CHAPTER 1

Introduction

With the recent developments in micro and nanotechnologies, there is a need for a robust and flexible tool, able to process various types of materials on a micrometric and submicrometric scale. To this extent, ultrafast laser sources appear as natural candidates for processing in the bulk of transparent materials, with the purpose of adding new functionalities to the induced structures. The continuous improvement of femtosecond laser sources triggers a considerable activity, based on the outstanding characteristics of the laser pulses. At first, the short duration of femtosecond laser pulses can be used as diagnosis tools to monitor ultrafast phenomena, like phase transitions or short lifetimes of excited states. Ultrashort pulses used in combination with focusing techniques constitute an exceptional source of intense, well localized light packets. This high intensity opens the door to the study of a plethora of nonlinear mechanisms, including nonlinear propagation and nonlinear ionization. Nonlinear ionization implies that light absorption and subsequent material photomodification is possible even in transparent materials. When nonlinear ionization is triggered, a population of free carriers builds up and can experience further laser heating via inverse bremsstrahlung on the pulse duration timescale. As a result, a material far from equilibrium characterized by a cold bulk coexisting with a hot electron gas is obtained. Upon energy relaxation, new material states emerge. With the help of adequate focusing methods, the spatial confinement of the energy deposition allows material modification on micrometric and submicrometric scales.

Ultrafast lasers offer the possibility to perform genuine 3D micromachining in the volume of transparent materials relying on nonlinear propagation and characterized by a localized energy deposition and minimal damage extension, overtaking the capabilities of alternative techniques. However, practical limitations appear. A few of them are summarized below. The magnitude of the laser-induced refractive index is generally low (typically of 10^{-3}), depending on the material of interest. Some type of glasses exhibit a negative refractive index change upon ultrafast laser irradiation, limiting the use of those glasses as substrates for writing of depressed cladding waveguides [10]. When operating deep in the bulk, spherical aberrations prevent from controlling the morphology of the photoprocessed region, with negative consequences on the micromachining capabilities. Those difficulties are not due to the method, but rather to the poor understanding of the fundamental nature of the induced modifications. To this extent, there are two challenges, both scientific and technological.

By understanding the sequence of relaxation mechanisms involved, and by controlling them via selective energy deposition, we aim at promoting ultrafast lasers as relevant and viable microfabrication tools for processing the bulk of transparent materials. This works provides thus the preliminary steps in applications of adaptive techniques to optimal laser-processing. To this extent, we focus on two aspects. We attempt to follow the physical processes associated to laser-induced modification of bulk transparent material, and we also seek for control factors, in order to generate optical structures with arbitrary morphologies and optical properties.

This thesis is organized as follows.

The second chapter aims at showing how light can be considered as a material structuring tool. A basic introduction to nonlinear phenomena, in particular nonlinear propagation and nonlinear ionization is presented. The consequence of the free-carrier generation on the transient refractive index is discussed. Additionally, a model for pulse propagation in transparent materials based on the nonlinear Schrödinger equation is introduced. This model takes into account nonlinear phenomena and allows to make predictions on the energy deposition. Finally, the end of the chapter deals with the energy relaxation mechanisms, and their consequences on the permanent optical properties of the irradiated material.

In the third chapter we present the laser sources employed all along this thesis, as well as the fundamentals of temporal pulse shaping, as this method will be employed in Ch. 6. A section is dedicated to the general problem of beam focusing in presence of a dielectric interface, and explanations as well as estimations regarding spherical aberrations and their minimization are provided. Furthermore, we present the fundamentals of phase contrast microscopy (PCM), as PCM is the main tool employed to analyze laser-induced refractive index changes in the bulk of transparent materials.

The fourth chapter is exclusively composed of experimental analysis in the bulk of irradiated borosilicate crown BK7 and amorphous fused silica (a-SiO_2), in PCM and in optical transmission microscopy (OTM) under different irradiation conditions. Moreover, observation results are correlated with simulation results provided by the model presented in Ch. 3.

In Ch. 5, the dynamics of the laser-induced refractive index changes at the first moments after material excitation is followed with time-resolved PCM and OTM techniques. In this way, information about the nature and the timescales of energy relaxation channels is obtained. Details are provided about the experimental realization and about the capabilities of the setup. Experimental results for high laser pulse energies and laser pulse energies close to the permanent modification threshold are shown. Additionally, at the end of the chapter, an estimation of the lifetime of free carriers in BK7 and in IOG-10 based on time-resolved OTM microscopy is presented.

Based on the temporal guideline provided by the previous chapter, Ch. 6 explores the possibility to control the rate and localization of energy deposition into the bulk of a-SiO_2 and BK7 via adaptive pulse shaping in the temporal domain. The heart of this adaptive pulse shaping apparatus is an evolutionary strategies. Therefore, an introduction to the principles of evolutionary algorithms is presented, as well as technical details regarding the implementation. Some selected experimental results are shown. The main driving force of this chapter is to demonstrate some control over the laser-induced refractive index changes via temporal pulse manipulation.

In Ch. 7, we use the results of the previous chapter to generate waveguides. We demonstrate the possibility to write embedded guiding structures at optical frequencies in the bulk of transparent materials which do normally not show a favorable response in standard ultrafast irradiation conditions.

Finally, Ch. 8 summarizes the main experimental results, and discusses about the possible evolutions that could take place in the field of laser processing of transparent materials.