

Analysis and Optimization of Ultrafast Laser-Induced Bulk Modifications in Dielectric Materials

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Abstract

In the recent years ultrafast laser technology has become a powerful tool for three-dimensional structuring of optical materials with the purpose of adding to the material functionalities in optics and photonics.

By focusing an ultrashort pulse of light in the volume of a transparent dielectric, free carrier generation takes place via nonlinear ionization. A substantial part of the laser energy is deposited into the free carrier gas, transferred into the bulk, and a material with new optical properties emerges upon energy relaxation.

Phase contrast (PCM) and optical transmission microscopy (OTM) techniques are employed to characterize the morphology of the laser-induced refractive index change in the bulk of amorphous silica (a-SiO₂) and BK7. The PCM observations are correlated with a theoretical estimation of the energy deposited in the vicinity of the focal plane based on the resolution of the nonlinear Schrödinger equation. In a-SiO₂, the formation of a void is connected with the appearance of a high energy exposure upon nonlinear propagation.

A time-resolved study of the laser-generated refractive index modifications with sub-picosecond and sub-micrometer resolution is performed. PCM and OTM images are recorded at different times after excitation, covering a time span from approximately 100 fs up to 10 ns. This analysis points out the importance of the thermal mechanisms and of the subsequent thermomechanical transformations in laser modification of bulk dielectrics.

By using an adaptive pulse shaping apparatus, we demonstrate that an optimal command of the energy deposition into the volume of the material can be obtained, even in presence of spherical aberrations. Furthermore, optical structures that do not normally appear in standard ultrafast irradiation conditions can be generated. In particular, we report the onset of large positive refractive index regions in BK7.

Finally, the flexibility offered by temporal pulse manipulation is exploited for microprocessing purposes. We demonstrate writing of embedded waveguiding structures at optical frequencies in the bulk of BK7.

Zusammenfassung

In den letzten Jahren hat sich die Technologie der ultrakurzen Laserpulse zu einem leistungsstarken Werkzeug entwickelt, das eine dreidimensionale Bearbeitung ermöglicht und es zudem gestattet, Materialien im Bereich der Optik und Photonik gezielt zu modifizieren und zu funktionalisieren.

Bei der Fokussierung eines ultrakurzen Laserpulses in das Volumen eines transparenten Mediums können aufgrund der hohen verfügbaren Intensitäten im Material Elektronen durch nichtlineare Wechselwirkungen in dessen Leitungsband angeregt werden. Ein signifikanter Anteil der verfügbaren Laserpuls-Energie kann zusätzlich durch diese bereits erzeugten quasi-freien Ladungsträger absorbiert werden ("free-carrier absorption"). Infolge des darauffolgenden Transfers der Energie des angeregten Elektronensystems auf das Gitter des Festkörpers kann somit ein Material mit neuen optischen Eigenschaften entstehen.

Zur Visualisierung wurden die Techniken der optischen Phasenkontrast- (PKM) und Transmissions-Mikroskopie (OTM) eingesetzt. Dadurch können laser-induzierte permanente Veränderungen der optischen Eigenschaften im Volumen von synthetischem Quarzglas ($a\text{-SiO}_2$) und dem technologisch relevanten Multikomponentenglas BK7 erfasst werden. Die PKM Ergebnisse werden mit den Resultaten eines auf der numerischen Lösung der nichtlinearen Schrödingergleichung basierenden theoretischen Modells korreliert, welches es erlaubt, die räumliche Verteilung der deponierten Laserpuls-Energie in der Nähe des Fokus zu berechnen. In $a\text{-SiO}_2$ kann mit Hilfe dieses Modells aufgezeigt werden, dass für die Bildung von laser-induzierten Hohlräumen ("voids") nichtlineare Propagationseffekte maßgebend werden.

Darüber hinaus wurden im Rahmen dieser Arbeit zeitaufgelöste Techniken der PKM und OTM entwickelt und charakterisiert, welche es gestatten, die Dynamik derartiger laser-induzierter Änderungen der optischen Eigenschaften (komplexer Brechungsindex) im Volumen von transparenten Materialien mit sub- μm räumlicher und sub-ps zeitlicher Auflösung bis in den ns-Bereich zu verfolgen. Diese Untersuchungen unterstreichen die Bedeutung von thermischen Mechanismen und den darauffolgenden thermomechanischen Prozessen bei der Laser-Modifikation im Volumen von Dielektrika.

Durch Verwendung von adaptiven Techniken der zeitlichen Pulsformung wird gezeigt, dass sogar in Anwesenheit von sphärischen Aberrationen eine optimale zeitliche Sequenz der Deponierung optischer Energie im Materialvolumen gefunden werden kann. Diese Sequenz gestattet es, Materialveränderungen zu realisieren, die sich unter den herkömmlichen Bestrahlungsbedingungen nicht erzeugen lassen. Zum Beispiel können in BK7 auf diese Weise stark positive Brechzahländerungen erzeugt werden, im Gegensatz zu den üblicherweise beobachteten negativen Brechzahländerungen.

Zusätzlich wird durch die zeitliche Pulsmanipulation die Flexibilität und Effizienz für Mikrobearbeitungszwecke erhöht. Damit wird erstmalig das Schreiben von eingebetteten Wellenleiterstrukturen für den optischen Frequenzbereich in BK7 demonstriert.

Résumé

Au cours des dernières années, les lasers à impulsions ultrabrèves se sont imposés comme des outils incontournables pour la micro structuration tri dimensionnelle et la fonctionalisation de matériaux pour des applications dans le domaine de l'optique et de la photonique.

En focalisant une impulsion lumineuse ultra brève dans la masse d'un matériau diélectrique transparent, un mécanisme d'ionisation non-linéaire peut conduire à la création de porteurs libres. L'énergie lumineuse peut alors être efficacement déposée. Après relaxation de l'énergie, un matériau avec de nouvelles propriétés optiques peut être obtenu.

Afin de caractériser les propriétés optiques de ce matériau transformé ainsi que la morphologie de la zone altérée, nous utilisons la microscopie à contraste de phase ainsi que la microscopie optique classique. Les échantillons étudiés sont principalement deux types de verres, la silice pure et le N-BK7.

Nous proposons de corrélérer les observations réalisées en microscopie à contraste de phase avec une estimation théorique de la densité d'énergie déposée obtenue en résolvant l'équation de Schrödinger non-linéaire. Dans la silice pure, l'apparition d'une micro cavité est ainsi associée à une région de forte exposition à l'énergie lumineuse.

Une étude basée sur un dispositif de microscopie de phase et de microscopie classique caractérisé par une résolution spatiale submicrométrique et une résolution temporelle subpicoseconde est également présentée. Une échelle de temps de la centaine de femtoseconde après excitation jusqu'à une dizaine de nanosecondes est ainsi explorée. Cette analyse révèle l'importance des phénomènes thermiques et des effets thermomécaniques.

En utilisant un dispositif d'optimisation de la forme temporelle de l'impulsion, nous démontrons la possibilité de contrôler le dépôt d'énergie dans la masse du matériau, y compris en présence d'aberrations sphériques. Il est ainsi possible de conduire le matériau de manière permanente dans des états inaccessibles lorsqu'on se limite à une irradiation ultra brève classique. En particulier, nous montrons l'existence de régions de densités élevées dans le BK7 après irradiation.

Enfin, la souplesse offerte par la mise en forme temporelle est employée afin de réaliser l'écriture de guides d'ondes enterrés dans le BK7.

Contents

1	Introduction	1
2	Light as a material structuring tool	4
2.1	Mathematical description of the laser pulse	5
2.2	Nonlinear propagation	6
2.2.1	Origin of the nonlinear refractive index	6
2.2.2	Self-focusing	7
2.2.3	Self-phase modulation	8
2.3	Nonlinear ionization and consequences on the transient optical properties . .	9
2.3.1	Nonlinear ionization mechanisms	9
2.3.2	Optical properties of an excited solid	12
2.3.3	Plasma defocusing	14
2.4	Pulse propagation and energy deposition	14
2.4.1	Evolution of the pulse envelope	14
2.4.2	Continuity equation	15
2.5	Energy relaxation and consequences on the refractive index	16
2.5.1	Introduction	16
2.5.2	Point defects	17
2.5.3	Matrix re-organization	19
3	Experimental apparatus and analysis tools	21
3.1	Output characteristics of lasers and pulse shaping apparatus	21
3.1.1	Radiation sources	21

3.1.2	Pulse shaping apparatus	23
3.1.3	Pulse characterization techniques	26
3.2	Beam manipulation for strong focusing in transparent materials	28
3.2.1	Focusing of Gaussian beams	29
3.2.2	Beam focusing in bulk materials: influence of spherical aberrations	31
3.3	Fundamentals of phase contrast microscopy	35
3.3.1	Overview of refractive index change detection methods	35
3.3.2	Qualitative study of the phase contrast technique	37
3.3.3	Practical realization	40
3.4	Experimental setup	41
3.4.1	Pulse picking	41
3.4.2	Beam focusing	42
3.4.3	Samples and sample displacement	42
3.4.4	Phase contrast apparatus	43
4	Direct observation of laser induced phase objects	44
4.1	Single pulse irradiation at various laser-pulse energies	45
4.1.1	Energy dependence study in amorphous fused silica	45
4.1.2	Energy dependence study in BK7	50
4.1.3	Influence of spherical aberrations	52
4.2	Multipulse irradiation	55
4.2.1	Irradiation at low repetition rates in a-SiO ₂ and BK7	55
4.2.2	Irradiation at high repetition rates (1kHz and 100 kHz) in a-SiO ₂ and BK7	56
4.2.3	Conclusion and perspectives	57
4.3	Irradiation with chirped pulses	58
4.3.1	Threshold dependence versus chirp	58
4.3.2	Energy deposition in the picosecond regime	59
4.4	Conclusion	61
5	Time-resolved imaging of the energy relaxation	63
5.1	Introduction	63
5.1.1	Ultrafast pulses for ultrafast images	63
5.1.2	State of the art	64
5.2	Experimental realization	66
5.2.1	Pump-probe setup	66
5.2.2	Choice of a criterion for zero delay	67

5.2.3	Temporal resolution of the imaging system	68
5.3	Time-resolved phase contrast pictures at high input energies	70
5.3.1	Experimental conditions and selected results	71
5.3.2	Discussion	72
5.3.3	Conclusion	76
5.4	Time-resolved phase contrast pictures at low input energies	76
5.4.1	Experimental conditions and selected results	77
5.4.2	Discussion	78
5.4.3	Conclusion	84
5.5	Free carrier lifetime in BK7 and IOG-10	85
5.5.1	Experimental conditions and presentation of the results	85
5.5.2	Quantitative estimate of the free carrier lifetime in BK7 and IOG-10	86
5.6	Conclusion	88
6	Optimal control for optical structures	89
6.1	In search of the optimal structuring tool	89
6.1.1	Short introduction to evolutionary algorithms	90
6.1.2	Building blocks of evolution strategies	91
6.2	Implementation of the adaptive pulse shaping strategy	95
6.2.1	Encoding excitation sequences into virtual individuals	95
6.2.2	Definition of a valuable solution	96
6.2.3	Experimental constraints on the maximum optimization time	98
6.3	Configuration of the algorithm	99
6.4	Optimization results in a-SiO ₂	100
6.4.1	High refractive index structures in a weak aberrations regime	100
6.4.2	High refractive index structures in a-SiO ₂ in presence of aberrations	101
6.4.3	Discussion	105
6.5	Optimization results in BK7	107
6.5.1	Preliminary experiments in single-shot regime	107
6.5.2	Optimization on high a repetition rate system	109
6.6	Conclusion	113
7	Waveguide writing in optimal conditions	115
7.1	Introduction	115
7.2	Waveguide laser-writing in longitudinal configuration	116
7.2.1	Experimental configuration	117
7.2.2	Experimental conditions and phase contrast microscopy analysis	118

7.2.3	Analysis of the guiding properties in far field	120
7.2.4	Conclusion	121
7.3	Waveguide laser-writing in transverse configuration	121
7.3.1	Experimental configuration	122
7.3.2	Phase contrast microscopy observations	122
7.3.3	Analysis of the guiding properties in far field	125
7.3.4	Conclusion	125
7.4	Perspectives toward low-loss waveguiding in BK7	126
7.4.1	Results of preliminary investigations	126
7.4.2	Discussion	127
7.4.3	Future work	127
8	Conclusion	128
A	Quantitative estimate of the free carrier lifetime from time-resolved microscopy images	130

List of Figures

2.1	Evolution of the real part of the refractive index (n^*) and of the extinction coefficient (κ^*) of a free electron gas in vacuum versus the density of free carriers, for an incident monochromatic wave with a wavelength of 400 nm. In order to illustrate the influence of the collision parameter, computations were carried out for $t_D = 0.2$ fs and $t_D = 1$ ns.	13
2.2	Principal points defects observed in fused silica [1, 2]. Large and small circles correspond to silicon and oxygen atoms, respectively.	18
2.3	Example of a m-membered ring in fused silica with $m = 3$. Taken from [3]	19
3.1	Structure of a zero dispersion stretcher unit.	23
3.2	Pulse shaping apparatus based on Fourier synthesis of spectral components.	25
3.3	Auto/cross-correlation apparatus	26
3.4	Overview of the building blocks for system I and system II	28
3.5	Figures of merit of a focusing system. The numerical aperture is simply defined as $NA = n \sin \Omega$. The working distance (WD), the radius of the back aperture (r) and the optical axis (O.A.) are also indicated.	29
3.6	Experimental layout emphasizing the effect of spherical aberrations, adapted from [4]. The position of the paraxial focal plane with respect to the air-glass interface (D), the focal plane in absence of interface (F) and the longitudinal extent of the focal volume (L.E.) as well as the location of the optical axis (O.A.) are indicated.	32

3.7	On axis intensity distribution generated by spherical wavefront distortion for $F = 0$ and $F = 1$ mm. F and D correspond to the position of the focal plane in absence and in presence of a air-glass interface, respectively (see Fig. 3.6).	34
3.8	Intensity peak on axis when varying the position of the paraxial focus (D) in presence of an air-glass interface.	35
3.9	Principles of optical phase contrast microscopy. Adapted from [5]. Sketch of the object to study. The surround (S) wave and a wave emerging from the object (P) are schematically represented.	37
3.10	Representative curves of the electric fields of the surrounding (S), particle (P) and diffracted (D) waves.	38
3.11	Artificial shift of the diffracted wave by $+\frac{\pi}{2}$ (left) or $-\frac{\pi}{2}$ (right).	39
3.12	Phase contrast microscope optical train, taken from [6]	40
4.1	Phase contrast microscopy (PCM) and optical transmission microscopy (OTM) observations of laser-induced traces by a single pulse in amorphous fused silica for various laser beam energies. On the right column, laser pulse energies in μJ are specified. The dot line represents the estimated focal plane.	46
4.2	Phase-contrast microscopy (PCM) and optical transmission microscopy (OTM) observation of a single-pulse laser modification trace in fused silica for an input energy of $1 \mu\text{J}$. The dot line represents the estimated focal plane. A black region is observed around the focal area and a white dot is formed on the tail of the structure.	46
4.3	Simulation of energy deposition into in fused silica at the end of the 120 fs irradiation pulse. (a) incident fluence (J/cm^2); (b) absorbed energy (J/cm^3); (c) peak intensity distribution (W/cm^3); (d) electronic density (cm^{-3}) 50 fs after the pulse center.	47
4.4	Phase-contrast observation of single-pulse laser modification trace in silica in the strong regime of interaction and comparison with optical transmission microscopy analysis (left side). The dotted line represents the estimated focal plane.	49
4.5	Phase contrast microscopy (PCM) and optical transmission microscopy (OTM) observations of laser-induced traces by a single pulse in BK7 for various laser beam energies. On the right column, laser pulse energies in μJ are specified. The dot line represents the estimated focal plane.	51

4.6	Aspect of fs laser-induced traces in BK7 in optical transmission microscopy (OTM) and in phase contrast microscopy (PCM). Aberrations are enhanced by focusing 0.5 mm into the bulk. On the right column, laser pulse energies in μJ are specified.	53
4.7	Aspect of fs laser-induced traces in a-SiO ₂ in phase contrast microscopy (PCM). The optical transmission microscopy observation was not recorded. Aberrations are enhanced by focusing 0.5 mm into the bulk. On the right column, laser pulse energies in μJ are specified.	54
4.8	Aspect of fs laser-induced traces in a-SiO ₂ (left) and Bk7 (right) for different numbers of shots per site in OTM and PCM. The maximal repetition rate is of 166 Hz. The laser pulse energy is 1 μJ . The number of shots per site (N) is indicated.	55
4.9	Aspect of fs laser-induced traces in a-SiO ₂ for different repetition rates. Laser pulse energies in μJ are specified. The number of shots (N) is also indicated.	56
4.10	Aspect of fs laser-induced traces in BK7 for different repetition rates. Laser pulse energies in μJ are specified. The number of shots is also indicated. Note the difference of scale bars.	57
4.11	Aspect of fs laser-induced traces in BK7 for different repetition rates. The number of shots is N=50000. The laser pulse energy is 1.2 μJ . The repetition rates are indicated.	58
4.12	Energy threshold for bulk modification versus pulse duration. Down- and up-chirped pulses have been focused in the bulk of fused silica.	59
4.13	Aspect of traces induced by down-chirped pulses in fused silica (left) and BK7 (right) in optical transmission microscopy (OTM) and phase contrast microscopy (PCM). The laser pulse energy is 1 μJ . The laser pulse durations (τ) are given in ps.	60
4.14	Excitation footprints in a-SiO ₂ at the end of a 2 ps irradiation pulse. (a) incident fluence (J/cm^2); (b) absorbed energy (J/cm^3); (c) peak intensity distribution (W/cm^2); (d) peak electronic density (cm^{-3}).	61
5.1	Principle of the time-resolved spectral interferometry, reproduced from [7]. M: mirror; BS: beam splitter; S: sample; SL: slit, SP: spectrometer	64
5.2	Principle of the transient lens method, reproduced from [8]. The pump beam is focused at $z = d$ and the probe beam at $z = 0$	65
5.3	Principle of the pump-probe experiment. SHG: second harmonic generation, SP: scattering element.	66

5.4	Experimental estimate of the temporal resolution of the pump-probe setup.	69
5.5	Time-resolved observations in optical transmission microscopy (OTM) and phase contrast microscopy (PCM) in fused silica for pump-probe delays of 1 ps, 10 ps and 500 ps. For the comparison, PCM and OTM pictures of the permanent imprint are shown. Phase contrast microscopy observations corrected from absorption (PCM corrected) are also presented for the time-resolved pictures. The laser pulse energy is $9.9 \mu\text{J}$. The regions permanently modified are indicated with dashed lines.	71
5.6	Time-resolved observations in optical transmission microscopy (OTM) and phase contrast microscopy (PCM) in BK7 for pump-probe delays of 1 ps, 10 ps and 500 ps. Otherwise as Fig. 5.5.	72
5.7	Time-resolved PCM picture in a-SiO ₂ (false colors) 500 ps after excitation. The location of the optical axis (O.A.), the location of the estimated focal plane and the half-angle of the focusing cone Ω_{SiO_2} are indicated. The laser pulse energy is of $9.9 \mu\text{J}$	73
5.8	Time-resolved observations in optical transmission microscopy (OTM) and phase contrast microscopy (PCM) in fused silica for pump-probe pulses delays of 1 ps, 10 ps and 400 ps. For comparison, PCM and OTM pictures of the permanent laser-induced traces are shown. Phase contrast microscopy observations corrected from absorption (PCM corrected) are also presented for the time-resolved pictures. The laser pulse energy is 220 nJ. The regions permanently modified are indicated with dashed lines.	77
5.9	Time-resolved observations in optical transmission microscopy (OTM) and phase contrast microscopy (PCM) in fused silica for pump-probe delays of 0.8 ns, 4.5 ns and 9.5 ns. Otherwise, as Fig. 5.8.	78
5.10	Estimate of the importance of the halo visible in phase contrast microscopy (PCM) observations. On the left, a time-resolved PCM observation corresponding to a laser energy of 220 nJ and a pump-probe pulse delay of 400 ps is presented. On the right hand side, we show the result of the Abel inversion in the analysis plane indicated by the dashed line. For comparison, the original grey-level profile in the analysis plane is plotted in the same graph.	79

5.11	Time-resolved PCM observations corresponding to a laser pulse energy of 220 nJ for different pump-probe delays ($\Delta\tau$) in bulk a-SiO ₂ . The colormap and the contrast were adjusted to enhance the intensity variations in the central region of energy deposition. The corresponding axial cross sections along the optical axis (O.A.) are overlapped with the original time-resolved PCM images. The region between the dashed bars corresponds to the zone where a low index cavity (void) remains visible in permanent regime ($\Delta\tau \rightarrow \infty$).	80
5.12	On the left, a time-resolved PCM observation corresponding to a laser energy of 220 nJ and a pump-probe delay of 100 ps is presented. On the right, the evolution of the region of interest (ROI I) is plot versus time. The value of ROI I in permanent regime is indicated on the graph.	82
5.13	On the left, a time-resolved PCM observation corresponding to a laser energy of 220 nJ and a pump-probe delay of 100 ps is presented. On the right, the evolution of the region of interest (ROI II) is plot versus time. The value of ROI II in permanent regime is indicated on the graph.	83
5.14	On the left hand side, a time-resolved PCM observation (false colors) corresponding to a laser pulse energy of 220 nJ and a pump-probe pulse delay of 800 ps is presented. The region where the pressure wave is visible is indicated. On the right hand side, the position of the pressure wave with respect to the optical axis (O.A.) is plotted versus time.	84
5.15	Example of transient absorption picture in Bk7.	85
5.16	Overview of the free carrier absorption versus time.	86
5.17	Free carrier density estimate 1 ps after excitation (left) in BK7. The region between the solid lines corresponds to the region of permanent modification. On the right, the free carrier density in the plane indicated by the dashed line is plotted for different values of $\Delta\tau$. The estimate of the free carrier decay rate (τ_{BK7}) is deduced from a fit of the experimental points by a first order exponential decay.	87
5.18	Same as Fig. 5.17 for an IOG-10 glass.	87
6.1	Adaptive closed-loop setup including a pulse shaping apparatus.	90
6.2	Evolutionary algorithm flowchart.	91
6.3	Illustration of discrete crossover between two parents. The random allele exchange results in the creation of two new offspring.	93
6.4	Scheme of the mutation operator. The strategy parameters $\bar{\sigma}$ are mutated first (a) and serve as a basis for mutating \bar{x} (b).	94

6.5	Procedure of fitness assignments based on pixel evaluation (left) and on target mask resemblance (right).	97
6.6	(a): Temporal intensity distribution produced by the optimal solution in a single irradiation sequence. (b): Comparison of the axial cross-sections induced by a short pulse (SP) with a FWHM duration of 200 fs and the optimal pulse (OP) with the target (b). (c): Phase contrast microscopy (PCM) observation of traces induced by a short pulse (SP) and an optimal pulse (OP) with a laser pulse energy of 0.4 μJ . Aberrations are minimized by focusing at $D=170 \mu\text{m}$ in the bulk.	102
6.7	Temporal intensity distribution produced by the optimal solution in a single irradiation sequence (left). The laser pulse energy is limited to 1.4 μJ . On the right, PCM observation of traces induced by a short pulse (SP) with a FWHM duration of 200 fs and an optimal pulse (OP). Aberrations are enhanced by focusing at $D=500 \mu\text{m}$ in the bulk.	103
6.8	Temporal intensity distribution produced by the optimal solution in a single irradiation sequence (left). The laser pulse energy is limited to 1.4 μJ . On the right, PCM observation of traces induced by a short pulse (SP) with a FWHM duration of 200 fs and an optimal pulse (OP). Aberrations are enhanced by focusing at $D=500 \mu\text{m}$ in the bulk.	104
6.9	Sequential energy deposition in free carriers in irradiated a-SiO ₂ for a single laser pulse energy of 1 μJ and 120 fs duration. The integration domains are written on the left side of the figure.	106
6.10	PCM observation of the traces imprinted by different individuals after 50 generations in a single shot regime ($N=1$). The selection criterion is based on a Gaussian mask. The laser pulse energy is limited to 350 nJ. Aberrations are minimized by focusing at $D=170 \mu\text{m}$ in the bulk.	108
6.11	Temporal intensity distribution produced by the optimal solution (left). On the right, PCM observation of traces induced by a short pulse (SP) and an optimal pulse (OP) in a single shot regime ($N=1$). The laser pulse energy is limited to 350 nJ. Aberrations are minimized by focusing at $D = 170 \mu\text{m}$ in the bulk.	109

6.12	(a): Temporal intensity distribution produced by the optimal solution. (b): Comparison of the axial cross-sections induced by a short pulse (SP) and the optimal pulse (OP) with the target (left). (c): Phase contrast microscopy (PCM) observation of traces induced by the accumulation of 50000 short pulses (SP) and 50000 optimal pulses (OP) at a repetition rate of 100 kHz. The energy per pulse is limited to 170 nJ. Aberrations are minimized by focusing at $D = 200 \mu\text{m}$ in the bulk.	110
6.13	Aspect of traces induced by the accumulation of $N=50000$ chirped pulses in BK7 in OTM (left) and PCM (right). The input energy is limited to $E=170$ nJ. The pulse durations τ in ps are given in the right column. Aberrations are minimized by focusing at $D = 200 \mu\text{m}$ in the bulk.	111
7.1	Experimental layout employed for longitudinal waveguide writing (adapted from [9]). The thickness of the sample (W) is of 5 mm in order to avoid beam clipping. Note the difference between the sample velocity V_{sample} and the velocity of the focal point V_{scan} inside the sample of refractive index n . The arrow indicates the direction of the sample movement.	117
7.2	Phase-contrast microscopy observation of an array of optical structures written longitudinally in a-SiO ₂ . The laser energy is indicated in microjoules. The scan speed is of $5 \mu\text{m/s}$. The laser source employed has a repetition rate of 100 kHz.	118
7.3	Optical transmission microscopy (OTM) and phase-contrast microscopy (PCM) observations of an array of optical structures written longitudinally in BK7 with a 150 fs pulse duration. The laser pulse energy is indicated in microjoules. The scan speed is of $50 \mu\text{m/s}$. The laser source employed has a repetition rate of 100 kHz. The contrast of the OTM picture was uniformly enhanced in order to render the structures visible.	119
7.4	Intensity distribution in a plane located at 115 cm far from the exit of a waveguide written longitudinally in BK7 with a laser energy of $1.1 \mu\text{J}$ and a pulse duration of 130 fs. The waveguide was injected with a HeNe laser through the same objective as we used for generating the structure ($NA = 0.45$). The radius at which the rings vanish (r) is indicated.	120
7.5	Experimental layout employed for transverse waveguide writing (adapted from [9]). The arrow indicates the direction of the sample movement.	122

7.6	Optical transmission microscopy (OTM) and phase contrast microscopy (PCM) observations of a waveguide generated in a-SiO ₂ with a laser pulse duration of 130 fs. The laser pulse energy is 160 nJ and the scan velocity is 100 μm/s. A PCM observation of the accumulation of 50000 pulses at the same laser energy is also shown.	123
7.7	Phase contrast microscopy (PCM) observations of imprints visible in BK7 after irradiation. The structures were not visible in optical transmission microscopy. The laser energy is 160 nJ per pulse. (a): Structure generated with a femtosecond pulse duration (SP) in a transverse writing configuration. The scan velocity is of 100 μm/s. (b): Structure generated with an optimal pulse (OP) provided by the optimization algorithm. The scan velocity is of 100 μm/s. (c): Accumulated effect of N = 50000 SP. (d): Accumulated effect of N = 50000 OP.	124
7.8	Intensity distribution in a plane located at 115 cm far from the exit of a waveguide written transversally in BK7 with a laser energy of 1.1 μm at a velocity of 100 μm/s. The waveguide was injected with a HeNe laser through a 20x objective. The radius at which the rings vanish (r) is indicated.	125
7.9	Optical transmission microscopy (OTM) (a) and phase contrast microscopy (PCM) (b) observations of imprints generated with an optimal pulse (OP) provided by the optimization algorithm in BK7. The scan velocity is of 50 μm/s. The laser energy is 520 nJ per pulse. (c): Accumulated effect of N = 50000 OP. The laser energy is 160 nJ per pulse. A gray level axial cross section is overlapped with the PCM capture.	126

List of Tables

3.1	Main characteristics of the output pulses for the different lasers	23
6.1	Standard parameters of the evolutionary strategies used for performing adaptive pulse shaping experiments.	100

