigating the effects of the choice of the material on the grism pair performances. The materials chosen for this
A parametric study is performed using a low dispersion glass (fused silica), a mildly dispersive glass (SF11), and a highly dispersive glass (SF58). The apex angle for this simulation was chosen to be the smallest angle allowing monotonous behavior of the group delay (see 4.5.1.3 for investigation of grism pair behavior away from the central wavelength). The grating groove density was set to 300 l/mm. The values of GDD, TOD, and FOD are reported in Table 10.

Table 10: Computations of GDD, TOD, and FOD for grism pairs featuring 300 l/mm, made of fused silica, SF11, and SF58 glass and adjusted to provide 35000 fs$^2$ of GDD

<table>
<thead>
<tr>
<th></th>
<th>Fused. Silica</th>
<th>SF11</th>
<th>SF58</th>
</tr>
</thead>
<tbody>
<tr>
<td>$G (cm)$</td>
<td>6.2</td>
<td>5.3</td>
<td>6.1</td>
</tr>
<tr>
<td>GDD (fs$^2$)</td>
<td>$-3.5\times10^4$</td>
<td>$-3.5\times10^4$</td>
<td>$-3.5\times10^4$</td>
</tr>
<tr>
<td>TOD (fs$^3$)</td>
<td>$-8.9\times10^4$</td>
<td>$-9.5\times10^4$</td>
<td>$-1.1\times10^5$</td>
</tr>
<tr>
<td>FOD (fs$^4$)</td>
<td>$4.5\times10^4$</td>
<td>$-1.9\times10^4$</td>
<td>$-2.6\times10^4$</td>
</tr>
</tbody>
</table>

Fused silica offers the smallest amount of TOD, a valuable feature since the TOD/GDD ratio needs to be as close as possible to 0.7 fs to ensure compression in bulk glass. Notice that grism pairs alone do not allow for such ratio to be reached with this set of parameters, for any material. Fused silica though offers an attractive TOD/GDD ratio and exhibits positive FOD, a feature incompatible with bulk glass compression since most glass exhibit positive FOD in the NIR (fused silica is a noticeable exception but is a poor candidate for bulk glass compression due to its limited dispersive power as discussed in section 4.4.3). SF58, the most dispersive glass of the investigated set offers negative dispersion at all orders but provides a large TOD/GDD ratio which is incompatible with bulk compression. The mildly dispersive glass, SF11, appears to be the best choice, it offers negative dispersion at all orders and mitigates the TOD/GDD ratio. Also notice that it offers the most compact, yet practically implementable footprint. In order to
investigate the validity of this study, similar simulations have been performed with BK7 (a low dispersion glass), SF10 (exhibiting dispersion power close to SF11) and SF57 (close to SF58) and similar trends were observed. The HERACLES grism pair features two SF11 prisms with apex angle $\alpha = 18^\circ$ and gratings with a groove density of 300 l/mm (highlighted column in Table 10).

### 4.4.1.4 Influence of misalignment

The formalism developed to describe the grism pair can readily handle the addition of a misalignment factor (by adding an error $\Delta \alpha$ or $\Delta \theta$). The error $\Delta \alpha$ is applied to the input angle onto the first grism, while $\Delta \theta$ is an error in the parallelism of the gratings. Assuming an optimum behavior for normal incidence, the effect of an experimental error $\Delta \alpha = \pm 0.5^\circ$ are investigated. Results are reported in Table 11.

Table 11: computations of GDD, TOD and FOD for an SF11 grism pairs featuring 300 l/mm gratings and adjusted to provide 35000 fs$^2$ of GDD assuming alignment error of the input beam

<table>
<thead>
<tr>
<th></th>
<th>$\Delta \alpha = 0^\circ$</th>
<th>$\Delta \alpha = 0.5^\circ$</th>
<th>$\Delta \alpha = -0.5^\circ$</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>G(cm)</strong></td>
<td>5.3</td>
<td>5.3</td>
<td>5.3</td>
</tr>
<tr>
<td><strong>GDD (fs$^2$)</strong></td>
<td>$-3.5x10^4$</td>
<td>$-4.6x10^4$</td>
<td>$-2.6x10^4$</td>
</tr>
<tr>
<td><strong>TOD (fs$^3$)</strong></td>
<td>$-9.5x10^4$</td>
<td>$-1.1x10^5$</td>
<td>$-8.6x10^5$</td>
</tr>
<tr>
<td><strong>FOD (fs$^4$)</strong></td>
<td>$-1.9x10^4$</td>
<td>$-3.7x10^4$</td>
<td>$-5.0x10^3$</td>
</tr>
</tbody>
</table>

The alignment of the input is critical to reach the design dispersion: a $0.5^\circ$ error on the alignment of the input beam results in $\sim$30% variation in GDD and dramatic increase in TOD. Consequently, the TOD/GDD ratio is affected by the error and therefore, pulse compression...
down to the a few percent of transform becomes impossible in a glass compressor. The effects of slight misalignment in the parallelism of the grisms (Δθ) are also investigated and results are reported in Table 12.

Table 12: computations of GDD, TOD and FOD for an SF11 grism pairs featuring 300 l/mm gratings and adjusted to provide 35000 fs² of GDD assuming slight misalignment of the parallelism of the grisms

<table>
<thead>
<tr>
<th>Δθ</th>
<th>G(cm)</th>
<th>GDD (fs³)</th>
<th>TOD (fs⁴)</th>
<th>FOD (fs⁵)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0°</td>
<td>5.3</td>
<td>-3.5x10⁴</td>
<td>-9.5x10⁴</td>
<td>-1.9x10⁴</td>
</tr>
<tr>
<td>0.5°</td>
<td>5.3</td>
<td>-3.9x10⁴</td>
<td>-9.8x10⁵</td>
<td>-2.5x10⁴</td>
</tr>
<tr>
<td>-0.5°</td>
<td>5.3</td>
<td>-3.2x10⁴</td>
<td>-9.3x10⁵</td>
<td>-1.4x10⁴</td>
</tr>
</tbody>
</table>

The parallelism of the grating has little effect on the dispersion orders; however this apparent neutrality is limited to time domain dispersion. Misalignment in the parallelism of the gratings results in spatial chirp, resulting in beam profile with a gradient of wavelength across the beam. Such gradient was experimentally found to be highly detrimental to successful optical parametric amplification.

4.4.2 Acousto-optic programmable dispersion filter

The acousto-optic programmable dispersion filter (AOPDF) introduced by Tournois in 1997 relies on the interaction between an acoustic wave and an optical wave in a birefringent crystal [192, 196-198]. The principle of operation can be summarized as follows: a chirped acoustic wave is launched into a birefringent crystal; the strain induced by the acoustic wave in the crystal creates a transient diffraction grating by modulating the index of refraction of the
material; this transient grating interacts with the optical wave simultaneously launched into the crystal which can be expressed mathematically by:

\[ S_1(\omega_1) e^{i(\omega_1 t - k_1 x)} \times S_{ac}(\Omega) e^{i(\Omega t - K x)} = S_2(\omega_2) e^{i(\omega_2 t - k_2 x)}, \]

where \( S_1(\omega_1) \), \( S_2(\omega_2) \) and \( S_{ac}(\Omega) \) are the complex spectral amplitude of the incoming pulse, the diffracted pulse and the acoustic wave respectively. The wave vector \( k_1 \) corresponds to the incoming optical wave, \( k_2 \), to the diffracted wave and \( K \) to the acoustic-wave. The speed of propagation of the optical wave exceeds that of the acoustic wave by several orders of magnitude, thus the grating generated by the acoustic wave appears static to the optical wave. The energy transfer from the wave at \( \omega_1 \) to the wave at \( \omega_2 \) occurs when the interaction between the three waves is phase matched such that:

\[ \omega_2 = \omega_1 + \Omega \]
\[ k_2 = k_1 + K \]

Phase-matching between the acoustic wave and the optical wave occurs at different depths in the crystal for different wavelengths due to the chirped nature of both pulses. The frequency of the acoustic wave is chosen such that upon phase-matching, the extra momentum provided by the acoustic wave allows phase-matching of the fast and slow axis of the birefringent crystal for the wavelength being diffracted. Such phase-matching can be represented as shown in Figure 36:
Figure 36: Phase-matching diagram of the interaction between a tailored acoustic wave and an optical wave in an acousto-optic programmable dispersion filter.

Upon diffraction, coupling coefficients in the photo-elastic tensor allow efficient transfer of the polarization axis from the fast axis to the slow axis. Figure 37 is a schematic of the principle of operation of an acousto-optic programmable dispersion filter [199].

Figure 37: Principle of operation of an acousto-optic programmable dispersion filter: wavelengths making up the input chirped pulse are switched from the fast-axis to the slow-axis of the birefringent crystal allowing the introduction of an arbitrary spectral phase upon the input pulse to cancel out the initial chirp [199].

When a spectral portion of the optical wave is phase matched with the acoustic wave, the wave is diffracted and switches polarization from the fast axis to the slow axis of the crystal, therefore introducing a programmable frequency dependent delay in the optical spectrum.
This section presents a basic approach to the theory of chirped mirrors. Accurate mathematical models have been developed [200-202] and are used to optimize the performances of chirped mirrors but will not be developed here. A review of the progress and successive improvement of chirped mirrors will also be presented in this section.

The basic idea of chirped mirrors consist in varying the reflectivity of the mirror along its depth [203, 204]. Spectral chirp is added to an incident pulse by adjusting the depth of penetration of the different wavelength contained in the pulse. Figure 38 shows a basic representation of the first implementation of chirped mirrors. Note that chirped mirrors are simply a special implementation of Bragg stack dielectric mirrors made of alternating layers of a high and low index of refraction [205]. In the case of chirped mirrors alternating layers of TiO$_2$ and SiO$_2$ are typically used.

![penetration depth dependence](image)

Figure 38: Representation of a chirped mirror showing the depth-dependent reflectivity of the mirror [206]

The group delay introduced by the chirped mirror can be evaluated by taking the derivative of the spectral phase introduced by the mirror. As mentioned earlier, each wavelength
experiences a different optical path in the mirror, which generates a spectral phase that can be defined as follows:

\[ \phi(\omega) = -\frac{2n}{c} \int_{\omega_0}^{\omega} L(\omega')d\omega', \quad (70) \]

where \( n \) can be approximated in this simple model by the average of the high index and the low index of refraction of the layers constituting the Bragg stack, \( n = \frac{1}{2}(n_{\text{low}} + n_{\text{high}}) \), \( L(\omega') \) is the optical path experienced by the frequency \( \omega' \) and \( \Omega_0 \) is the lowest frequency reflected by the mirror. Similarly, \( \Omega_1 \) can be defined as the highest frequency reflected by the mirror and is used in the forthcoming calculations. The GD introduced by the mirror can thus be written as the derivative of the spectral phase.

\[ GD(\omega) = \frac{2n}{c} L(\omega) \quad (71) \]

Models introducing nonlinear chirp are typically calculated via algorithms implemented on computers due to their complexity. The chirped mirrors implemented in the Octavius-85, the front end Ti:Sapphire oscillator of HERACLES feature over 100 variables that need to be adjusted to reach a successful design.

For illustration purposes, the following describes a chirped mirror that would compensate for linear dispersion. Note that proper engineering of \( L(\omega) \) is a key to the dispersion compensation via chirped mirrors. For constant dispersion compensation, the function \( L(\omega) \), representing the optical path length experienced by the frequency \( \omega \), should be defined as follow.

\[ L(\omega) = L_{\text{max}} \frac{\omega - \Omega_0}{\Omega_1 - \Omega_0}, \quad (72) \]
where $L_{\text{max}}$ is the optical path experienced by $\Omega_1$, the frequency travelling the fastest. The GDD obtained with such a linear dependence is a constant.

$$GDD = \frac{2}{\Omega_1 - \Omega_0} \frac{nL_{\text{max}}}{c}$$  \hspace{1cm} (73)

It can be observed that an $\omega^n$ dependence of $L(\omega)$ would lead to an $\omega^{n-1}$ dependence of the GDD, therefore allowing for compensation of nonlinear dispersion. Computer algorithms can therefore design the Bragg stack by adjusting the parameters such as layer thickness, duty cycle, number of layers and depth of each layer to obtain the targeted function $L(\omega)$.

The first implementations of chirped mirrors therefore consisted of alternative layers of high and low index of refraction. In order to produce an efficient dielectric mirror having a peak reflectivity for the wavelength $\lambda$, the thickness of the layers should be $\lambda/4$. Therefore, varying the thickness of the alternating layers with the depth of the mirror allows reflection of different wavelengths at different depth. Unfortunately, such implementation led to unwanted fluctuations in the GDD that created a significant drift from the targeted function $L(\omega)$ [206]. The cause of these oscillations is due to the Gires Tournois effect in the mirror leading to variation in the GDD.

A Gires Tournois Interferometer (GTI) is a particular type of Fabry Perot interferometer. It is made from a partially reflective surface and a highly reflective surface. Similar to a Fabry Perot interferometer, a standing wave resonates between the two surfaces and a wavelength dependent phase is generated leading to constructive and destructive interferences for different wavelengths thus leading to strong wavelength dependent oscillations in the GDD. Figure 39 shows the typical wavelength dependence of the GDD of a GTI [157]. Gires Tournois Interferometers have been successfully used to recompress mode-locked pulses from lasers.
having a narrow bandwidth such as Nd:Glass but are not appropriate for broadband sources since the variations of GDD are very pronounced and occur frequently.

Figure 39. Evolution of the GDD in a Gires Tournois Interferometer [157]

The design of chirped mirrors led to a Gires Tournois effect within the structure of the mirror itself. The reflectivity at an interface between two media with index of refraction \( n_1 \) and \( n_2 \) is given by equation Eqn.(74).

\[
R = \left( \frac{n_2 - n_1}{n_2 + n_1} \right)^2
\]  

(74)

The reflectivity of the interface between the air and the first layer acts as a partial reflector while the light reflected by the underlying layer is the high reflective mirror, creating a GTI mirror and leading to oscillations of the GDD around the targeted function.
Figure 40. GTI effect due to front reflection between air and mirror [206]

A quick analysis of the GTI effect in chirped mirrors can be made based on the previous derivation by slightly modifying the phase introduced by chirped mirror. From Figure 40, it can be observed that the GTI introduces an extra phase and furthermore that the introduced phase is wavelength dependent. The phase introduced by the GTI can be written as:

\[ \Phi(\omega) = \arctan \left( \frac{(1-r^2)\sin \phi}{2r - (r^2 + 1)\cos \phi} \right) \]  

(75)

where \( r \) is the reflectivity of the interface between the air and the mirror and \( \phi \) is defined by Eqn.(70). The group delay can be obtained by taking the derivative of Eqn.(75) with respect to frequency.

\[ GD(\omega) = \frac{(r^2-1)\frac{d\phi}{d\omega}}{r^2 + 1 - 2r \cos \phi} \]  

(76)

Figure 41 illustrates two concepts: first, the numerical derivative of the group delay is taken for the simple case developed earlier where \( \phi \) was given by Eqn.(70) and Eqn.(72); second the GTI effect on the group delay (GD) and the group delay dispersion (GDD).
In order to suppress the GTI effect, a wedged anti reflection coating was applied to the mirror, on top of the chirped Bragg stack. One of the advantages of chirped mirrors is their ability to be highly reflective over wider bandwidths than regular dielectric mirrors [207]. Applying an anti-reflection coating partially solved the issue of GTI effect, but a perfect match could not be achieved and oscillations were still observed in the GDD [208, 209].

In order to handle the wide bandwidth and further attenuate the oscillations in the GDD, another solution was proposed by Kärtner et al. [210]. As mentioned earlier, to create a highly reflective dielectric mirror, one alternates layers of high and low refractive index material with each layer having a thickness $L_{low} = L_{high} = \lambda/4$. It was found that by keeping the sum of the thicknesses of two consecutive layers equal to $\lambda/2$ but by changing the duty-cycle, reflections from the interface air/mirror can be limited and even controlled. Mathematically, calling the duty-cycle $\eta$ this is expressed by $L_{low} + L_{high} = (1-\eta)\lambda/2 + \eta\lambda/2$. Ramping up the duty-cycle over
the first few layers in the direction of propagation lead to chirping of the chirped mirror, hence the name ‘double chirped mirror’ (DCM) [200, 208]. The typical structure of a chirped mirror is compared to the one of double chirped mirror on Figure 42.

Figure 42. Comparison between a chirped mirror and a double chirped mirror [209]

The use of double chirped mirrors improved the way dispersion could be handled in femtosecond resonators. Nevertheless, in order to handle octave spanning spectra, further care needs to be taken to remove the remaining oscillations in the GDD. This has been achieved by using pairs of chirped mirrors whose oscillations are slightly shifted so that they cancel each other out. Remembering that the source of the oscillations is a Fabry Perot-like effect, the phase can be shifted slightly by changing the angle of incidence on the mirror, which leads to an effective change of the thickness of the interferometer and thus shifts the extra phase introduced by the GTI allowing for the possibility to cancel out the phase. Pulses in the sub-5 fs regime can be produced using double chirped mirror pairs [211].
4.5 The HERACLES stretcher compressor assembly

The previous sections describe the performance of traditional stretcher compressor assemblies and introduced the model and simulation for the grism pair implemented in the HERACLES dispersive line. Two aspects have been purposely overlooked in the previous sections:

- the balance between TOD and GDD has been mentioned but no efforts have been made to match the stretcher to a potential glass compressor.
- the computations of GD, GDD and TOD have been performed only at the center wavelength. This approach is usually suitable for pulses with limited spectral bandwidth but insufficient for ultra-broadband pulses where the tail of the spectrum extends over a hundred nanometers away from the center wavelength.

This section addresses these issues and details the stretcher-compressor assembly of HERACLES. The code developed for this purpose is introduced, simulations are performed and the results are presented.

4.5.1 Hybrid grism-grating stretcher

4.5.1.1 Introduction of the dispersion code

Each element in the optical path of the few cycle pulse between the output of the oscillator and the final output of the system needs to be taken into account to ensure optimum pulse compression. The elements taken into account in the code developed to model the path of few-cycle pulse throughout the system are separated as follow:

- The stretcher assembly is split into three separate sub-systems: a grism pair, a grating pair and an AOPDF. These three components are modelled independently and
parameters such as the grism separation, prism separation, grating groove density, grism material, grism apex angle, dispersion coefficient of the AOPDF are accessible to the user.

- The AOPDF implemented in HERACLES consists of a **4.5 cm long piece of TeO₂** providing a constant GDD, TOD and FOD. Their values for this element are included in the simulations.

- Optical parametric amplification is performed in three consecutive OPA stages consisting each of a 5 mm long piece of BBO, resulting in a **1.5 mm long effective BBO piece**. The constant dispersion experienced by the few-cycle pulses in these OPA stages is also added to the total dispersion.

- The compressor, consisting of **a large diameter piece of highly dispersive glass** (see section 4.4.3) is added to dispersion as it compensates for the dispersion provided by the stretcher.

4.5.1.2 Contribution of BBO and TeO₂ to the dispersion

The effects of the three 0.5 cm long BBO optical parametric amplifier crystals and the 4.5 cm TeO₂ crystal in the AOPDF are modelled first since they together provide a dispersion offset that offers no adjustment. The choice of the length of the BBO crystal is dictated by the OPA design and the length of the TeO₂ crystal is dictated by the interaction length required to acousto-optically modulate a 400 nm wide spectrum (the AOPDF mounted into HERACLES can control spectral width slightly wider than the typical 300 nm wide gain bandwidth of a BBO non-collinear OPA (NOPA) operated at 850 nm). Group delay, GDD, TOD and FOD are calculated.
over the 300 nm bandwidth spanning from 700 to 1000 nm for these two elements. Results are shown in Figure 43.

Figure 43: a. Computed group delay (set to zero at the center wavelength of 850 nm) produced by a 1.5 cm effective BBO crystal (blue line) and a 4.5 cm TeO$_2$ crystal (black line) normalized to the center wavelength $\lambda_0 = 850$ nm; b. computed GDD produced by a 1.5 cm effective BBO crystal (blue line) and a 4.5 cm TeO$_2$ crystal (black line); c. computed TOD by a 1.5 cm effective BBO crystal (blue line) and a 4.5 cm TeO$_2$ crystal (black line); d. computed FOD produced by a 1.5 cm effective BBO crystal (blue line) and a 4.5 cm TeO$_2$ crystal (black line)

It is noticeable that the dispersion provided by the TeO$_2$ crystal is beyond the range of dispersion that can be compensated via the acousto-optic interaction (~1.5 ps). The AOPDF therefore introduces a constant positive offset in GDD and TOD with modulation provided by the acousto-optic effect. The dispersion introduced by the ordinary axis of the BBO crystals (the
polarization of the signal beam during the optical parametric amplification), is largely negligible compared to that of the TeO$_2$ crystal and within the range of compensation of the AOPDF.

The TeO$_2$ crystal also provides a group delay of nearly 18 ps via positive GDD and strong TOD. The TOD provided by the TeO$_2$ crystal is positive and therefore partially compensates for the excess negative TOD provided by the grism pair (see section 4.3.1). The strongly dispersive properties of TeO$_2$ are known since the principle of operation of the AOPDF relies on them. As a remark, these computations have been performed considering only the ordinary axis of the TeO$_2$. This is an approximation since diffracted wavelengths in the AOPDF experience both ordinary and extraordinary axis propagation depending upon the depth at which they are diffracted. Nevertheless, the dispersion behavior is similar for light polarized along either axis.

4.5.1.3 Contribution of the grism pair, grating pair and AOPDF

Grism pairs offer negative GDD and excessive negative TOD while grating pairs offer negative GDD and positive GDD. In this section, the potential for compensating the excess TOD provided by a grism pair via a grating pair is investigated and this implementation in HERACLES is discussed.

The OPA pump pulses of HERACLES are set to 85 ps duration (Chapter 2 and 5). This duration is fixed by the design of the pump beam generation amplifier line and offers little adjustment. In order to obtain both broadband amplification and high gain, it is shown in chapter 5 that a stretched signal pulse with duration up to 30% that of the pump pulse results in no spectral narrowing. Increasing the signal pulse duration improves the temporal overlap between
the pump and signal pulse and results in higher gain but reduced bandwidth. Therefore a stretched pulse duration of ~26 ps is chosen for the simulation. Stretching a broadband pulse with 250 nm of bandwidth FWHM from its transform-limited duration to 26 ps requires an amount of GDD = -40000 fs². A grating pair and a grism pair are used in conjunction to provide the total amount of GDD, but the contribution of each pair to the overall GDD is tuned by numerically adjusting the distance between the two grisms and the distance between the two gratings. A grism pair made of SF11 glass with an 18° apex angle with a grating groove density of 300 l/mm, similar to the one discussed in 4.4.1, is chosen. This design described in the preliminary study of section 4.3.1 the TOD/GDD ratio closest to 0.7 fs, therefore limiting the amount of GDD. A grating pair with 300 l/mm is also chosen to provide dispersion of the same order as the grism pair with reasonable spacing. Simulation results for this arrangement are shown in Figure 44.

Figure 44: a. Computed GDD and TOD behavior for a grating and grism pair adjusted such that the total GDD provided by both pairs equal -40000 fs² for a range of grism separation from 0.5 to 6 cm; b. computed TOD/GDD ratio of a grating and grism pair adjusted such that the total GDD provided by both pairs equal -40000 fs² for a range of grism separation from 0.5 to 6 cm.

Figure 44.a shows the evolution of the GDD of both pairs as a function of the grism pair spacing. As the grism pair spacing is increased, the grating pair spacing is reduced and the total
amount of GDD is maintained constant and equal to -40000 fs², and, the TOD of the grism pair and that of the grating pair are plotted. Three regions appear in the plot: on the left, for small grism separations, the overall TOD is positive and exceeds that of the GDD, reminiscent of the behavior of a grating pair. At the other extreme, for grism separation greater than 4 cm, the negative TOD of the grism exceeds the TOD provided by the grating pair and the double pair exhibits the behavior similar to a single grism pair. The region between these two extreme cases is therefore most interesting: in the region for grism separations comprised between 2 and 4 cm, the TOD provided by the grating pair balances the TOD provided by the grism pair and the double pair exhibits a behavior hybrid between that of a grism pair and that of a grating pair. The advantages of such design are more evident on Figure 44.b.

The TOD/GDD ratio has been calculated for the same range of adjustment of the grism pair spacing. Again, while the distance between grism pair was changed, the grating pair was also tuned to maintain a constant GDD = -40000 fs². The same three regions as previously appears: for small grism spacing, the TOD/GDD ratio is negative resembling the behavior of a grating pair, for large grism spacing, the TOD/GDD is greater than one resembling a grism pair behavior and the region in between offers interesting properties. For grism spacing between 2 and 4 cm, the TOD/GDD ratio varies linearly from 0 to 1.2 fs and crosses the 0.7 fs line. This is a highly valuable property since it enables compression in any standard piece of glass. Furthermore, the concept is scalable: for any desired value of GDD, it is always possible to tune the respective distances between the gratings and the grisms to reach a TOD/GDD ration of 0.7 fs.

The previous study was performed at the central wavelength of 850 nm. In order to investigate if this concept is suitable for ultra-broadband applications, the GD, GDD and TOD of
the grism and grating pairs used in the previous simulation are computed for wavelengths ranging from 700 to 1000 nm. Results for the grism pair are shown in Figure 45.

![Figure 45: a. Calculated group delay (set to zero at the center wavelength of 850 nm) and GDD provided by an SF11 grism pair featuring 300 l/mm gratings and a 5.4 cm grism separation; b. calculated TOD and FOD provided by an SF11 grism pair featuring 300 l/mm gratings and a 5.4 cm grism separation](image)

Notice that the representation of the GD, GDD, TOD, and FOD with respect to wavelength is misleading – yet correct. For example, the group delay curve appears to have a positive slope and yet the GDD has negative values. This is attributed to the fact that computations are performed in the frequency domain but greater insight is provided by plotting over wavelength, thus the slopes of the curves appear reversed by the choice of representation. The group delay curve has been set to zero for the delay experienced by the central wavelength of 850 nm to provide a better reading of the dispersion. The group delay curve shows that the pulse is stretched to ~33 ps full width, corresponding to the delay introduced by the grating pair between 700 nm and 1000 nm. The shape of the group delay curve gives information on the higher order terms: a linear curve would indicate pure GDD or dispersion dominated by the GDD. In the case of the grism stretcher, the curve exhibits a parabolic shape showing the domination of TOD, especially for the shortest wavelengths of the spectral band under
consideration. Looking at the order of magnitudes of GDD and TOD, it is noticeable that both GDD and TOD are negative over the entire bandwidth, a valuable feature to match the dispersion of a piece of glass. Nevertheless the TOD exceeds the GDD over the entire bandwidth, an observation consistent with the shape of the group delay curve leading to TOD/GDD > 1. The magnitude of the FOD falls within the capabilities of compensation of the AOPDF implemented on HERACLES and is therefore of little interest for this study. Tavella et al. [190] reported the use of a specially designed chirped mirror, providing a large excess of positive TOD to compensate for the grism pair’s excessive negative TOD. A theoretical study by Zhend et al. reports that such a limitation can be avoided by adjusting the parameters of the grism pair [212]. Our study, in agreement with Tavella et al. [190], did not confirm the model of [212] and we proposed an alternative solution. Similarly to the grism pair, computations of the GD, GDD, TOD and FOD over the 700 to 1000 nm spectral band for a grating pair are gathered in Figure 46.

Figure 46: a. Calculated group delay (normalized to the center wavelength) and GDD provided by a 300 l/mm grating pair and a 17.8 cm grating separation; b. calculated TOD and FOD provided by a 300 l/mm grating pair and a 17.8 cm grating separation
The group delay curve for the grating pair follows a curve opposite to that of the grism pair, confirming the potential of both pairs to compensate each other. Consequently, the GDD curves have opposite slopes and the GDD remains negative over the entire bandwidth. Interestingly, the values of GDD are in the same order of magnitude for a grism pair with a grism separation of 5.4 cm and a grating pair with 17.8 cm separation, and the sum of both curves suggests a quasi-constant GDD curve over a broad spectral range. The TOD of the grating pair is positive over the entire bandwidth and, even though the match is not as perfect as for the GDD curves, can partially compensate for the excess TOD provided by the grism pair. Finally, the FOD provided by the grating pair is found to be negative over the entire bandwidth and can therefore compensates the FOD provided by the grism pair.

The value of the technique has been investigated by summing the GDD of the grating pair and the GDD of the grism pair, summing the TOD of the grating pair and the TOD of the grism and taking the ratio TOD/GDD of the resulting sums over the bandwidth under consideration. The result is compared to the TOD/GDD ratio of the grism pair alone and the TOD/GDD ratio of the grating pair alone.

Figure 47: Calculated evolution of the TOD/GDD ratio over a 300 nm spectral band ranging from 700 to 1000 nm for a grating pair, a grism and grism/grating pair combination showing the superior behavior of the combination
The example for this calculation is not optimized for the flattest GD when combining only the grating pair and the grism pair. Nevertheless, it shows an excellent reduction of the TOD/GDD ratio compared to that of the grism pair alone and it brings and maintains the TOD/GDD ratio in the vicinity of 0.7 fs over the entire bandwidth (insert). Therefore a grism and grating pair used in conjunction provides an additional “knob” to tune the TOD independently from the GDD, and can dramatically reduce excessive TOD provided by a grism pair alone. Furthermore such combination is suitable for ultra-broadband applications and provides negative GDD and negative TOD. This combination has been investigated for the first time in the frame of this thesis.

4.5.2 Bulk glass pulse compression

4.5.2.1 Requirements for bulk glass compression

The feasibility of ultra-broadband pulse stretching while maintaining a TOD/GDD ratio in the vicinity of the target value of 0.7 fs has been demonstrated in the previous section. Additionally, the constant and uncompressible dispersion provided by the TeO₂ crystal making the AOPDF and the BBO crystals of the optical parametric amplifier has been quantified. Summing the effect of these components provides the stretched pulse duration and the amount of dispersion that needs to be compensated for in the compressor. Table 13 summarizes the values of the dispersion of the components discussed in the previous section at the center wavelength of 850 nm and under the conditions for a grating separation of 13.8 cm and a grism separation of 5 cm.
Table 13: Total amount of dispersion provided by an SF11 grism pair featuring 300 l/mm gratings separated by 5 cm, a grating pair featuring 300 l/mm gratings separated by 13.8 cm, a 4.5 cm long TeO2 piece modeling an AOPDF and a 1.5 cm BBO crystal modeling three OPA stages.

<table>
<thead>
<tr>
<th></th>
<th>GDD (fs^2)</th>
<th>TOD (fs^3)</th>
<th>FOD (fs^4)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Grism pair</td>
<td>-28.5x10^3</td>
<td>-89x10^3</td>
<td>-9.8x10^3</td>
</tr>
<tr>
<td>Grating pair</td>
<td>-27.3x10^3</td>
<td>35.7x10^3</td>
<td>-63x10^3</td>
</tr>
<tr>
<td>TeO2</td>
<td>20.4x10^3</td>
<td>13.6x10^3</td>
<td>7.5x10^3</td>
</tr>
<tr>
<td>BBO</td>
<td>1x10^3</td>
<td>0.8x10^3</td>
<td>-0.25x10^3</td>
</tr>
<tr>
<td>Total</td>
<td>-34400</td>
<td>-38900</td>
<td>-69750</td>
</tr>
</tbody>
</table>

This configuration was intentionally chosen to be imperfect to illustrate the flexibility of the grating/grism combination. The configuration offers a total GDD slightly lower than the target value of -40000 fs^2 but remains in an acceptable range. The TOD/GDD ratio is not at an acceptable level: the excess TOD leads to a ratio of ~1.1 fs. In order to reduce the ratio while increasing the amount of GDD, the grating pair separation is extended to provide excess GDD and reduce the TOD/GDD. With a 5 cm separation grism pair (similar to previously) and a 17.8 cm separation grating pair (previously 13.8 cm) the new dispersion total is shown in Table 14.
Table 14: Total amount of dispersion provided by an SF11 grism pair featuring 300 l/mm gratings separated by 5 cm, a grating pair featuring 300 l/mm gratings separated by 17.8 cm, a 4.5 cm long TeO2 piece modeling an AOPDF and a 1.5 cm BBO crystal modeling three OPA stages.

<table>
<thead>
<tr>
<th></th>
<th>GDD (fs$^2$)</th>
<th>TOD (fs$^3$)</th>
<th>FOD (fs$^4$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Grism pair</td>
<td>-28.5x10$^3$</td>
<td>-89x10$^3$</td>
<td>-9.88x10$^3$</td>
</tr>
<tr>
<td>Grating pair</td>
<td>-35.2x10$^3$</td>
<td>46.4x10$^3$</td>
<td>-83.4x10$^3$</td>
</tr>
<tr>
<td>TeO$_2$</td>
<td>20.4x10$^3$</td>
<td>13.6x10$^3$</td>
<td>7.5x10$^3$</td>
</tr>
<tr>
<td>BBO</td>
<td>1x10$^3$</td>
<td>0.8x10$^3$</td>
<td>-0.25x10$^3$</td>
</tr>
<tr>
<td>Total</td>
<td>-42300</td>
<td>-28200</td>
<td>-86050</td>
</tr>
</tbody>
</table>

The overall amount of GDD provided by this configuration is close to the target value of -40000 fs$^2$ and offers values that are practically reasonable to implement. Sub-millimeter adjustment of the grism pair separation is required to reach the exact value of -40000 fs$^2$ but such fine adjustment must be performed in the laboratory while monitoring the pulse duration at the output of the system. Similarly, the TOD/GDD ratio is now brought to 0.67 fs, in the close vicinity of the target value. The previous argument holds for fine adjustment of the TOD/GDD ratio, but likewise such refined tuning can only be performed in a meaningful way in the laboratory and the stretcher embodiment enables such fine tuning. The profile of the stretched pulse duration computed with the total dispersion coefficient of Table 14 is shown in Figure 48.
The optimization of the TOD/GDD ratio allows for a quasi-Gaussian pulse (the slight di-
symmetry is attributed to the TOD) to interact with the pump pulse in the amplification process. For a TOD/GDD ratio exceeding one, typical pre-pulses appear at the front of the pulse, which result in reduced efficiency of the OPA process via reduction of the peak power. The optimization of the TOD/GDD ratio therefore not only facilitates proper lossless compression but also increases gain in the optical parametric amplifier. Finally, the target pulse duration of 26 ps can be reached enabling close to optimum nonlinear interaction in the OPA stages.

4.5.2.2 Choice of material

As the stretcher design is built to match a glass compressor, little design work is required for the compressor. The choice of the glass, its availability and the size of the glass piece are the only parameters to be determined. Table 15 shows the dispersion coefficients of three type of glass providing little dispersion, mild dispersion and large dispersion along with their TOD/GDD ratio.
Table 15: Dispersive properties of fused silica, SF11 and SF59 glass

<table>
<thead>
<tr>
<th></th>
<th>GDD ($f^2\cdot cm^{-1}$)</th>
<th>TOD ($f^3\cdot cm^{-1}$)</th>
<th>FOD ($f^4\cdot cm^{-1}$)</th>
<th>TOD/GDD (fs)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fused silica</td>
<td>346</td>
<td>282</td>
<td>-134</td>
<td>0.81</td>
</tr>
<tr>
<td>SF11</td>
<td>1821</td>
<td>1243</td>
<td>284</td>
<td>0.68</td>
</tr>
<tr>
<td>SF59</td>
<td>2782</td>
<td>2052</td>
<td>514</td>
<td>0.73</td>
</tr>
</tbody>
</table>

The GDD provided by a 1 cm piece of SF59 exceeds that provided by a fused silica piece of the same size by almost an order of magnitude. Dispersion being a linear effect, this results in a glass block about ten times shorter if SF59 is used rather than fused silica. In order to investigate the three options, the length of the glass piece required to compensate exactly the GDD of the stretcher, TeO$_2$ and BBO is computed and the remaining TOD and FOD are calculated (based on Table 14).

Table 16: Computed required glass length to compensate the GDD introduced by a 5 cm grism pair, a 17.8 cm grating pair, a 4.5 cm TeO$_2$ crystal and a 1.5 cm effective BBO crystal assuming a fused silica, SF11 or SF59 glass compressor. Residual TOD and FOD upon matching GDD are also computed

<table>
<thead>
<tr>
<th></th>
<th>Glass length (cm)</th>
<th>Residual TOD ($f^3$)</th>
<th>Residual FOD ($f^4$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fused silica</td>
<td>122.38</td>
<td>6322</td>
<td>-102450</td>
</tr>
<tr>
<td>SF11</td>
<td>23.23</td>
<td>667</td>
<td>-79460</td>
</tr>
<tr>
<td>SF59</td>
<td>15.21</td>
<td>3010</td>
<td>-78240</td>
</tr>
</tbody>
</table>

The small amount of dispersion provided by fused silica results in the need for a 1.2 m path length in this type of glass to compensate for the dispersion provided in the stretcher. Such length could be achieved by passing several times through a shorter piece but complicates the practical implementation of the compressor. SF11 on the other hand requires only a 23.2 cm long
glass piece to achieve pulse compression and provides a close match in TOD/GDD ratio resulting in small residual TOD. Nevertheless SF59 is even more efficient at compressing the stretched pulse, even though the residual TOD provided by the SF59 is larger than that provided by SF11. However this is only due to the use of the current design: as was shown in previous sections, slight adjustments to the stretcher allow close matching between the stretcher and the compressor.

The initial design of HERACLES featured SF59 as a glass compressor. Nevertheless, after inquiring from Schott the feasibility of such a large piece of SF59, it was found that this glass cannot be produced in such large pieces. The design was then modified to operate with SF57 that features dispersion coefficient on the same order as SF59 (GDD = 2122 fs².cm⁻¹, TOD = 1533 fs³.cm⁻¹, FOD = 304 fs⁴.cm⁻¹) and can be produced in larger dimensions. Computation of the residual GDD featuring a 21.2 cm long SF57 glass compressor and taking into account all parameters in the system (assuming that the AOPDF is turned off), performed at the center wavelength of 850 nm are gathered in Table 17.

<table>
<thead>
<tr>
<th></th>
<th>GDD (fs²)</th>
<th>TOD (fs³)</th>
<th>FOD (fs⁴)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Grism pair</td>
<td>-28.5x10³</td>
<td>-89x10³</td>
<td>-9.88x10³</td>
</tr>
<tr>
<td>Grating pair</td>
<td>-35.2x10³</td>
<td>46.4x10³</td>
<td>-8.34x10³</td>
</tr>
<tr>
<td>TeO₂</td>
<td>20.4x10³</td>
<td>13.6x10³</td>
<td>7.5x10³</td>
</tr>
<tr>
<td>BBO</td>
<td>1x10³</td>
<td>0.8x10³</td>
<td>-0.25x10³</td>
</tr>
<tr>
<td>Compressor</td>
<td>42.2x10³</td>
<td>29.4x10³</td>
<td>7.7x10³</td>
</tr>
<tr>
<td>Total</td>
<td>-100</td>
<td>-1200</td>
<td>-78300</td>
</tr>
</tbody>
</table>

Table 17: Residual GDD, TOD and FOD when a 21.2 cm long SF57 compressor is used to compensate the dispersion induced by a grism pair with 5 cm separation, a grating pair with 17.8 cm separation, a 4.5 cm long TeO₂ AOPDF and an effective 1.5 cm long BBO crystal.
The residual dispersion after compression is enough to prevent compression down to the few-cycle pulses as discussed in section 4.4. Nevertheless, the previous simulation does not include the spectral phase the AOPDF can provide when properly programmed. A plot of the residual group delay generated by the residual GDD, TOD and FOD is shown in Figure 49.

![Figure 49: Calculated residual group delay, set to zero at the center wavelength of 850 nm, after propagation through the stretcher and compressor assemblies](image)

The AOPDF implemented on HERACLES is capable of compensating an overall group delay of ±1.5 ps. The residual group delay after compression, shown in Figure 49 is for the most part contained within this temporal window (aside from wavelengths below 725 nm) and should therefore allows successful compensation if the right radio frequency signal is applied to the AOPDF. Simulations have been performed, over the entire bandwidth to compute the group delay, GDD, TOD and FOD and optimize to achieve the flattest group delay over the widest spectral band after optimizing the AOPDF signal. Results of the optimized computations are shown in Figure 50.

119
The residual group delay is contained within 10 fs over 275 nm. For a Gaussian spectrum, such limited group delay would result in a transform-limited pulse with a duration shorter than 5 fs. Nevertheless, the spectrum provided by the Octavius 85 is modulated and therefore, the time bandwidth product of the pulses is \(~0.55\). A 150 nm FWHM wide spectrum (assuming that the FWHM of the amplified spectrum is \(~150\) nm) of a pulse featuring a time bandwidth product of 0.55 results in a 8.8 fs pulse duration, the target value for HERACLES.
Shorter pulse duration can be achieved by adding custom made chirped mirrors at the output of the system providing pulses with duration in the 7 fs range.

4.5.2.3 **B-integral management**

An important parameter to ensure good beam profile and prevent damages in the compressor is the B-integral. The B-integral is a measure of the accumulated nonlinear effects, accumulated during the propagation in a medium. The mathematical expression for the B-integral is given by [157]:

\[
B = \frac{2\pi}{\lambda} \int n_2 I(z) dz
\]  

(77)

where \(n_2\) is the nonlinear refractive index of the medium in which the pulse propagates (along the \(z\)-axis) and \(I(z)\) is the intensity of the pulse at location \(z\). In order to prevent pulse self-action such as self-focusing or self-phase modulation, the value of \(B\) needs to remain below ~1. The value of the B-integral has been computed for a stretched 2 mJ pulse passing through a 21.2 cm long piece of SF57 and reaching its transform limit at the end of the glass block. Computations of the B-integral were performed for both 1 cm and 5 cm beam diameters.
The evolution of the pulse duration as the pulse propagates in the compressor is shown in Figure 51a. The exponential evolution of the pulse duration, and consequently of the peak power, results in an exponential-like increase in the B-integral value. The beam diameter is therefore the only parameter that can be adjusted to limit the B-integral. A dramatic reduction in the B-integral value is observed as the beam diameter is increased from 1 cm to 5 cm. The beam diameter chosen for the HERACLES compressor is 5 cm since it maintains the B-integral below one for pulses with energy as high as 2 mJ.
5 NON-COLLINEAR OPTICAL PARAMETRIC AMPLIFICATION: THEORY AND SIMULATIONS

Optical parametric amplification (OPA) is chosen as an alternative to chirped-pulse amplification in the HERACLES design for its superior abilities to amplify sub-10 fs pulses (see section 2.2 for a discussion on the advantages of optical parametric amplification). Nevertheless, the design of such amplifier implies a great deal of design work since phase-matching needs to be achieved over 300 nm of bandwidth. Furthermore, a successful experimental implementation requires understanding and modeling the involved processes such as optical parametric generation, beam walk-off or saturation.

This chapter presents an analytical approach to the theory of optical parametric amplification. This rigorous mathematical approach is seen to be rather involved and to provide little insight on the experimental parameters at stake due to the large number of assumptions required to obtain an analytical solution. The rest of the chapter details the 3D code implemented in the frame of this thesis to model optical parametric amplification and shows the results of the model. A parametric investigation of critical parameter designs has been performed in order to optimize the experimental performances of the HERACLES parametric amplifier stages.

5.1 Theory of optical parametric amplification

The nonlinear-coupled wave equations ruling the optical parametric amplification process are first derived from the nonlinear polarization and Maxwell’s equations. Analytical solutions are then derived in a simplified case. The derivation performed in this section follows that proposed by Baumgartner et al. [120] and some simplification proposed by Witte [213] and benefits from the analysis that can be found in Abramowitz et al. [214].
5.1.1 Nonlinear polarization and introduction of coupled wave-equations

In any media subjected to a propagating electric field, a polarization $\mathbf{P}$ induced by the motion of the electrons under the effect of the oscillating electric field is generated with a vectorial expression given by:

$$\mathbf{P}(\omega) = \varepsilon_0 \varepsilon \mathbf{E}(\omega)$$

(78)

where $\mathbf{E}(\omega)$ is the optical electric field, $\varepsilon_0$ is the vacuum permittivity and $\varepsilon$ is the permittivity tensor describing the optical properties of the material in which the wave propagates. In the linear regime, the frequency of oscillation of the polarization is the same as that of the driving electric field. In the case of a nonlinear interaction between multiple waves, the resulting nonlinear polarization does not oscillates at the same frequency as the incident fields and is in the general case given by:

$$\mathbf{P}_p(\omega_p) = \varepsilon_0 \chi(-\omega_p, \omega_s, \omega_i) \mathbf{E}_s(\omega_s) \mathbf{E}_i(\omega_i),$$

(79)

where $\mathbf{P}_p(\omega_p)$ is the nonlinear polarization resulting of the interaction of the optical field $\mathbf{E}_s(\omega_s)$ and $\mathbf{E}_i(\omega_i)$ in a nonlinear medium with a nonlinear susceptibility tensor $\chi(-\omega_p, \omega_s, \omega_i)$. In the case of optical parametric amplification, the nonlinear polarization $P_s$, $P_i$ and $P_p$ resulting from the superposition of the signal (subscript $s$), the idler (subscript $i$) and pump (subscript $p$) waves can simply be written as:

$$P_s = 2\varepsilon_0 d_{eff} E_i^* E_p$$
$$P_i = 2\varepsilon_0 d_{eff} E_s^* E_p,$$
$$P_p = 2\varepsilon_0 d_{eff} E_s E_i$$

(80)

where the explicit frequency dependence has not been written to simplify the notation and the susceptibility has been replaced by the effective nonlinear coefficient $d_{eff}$. Under the slowly
The varying envelope approximation – valid in the case of picosecond pulses at NIR wavelengths –
the optical electric field $E_k$ and polarization $P_k$ (where $k = s, i$ and $p$) can be written as sums of
harmonic waves with the expression:

$$
E_k(z,t) = \sum_k \text{Re}(E_k e^{j(k_0 t - k_z z)})
$$

$$
P_k(z,t) = \sum_k \text{Re}(P_k e^{j(k_0 t - k_z z)})
$$

which gives, under the envelope approximation and once plugged into the nonlinear polarization
equation:

$$
P_s = 2\varepsilon_0 d_{sf} E_s^* E_p e^{-jkz}
$$

$$
P_i = 2\varepsilon_0 d_{if} E_s^* E_p e^{-jkz}
$$

$$
P_p = 2\varepsilon_0 d_{pf} E_s E_i e^{jkz}
$$

where the $\Delta k$, the phase mismatch term, given by:

$$
\Delta k = k_p - k_s - k_i
$$

is naturally appearing. With the expression of the nonlinear polarization at hand, the propagation
equation involving polarization can be derived from Maxwell’s equations:

$$
\nabla \cdot \vec{D} = 0
$$

$$
\nabla \cdot \vec{B} = 0
$$

$$
\nabla \times \vec{E} = -\frac{\partial \vec{B}}{\partial t}
$$

$$
\nabla \times \vec{H} = \vec{J} + \frac{\partial \vec{D}}{\partial t}
$$

The relationships between the electro-magnetic field in vacuum and in the medium under
consideration are given by:
\[
\vec{D} = \varepsilon_0 \varepsilon \vec{E} + \vec{P} \\
\vec{B} = \mu_0 \vec{H}
\]  
(88)

where \( \mu_0 \) is the magnetic permeability. Substituting Eqn.(88) into Eqn.(86) and combining the Maxwell’s equation, the wave equation describing the propagation of an optical wave in a media with permittivity \( \varepsilon_0 \) and permeability \( \mu_0 \) can be expressed.

\[
\frac{\partial^2 E}{\partial z^2} - \mu_0 \sigma \frac{\partial E}{\partial t} - \mu_0 \varepsilon_0 \varepsilon \frac{\partial^2 E}{\partial t^2} = \mu_0 \frac{\partial^2 P}{\partial t^2} .
\]  
(89)

Substituting the expression of the electric field of the signal, idler and pump wave under the slowly varying approximation into Eqn.(89), leads to [120]:

\[
\frac{\partial E_{k}}{\partial z} + \frac{n_k}{c} \frac{\partial E_{k}}{\partial t} + \alpha_k E_{k} = -j \frac{\mu_0 c \omega_k}{2n_k} P_k ,
\]  
(90)

where \( n_k \) is the index of refraction of the material at the wavelength of wave \( k \) (\( k = s, i \) or \( p \)), and \( \alpha_k = 1/2 \mu_0 \sigma c / n_k \) is the electric field absorption coefficient. Plugging Eqn.(82) into Eqn.(90) for the signal, idler and pump beam yields the set of coupled partial differential equations describing optical parametric amplification.

\[
\frac{\partial E_s}{\partial z} + \frac{n_s}{c} \frac{\partial E_s}{\partial t} + \alpha_s E_s = -j \frac{\omega_s d_{\text{eff}}}{n_s c} E_s^* P_p e^{-j\Delta z} \\
\frac{\partial E_i}{\partial z} + \frac{n_i}{c} \frac{\partial E_i}{\partial t} + \alpha_i E_i = -j \frac{\omega_i d_{\text{eff}}}{n_i c} E_i^* E_p e^{-j\Delta z} \\
\frac{\partial E_p}{\partial z} + \frac{n_p}{c} \frac{\partial E_p}{\partial t} + \alpha_p E_p = -j \frac{\omega_p d_{\text{eff}}}{n_p c} E_s E_p e^{+j\Delta z}
\]  
(91)

These equations can be further simplified under practical consideration. The nonlinear material used for the optical parametric amplification is typically chosen so that absorption is negligible at the wavelength of operation of the OPA, therefore setting \( \alpha = 0 \) (for \( k = s, i \) or \( p \)).
Furthermore, the crystals used for ultra-broadband amplification are typically short in length, such that dispersion effects can be neglected. Therefore, it is reasonable to assume that the terms $n_k/c$, invert of the phase velocity can be assumed to be equal for each equation. If the equations are expressed in the referential of the three waves, the time dependence can be removed leading to a set of ordinary differential equations which is the usual expression for the mixing of three waves in a nonlinear medium.

$$\frac{\partial E_s}{\partial z} = -j \frac{\omega_s d_{eff}}{n_s c} E^*_p E_p e^{-j\Delta z}$$
$$\frac{\partial E_i}{\partial z} = -j \frac{\omega_i d_{eff}}{n_i c} E^*_s E_p e^{-j\Delta z}$$
$$\frac{\partial E_p}{\partial z} = -j \frac{\omega_p d_{eff}}{n_p c} E_s E^*_i e^{+j\Delta z}$$

(92)

5.1.2 Analytical solutions of the coupled wave equations

Solving the coupled partial differential equations describing a three-wave interaction in a nonlinear medium requires writing these equations under a form that has a known analytical solution. In the case of these equations, the aim is to identify the analytic solution of Eqn.(92) to an elliptic integral of the first kind which takes a Jacobi elliptic function as a solution. The set of equations is first modified by setting the complex electric field amplitudes $E_{k=s,i,p} = \rho_k e^{+j\phi_k}$ and separating real and imaginary parts. Such substitution leads to the modified set of equations:
where the phase variable $\theta$ is given by:

$$\theta = \Delta k + \phi_p - \phi_i - \phi_s, \quad (94)$$

The set of modified equations can be further normalized by introducing the set of invariants and normalized constants proposed by Baumgartner et al. [120]:

$$u_s = \left( \frac{\varepsilon_0 \lambda_s n_s}{4\pi W} \right)^{1/2} \rho_s$$

$$u_i = \left( \frac{\varepsilon_0 \lambda_i n_i}{4\pi W} \right)^{1/2} \rho_i$$

$$u_p = \left( \frac{\varepsilon_0 \lambda_p n_p}{4\pi W} \right)^{1/2} \rho_p$$

where $W$ is the total power flow per unit area in the direction of propagation of the three waves involved in the optical parametric amplification process and is defined by:

$$W = \left( \frac{\varepsilon_0 c}{2} \right) \left( n_s \rho_s^2 + n_i \rho_i^2 + n_p \rho_p^2 \right) = I_1(0) + I_2(0) + I_3(0) \quad (96)$$

where $I_{k=s,i,p}(0)$ are the intensity of the signal, idler and pump wave at $z = 0$. With the definition of $W$ at hand, $u_{k=s,i,p}$ can be written as the square root of the normalized intensity for each wave ponderated by the inverse of the optical frequency of the wave, a form providing more physical insight:
The final reduction of the set of equation is achieved by transferring the coordinates from the physical variable $z$ to the normalized variable $\xi$ [120] where $\xi$ is given by:

$$\xi = 4\pi d_{eff} z \sqrt{\frac{\pi W}{\varepsilon_0 \lambda_s^2 \lambda_i^2 n_s n_i n_p}}.$$

(98)

Equipped with this new set of variables and normalizations, the set of coupled partial differential (Eqn.(92)) can be written in the simple form:

$$\frac{du_s}{d\xi} = -u_s u_p \sin \theta$$

$$\frac{du_i}{d\xi} = -u_i u_p \sin \theta$$

$$\frac{du_p}{d\xi} = u_s u_i \sin \theta$$

(99)

The phase term $\theta$ is of major importance since it is the key to solve the set of equations. An solution for $\theta$ is first found and later plugged back into the equations to find expressions for $u_{k=p,i}$. The evolution of the phase term is given in the $\xi$-referential by:

$$\frac{d\theta}{d\xi} = \frac{\Delta k}{\xi} + \left( \frac{u_s u_i - u_i u_p}{u_p u_s} - \frac{u_s u_i}{u_i} \right) \cos \theta = \frac{\Delta k}{\xi} + \frac{1}{\tan \theta} \cdot \frac{1}{d\xi} \ln(u_s u_i u_p)$$

(100)

Solving Eqn.(100) for $\theta$ can be achieved ‘independently’ from the amplitude equations by performing a few manipulations suggested by Witte [213]. Eqn.(100) is first written as:
\[
\frac{\partial \cos \theta}{\partial \xi} = -\sin \theta \frac{\Delta z}{\xi} - \left( \frac{u_s u_i}{u_p} - \frac{u_s u_{i_p}}{u_i} \right) \sin \theta \cos \theta . \tag{101}
\]

Substituting the last equation of the set of Eqn.(99) into the previous equation to remove the \( \sin \theta \) dependence provides:

\[
\frac{\partial \cos \theta}{\partial \xi} = -\frac{\Delta z}{\xi} \frac{1}{u_s u_i} \frac{\partial u_p}{\partial \xi} = \left( \frac{1}{u_i} \frac{\partial u_s}{\partial \xi} + \frac{1}{u_s} \frac{\partial u_i}{\partial \xi} + \frac{1}{u_p} \frac{\partial u_p}{\partial \xi} \right) \cos \theta \tag{102}
\]

which can be written as:

\[
\frac{\partial \cos \theta}{\partial \xi} = -\frac{1}{u_s u_i u_p} \left[ \frac{\Delta z}{\xi} \frac{\partial u_p}{\partial \xi} + \left( u_s \frac{\partial u_s}{\partial \xi} + u_s u_p \frac{\partial u_i}{\partial \xi} + u_s u_i \frac{\partial u_p}{\partial \xi} \right) \cos \theta \right] \tag{103}
\]

Multiplying both sides by the product \( u_s u_i u_p \) and gathering all the cosine terms on the left hand side leads to:

\[
u_s u_i u_p \frac{\partial \cos \theta}{\partial \xi} + \cos \theta \frac{\partial u_s u_i u_p}{\partial \xi} = -\frac{1}{2} \frac{\Delta z}{\xi} \frac{\partial u_p^2}{\partial \xi} \tag{104}
\]

The left hand-side can be written as a simple derivative of the product \( u_s u_i u_p \cos \theta \) with respect to \( \xi \):

\[
\frac{\partial u_s u_i u_p \cos \theta}{\partial \xi} = -\frac{1}{2} \frac{\Delta z}{\xi} \frac{\partial u_p^2}{\partial \xi} \tag{105}
\]

which can be integrated and provide the expression for \( \theta \):

\[
\cos \theta = -\frac{1}{2} \frac{\Delta z}{u_s u_i u_p} u_p^2 \tag{106}
\]

with the integration constant:
The set of coupled equations requires a more sophisticated approach in order to be solved. A set of constant parameters relating $u_p$, $u_s$ and $u_i$ can enable plugging up, $u_s$ and $u_i$, into each other’s partial differential equation allowing partial decoupling of the equations. The Manley-Rowe equations, expressing the conservation of the number of photons in any nonlinear process provide such relationship. They are expressed in the $\xi$-referential by substituting the equation expressing the conservation of energy ($\omega_s + \omega_i = \omega_p$) and the equation expressing the conservation of momentum ($\omega_s u_s^2 + \omega_i u_i^2 + \omega_p u_p^2 = 1$). After a few algebraic manipulations, the Manley-Rowe equations are obtained:

\[
\begin{align*}
  m_s &= u_s^2 + u_p^2 \\
  m_i &= u_i^2 + u_p^2 \\
  m_p &= u_s^2 - u_i^2 
\end{align*}
\]  

where $m_s$, $m_i$ and $m_p$ are constants. Implications of these equations on the power flow in the optical parametric amplification process are further discussed in section 5.3. The integration of the system of coupled differential equation is performed by solving the last equation of the set Eqn.(99) and deducing the other variables from the knowledge of $\rho_p$. Integrating this equation is achieved by solving the following integral in which the Manley-Rowe equations have been substituted as well as the expression of $\cos \theta$ the details of the manipulation can be found in [120].

\[
\xi = \frac{1}{2} \int_{u_p(0)}^{u_s(\xi)} \frac{du_p^2}{\sqrt{u_p^2(m_s - u_p^2)(m_s - u_p^2) - \left( \Gamma - \frac{1}{2} \frac{\Delta k_z}{\xi} u_p^2 \right)^2}}.
\]
The form of the integral in Eqn.(109) suggests an elliptic integral of the first kind as a solution and will therefore lead to an expression of \(u^2_p(\xi)\) as a Jacobi integral (sn-function), the Jacobi integral being the inverse operation of an elliptic integral [214]. This expression is general and applies to frequency up-conversion, sum frequency generation and optical parametric amplification. From this point on, the derivation applies solely to the latter case. In the case of optical parametric amplification, the initial conditions are such that the pump is much more intense than the signal and the idler is non-existent which is mathematically expressed by \(u_s^2(0) << u_p^2(0)\) and \(u_i^2(0) = 0\). From these initial conditions, the solution of the system of equations is [120, 214]:

\[
\begin{align*}
\mathcal{K}(\xi) & = \left(\frac{\Delta k z}{2 \xi}\right) + \left[u^2_p(0) - \left(\frac{\Delta k z}{2 \xi}\right)^2\right]^{\frac{1}{2}}\left[u^2_p(0) + u^2_s(0) - \left(\frac{\Delta k z}{2 \xi}\right)^2\right]^{\frac{1}{2}} \\
u_s^2(\xi) & = u_s^2(0) + u_p^2(0) - u_p^2(\xi) \\
u_i^2(\xi) & = u_i^2(0) + u_p^2(0) - u_p^2(\xi)
\end{align*}
\]

(110)

The function \(\mathcal{K}\) is a complete elliptic integral and ensures in the expression of \(u^2_p(\xi)\) that at \(z = \xi = 0\), \(\text{sn}^2 = 1\). The expression of the constant \(\gamma\) is given by:

\[
\gamma^2 = \frac{u^2_p(0) - \left(\frac{\Delta k z}{2 \xi}\right)^2}{u^2_p(0) + u^2_s(0) - \left(\frac{\Delta k z}{2 \xi}\right)^2},
\]

(111)

The expression of \(u^2_s(\xi)\) can be expressed as a function of the inverse elliptic integral sn and cd by using the properties of the elliptic integrals [213, 214]:
The sd-function takes the simple form of a hyperbolic cosine function when the parameter $\gamma^2$ goes to 1. In the case of optical parametric amplification, the pump largely exceeding the seeded signal at $z = \xi = 0$, the value of $\gamma^2 = 1$. This condition is maintained as long as the pump is not depleted. Expressions for the intensity of the signal $I_s$, the intensity of the idler $I_i$ and the intensity of the pump $I_p$ can be deduced under the approximation of non-depletion and are given by:

$$I_s(z) = I_s(0)\cosh^2(gz)$$

$$I_i(z) = \frac{\omega_i}{\omega_s} I_s(0)\sinh^2(gz)$$

$$I_p(z) = I_p(0)$$

where the coefficient $g$ is the parametric gain encountered in textbooks with the expression:

$$g = \sqrt{\frac{\Delta k^2}{2\xi}} - \left(\frac{\Delta k^2}{2\xi} \right)^2 \frac{\xi}{z} = \sqrt{\chi^{(2)}} \frac{\omega_s \omega_i I_p(0)}{2\epsilon_0 n_s n_i n_p c^3} - \left(\frac{\Delta k^2}{2}\right)$$

The evolution of the signal, idler, and pump is plotted in Figure 52 for a parametric gain of 2000 using the expressions derived above.
These expressions provide very little insight on the effects of experimental parameters other than the nonlinear medium length. Furthermore, the model validity is limited as illustrated in Figure 52: for a limited gain of 2000 that can readily be achieved experimentally, the signal output energy after 5 mm interaction is exceeding that of the pump, a situation that is physically impossible. Such inconsistencies are due to the number of approximations (no depletion, no loss, no dispersion, no spatial walk-off, no temporal or spectral dependence) that are required to provide an analytical solution. This simple model required resorting to advanced mathematical functions showing that a more realistic model requires numerical modeling.

5.2 Broadband phase-matching

5.2.1 Broadband phase-matching: conceptual approach

In the case of optical parametric amplification, the energy of the pump, signal and idler beams are instantaneously exchanged between with the three waves. As long as the pump and signal overlap in space and time in the nonlinear crystal, the energy exchange can occur.
Therefore, for successful amplification to occur, it is necessary that the pump and signal beam overlap in time, a condition that requires a careful design to be met.

Before tackling the case of optical parametric chirped-pulse amplification, it is insightful to introduce the case of a femtosecond optical parametric amplifier operated in collinear geometry. In this case, an energetic femtosecond pump pulse and a low energy signal pulse are incident on a nonlinear crystal. The wavelength mismatch between the pump and signal (or idler) can lead to group velocity mismatch due the difference in refractive index experienced between the two waves. For example, for a signal at 800 nm and an idler at 1.8 µm, both polarized along the ordinary axis of a 5 mm long BBO crystal ($n_{o800} = 1.6606$, $n_{o1800} = 1.6425$), the temporal walk-off between these two waves is as large as 300 fs. In the amplification process, while the two waves are walking away from each other the leading edge of the slow signal is feeding into the trailing edge of the fast idler resulting in a decrease in efficiency and lengthening of the pulse. This effect is illustrated in Figure 53 [129].

![Figure 53: Schematic of the temporal walk-off between an idler and signal wave during a collinear nonlinear interaction](image)

In the case of the optical parametric chirped-pulse amplification of ultra-broadband pulses, the effects of phase velocity mismatch are twofold. First, the temporal walk-off of the signal with respect to the idler results in distortion of the seed spectrum: indeed, since the signal being chirped, a temporal walk-off of the idler with respect to the signal lead to a sweep of the gain across the spectrum that can modify the shape of the seed spectrum and shorten it. Second,
the spectral width of the signal spectrum, typically spanning several hundreds of nanometers, requires phase-matching over a large range of wavelength, unlike the case of traditional femtosecond OPAs where the signal pulses rarely exceed 50 nm of bandwidth at 800 nm signal center wavelength.

Non-collinear phase-matching was introduced in order to address this issue [127-129]. In this configuration, the signal and idler waves propagate with a slight angle with respect to each other in the amplification medium as shown in Figure 54 [215].

![Figure 54: Schematic of the non-collinear geometry allowing compensation of the temporal walk-off between idler and signal wave during nonlinear interaction](image)

This configuration is valid as long as the beam displacement along the direction normal to propagation is small enough to ensure good spatial overlap between the pump, signal and idler along the propagation as illustrated in Figure 55.

![Figure 55: Effects of beam size correlated with non-collinear geometry on the spatial overlap and consequently on the parametric gain](image)

An adverse effect of this configuration is that the idler is heavily spatially chirped. This consequence is of minor importance in the case of OPCPAs since the idler is usually discarded.
5.2.2 Broadband phase-matching: mathematical description

The non-collinear geometry introduces an angle $\alpha$ between the pump and the signal and results in an angle $\Omega$ between the idler and the signal. In this work, the pump beam is chosen as a reference to project other vectors onto. The arrangement and notations used for deriving the condition for ultra-broadband phase-matching are gathered in Figure 56.

![Figure 56: Schematic of the arrangement used in a non-collinear optical parametric amplifiers. The notation used for the derivation of the phase-matching condition are gathered on the schematic.](image)

The condition for phase-matching is generally expressed via the conservation of momentum:

$$\Delta k_0 = k_p - k_s - k_i$$  \hspace{1cm} (115)

This formula needs to be valid at all desired wavelengths in the case of broadband phase-matching. In order to ensure that $\Delta k$ is equal to zero over a large frequency range, it is useful to write the mismatch as a Taylor expansion around $\omega_0$:

$$\Delta k = \Delta k_0 + \left. \frac{\partial \Delta k}{\partial \omega} \right|_{\omega_0} \Delta \omega + \frac{1}{2} \left. \frac{\partial^2 \Delta k}{\partial \omega^2} \right|_{\omega_0} \Delta \omega^2 + \ldots$$  \hspace{1cm} (116)
Setting the first order term $\Delta k_0 = 0$ ensures phase-matching at the center wavelength the term $\left. \frac{\partial \Delta k}{\partial \omega} \right|_{\omega_0} = 0$ (with proper choice of non-collinear angle $\alpha$) guarantees broadband phase-matching since it guarantees no variations of the phase mismatch with respect to frequency. Equating higher order terms to zero enables finer adjustment of the phase mismatch. In order to express the condition between the wave vector of the signal and that of the idler ensuring broadband phase-matching, the three wave vectors are projected onto the pump beam and an expression for the phase mismatch is given for the direction collinear to the pump beam and the direction normal to the pump beam (see Figure 58).

$$\Delta k_\parallel = k_p - k_s \cos \alpha - k_i \cos \Omega$$
$$\Delta k_\perp = k_i \sin \Omega - k_s \sin \alpha \quad (117)$$

The derivative of both expressions is then expressed. Considering that the pump is monochromatic $\left( \frac{\partial k_p}{\partial \omega} = 0 \right)$ and that the angle between the pump and the signal is set and independent from the wavelength $\left( \frac{\partial \alpha}{\partial \omega} = 0 \right)$, the derivatives can be expressed as:

$$\frac{\partial \Delta k_\parallel}{\partial \omega} = -\frac{\partial k_s}{\partial \omega} \cos \alpha - \frac{\partial k_i}{\partial \omega} \cos \Omega + k_i \frac{\partial \Omega}{\partial \omega} \sin \Omega \quad \quad (118)$$
$$\frac{\partial \Delta k_\perp}{\partial \omega} = -\frac{\partial k_i}{\partial \omega} \sin \alpha - \frac{\partial k_s}{\partial \omega} \sin \Omega + k_i \frac{\partial \Omega}{\partial \omega} \cos \Omega \quad \quad (119)$$

The derivative of $\Omega$ with respect to $\omega$ is non-zero and therefore provides the variable of adjustment which enables broadband phase-matching. In addition, it results in spatial chirp in the generated idler beam.

138
The presence of the terms \( \frac{\partial k_m}{\partial \omega} \) corrected by cosine and sine term express the group velocity matching. In order to obtain an expression for the phase-matching condition, each equation is equated to zero. Eqn.(118) is multiplied by \( \cos \Omega \) and Eqn.(119) is multiplied \( \sin \Omega \) and both equations are added. The resulting expression is:

\[
\frac{\partial k_i}{\partial \omega} (\cos^2 \Omega + \sin^2 \Omega) - \frac{\partial k_i}{\partial \omega} (\sin \alpha \sin \Omega + \cos \alpha \cos \Omega) = 0
\]

which further reduces to:

\[
\frac{\partial k_i}{\partial \omega} = \frac{\partial k_s}{\partial \omega} \cos(\Omega(\omega) - \alpha)
\]

and can be expressed in a form having more physical meaning as the relationship between the group velocity of the signal and that of the idler:

\[
v_{s_r} = v_{s_i} \cos(\Omega - \alpha)
\]

It appears from the latter expression that under the right choice of material, input polarization and non-collinear angle \( \alpha, \Omega \) plays the role of adjustment variable and therefore enables broadband phase-matching over hundreds of nanometers.

For completion, a similar expression can be derived for the residual second order term. The second derivatives of \( k_{//} \) and \( k_{\perp} \) can be expressed as:

\[
\frac{\partial^2 k_//}{\partial \omega^2} = -\frac{\partial^2 k_i}{\partial \omega^2} \cos \alpha - \frac{\partial^2 k_i}{\partial \omega^2} \cos \Omega + 2 \frac{\partial k_i}{\partial \omega} \frac{\partial \alpha}{\partial \omega} \sin \Omega + k_i \frac{\partial^2 \Omega}{\partial \omega^2} \sin \Omega + k_i \left( \frac{\partial \Omega}{\partial \omega} \right)^2 \cos \Omega
\]

\[
\frac{\partial^2 k_{\perp}}{\partial \omega^2} = -\frac{\partial^2 k_i}{\partial \omega^2} \sin \alpha + \frac{\partial^2 k_i}{\partial \omega^2} \sin \Omega + 2 \frac{\partial k_i}{\partial \omega} \frac{\partial \alpha}{\partial \omega} \cos \Omega + k_i \frac{\partial^2 \Omega}{\partial \omega^2} \cos \Omega - k_i \left( \frac{\partial \Omega}{\partial \omega} \right)^2 \sin \Omega
\]
Setting these two equations to zero, multiplying Eqn.(123) by \(\sin\Omega\) and Eqn.(124) by \(\cos\Omega\) and adding the two equations leads to the condition for setting the phase-matching to zero up to the second order:

\[
-\frac{1}{v_{s,\omega}} \sin(\alpha + \Omega) + \frac{2}{v_{s,\omega}} \frac{\partial \Omega}{\partial \omega} + k_i \frac{\partial^3 \Omega}{\partial \omega^3} = 0
\]  

(125)

This expression has low physical insight but can be useful for phase-matching of spectra with bandwidth exceeding several hundreds of nanometers.

5.3 Phase evolution of the optical parametric process

The design of an efficient OPA requires careful computation of the phase-matching condition as discussed in the previous section. It is necessary to investigate the effects of the phase \(\theta\) (Eqn.(94)) between the three waves acquired during the amplification process. Considering the set of equation Eqn.(99) maximum amplification is achieved when \(\sin\theta = -1\) or \(\theta = -\pi/2\). The phase of the idler initially adjusts such that this condition is met to maximize the gain. It can be seen from the expression of \(\theta\) (in Eqn.(106)) that as the pulse propagates in the amplifying medium, some phase builds up if \(\Delta k\) is non-zero causing gain reduction. Eventually, the accumulation of phase can be such that \(\sin\theta\) becomes positive resulting in an effective energy flow from the signal and idler back to the pump which is not desired in NOPA. Practically, in the case of ultra-broadband spectra, \(\Delta k\) is not constant and oscillates around zero across the spectrum to be amplified which can result in energy flow back occurring at certain wavelength while other wavelengths are still being amplified. Distortions of the seed spectrum and potentially distortion...
of the output pulse shape in the time domain can be a consequence. It is then necessary to investigate the parameters responsible for the evolution of the phase $\theta$.

A procedure similar to that performed in section 5.1.2 can be executed to express a set of differential equations for the phase $\phi_p$, $\phi_s$ and $\phi_i$ the acquired phase of the pump, signal and idler. This set of equations is given by [127]:

$$\frac{d\phi_s}{dz} = -\frac{2\pi}{c^2} \chi_{\text{eff}} \frac{\omega_s^2}{k_s} \rho_p \rho_s \cos \theta$$  \hspace{1cm} (126)

$$\frac{d\phi_i}{dz} = -\frac{2\pi}{c^2} \chi_{\text{eff}} \frac{\omega_i^2}{k_i} \rho_p \rho_s \cos \theta$$  \hspace{1cm} (127)

$$\frac{d\phi_p}{dz} = -\frac{2\pi}{c^2} \chi_{\text{eff}} \frac{\omega_p^2}{k_p} \rho_s \rho_i \cos \theta$$  \hspace{1cm} (128)

where $\chi_{\text{eff}}$ is the effective nonlinear susceptibility and $\rho_m (m = p, s, i)$ are the amplitude of the three waves as defined in section 5.1.2. These equations can be written in a form carrying more physical insight by using the Manley-Rowe equations and assuming that the idler is initially zero. Under these assumptions, the equations are expressed as [127]:

$$\frac{d\phi_s}{dz} = -\frac{\Delta k}{2} \left[ 1 - \frac{\gamma_s^2}{f + \gamma_s^2} \right]$$  \hspace{1cm} (129)

$$\frac{d\phi_i}{dz} = -\frac{\Delta k}{2}$$  \hspace{1cm} (130)

$$\frac{d\phi_p}{dz} = -\frac{\Delta k}{2} \frac{f}{1 - f}$$  \hspace{1cm} (131)

where $\gamma_s = \frac{\omega_p}{\omega_s} \frac{I_s(0)}{I_p(0)}$ is the input photon intensity ratio and $f = 1 - \frac{I_p}{I_p(0)}$ is the fractional depletion of the pump beam [127]. From these equations, the initial phase of the idler, automatically set to maximize the gain, can be found to be $\phi_i(0) = \phi_p(0) - \phi_s(0) - \pi/2$. Integrating
the set of differential equations yields the expression of the phase acquired (via the increase of the $\Delta k z$ term) by each beam during the amplification process.

$$\phi_s = \phi_s(0) - \frac{\Delta k z}{2} - \frac{\Delta k \gamma_s^2}{2} \int \frac{dz}{f + \gamma_s^2}$$

$$\phi_i = \phi_i(0) - \phi_s(0) - \pi - \frac{\Delta k z}{2}$$

$$\phi_p = \phi_p(0) - \frac{\Delta k z}{2} \int \frac{f dz}{1 - f}$$

From these equations, it can be seen that the signal is independent of phase variations of the pump. Therefore the presence of spatial phase on the pump beam has little effects on the signal beam. Secondly, these equations confirm that the phase acquired across the spectral range during the amplification is non-uniform since the phase of each wave depends on $\Delta k$ and $\Delta k$ is not uniform across the amplified spectrum. Finally and most importantly, the acquired phase increases with propagation and, at maximum depletion, the energy starts to flow back from the signal and idler to the pump. Since maximum depletion can happen faster for some frequencies than others, dramatic spectral distortions can occur in the amplification process if saturation effects are not monitored.

5.4 Numerical modeling of a broadband non-collinear optical parametric amplifier

The modeling of the HERACLES optical parametric amplifier stages was performed in two steps:

- Development of a code for calculation and optimization of the phase-matching condition and
development of a code for solving the set of nonlinear partial differential equations ruling the parametric amplification process

This section is split into two sub-sections following the same logic. The calculation of $\Delta k$ under different conditions is first detailed and results of simulation displayed. In the second subsection, the code developed for solving the set of nonlinear partial differential equations is briefly discussed and the parametric study of the amplification process performed to develop the HERACLES OPA stages is presented.

5.4.1 Broadband phase-matching and non-collinear geometry

The phase-matching conditions and equations derived in the previous section have been implemented into MATLAB to design the HERACLES optical parametric amplifier. This investigation was motivated by the need to know both the range of phase-matching and non-collinear angles in which broadband amplification could be achieved and the angular sensitivity of the nonlinear process for experimental purposes.

Phase-matching is achieved by making use of the birefringence nature of nonlinear crystals. The crystals used for the optical parametric amplification of ultra-broadband pulses are crystals of $\beta$-BaB$_2$O$_4$ ($\beta$-BBO). BBO is a negative uniaxial crystal with trigonal crystal structure. The transparency window of BBO spans the spectral range from 190 nm to 3500 nm making it suitable for optical parametric amplification of optical pulses with center wavelength around 800 nm featuring an idler extending no further than ~2000 nm. The nonlinear coefficient of BBO is ~6 times higher than that of KDP allowing for short interaction length, therefore relaxing constraints on phase-matching conditions. BBO can be produced in large volume (up to
1.5x1.5 cm² aperture) and has a damage threshold of ~7GW.cm⁻² at 250 ps. Figure 57 shows the evolution of the ordinary and extraordinary refractive indices over the 200 nm to 1400 nm spectral range.

![Graph showing the evolution of ordinary and extraordinary refractive indices](image)

Figure 57: Computed evolution of the ordinary and extraordinary refractive index of BBO as a function of wavelength [216]

Since the ordinary index of refraction is higher than the extraordinary index over the entire range of amplification (700 nm to 1000 nm), it is necessary to polarize the 532 nm pump beam in the plane of the crystal optical axis. The signal and idler are polarized perpendicularly, and experience the ordinary refractive index. Setting the polarization to achieve phase-matching leaves two possible configurations: the signal beam can either be between the optical axis and the pump (tangential phase matching or TPM configuration) or the idler can be in this position (Poynting vector walk-off compensation scheme or PVWC) configuration as shown in Figure 58.
The pump beam is in the plane of birefringence. It therefore experiences spatial walk-off in which the Poynting vector and the wave vector are no longer collinear. The angle between the pump wave vector and its Poynting vector is given by:

$$\theta_{\text{walk-off}} = \frac{n_{\text{eff}}(\omega_p, \theta)}{2}\left(\frac{1}{n_o(\omega_p)^2} - \frac{1}{n_e(\omega_p)^2}\right)\sin(2\theta),$$

(135)

where $n_{\text{eff}}$ is the effective refractive index experienced by the pump beam as it propagates in the BBO crystal, $\theta$ is the phase-matching angle and $\omega_p$ the center wavelength of the pump. In the case of a phase-matching angle $\theta = 23.8^\circ$ and a center frequency $\omega_p = 3.54 \times 10^{15}$ Hz (center wavelength $\lambda_p = 532$ nm), the walk-off angle is approximately $\theta_{\text{walk-off}} = 3.3^\circ$, which is close to the value of the non-collinear angle $\alpha$ with maximum gain bandwidth. Experimentally, inputting the pump and signal beams in the wrong direction with respect to each other results in limited gain since the pump walks away from the signal, which is limiting the interaction region. This can be readily corrected by placing the crystal upside down.
In order to address the question of the sensitivity of the alignment and find the optimum phase-matching and non-collinear angles, a parametric simulation has been performed (Figure 59). The optimum non-collinear angle was found to be $\alpha = 2.36^\circ$. The phase-matching angle that minimizes the phase mismatch along propagation is $\theta = 23.83^\circ$. Due to the S-shape of the phase-matching curve, the angle $\theta = 23.8^\circ$ results in the broadest amplification bandwidth. The parameters chosen for the HERACLES optical parametric amplifier stages are therefore $\alpha = 2.36^\circ$ (internal angle) and $\theta = 23.8^\circ$. Through this parametric simulation, the amplification process was found to be extremely alignment sensitive: a deviation of $\sim 0.5^\circ$ from the optimum angular values would result in dramatic alteration of the spectral gain shape and width.
Figure 59: Simulations of the effects of phase-matching angle tuning (a, $\theta = 23.83^\circ$, c, $\theta = 23.86^\circ$, e, $\theta = 23.89^\circ$) and non-collinear angle (b, $\alpha = 2.36^\circ$, d, $\alpha = 2.39^\circ$, f, $\alpha = 2.42^\circ$) tuning on the phase mismatch and gain spectral profile.
5.4.2 Introduction of the model

The considerations on phase-matching presented in the previous section give a first order approximation to the optical parametric amplification process. In order to accurately predict the performances of the OPA stages, spectrally, spatially and temporally, it is necessary to solve the set of nonlinear partial differential equation (Eqn.(92)). The computing strategy adopted for this model is to define, for each wavelength a spatial profile and numerically propagate this 2D array along with the pump beam through the nonlinear medium. The process is repeated for each wavelength and the spectrum and overall spatial profile are reconstructed at the end of the numerical amplification by concatenating the different spectral slices and adding the beam profiles together. The overall code scheme is represented on Figure 60.

![Figure 60: Schematic representation of the strategy employed to solve the partial differential equations ruling the optical parametric amplification process in space, time and spectral domains](image)

This approach allows simulation of spatial walk-off, taking into account potential spatial chirp on the signal beam and to define the spatial and temporal profile of the pump and signal beams. The code requires simply the experimental spectrum of the signal as in input: spectral...
phase can be added to the signal to simulate the effect of a pulse stretcher on the signal beam and provide an additional degree of freedom in the modeling. In order to limit the computational requirements, the code neglects the effects of spectral phase acquired during the propagation. As an indirect consequence, the code is unable to operate in strongly saturated regimes.

5.4.2.1 Influence of walk-off

The influence of the pump spatial walk-off has been numerically investigated. In the previous section detailing the phase-matching condition, the spatial effects were not taken into account and the sign of the non-collinear angle $\alpha$ was irrelevant. A simulation was run in relatively low gain conditions for the first amplifier stage where the pump pulse energy $E_p$ was 250 $\mu$J, the pump beam diameter was 400 $\mu$m and the pump pulse duration was $\sim$85 ps FWHM. The seed energy was chosen to be 20 pJ, a value preventing any saturation effects (see section 5.4.2.4) and close to experimental values for the first OPA stage. The beam diameter was matching that of the pump beam. The pulse duration of the signal pulse was $\sim$13 ps, obtained by linearly chirping a 5 fs pulse with 20000 fs$^2$ second order dispersion and the amplification was performed in a 5 mm long BBO crystal. The resulting effects on the gain are shown in Figure 61 and the two configurations are schematically depicted in Figure 58.
In the case of a negative birefringent crystal such as BBO, the beam in the plane of birefringence walks away from the optical axis. In the configuration where the wave vector of the signal beam lies between the optical axis of the crystal and the pump beam wave vector \((\theta - \alpha)\) the maximum gain over the entire amplification bandwidth is \(\sim 4.5\). On the other hand, when the signal beam is placed at an angle \(\theta + \alpha\) with respect to the optical axis, the spectral gain reaches up to 200 at 900 nm, a 50-fold improvement with respect to the previous configuration.
5.4.2.2 Influence of pump-seed synchronization

The pump and signal pulses involved in the parametric process are both a few tens of picosecond long. The optical parametric amplification process provides gain as high as $10^4$ in the first stage and is therefore extremely sensitive to timing jitter. The optical synchronization design of HERACLES ensures jitter below the picosecond range (the jitter depends only on thermal expansion of components and instability of the optical mounts) and is therefore kept in check. Furthermore, a number of operating systems have been reported relying on such design [134, 136, 217-219]. Nevertheless, the optical path experienced by the pump pulse between the beam splitter that separated the pump and signal beam is significantly longer (fiber amplifier, regenerative amplifier, single pass amplifier and multi-pass booster amplifier) than that experienced by the signal pulse (stretcher). The signal pulse interacting with the pump pulse in
the OPA has been produced by the Ti:Sapphire master oscillator a few nanoseconds after the pulse used in the pump beam line. These pulses are intrinsically synchronized by the high stability of the repetition rate of the Ti:Sapphire oscillator but need to be timed properly via a delay line. Figure 63 shows the effects of improper timing of the arrival time of the pump pulse with respect to the signal pulse. The simulation was performed assuming 250 $\mu$J of pump energy, a beam diameter of 400 $\mu$m and pump pulse duration of ~85 ps. The signal beam had an energy of 20 pJ, a beam diameter of 400 $\mu$m and the pulse was linearly chirped to 13 ps. The amplification was performed in a 5 mm long BBO crystal.
Figure 63: Simulated effects of time delay between the pump and signal pulses on the spectral gain profile for -100 ps (a), -50 ps (c), 0 ps (e), 50 ps (g) and 100 ps (i) delays and effects of time delay between the pump and signal.
pulses on the amplified spectrum (dark line) for -100 ps (b), -50 ps (d), 0 ps (f), 50 ps (h) and 100 ps (j) delays compared to the seeded spectrum (grey area).

The effects of delay mismatch between the two interacting pulses are twofold: first due to the reduced overlap of the pulses, the overall gain decreases. Second, and more critically, since the signal pulse is chirped, a time mismatch signifies that some wavelengths might be located in the tail of the pump pulse or even outside the pump pulse resulting in reduced or no gain. Therefore, a time offset can translate into spectral distortions, spectral narrowing and eventually lengthening of the amplified pulse.

5.4.2.3 Influence of seed chirp and pump to signal pulse duration ratio

The design of an OPA requires choosing the pulse duration of the pump and signal pulses appropriately. Optimizing the amplification process requires stretching the signal pulse enough to prevent damages upon amplification and ensure temporal overlap between the pump and signal pulse during the amplification process. If the signal pulse is short compared to the pump pulse, the non-overlapped part of the pump pulse is not contributing to the optical parametric amplification of the signal pulse but to the generation of superfluorescence, or amplification of quantum noise, a parasitic process inherent to the large gain of parametric amplifiers [118, 121, 220-223]. The pump pulse being Gaussian, if the pump and signal pulse have comparable durations this results in lower gain for the wavelength in the tail of the spectrum. A simulation has been performed where the signal pulse duration was adjusted from 6.5 ps to 75 ps – by adjusting the amount of initial chirp provided to the ultra-broadband experimental seed spectrum – and the pump pulse was set to 70 ps. The amplification was performed in a 5 mm long BBO crystal.
The FWHM amplified spectral width is not affected by the signal pulse duration until the pulse duration reaches ~30% of that of the pump pulse. Beyond this point and until the signal pulse reaches 80% of the duration of the pump pulse, the spectral bandwidth being amplified shrinks linearly from 300 nm to ~200 nm. The effect of such spectral narrowing in the temporal domain have been investigated by computing the transform-limited pulse generated by the extreme case where the signal pulse fills only 20% of the pump pulse and the case when the signal is 10% longer than the pump pulse. The result is less dramatic than could be anticipated from the spectral standpoint: an increase of only 17% from 7 fs to 8.3 fs in pulse duration is observed while the spectral bandwidth is reduced by over 30%. Such a discrepancy is resolved by observing the shape tail of both compressed pulses: in the case where $\tau_s/\tau_p = 0.2$, the broader amplified spectrum results in pre- and post-pulses in the time domain. The spectrum of the seed pulse is amplified relatively uniformly and the wider spectrum only contributes to generate side pulses. The time bandwidth product of this pulse is ~0.8. On the other hand, when the seed pulse is stretched to match that of the pump pulse ($\tau_s/\tau_p = 1.1$) the seed spectrum is heavily shaped by

Figure 64: a. Calculated fraction of the seeded bandwidth being amplified as a function of ratio between the pump pulse and signal pulse duration; b. corresponding time domain FWHM pulse duration for the extreme cases of $\tau_s/\tau_p = 0.2$ and $\tau_s/\tau_p = 1.1$. 
the gain and converges toward a 200 nm wide Gaussian spectrum which results in smaller pre- and post-pulses and a time bandwidth product close of 0.69, closer to 0.441, the theoretical limit.

5.4.2.4 Influence of pump depletion on spectral properties

Optical parametric amplifiers can provide several order of magnitude of gain over a few millimeters interaction length. Depletion of the pump beam can happen when the seed energy is high enough. This effect is illustrated on Figure 65 for a pump beam with 250 µJ energy, 300 µm diameter and ~85 ps duration and a seed pulse with energy adjusted from 20 pJ to 20 nJ, a constant beam diameter of 300 µm and a constant pulse duration of 13 ps.

Figure 65: Simulated pump depletion for 20 pJ (red line), 200 pJ (green line), 2 nJ (black line) and 20 nJ seed energy.

The depletion process was investigated by looking at the effects of increasing seed energy on the pump pulse. The pump pulse temporal profile at the output of the 5 mm OPA crystal is plotted for the same seed energies considered in the previous study (20 pJ, 200 pJ, 2 nJ and 20 nJ). The increase in seed energy results in an increasing depletion of the pump where the signal and pump pulses overlap in time. Saturation of the final OPA stage is a way to ensure
maximum output amplified bandwidth but also results in efficient amplification of any superfluorescence that could have been generated in the preceding parametric amplifier stages.

### 5.4.2.5 Influence of crystal length on gain and amplified spectral width

The design of the OPA requires choosing the length of the nonlinear crystal. Simulations were performed to compute both the gain and spectral bandwidth for crystal lengths ranging from 2 mm to 10 mm. The code used for the spectral bandwidth simulation was that used in section 5.2 while the code used for the gain computation was the one used in this section (5.4). The values of the gain have been normalized to the maximum achieved gain since the trend is independent from parameters such as pulse energy, beam diameter and pulse duration – assuming no saturation effects.

The FWHM spectral linewidth increases from 150 nm to over 350 nm when the crystal length is reduced from 10 mm to 1 mm (Figure 66.a). The dramatic increase in spectral

![Figure 66: a. Simulated effects of nonlinear crystal length on spectral gain profile and bandwidth; b. effects of nonlinear crystal length on parametric gain shown on a logarithmic scale (blue curve) and linear curve (black curve). The linear curve has been normalized to the maximum gain.](image-url)
bandwidth is explained by a reduced acquired spectral phase during the amplification process as the crystal length is reduced. The phase mismatch $\Delta k$ is constant regardless of the crystal length but the acquired phase between the three waves is the product of the phase mismatch $\Delta k$ and the crystal length. As the amplified spectral bandwidth is increased by a factor $\sim 2.4$, for a crystal length reduction from 10 mm to 1 mm, the gain is reduced by five orders of magnitude. In the HERACLES design, 5 mm BBO crystals have been chosen as a trade-off between gain and bandwidth and transform-limited pulses as short as $\sim 7$ fs could be achievable.

Reducing the interaction length results in a drastic reduction of the gain. This reduction can be overcome by increasing the peak power of the pump and signal pulses to drive the nonlinearity harder and compensate the gain loss inherent to a shorter crystal. Such an approach permits the achievement of a wide amplified bandwidth without sacrificing gain. This can be achieved by reducing the beam diameter or increasing the pump energy, to the limits set by the damage threshold. Another approach to increasing the peak power density in the OPA crystal is to deliver pulses with duration of a few picoseconds rather than a few tens of picoseconds, the damage threshold of optical coating typically increase for shorter pulse durations.

5.5 **Beyond the phase-matching condition: two-color pumping**

In the prospect of even shorter pulses, broadband phase-matching via non-collinear geometry in BBO offers the possibility to generate pulses with transform-limited duration $\sim 7$ fs. Using shorter pump pulse duration allows the use of shorter crystals therefore allowing for pulses with duration in the sub-5 fs. Nevertheless the extremely short length of the crystals limits the
gain (section 5.4.2.5) and is therefore becomes impractical to generate high energy few cycle pulses.

Another approach was recently proposed and experimentally demonstrated by [224] featuring a two-color pumping scheme: a high-energy pump beam at the fundamental frequency of 1064 nm is frequency-doubled and -tripled, generating two OPA pump beams at respectively 532 nm and 355 nm. The gain bandwidth provided by the 532 nm beam ranges from 750 nm to 1050 nm while the 355 nm, also in a non-collinear arrangement, provides amplification in the spectral range spanning ~575 nm to 750 nm. The slight overlap between the two amplified spectral regions enables uniform amplification over almost an octave. Simulations have been performed using the code used in section 5.4.1 and the results are shown in Figure 67.

![Figure 67: a. Simulated amplified spectrum under two-color pumping (532 and 355 nm) conditions (blue line) compared to seed spectrum (gray shaded region); b. simulated comparison of minimum achievable pulse duration between single-color pumping at 532 nm (black curve) and two-color pumping at 532 nm and 355 nm (blue curve).](image)

Pulses as short as 3.8 fs (corresponding to ~1.4 electric field cycle) could be generated via this technique: a two-folds improvement in pulse duration with respect to single beam pumping. This technique is scalable in energy, does not require the use of thinner crystals and is simple to implement, unlike the other proposed schemes [225, 226]. It therefore appears as a
good candidate for the generation of quasi-single cycle pulses with energy in the multi millijoule level. Nevertheless, technological developments will be required to ensure proper dispersion management and successful compression of pulses with such large bandwidth: currently, only chirped mirrors appear to be capable of handling the dispersion management of ultra-wide spectra.

5.6 Conclusions for HERACLES optical parametric amplifier stages

The theoretical study performed in the previous sections aims at facilitating the experimental implementation of the HERACLES optical parametric amplifiers. Several conclusions can be drawn from the simulations.

- The spatial effects need to be carefully monitored, particularly the spatial walk-off of the pump that can hinder the gain if the crystal is placed upside down.
- The trade-off between tight focusing and spatial walk-off needs to be taken into account when setting the beam sizes at the different stages of the amplification: damage threshold considerations are not the only concern, too small of a beam results in greater sensitivity to spatial-walk off.
- The angular alignment of the optical parametric amplifier is critical. Alignment needs to be performed with great care to optimize both gain and amplified bandwidth.
- The alignment of the delay line implemented to provide temporal overlap between the pump and signal pulse needs to be achieved within 1 cm accuracy over up to 1.2 m.
- The choice of the crystal length is a trade-off between maximum achievable gain and amplified spectral bandwidth.
Saturation effects need to be monitored in order to not distort the amplified spectrum. It is nevertheless valuable to saturate the final optical parametric amplifier stage since it allows amplification of the full gain bandwidth efficiently.
6 AMPLIFIER SYSTEM PERFORMANCE

6.1 Ti:Sapphire oscillator front-end

6.1.1 Layout and performances

The main oscillator of the system is a carrier envelope phase (CEP) stabilized Ti:Sapphire oscillator providing an octave spanning spectrum and 5 fs pulses. The 1.8 m long cavity leads to an 85 MHz repetition rate. The energy per pulse is typically ~1.5 nJ. Figure 68 shows the layout of the oscillator.

![Figure 68: Layout of the sub 6 fs Ti:Sapphire oscillator.](image)

A typical spectrum obtained from the oscillator is shown in Figure 69. In order to represent the full bandwidth and the shape of the spectrum, both logarithmic and linear scales are shown. Even though the spectrum is ultra wide and spans an octave, two main peaks can be observed on both edges of the emission bandwidth.
The shape of the spectrum can be explained by the 1% transmission of the output coupler; the large peaks in the emitted spectrum correspond to the wavelength for which the reflectivity of the output coupler decreases on both side of the spectrum. The choice of reducing the reflectivity of the output coupler for wavelengths below 750 nm and beyond 1000 nm is dictated by the fact that the gain bandwidth of Ti:Sapphire starts dropping significantly for these spectral ranges. Furthermore, most of the spectral bandwidth produced in the wings of the spectrum are produced via self-phase modulation in the Ti:Sapphire crystal and experience more loss than gain in the cavity. The reflectivity of the output coupler, is kept as high as 99% to maintain a high peak power in the cavity to drive the self-phase modulation. Nevertheless, this results in limited output power and numerous instabilities such as Q-switch-mode-locking.

In order to mitigate this issue, MenloSystems Inc. have developed a new 2% output coupler [227] with increased reflectivity in the spectral wings. Such an output coupler, still undergoing tests, has proven to significantly smoothen the spectrum but did not allow for the emission of an octave spanning spectrum since hardly any emission could be observed beyond 900 nm.
The output pulse duration of the Octavius was measured to be ~6 fs by interferometric autocorrelation (Figure 70). The home-made autocorrelator was in a Michelson-type collinear configuration and the dispersion of the two beam splitters inserted in the assembly was pre-compensated by chirped mirrors before performing the measurement. The nonlinear medium employed in the measurement was a 30 µm thick Type-I BBO crystal (Quantum Technology).

![Figure 70: Measured interferometric autocorrelation trace showing the 6 fs pulse produced by the Octavius oscillator](image)

The output pulse shows a slight amount of residual chirp. This chirp is attributed to imperfect pre-compensation of the dispersion of the output coupler of the oscillator and of the beam splitters placed in the autocorrelator. The output profile of the oscillator was also recorded at the output of the oscillator and is shown in Figure 71.
The output profile of the Ti:Sapphire oscillator is far from ideal for amplification purposes: in the near field (~50 cm away from the output of the laser), a shape as shown in Figure 71 is typically obtained regardless of alignment. For distances greater than ~50 cm the spatial phase probably acquired in the Ti:Sapphire crystal while driving the self-phase modulation leads to complex, modulated beam patterns, strongly varying with alignment. It was also found experimentally that the beam exhibits spatial chirp. These issues have been solved by implementing a spatial filter at the output of the oscillator as discussed in section 6.4.1.

6.1.2 Optimization of mechanical design

The initial mechanical design of the laser has been significantly modified to make it more user-friendly and reliable. The initial design consisted of a base plate onto which a rigid cover could be placed. The stiffness of the cover was such that, when placing the cover on the oscillator, distortions to the base plate led to systematic misalignment of the oscillator. Furthermore, the tight fit between the cover and the base plate occasionally resulted in contact between the plate and the cover, also leading to alteration of the laser performances. Finally no external access was provided to adjustment knobs when the laser was fully encapsulated which
forced the user to remove the cover to make minor adjustments to the cavity, leading to increased contamination – e.g. dust into the cavity.

Most of the initial mechanical defects in the design of the laser have been fixed by MenloSystems Inc. based on our recommendations. The dimensions of the base plate and the cover were slightly modified to allow a 2 mm gap between the plate and the sides of the cover. The encapsulation was redesigned so that the stiffness of the cover was less than the stiffness of the base plate. This has been achieved by making the sidewalls of the laser encapsulation part of the base plate. Finally holes have been drilled in the sidewalls to allow easy access to the main knobs without opening the encapsulation allowing better contamination management.

6.1.3 Optimization of thermal management

A significant part of our work on the oscillator has been to make it a turnkey system and increase its long term stability. The initial design of the thermal management did not take into account long-term drift of the mechanical parts and led to significant changes in performance during day-long operation and inconsistent behavior from day to day.

In order to avoid the vibrations typically induced by water-cooling, the crystal and the base plate (on which the laser is mounted) were both thermo-electrically cooled. The thermal management of the base plate was used to isolate the laser from room temperature fluctuations. A thermal sensor was placed at the center of the plate holding the laser and was located under the crystal holder. The crystal holder consists of a block of aluminum used as a heat sink extracting the heat generated by the pump light in the crystal. The heat was then transferred to the optical table via the base plate. For mechanical stability the base plate was held on three stainless steel feet that mechanically isolate the oscillator from the table. Since stainless steel has a poor
thermal conductivity, the heat was not efficiently transferred to the table and accumulated in the plate holding the laser. Slight expansion of the aluminum base plate led to systematic misalignment of the oscillator after a few hours of operation. Furthermore the location of the thermal sensor, precisely under the heat sink, used for feedback to maintain the base plate temperature constant led to the creation of a temperature gradient in the plate.

Since the cause of the long time drift of the laser performance was initially unknown, the temperature of the base plate was monitored in tests in our laboratory under several experimental conditions. Operation of the laser in the mode-locked regime results in a much more efficient extraction of the gain, leading to higher output power and consequently reduced heat generated in the crystal. The typical non-mode-locked output power is ~30 mW while up to 130 mW can be obtained in the mode locked regime. Therefore a parametric study was necessary to optimally set the pump power, the temperature of the thermo-electric controller of both the crystal and the base plate and record both the temperature of the base plate and the output power.

A first experiment was performed by setting the crystal temperature $T_{\text{crystal}} = 14$ °C and the base plate temperature $T_{\text{base plate}} = 25$ °C, slightly above room temperature and measure the temperature and power evolution over several hours in both mode locked and non-mode locked regimes. In the mode locked regime, a less dramatic drift is expected since a larger percentage of the pump power is extracted. The results are shown in Figure 72 and are in good agreement with the expectations: in the non-mode-locked regime the base plate experienced a ~2.2 °C drift resulting and a relative power fluctuation of 25 %. In the mode locked regime the base plate experienced a thermal drift of only ~1 °C which resulted in a relative power fluctuation less than 4%.
Another set of experiments were run where the pump power was adjusted in the non-mode-locked regime. Increasing the pump power in its most inefficient regime should lead to a higher thermal strain on the system. Therefore, if the long term drift is due to poor thermal management, increasing the pump power should lead to more unabsorbed and unused power converted into heat. The experiment was run with the laser non-mode locked, the crystal temperature set to $T_{\text{crystal}} = 14 \, ^{\circ}\text{C}$ and the temperature of the base plate $T_{\text{base plate}} = 25 \, ^{\circ}\text{C}$. In the first experiment, the pump power was set to 3.5 W. The non absorbed pump power transmitted
through the Ti:Sapphire crystal was measured to be 1.5 W, therefore the power converted into heat is estimated to be ~1 W. In the second experiment, the pump power was set to 5.5 W. The non absorbed power measured behind the crystal was 2 W, leading to an estimate power converted to heat of ~3 W. Figure 73 shows the results obtained when monitoring the temperature of the base plate and the output power over several hours under these conditions.

Figure 73: Left column – evolution of temperature and power as a function of time for 3.5W pump power, $T_{\text{crystal}} = 14 \degree C$ and $T_{\text{base plate}} = 25\degree C$ in non-mode locked regime, right column – evolution of temperature and power as a function of time for 5.5W pump power, $T_{\text{crystal}} = 14 \degree C$ and $T_{\text{base plate}} = 25\degree C$ in non-mode locked regime

For a pump power $P_{\text{pump}} = 3.5$ W, the temperature eventually stabilizes around 27.5 °C. If the laser was kept on permanently, a steady state could be reached. In the case $P_{\text{pump}} = 5.5$ W,
there is no such convergence and the temperature of the base plate keeps increasing. After 250 minutes, since the power kept going down, the experiment was stopped to prevent damage to the crystal. Indeed, the heat sink controlling the crystal temperature was hot to the touch, proving that the heat was flowing back from the plate to the crystal and proving that the system had become unstable and that the heat was not being removed efficiently.

In order to solve these thermal issues and improve both day-to-day and long term stability of the laser, MenloSystems Inc. made several improvements based on our recommendations. The stainless steel feet mechanically isolating the oscillator from the table were replaced with aluminum feet providing improved heat transfer to the optical table. The oscillator and the pump are also now both mounted on a common water cooled base plate ensuring efficient heat removal. The water cooled base plate is specially designed to mitigate vibrations. The thermal sensor of the base plate has been moved away from the crystal heat sink. These measures improved the long term stability and prevented the laser performance from drifting over several hours. In order to improve day-to-day stability, the thermal management of the crystal has been modified. The initial design consisted of a single thermo-electric cooler with a feedback loop. The newly designed temperature controller of the crystal consists of two competing thermo-electric controllers: one controller is permanently set to cool the crystal down to ~13 °C but has a constant current available while a second controller is permanently set to warm up the crystal to 20 °C and can adjust its current to reach the targeted temperature. Having one thermo-electric cooler permanently maintaining the crystal down to 13°C results in flowing a constant amount of heat from the crystal heat sink to the base plate. Since the second thermo-electric cooler, responsible for warming up the crystal has adjustable current, it can be used to buffer the influence of the optical pump power: when the pump is on, the current is reduced and vice versa.
Therefore the amount of heat flowing from the crystal to the base plate is essentially constant. With these improvements, the oscillator now maintains mode locking all day long without any adjustment and is close to turn key operation. A warm up time of 20 minutes is usually required in the morning with minor adjustment to reach optimum performances.

6.1.4 Carrier-envelope phase stabilization of the front-end oscillator

HERACLES is designed to enable field-sensitive experiments. It is therefore necessary to have controlled over the carrier-envelope phase, the relative lag between the electric field and the Gaussian pulse envelope. This lag arises from the difference between the propagation speed of the envelope (group velocity) and the propagation speed of the carrier (phase velocity).

The frequency $f_{CEO}$ at which the carrier sweeps under the envelope can be readily extracted by an f-2f interferometer. The IR-tail ($\lambda > 1100$ nm) and the green-tail ($\lambda < 600$ nm) of the ultra-broadband spectrum provided by the Octavius are sent to this interferometer. The IR-tail is frequency-doubled in a thin LBO crystal cut to phase-match second harmonic generation of wavelength around 1140 nm. The frequency-doubled signal beats with the fundamental at 570 nm and the beat signal, directly providing $f_{CEO}$, is recorded on a photodiode. The layout of the f-2f interferometer is shown in Figure 74 and the underlying principle of the system is explained in Appendix A.
In the case of HERACLES, the oscillator is designed such that $f_{CEO}$ is equal to ~21 MHz, a quarter of the repetition rate. Upon its measurement, the value of $f_{CEO}$ is fed into an electronic feedback loop that compares the value of the beat signal to the exact value of the quarter of the repetition rate (this value is measured directly by a photodiode placed in the system and divided by four by a frequency divider). The error signal generated by the difference between the beat signal and the exact quarter of the repetition rate is treated to generate an output signal being fed into the power supply of an acousto-optic modulator placed in the pump beam of the Ti:Sapphire oscillator. Slight adjustments of the pump power are sufficient to modify the properties of the gain medium and control the group velocity and phase velocity, therefore enabling control of $f_{CEO}$. Measurements of the error signal between the exact quarter of the repetition rate and the beat signal have been performed when the phase was locked and unlocked over ~1 minute and are reported in Figure 75.
The width of the error signal is \( \sim 100 \) kHz and no excursion could be observed when the feedback loop was enabled (aside from transient unlocking at time 30 s and 40 s). On the other hand, when the feedback loop is disabled, the error signal wanders over \( \sim 100 \) kHz. Improvements have been made and are currently being added to enhance CEP phase stability and detection of HERACLES.

6.2 Pump beam generation amplifier line

6.2.1 Fiber pre-amplifier – motivation

The fiber pre-amplifier of HERACLES ensures optimal seeding of the subsequent amplifiers of the chain and fulfills four tasks:

- reaching a few nanojoules of energy
- maintaining a high signal to ASE ratio
• providing an excellent beam profile
• buffering energy variations from the ultra-broadband seed laser.

**Reaching a few nanojoules of energy** enables efficient seeding of the regenerative amplifier (Figure 7). The influence of the seed energy level has been investigated by Ishii *et al.* [219] and shows that a reduced seed energy results in dramatic increases in ASE levels (Figure 76).

![Figure 76: Influence of the seed energy level on the output signal to ASE ratio for high gain amplifier [219].](image)

Maintaining a high signal to ASE ratio ensures the generation of high contrast pulses throughout the amplifier chain. The presence of an ASE pedestal reduces the efficiency of the amplifier chain since a fraction of the overall gain is provided to the pedestal.

Providing an excellent beam profile is necessary to prevent damage in the subsequent amplifiers, maximize energy extraction in the chain and achieve optimum optical parametric amplification.

Buffering the energy variations from the ultra-broadband seed laser enables low jitter output of the entire amplifier chain. The seed of the amplifier chain is a small fraction of the tail of the spectrum of the ultra-broadband Ti:Sapphire oscillator. This seed is not stable in
time, neither on a few hour time scale nor on a day to day time scale. It is therefore required that the pre-amplifier buffers these fluctuations.

6.2.2 Fiber pre-amplifier implementation

Amplification in the fiber pre-amplifier is performed in two consecutive and identical stages. Such splitting of the amplification limits the gain in each amplifier and filters undesired amplification byproducts between the amplifiers. The first stage is designed to bring the signal above the ASE level while efficient amplification is provided in the second stage. This approach enables reaching the nanojoule level of energy while providing a high contrast pulse train. Single mode fibers are used in order to provide TEM\(_{00}\) spatial output. The second stage is strongly saturated thereby decoupling the variations of the seed energy to the rest of the chain.

Half of the output beam of the Ti:Sapphire oscillator is sampled and directed toward the fiber pre-amplifier by two mirrors HR at 1064 nm. The unused light leaking through the mirrors is sent onto a fast photodiode and used to provide trigger signals for the entire system. The \(~50\) nm wide spectral band filtered in this process is coupled into a 5 \(\mu\)m core, 125 \(\mu\)m cladding, single mode, polarization maintaining (PM) undoped fiber (Figure 77) by a 1.1 cm focal length lens. The coupling efficiency is limited to \(~10\%) due to the poor beam quality of the Ti:Sapphire oscillator, resulting in a seeded energy in the sub-picojoule range.
Figure 77: Cross-section of the polarization maintaining Yb fiber used in the regenerative amplifier. The 5 µm core appears between the two large stress rods ensuring horizontally polarized output.

The seeded pulses pass through a fiber coupled isolator (OFR) providing a ~30 dB isolation. This isolator prevents ASE from propagating back to the Ti:Sapphire oscillator, reflect on the 99% reflective output coupler of the oscillator and couple back into the fiber pre-amplifier. The seed pulses are then sent into the gain medium consisting of a 15 m long single mode, Yb-doped PM fiber (Figure 77) via a three way coupler (Novawave). The choice of Yb as a doping ion was dictated by the low gain of Yb at 1064 nm resulting in limited ASE generation, providing a favorable signal-to-ASE ratio at the input of the amplifier. The pump power is provided by a 750 mW single emitter diode operating at a fiber-Bragg-grating-stabilized wavelength of 976 nm (Bookham) coupled to the gain fiber via the three way coupler. The pulses amplified in the first pre-amplifier stage are sent into the second, identical stage via a spectral filter with a spectral linewidth of 8 nm centered at 1064 nm (Novawave). The second pre-amplifier stage is terminated by an inline isolator preventing high energy pulses from the regenerative amplifier to couple back into the pre-amplifier and a fiber coupled collimator (Thorlabs) ensuring excellent collimation of the output beam. Figure 78 shows the layout of the two fiber pre-amplifier stages.
6.2.3 Fiber pre-amplifier performances

The 85 MHz pulse train seeded into the pre-amplifier with an average power of ~8.5 µJ is amplified to an average power of ~2 mW after the first pre-amplifier stage, resulting in pulses with 23 pJ energy. The gain of the first stage is ~200 at full pump power. The average output power provided by the second stage, seeded by the maximum output of the first stage reaches up to 240 mW, or correspondingly 2.8 nJ energy per pulse. The nonlinear relation between pump power and output energy (Figure 79.a) shows the strong saturation of the second pre-amplifier stage, therefore isolating the variations of the seed from the remaining of the amplifier chain. The gain of the second stage at full pump power is comparable to that of the first stage (~200). The effectiveness of the dual stage approach is illustrated in Figure 79.b: the output spectrum of the second stage was measured a function of the pump power in the first amplifier, therefore adjusting the seed energy level of the second stage. For low seed energy levels, the output spectra show large ASE peaks at 1030 nm. As the seed energy level is increased, the ASE level is decreased and the peak at 1064 nm is increased, eventually leading to complete suppression of the ASE.
Spectral broadening, attributed to self phase modulation in the fiber is observed at the output of the second stage. A complex spectral phase is expected at the output of the pre-amplifier due to the conjugated effects of self phase modulation and normal dispersion acquired along the propagation in the 30 m long pre-amplifier. The pulse duration was measured via intensity autocorrelation at the output of the pre-amplifier and showed a 25 ps pulse (Figure 80).

Figure 80: Autocorrelation trace of the output pulses of the fiber pre-amplifier.
6.2.4 Regenerative amplifier implementation

The pulse train provided by the fiber pre-amplifier is prepared for injection into the regenerative amplifier in the seed injection line. The axis of polarization of the linearly polarized output of the pre-amplifier is rotated to vertical via a half wave plate placed at the output of the pre-amplifier. A collimator is mounted on the output of the fiber pre-amplifier providing a collimated beam of ~4 mm diameter. A telescope assembly made of a 10 cm and a -5 cm AR coated lenses reduces the beam size to ~2 mm, the size of the Nd:YVO₄ rod in the regenerative amplifier. The 85 MHz pulse train is then passed into an RTP pulse picker enabling reduction of the repetition rate to frequencies between single shot up to 25 kHz. The pulse picker is operated in the quarter wave voltage regime: the pulses picked experience a quarter wave delay, resulting in circular polarization output while the remaining of the pulse train remains vertically polarized. A thin film polarizer is placed behind the pulse picker, reflecting the vertically polarized pulses and transmitting 50% of the picked pulses. Operating in the half wave voltage would result in complete transmission of the picked pulses but limitations in the high voltage switch prevent operation at high repetition rate at half wave voltage. The horizontally polarized pulses transmitted through the TFP are sent to another half wave plate to rotate their polarization by 45° before being sent through a Faraday rotator that provides an additional 45° rotation of the plane of polarization resulting in vertical polarization. The prepared pulses are then sent into the regenerative amplifier via reflection off a thin film polarizer placed in the regenerative amplifier cavity Figure 81.
The regenerative amplifier layout is based upon the previous simulations. The gain medium is a water cooled 4 cm long, 2 mm diameter Nd:YVO₄ rod with a 0.3 at.% dopant concentration. The rod is side pumped by three sets of diode bar arrays providing a maximum total output power of 180 W (RBA module, Northrop Grumman Cutting Edge Optronics, Figure 82.a). The 4 cm long rod held by two glass sleeves glued at each end of the rod.

The profile of the fluorescence is shown in Figure 82.b. The image was acquired by imaging the end of the rod onto a CCD camera. The blue circle surrounding the center region is
the image of the holding sleeve, slightly illuminated by the pump light coupled in the sleeve. The image was recorded at full pump power and shows a rather uniform gain distribution.

The resonator is the ~1 m long flat-flat cavity described in 3.4.4. The chosen cavity length allows pumping up to ~100 W without risking damages on the cavity end mirrors and allows for output pulses with energy in the 800 µJ range. A thin film polarizer, quarter wave plate and Pockels cell are added to the cavity to allow injection and ejection of the seed pulses. \( \beta \)-\( \text{BaB}_2\text{O}_4 \) (BBO) was chosen for the material of the Pockels cell (Quantum Technology) as it exhibits a higher damage threshold than RTP. The main inconvenient of BBO compared to RTP is its higher quarter voltage requiring more complex switching electronics.

The pulse duration and signal to ASE ratio are controlled by a VBG (Optigrate Corp.) as described in 3.4.5. The VBG (5x5x10 mm³) has a spectral reflectivity linewidth of 50 pm and a reflectivity of 70 % (Figure 83).

![Figure 83: Transmissivity of the VBG used in the HERACLES regenerative amplifier (source: specification sheet from Optigate Corp.).](image_url)

The VBG is AR coated and the holographic grating is recorded at a slant angle with respect to the glass surface. Both of these measures prevent parasitic Fresnel reflections from the glass surface that would result in multiple pulses oscillating in the amplifier cavity. The choice of
a 70 % reflectivity was initially chosen to prevent intra-cavity damages since early testing with HR mirrors had resulted in damages to the Nd:YVO₄ rod coating. Nevertheless, with the in-depth testing that has been performed, it is now proven that a higher reflectivity would be possible and result in faster ejection of the pulse and possibly higher output energy. The beam transmitted by the VBG is used for monitoring purposes.

6.2.5 Regenerative amplifier performances

The regenerative amplifier is seeded with the 2 nJ pulses produced by the fiber pre-amplifier. The output energy was measured as a function of pump power for repetition rates of 1 kHz, 2 kHz, 5 kHz and 10 kHz (Figure 84). Similarly the number of round trips required to extract the pulse with maximum energy has been recorded.

![Figure 84: Characteristics output energy versus input power (a) and corresponding number of round trips required to reach saturation at 1 kHz repetition rate (black squares), 2 kHz (red circles), 5 kHz (blue upward triangles) and 10 kHz (pink downward triangles).](image)

Energies up to 880 µJ have been extracted at 1 kHz for pump powers up to 90 W. The pump power was not further increased to prevent damages to the dielectric coating of the Pockels
cell or of the VBG. For energies beyond 500 µJ, the BBO Pockels cell, even though not phase matched for second harmonic generation, started to produce green light, witnessing the magnitude of the peak intensity. The experimental results of output energy show good agreement with the simulations performed in chapter 3. In the case of increased repetition rate, a slight discrepancy appears on the evaluation of the number of round trips, potentially attributed to the neglecting the spontaneous emission in the model.

One of the main constraints for the amplifier line is to provide clean pulses with large signal to ASE ratio. The role of the VBG to achieve this goal was highlighted in section 3.4.5. In order to practically verify the efficiency of the VBG, the regenerative amplifier was modified to reduce the beam size from 1.2 mm to 600 µm in the rod resulting in a large pumped volume of the rod being unseeded. The inversion produced in the unseeded volume turns either to heat or ASE, therefore reducing the size of the resonating seed in the rod increases the likelihood of producing ASE. This detuned cavity was operated either with the VBG as an end mirror or with an output coupler with a reflectivity $R = 40\%$. The cavity operated with the VBG shows a linear evolution of output energy with pump power. When the VBG is replaced by the output coupler, the output energy evolves linearly with pump power for pump powers up to 100 W. In the 100 to 140 W range, the output energy slightly decreases and is accompanied by instabilities in the time domain when the pulse is observed on an oscilloscope. This decrease in energy is attributed to competition for the gain between the ASE and the signal. The increased gain in the unseeded region of the rod for pump power beyond 140 W, coupled with the high-Q of the cavity allows for an increasing amount of ASE to be generated in the absence of VBG.
Figure 85: a. Characteristics output energy versus input pump power with an intra-cavity VBG (black squares) or a R = 40% output coupler (red circles) for a modified regenerative amplifier design enhancing the regeneration of ASE; b. output spectrum from the HERACLES regenerative amplifier operated with a VBG or a R = 40% output coupler (inset).

The spectral performance of the amplifier was also investigated by directly measuring the output spectrum with an optical spectrum analyzer (OSA). The spectrum measured when the VBG was inserted in the cavity shows a FWHM of ~40 pm in close agreement with the prediction made in 3.4.5. The signal to ASE ratio measured by the spectrum analyzer was as high as five order of magnitudes, limited by the dynamic range of the measuring instrument. The output spectrum was also measured when the R = 40% replaced the VBG in the original cavity. A spectral width of ~3.5 nm was recorded with a large pedestal attributed to ASE.

The effects of the VBG on the pulse duration have been investigated. The pulse durations at the input and at the output of the amplifier have been measured by intensity autocorrelation and are reported in Figure 86. The experimental results show again good agreement with the calculations presented in 3.4.5.
The cleanliness of the pulse was further investigated by frequency doubling the pulses produced by the regenerative amplifier and measuring the second harmonic generation efficiency. The beam was loosely focused to a measured diameter of 1.4 mm into a non-critically-phase-matched 2 cm long LiB₃O₅ (LBO) crystal heated up to 148°C. The loose focusing resulted in a limited peak power of 1 GW.cm⁻². Efficiencies up to 60% could be achieved and energies as high as 450 µJ in the green could be reached.

The final critical aspects of the regenerative amplifier relate to its ability to maintain the excellent beam profile of the seed pulses. As discussed in 3.4.4, the amplifier design, when operated as continuous wave or Q-switched laser allows slight excitation of higher order modes. Seeded with an excellent beam profile, it was experimentally proven that the regenerative amplifier operates only on the TEM₀₀ mode, producing a Gaussian profile output at any repetition rate and at pump powers up to ~80 W. For powers beyond 80 W the beam shapes become oval and side lobes starts appearing on the spatial profile. Implementing an intra-cavity aperture located at the VBG could address this issue. Nevertheless, the output energy produced
by the regenerative amplifier at 80 W was sufficient for seeding the following amplifiers and further increase of the pump power, beyond 90 W could result in catastrophic damage to optical components.

Figure 87: Beam profile at the output of the HERACLES regenerative amplifier at 1 kHz (upper left hand corner), 2 kHz (upper right hand corner), 5 kHz (lower left hand corner) and 10 kHz (lower right hand corner).

6.2.6 Post-amplifier implementation

The geometry chosen for the practical implementation of the post-amplifier is a single pass design featuring the same RBA module as the regenerative amplifier (see section 6.2.4). The slightly diverging beam provided by the regenerative amplifier is collimated and down sized to ~1.75 mm diameter to fill the amplifier rod.
A half wave plate is inserted in the beam path to adjust the beam polarization with the axis of the YVO₄ rod to maximize the gain. The ability to adjust the input polarization allows adjusting the output energy by reducing the effective gain. The collimated input beam is strongly affected by the thermal lens and a focused beam is produced at the output of the post amplifier. The beam is collimated by 17.5 cm focal length AR coated lens. Figure 88.b shows the layout of the single pass amplifier. The collimated beam is then spatially filtered to ensure a perfect beam profile for the following amplifier (Figure 88.a).
Figure 89: Characteristics output energy versus pump power at 1 kHz (a), 2 kHz (c) and 5 kHz (e) and output energy in the second harmonic as a function of amplifier pump power at 1 kHz (b), 2 kHz (d) and 5 kHz (f) for various IR seed energies.
The pointing stability and beam profile quality have been investigated. It is necessary that the beam quality to be excellent in order to achieve high efficiency in the optical parametric amplification process and an excellent pointing stability is required to prevent fluctuations at the output of the OPCPA generated by inconsistent overlap between the pump and signal beams.

Figure 90: a. Measurement of the pointing stability over 1000 shots on a CCD camera; b. beam profile in the far field measured by relay imaging the end of the amplifier rod.

The beam quality reported in Figure 90 was recorded by relay imaging the end of the Nd:YVO₄ rod at maximum pump power and see energy. A perfect output Gaussian output is provided. A few diffraction rings are visible due to slight overlap between the tail of the Gaussian and the edge of the rod. These ring can be suppressed by reducing the beam size but lead to a decrease in output energy. Spatial filtering can also achieve removal of the diffraction rings. Finally the, pointing stability was recorded over 1000 shots on a CCD camera ~3 m away from the output of the regenerative amplifier. A beam wandering of ~20 µm was recorded corresponding to a wandering angle of 6 µrad. The beam wandering represents ~5% of the beam diameter. Post-amplifier performance
The output energy of the single pass amplifier has been measured as a function of pump power and seed energy at 1 kHz, 2 kHz and 5 kHz repetition rate (Figure 89.a, c and e). In order to investigate the cleanliness of the pulse, the output has been frequency doubled in the same 2 cm, non-critically phase match LBO crystal used in 6.2.5 (Figure 89.b, d, e).

The YVO₄ single pass amplifier provides energy as high as 2.2 mJ at 1 kHz, limited by the damage threshold of the end facet of the YVO₄ rod. An increase in the repetition rate to 2 kHz and 5 kHz results in a decrease of maximum energy to 2.1 mJ and 1.8 mJ, mostly attributed to the slight decay of the seed energy. Up to 1.3 mJ, 800 µJ and 600 µJ of energy per pulse can be produced at respectively 1 kHz, 2 kHz and 5 kHz, making the output of the single pass amplifier a potential OPCPA pump. Energy conversion up to 70 % has been achieved confirming the excellent cleanliness of the pulses produced by the regenerative amplifier.

6.2.7 Booster amplifier implementation

Boosting the pump energy to the 10 mJ level requires increasing the diameter of the gain media to prevent catastrophic optical damage to the gain medium. Despite the numerous advantages of using Nd:YVO₄ as a gain medium, Nd:YAG was chosen for the booster amplifier. This choice was dictated by the unavailability of large diameter Nd:YVO₄ rods with satisfying quality at the time of the implementation of the booster amplifier. The use of Nd:YAG as a gain medium offers superior storage capability as compared to Nd:YVO₄ due to its higher saturation energy (see table 2). As a drawback, the small signal gain of Nd:YAG is smaller than that of Nd:YVO₄ and Nd:YAG being an isotropic crystal, depolarization effects are expected to occur. Additionally, an increase in rod diameter is typically accompanied by a reduction in small signal gain.
The geometry of the layout chosen for the booster amplifier takes into account the shortcomings of Nd:YAG, particularly that of the depolarization. Two identical heads equipped with 1 cm diameter, 12.5 cm long, 0.6 at. % dopant, radially pumped by up to 1 kW of continuous power provided by diode bar arrays constitute the gain medium of the booster amplifier (Northrop Grumman Cutting Edge Optronics – model REA). The saturation energy of each head is evaluated to \(~50\) mJ. The experimental arrangement of the heads is shown in Figure 91.

![Figure 91: Layout of the booster amplifier featuring two pump heads relay imaged onto each other for depolarization mitigation](image)

The end of each rod is relay imaged onto the other rod via a 1:1 telescope consisting of two AR-coated 10 cm focal length lenses placed between the heads. A half wave plate ensures rotation of the plane of polarization from one head to the next. Such arrangement is chosen to provide efficient depolarization compensation (see theory in section 3.6.3). Due to the limited gain available from each head, a polarization switched cavity built around the two heads was
investigated. Such a cavity allows multi-passing through the head to overcome the limited gain and resembles a regenerative amplifier.

6.2.8 Booster amplifier performances

The first issue addressed when assembling and testing the booster amplifier was to determine if the twin-head approach was satisfactory in terms of depolarization reduction as modeled in section 3.6.3. In order to investigate the efficiency of the depolarization compensation scheme, a linearly polarized, cw Nd:YAG laser providing ~100 mW output power at 1064 nm was setup to seed the two heads implemented either with the depolarization compensation scheme or without. The setup is shown in Figure 92.

![Figure 92: (top) Layout of the depolarization-compensation-free setup and typical beam profile retrieved at mid-range pump power – the beam profile was measured through parallel polarizers; (bottom) amplifier layout featuring the dispersion compensation scheme – the beam profile was measured through crossed-polarizers](image)

The measured spatial profiles of Figure 92 (on the same intensity scale) provide a qualitative measure of amount of depolarization for each case suggesting the validity of the depolarization compensation scheme. A quantitative measure of the depolarization was performed for both setups using the same seed laser. Upon propagation through the twin heads, the beam was propagated ~1.5 m away from the second head, passed through a polarizer and
through a filter designed to absorb the 808 nm pump light. The power available in each polarization was recorded for pump powers ranging from 300 to 650 W. It was chosen to place the power meter as far from the amplifier as possible to limit the amount of measured ASE. In order to make the measurement accurate, the cw seed was blocked for each measurement and the amount of remaining ASE subtracted from the measured values. The approximation that the seed is not modifying the amount of ASE is valid given the low amount of seed used for the experiment. The experimental results of this measurement are presented in Figure 93.a.

![Figure 93](image)

**Figure 93:** (a) Measured depolarization for a twin-head setup featuring a depolarization compensation scheme (black line) and without depolarization compensation scheme (red line); (b) measured small signal gain of a single head as a function of pump power.

Depolarization levels as high as 25% have been measured when the depolarization compensation was not put in place, in close agreement with the theoretical predictions (section 3.6.3). Upon implementation of the compensation scheme the depolarization could be reduced to ~10%. The small signal gain of a single head was also measured using the same seed laser. Small signal gain values up to 1.35 were measured for pump powers up to 650-700 W, proving that with proper depolarization compensation and seeding, net gain can be achieved from a twin-head setup.
The multi-pass cavity was then implemented and seeded by the output of the HERACLES post-amplifier. Stable operation of the amplifier proved to be challenging for at least two main reasons:

- the tested cavity (Figure 91) successfully controls depolarization effects but offers no control over the accumulated effects of thermal lensing. It was therefore found that this cavity could not be maintained stable for pump powers beyond ~500 W.
- the addition of the Pockels cell in the cavity led to increased losses which, added to the depolarization losses, resulted in a further reduced single pass gain. This drawback could be overcome by increasing the pump power or increasing the number of round trips in the cavity. Nevertheless both of these options result in an unstable cavity.

Overcoming this issue could be achieved by reducing the rod diameter and/or increasing the dopant concentration. An increase in dopant concentration would result in higher small signal gain and therefore relax the constraints on the pumping level while a reduction in diameter would also lead to an increase in small signal gain for similar pump power.

6.2.9 Alternative pump beam amplifier line design

Several promising alternative pathways have been reported that generate pulses with a few tens of picosecond duration and multi-mJ levels of energy at multi-kHz repetition rate:

- a system featuring a thin-disk regenerative amplifier that delivers picosecond pulses with energy up to 25 mJ at 3 kHz repetition rate was recently reported [149]. As compared to the one implemented on HERACLES this system exhibits a higher level
of complexity since the amplifier line relies on the CPA technique and only a few nanometers of bandwidth are available for stretching of the pulse. The system therefore requires cumbersome and alignment-challenging stretcher and compressor. Additionally the low single pass gain provided by the thin-disk results in a large number of passes required to reach saturation which yields accumulation of B-integral (attributed to the presence of the necessary intra-cavity Pockels cell). If a similar setup was to be implemented on HERACLES, it would be valuable to design a cavity featuring a sub-cavity around the thin-disk: in doing so, the gain-per-cavity-round-trip would be enhanced therefore limiting the number of passes through the Pockels cell. Similarly, the concept of using a VBG to control the pulse duration could be carried to a thin-disk regenerative amplifier and could prevent from using a stretcher/compressor assembly.

- another system relying on Yb:YAG technology has been reported and promises possibilities of reaching the 100 mJ mark at kHz repetition rate via cryo-cooling of solid-state Yb:YAG amplifiers. The current level of performance reported from this system is ~40 mJ of energy at 2 kHz repetition rate with 15 ps pulse duration [148]. This system nonetheless presents a series of inconveniences: first the cryogenic cooling brings additional complexity since the gain media needs to be maintained under vacuum. Second, if a recycling pump is being used to recycle the liquid nitrogen used to cool down the gain medium, vibrations from the pump will be transferred to the optical table and affect the CEP stabilization of the system. Finally, this system also operates as a CPA system and offers even less bandwidth (because of narrowing of the emission bandwidth at cryogenic temperature) than the previously
mentioned system which results in even more demanding stretching schemes. This scheme though promising for high energy at high repetition rate requires refinement of the engineering to be implemented on a system such as HERACLES.

- a last alternative approach that could be considered is the so-called INNOSLAB amplifier scheme. The architecture relies on multi-passing the seed pulses through a slab piece of gain material. The regimes of operation of INNOSLABS systems are not exactly matching that of HERACLES since either nanosecond or femtosecond pulses have been amplified. In the femtosecond regime up to 400 W of average power have been reported at 76 MHz repetition rate for pulses with duration as short as 680 fs [228]. Amplification was performed in Yb:YAG, therefore control of the spectral phase could enable the production of few tens of picosecond pulses. It is nonetheless unclear whether these systems can successfully maintain their average power when operated at lower repetition rate.

6.3 Grism stretcher and glass compressor performance

6.3.1 Implementation

The stretcher implemented on HERACLES features a grism pair and an AOPDF. The gratings used for the grism pair are 5x5 cm², gold coated 300 l/mm gratings (Richardson gratings), designed for maximum efficiency at 850 nm. At this wavelength a diffraction efficiency of ~80% is guaranteed by the manufacturer and the efficiency rolls over to ~70% for wavelengths beyond 950 nm. The prisms used for the grism pair are made of SF11 glass with a design apex angle of 18° (EROptics). The angle was experimentally checked to be 17.78°, the
slight deviation from the design being within the manufacturer tolerance. Nonetheless this 0.2° is worth noticing as it creates a significant deviation from expected performances if this offset is not considered in the model. The prisms and gratings are mounted together on a custom made mount and brought in direct contact. The whole assembly is then mounted on standard 1” tip-tilt mounts, therefore enabling maximum freedom of adjustment of the pair. Due to the relative weight of each grism pair, the overall height of the grism compressor is kept as low as possible to prevent adverse mechanical shifts that can contribute to slow drifting of the carrier-envelope phase. It was chosen to not anti-reflection coat the prisms upon manufacturing to enable testing of the assembly at a reduced cost. Figure 94.a shows one of the grism and Figure 94.b shows the compact layout of the stretcher.

![Figure 94: a. Picture of one of the grisms of the HERACLES stretcher; b. Picture of the compact layout of both the grism pair stretcher and AOPDF. This layout enabled stretching ratio of up to 10000.](image)

The experimental layout of the grism pair has tight tolerances as minor misalignments potentially lead to large drifts in expected performances. The ideal performances are obtained for an input angle of 7.5° with respect to the input phase of the grism, a shift between grisms of 3.5 cm measured from the apex of the bottom grism and a separation of 2.1 cm. The constraint on
this last figure is less stringent as the top grism is placed on a translation stage. An error in the input angle typically leads to inappropriate ratio TOD/GDD while an inexact grism shift can result in clipping of the high frequency part of the spectrum and improper amount of GDD. The optimal layout is shown in Figure 95.

![Figure 95: Optimal layout of the HERACLES grism pair](image)

The parallelism of the grism is also a critical aspect of the alignment but can readily be addressed by imaging the beam profile at the output of the grism pair: a vertical misalignment will translate into a vertically elongated spatially chirped beam. Similarly, a horizontal misalignment will translate into a horizontally elongated beam. Corrections of the parallelism are performed by adjusting the tip and tilt knobs of the top grism and the retroreflector mirror.

After propagating through the grism pair, the beam is sent into the AOPDF. The alignment of the AOPDF is performed by simply retroreflecting the front surface Fresnel reflection. Optimization of the alignment is performed by observing the pulse train on an oscilloscope via a fast photodiode at the output of the Dazzler and maximizing the diffraction efficiency. A typical diffraction efficiency of ~15% is expected for a loaded GDD of 3500 fs² over a 400 nm spectral width. The width of the diffracted window is typically 40 to 80 µs.
The HERACLES compressor consists of a simple $7\times7\times5$ cm$^3$ piece of SF57 (Schott). The two parallel $7\times7$ cm$^2$ surfaces are polished to optical quality and are expected to receive an AR-coating covering the 700 to 1100 nm spectral range. The beam passes through the glass piece three times at quasi-normal incidence making the total optical path in the glass $\sim15$ cm. The glass piece is shown in Figure 96.

![Figure 96: SF57 glass compressor of HERACLES](image)

### 6.3.2 Measured performance

The required performance of the grism stretcher are twofold: first, the assembly must transmit the entire seeded spectrum as efficiently as possible. Second the dispersion provided by the grism pair should closely match that of the glass compressor. Tests have been performed to address both these issues.

The throughput of the grism pair was investigated by building a pair with 7 cm separation, close to the maximum separation – at normal incidence – enabled by the size of the
gratings. The measured spectra before and after the stretcher are shown on a logarithmic scale in Figure 97.

A reduction in spectral intensity is observed for wavelengths beyond 1000 nm and is attributed to the reduced reflectivity of the gratings. This part of the spectrum does not fall within the gain bandwidth of the optical parametric amplifier it is therefore a non-critical loss. The current overall throughput of the grism pair assembly is ~15%. It is expected that the throughput will increase beyond 40% when an anti-reflection coating will be applied to the prisms. The amount of losses in the stretcher must be minimized but are not critical due to the high gain of the optical parametric amplifier stages.

The main test of the performance of the stretcher is its ability to stretch the pulse to match the glass compressor and therefore ensure recompression of the pulses after amplification. The pulse was sent through the stretcher featuring the specifications given in the previous section and the AOPDF loaded with 1500 fs² of GDD, 1000 fs³ of TOD and -500 fs⁴ of FOD. It was then amplified in two OPA stages to ~0.8 μJ and compressed by passing through the glass
compressor. The output performance of the stretcher/compressor assembly is shown in Figure 98.

![Figure 98: a. Measured intensity autocorrelation trace of the output of HERACLES and b; corresponding spectrum](image)

Only a limited portion of the available spectrum was used to achieve the shortest pulse as can be seen from Figure 98.b. The spectral width was purposely limited by blocking the section of the spectrum below ~770 nm in an attempt to shorten the pulse via optimization of the time bandwidth product rather than by increasing the spectral width. The part of the spectrum below 770 nm is attributed to superfluorescence and did not contribute to the measurement of the pulse duration (when the seed pulses were blocked, no signal was measured from the autocorrelator). An 18 fs pulse duration (6.5 cycles) was measured with an estimated spectral width of 60 nm corresponding to a time bandwidth product of 0.470 confirming the ability of the stretcher/compressor assembly to handle few-cycle pulses. This experiment also shows the importance of maintaining the spectral shape in achieving the shortest pulses, a feature that has not been investigated by other reported work. Pre-shaping of the spectral intensity to achieve a close-to-Gaussian amplified spectrum can be performed in the AOPDF by controlling the...
diffraction efficiency for each wavelength and can be a simple way to reduce the stress placed on both the OPAs and the AOPDF and ensure greater robustness in achieving few-cycle pulses.

6.4 Optical parametric amplifier performance

6.4.1 Implementation

The implementation of the optical parametric amplifier of HERACLES follows the conclusion of chapter 3. The amplification is performed in three cascaded stages consisting each of a 5 mm thick BBO crystal with a clear aperture of 1 cm² (Quantum technology). The phase matching angle was set to 23.8° with a ±0.1° manufacturing tolerance. Uncoated BBO crystals were preferred to perform initial tests in order to reduce the costs in case of damages or errors in the model. It was decided to alternate stretching and amplification stages in order to optimize the output performances. The layout of the optical parametric amplifier is shown in Figure 99.

![Figure 99: Layout of the HERACLES parametric amplifier and stretcher stages](image)

The energy available to seed the parametric amplifier is ~0.5 nJ since a fraction of the total energy is used to achieve CEP stabilization and another fraction is used to seed the pump
beam amplifier line. It was found, as discussed in section 6.1 that the beam profile was rather poor in the far field and suffered from spatial chirp which prevented broadband amplification in the optical parametric amplifier. A spatial filter was therefore implemented directly at the output of the oscillator and allowed the generation of Gaussian beam at the expense of a 40% loss in available energy. It was nevertheless found that the spatial filter contributed to reducing the spatial chirp. Figure 100 shows the spatial profile at different positions before amplification.

![Figure 100: a. Measured spatial profile at the output of the Octavius oscillator; b. measured spatial profile at the output of the spatial filter; c. Measured output profile at the output of the grating stretcher.](image)

After being spatially cleaned the pulses are sent into the grating stretcher to be temporally spread to ~15 ps duration. They are then directed into the first OPA pre-amplifier and amplified to the microJoule level of energy. The beam is spatially cleaned again to suppress the superfluorescence generated in the first OPA stage. Fluorescence is likely to be generated due to the low amount of seed energy. The beam is then passed through the AOPDF (with 10% to 20% efficiency depending on the spectral phase required) and the grism pair (~15% throughput) where the pulse duration is further stretched to duration comprised between 30 and 45 ps. These pulse durations enable maximum energy extraction while preventing excess spectral narrowing during the amplification process (see section 5.4.2.3). The pulses are then sent into the two power amplifiers. Currently, the system is set up with the AOPDF and grism stretcher placed
after the second OPA stage since all studies have required as much output power as possible and since the booster amplifier is still under development.

### 6.4.2 Optical parametric amplifier performances

The performance of the parametric amplifier have been investigated on the first OPA stage under the assumption that if the performances of the first OPA match the model described in section 5.4, the following stages could be predicted accurately and the output performances would match the design requirements. The first aspect that was investigated was ability for the amplifier to amplify spectra wide enough to withstand pulse with sub-10 fs duration. Figure 101 shows a typical spectrum obtained when pumping the first OPA stage with ~400 µJ energy with a beam diameter of ~100 µm for both the signal and pump. The signal energy is typically ~100 pJ.

![Amplified spectrum and Seed spectrum](Image)

**Figure 101:** Measured seeded (black line) and amplified (blue line) spectra at the input and output of the first ultra-broadband OPA

Amplification is obtained from ~725 to 975 nm closely agreeing with the model. Furthermore, the spike in the seed spectrum at 900 nm is much more amplified than the spike at
800 nm suggesting that the gain is higher at longer wavelength than shorter ones as predicted by the model. The FWHM of the amplified spectrum, though challenging to define due to the shape of the spectrum, can conservatively be estimated to exceed 150 nm of bandwidth which corresponds to an ideal Gaussian transform-limited (time-bandwidth product (TBP) $\Delta \nu \tau = 0.441$) pulse of 6.6 fs. Assuming a pessimistic TBP of 0.6, results in a compressed 9 fs pulse.

The effects of angular tuning have also been investigated. It was found in theory and experimentally confirmed that ultra-broadband phase matching can only be achieved for very well defined angles. The parametric amplifier was aligned and slightly detuned away from normal incidence. The angular detuning was evaluated by measuring the deviation of the reflected pump beam from the uncoated surface of the BBO crystal.
Figure 102: Measured normalized amplified spectra (blue line) and seeded spectrum (grey shaded region) for an angular detuning of -0.25° (a), -0.16° (b), +0.25° (c) and +0.16° (d).

In this experiment, the sign of the angle $\alpha$ is simply used to symbolize that the pump beam was retro-reflected either to the right or to the left of the incoming beam but is not related to any other physical measurements. In the case of $\alpha$ negative, the gain bandwidth shifts toward the IR. The lack of seed in that spectral region simply results in a reduction of the gain for wavelengths 850-900 nm region. For $\alpha$ positive, the gain bandwidth clearly shifts toward visible wavelengths but is also accompanied by a reduction in the magnitude of the gain. Both these behaviours were predicted by the code as a consequence of an increased acquired $\Delta k z$.

After investigating the spectral properties of the parametric amplification process, the gain properties were investigated as a function of temporal overlap and pump energy (Figure 103). In order to ensure optimum temporal overlap between the pump and signal pulses in the OPA, the pump beam is directed toward an adjustable delay line before pumping the OPA. This delay line was adjusted and the resulting gain on the signal beam measured for several delays. The reference delay is taken as the delay enabling the highest parametric gain. In all case, the seed energy was $\sim$100 pJ, the seed pulse duration was $\sim$15 ps and the signal beam diameter in the
OPA crystal \( \sim 100 \, \mu \text{m} \). The pump pulse duration was 85 ps and the pump beam diameter was matching that of the signal. Measurements were performed at 1 kHz repetition rate. For this experiment the pump energy was limited to \( \sim 420 \, \mu \text{J} \) to ensure the absence of superfluorescence.

The measured behaviour of the parametric amplifier as the delay between pulses is being scanned meets theoretical expectations. As the overlap between the two pulses and the signal pulse moves toward the FWHM of the pump pulse, the gain increases exponentially. In a delay window of \( \sim 10 \, \text{ps} \), located around the zero delay point, the relative delay is of little importance since the signal pulse remains within the FWHM of the pump pulses (notice that this is achieved because of the favourable pulse duration ratio). Passed the maximum overlap point, the gain decreases exponentially as the signal pulse is moved away from the FWHM of the pump pulse. No superfluorescence could be detected for any delay.

Finally, the influence of pump energy on the parametric gain was investigated. For that purpose, a light valve composed of a half-wave plate and polarizer beam splitter was inserted.

![Figure 103: Measured parametric gain versus delay between pump and signal pulse.](image)
into the beam path of the pump beam allowing control over the pump beam energy. Results are gathered in Figure 104.

![Graph showing parametric gain versus pump energy for a constant seed energy level.](image)

Figure 104: Measured parametric gain versus pump energy for a constant seed energy level

The experimental behaviour matches the theoretically expected one confirming the proper design of the OPA stages: an exponential gain is obtained as the pump power is linearly increased. OPA gains up to 35000 could be obtained (at that point, the onset of superfluorescence started appearing), yielding an estimated energy from the first OPA stage of 3.3 μJ with no depletion of the pump. The pump was recycled to pump the second OPA stage and led to a further gain of ~60 yielding an output energy of ~200 μJ currently at up to 2 kHz repetition rate. Further improvement can be achieved by proper design of the relay imaging performed between the first and second OPA stage.

6.5 Safety features and environment control

A large system as HERACLES featuring high average optical powers, high repetition rates, high pump powers, high DC driving currents, etc requires careful monitoring, interlocking
and containment. A large amount of time was devoted to ensure maximum safety to both the user and the components of the system.

The pump generation line relies exclusively on diode pumping. It is well-known that a major cause of failure of laser diodes is operation without cooling. To address that issue, interlocking between the power supplies providing the DC current to the diodes and the chillers cooling each stage have been put in place. For the fiber-amplifier, where thermo-electric cooling is used an internal interlock already was already put in place by the manufacturer of the power supply. To further broaden the safety of the diodes, all chillers are plugged to a computer interface that allows monitoring at all time of pressure, water flow and temperature for each cooling line. Sound alarms have been implemented in case of failure of one of the chillers.

The master-clock that controls all triggers in the system has been re-programmed to enable higher safety to the equipment. In case of failure of the Ti:Sapphire oscillator to provide the 85 MHz clock signal, all triggers are being shut down, preventing amplification of random noise in the amplifier line that could lead to damages to components. Furthermore monitoring of the key stages of the facility is either hard-wired in the system or readily available to be achieved within minutes to track potential failure of a stage or drifts in performance.

Control of the environment has also been put into place. In order to ensure optimum operation of HERACLES it is necessary to isolate the system from dust and air-flow. The air-conditioning of the laboratory typically generates a large amount of dust that directly falls onto the system. To address this issue, several encapsulations have been built. The most sensitive component of the system, the Ti:Sapphire oscillator is enclosed in two boxes. The positive effect of these boxes on, for example, CEP stabilization have been clearly observed. In order to isolate
the rest of the system from the environment, the entire 1.5x3 m² table has been encapsulated in a modular enclosure that enables access to each part of the system separately.

Finally, the layout of the laboratory is such that entrance is limited to authorized personnel only. A software was developed to inform visitors of the wavelength being used and the power levels involved before entering the laboratory. An antechamber to the laboratory was devised where appropriate laser goggles are made available.
The future of ultra-short laser science is bright both on the laser and application side. Systems enabling unprecedented energy levels with unprecedented level of control on the electric-field are currently available only in a few laboratories worldwide but are in the process of becoming widely available and will enable exciting new fundamental physics. The availability of carrier-envelope phase controlled systems with energy exceeding the millijoule at the kHz repetition rate can enable, if coupled to a high harmonic generation facility, research at the atomic space-scale and in the attosecond timescale.

This thesis is focused on the modeling and building of a turnkey facility (HERACLES) that delivers sub-10 fs pulses with millijoules of energy at repetition rates up to 10 kHz. The facility is currently awaiting the completion of its final booster amplifier. It therefore currently provides pulses with \( \sim 1 \mu \text{J} \) of energy before compression (\( \sim 400 \text{ nJ} \) after compression). All key components have been proven to work consistently from day-to-day and for up to 12 hours at a time and interact with each other properly. This thesis covers a number of areas of laser engineering and has enabled some contribution to the current state of knowledge in the laser development field.

A hybrid amplifier line has been modeled and built that amplifies pulses with sub-picojoule energy up beyond 10 mJ while providing excellent beam profile, control spectral content and controlled pulse duration in the few tens of picosecond duration. This amplifier line was one of the first merging fiber and solid-state technology to take advantage of both architectures. The novelties included in this amplifier line also include the design and successful building of a regenerative amplifier featuring both a VBG and a side-pumped Nd:YVO\(_4\) crystal.
The behavior of this amplifier has been thoroughly investigated in a wide range of operation. The positive effects of the insertion of the VBG and the advantages of the side-pumping geometry have been proved by the reduced ASE, the perfectly tailored and transform-limited pulse emitted from the amplifier and the outstanding beam quality and stability. Behaviours predicted by other research group such as chaotic amplification at high repetition rate have also been observed and modeled.

The stretcher implemented on HERACLES is also featuring a few novelties that have been developed in the frame of this thesis. First, a complete semi-analytical model has been developed to predict the behavior of a grism pair able to handle spectra wider than 300 nm. Such model had not been reported at the time when it was being developed at LPL. The accuracy of the code developed to model such stretcher has been validated experimentally in the frame of this thesis.

The last major contribution of this work to the laser development community is the development of a 3D simple code for modeling the temporal, spectral and spatial properties of optical parametric amplifiers. The code can run on a simple laptop and provide accurate values for output energy, output spatial and temporal profile, spectral distortions due to local pump depletion. The results predicted by the program have successfully been confronted to experimental results. The code has also been tested to investigate two-color pumping and has proved to be modular enough to predict in theory the results obtained experimentally by other groups in such geometry. Further refinement to the code could be performed to predict more accurately saturation effects. These refinements would need to be performed at the expense of the time of execution of the code and would require the use of a computer cluster.
The contribution of this thesis is not limited to the research area but has also enabled the development of a few commercial system in via fruitful collaborations. The Octavius Ti:Sapphire oscillator was partially re-engineered in the frame of this work to take it from an unstable system unable to operate for time periods greater than two hours to a rugged, turnkey system that has been operating for the past two and half year for ten to twelve hours a day without major alignment or modification to it. This collaboration with IdestaQE is now being extended to further stabilize the Octavius in particular its carrier-envelope phase. Another major constructive collaboration put in place during the building of HERACLES is linked to development of the Nd:YVO₄ modules used in the regenerative amplifier and the single-pass post-amplifier. These devices were not yet commercialized when the first one was sent to LPL and extensive testing has been performed in continuous, Q-switch and regenerative amplifier configurations. Our work has contributed to pinpoint the poor quality of the polishing of the Nd:YVO₄ rods that led to damages at peak powers well below typical damage thresholds levels. Finally, some existing relationships between LPL and Fastlite (on the AOPDF) and Quantum Technologies (on the Pockels cells and nonlinear crystals used measurement devices and OPAs) have been strengthened.

The availability of HERACLES as a quasi-hands-off facility will broaden the scope of the research on-going in the LPL. Current research on plasma studies and filaments will benefit from a few-cycle system providing millijoule level of energy. The material processing team of LPL will also benefit from a state-of-the-art tool that will enable research in regimes that have not been tackled so far. Finally, HERACLES will enable the beginning of completely new research program on high-harmonic generation and attosecond pulse generation, the application of which appear to be endless.
APPENDIX – PRINCIPLE OF CEP STABILIZATION
In order to achieve CEP stabilization, it is necessary to measure the frequency at which the carrier sweeps under the envelope. For this it is necessary to represent the spectrum of the oscillator as a frequency comb; each wavelengths contained in the spectrum is represented as a longitudinal mode separated by a fix frequency offset \( \Delta \nu = c/2L \), where \( L \) is the laser cavity length and \( c \) the speed of light, from its neighboring modes. The spectrum can then be represented as [209]:

![Figure 105: Representation of the spectrum as a frequency comb and highlighting the role of \( f_{CEO} \)](image)

The spacing between longitudinal modes is assumed to be a constant. Nevertheless, there is no guarantee on the exactness of the center frequency of the laser output. This offset of the frequency comb from the zero-frequency is called \( f_{CEO} \). In the time domain it exactly translates into a shift of the carrier with respect to the envelope. \( f_{CEO} \) is measured in a \( f-2f \) interferometer as described in section 6.1.4. A simple mathematical derivation of the functioning of this type of interferometer can be obtained by writing the fundamental frequency, the IR frequency and the frequency double IR frequency in terms of longitudinal modes using the notation of Figure 105:

\[
\omega_{\text{fund}} = f_{CEO} + 2mf_{\text{rep}} \tag{136}
\]

\[
\omega_{\text{IR}} = f_{CEO} + mf_{\text{rep}} \tag{137}
\]
The beating of two sinusoidal signals at frequency $\omega_1$ and $\omega_2$ is given in the general case by:

\[
\sin(\omega_1 t) + \sin(\omega_2 t) = 2 \cos\left(\frac{\omega_1 - \omega_2}{2} t\right) \sin\left(\frac{\omega_1 + \omega_2}{2} t\right)
\]  

(139)

The term of interest in the case of CEP-stabilization is the difference frequency term. Indeed, when beating the $\omega_{fund}$ and $\omega_{SHG}$ signal, the resulting signal has the frequency $f_{CEO}$:

\[
\omega_{SHG} - \omega_{fund} = 2\left(f_{CEO} + mf_{rep}\right) - f_{CEO} + 2mf_{rep} = f_{CEO}
\]  

(140)

The measured value of $f_{CEO}$ is then sent into a feedback loop that controls parameters of the oscillator to ensure stability of the $f_{CEO}$ value, guaranteeing the locking of the carrier with respect to the envelope.
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