Chapter 1

Introduction

The nature of light is one of the most fascinating key problems in modern physics. It was studied by Sir Isaac Newton [1] who proposed a corpuscular theory, and Christiaan Huygens [2] who developed a competing wave theory. The high prestige and position of Newton caused that his theory remained unchallenged until the discovery of interference, when the wave theory was revived. Afterwards, Maxwell described light as a propagation of electromagnetic waves [3]. The continuous nature of light was again questioned when the photoelectric effect was observed along with the Compton effect. An explanation was finally provided by Albert Einstein [4, 5, 6, 7], who restructured existing theories and created a basis for the presently accepted wave-particle duality [8] in modern quantum mechanics.

One of the greatest achievements and practical applications of quantum mechanics is the LASER [9, 10, 11], which is an acronym originating from **L**ight **A**mplification by **S**timulated **E**mission of **R**adiation. This new light source completely revolutionized not only research but also everyday life. It became this common, that it entered the English vocabulary as "laser", losing its capitalization. The variety of today lasers span from narrow bandwidth, continuous wave lasers to devices delivering ultrashort pulses with tremendous instantaneous intensities. The use of short pulses turned out to be extremely useful in numerous applications. The instantaneous intensity of the laser pulse depends inversely from its duration, thus for relatively low energies one can achieve huge intensities. Such short pulses are one of the most rapid controllable processes so far. The time scales are now on the order of attoseconds [12], and the commercially available sources are capable of delivering pulses as short as few femtoseconds on daily bases, which for infrared wavelengths corresponds to less then two oscillations of the electric field within the duration of the pulse [13, 14, 15]. Generation of such short pulses is a task which due to the Uncertainty Principle requires a very wide spectrum of the laser [16, 17]. The broad bandwidth, although necessary, is not the only condition for an ultrashort pulse duration. Group velocity differences between the spectral components causes the pulse to spread in time, therefore it is absolutely essential to balance this dispersion already in the cavity of the laser [16, 17, 18, 19]. In this way the laser pulse remains short in the cavity, but as it propagates outside, the dispersion of the transmitting optics or even air stretches the pulse in time. In order to counterbalance for the dispersion various designs have been introduced, starting from passive grating [20, 21, 22] or prism compressors [23, 22] to more elaborate active phase control devices involving liquid crystals [24, 25], acousto-optic modulators [26, 27], or deformable mirrors [28]. The original motivation behind the construction of these devices was mostly pulse compression [29, 30, 31], but the great potential for changing the phase independently for each spectral component opened the possibilities of pulse shaping [32]. Among other things it triggered a fascinating and rapid development in the field of coherent control. The huge advantage of pulse shaping techniques is the ability to gather information about a system while controlling it with the use of shaped pulses [33]. The type and quality of the collected information depends on the level of control over the pulse structure.

Soon after the introduction of a single array Spatial Light Modulator (SLM) capable of phase control, double arrays SLM became available [34, 35, 36] with optional amplitude modulation. The amplitude modulation is achieved in the commercial SLM [37] by cutting out one polarization component of the shaped beam, thus removing the polarizer results in a modulation of phase and polarization [38, 39, 40]. The arrangement of the two arrays unfortunately limits the spectral polarization states available from such a device to ellipses with their principal axes fixed. The rotation of the polarization, is obtained when a third array is employed [41].

In the first chapters of this thesis the simultaneous and independent shaping in the frequency domain of the phase, amplitude, and polarization is presented. In Chapter 2 the Jones formalism is used for showing the dependency between the elliptical polarization parameters, the principal axes ratio and orientation, and the amplitudes of the orthogonal polarization components and their relative phase shift. This important relation permits to generate a desired polarization state by changing the amplitudes and the relative phase shift. In Chapter 3 a shaper scheme that fully controls the spectral phase, amplitude, and polarization of femtosecond laser pulses is presented. In particular, it allows for independent manipulation over the major axis orientation and the axis ratio of the polarization ellipse. This is accomplished by integrating a 4f-shaper setup in both arms of a Mach-Zehnder interferometer and rotating the polarization by 90° in one of the arms before overlaying the beams. Next, in Chapter 4 a different type of shaper setup is introduced which takes advantage of laser pulses passing through a spatial light modulator twice, thereby effectively utilizing a four array configuration. This approach also grants control over the phase, amplitude, and polarization without facing problems of interferometric stability but with certain limitation concerning the rotation of the polarization ellipse orientation. The Jones vector of the light wave after passing through the setup is considered in detail including polarization sensitive grating efficiency. A new method of counteracting the polarization dependent grating transmission is described and a comparison between the desired and recorded pulses is presented. Chapter 5 discusses various alignments of the optical axes of the crystal arrays placed inside the Spatial Light Modulator and the interaction of such modulators with light. A compact four array modulator is proposed, which is capable of complete phase, amplitude, and polarization control, as the parallel shaper setup.

The next Chapter of this thesis describes the coherent control experiment involving the application of phase, amplitude, and polarization shaping performed on the sodium potassium molecule in a gas phase produced in the molecular beam apparatus.

The coherent control experiment performed and described in the frame of this work involves a closed learning loop, a concept, which is based on an idea of Herschel Rabitz [33]. It allows to circumvent the extremely complicated problem of the calculation of the electromagnetic field, which is optimal for achieving the desired goal. This approach has been successfully applied in optimization of nonlinear processes [42], X-ray generation, [43], femtochemistry [44], control of chemical reactions [45, 46], femtobiology [47], and many more.

Optimal control of the ionization described in Chapter 6 compares various types of optimizations, where different pulse parameters are modified. These types are phase, or phase and amplitude, or phase, amplitude, and polarization optimization. The impact of each parameter on the achieved efficiency is discussed in detail together with the temporal and spectral structures of an optimal pulse. This chapter closes the part of this thesis concerning the polarization shaping and coherent control.

Chapter 7 is dedicated to the photoassociation of cold Rubidium molecules from atoms trapped inside of a Magneto-Optical Trap (MOT). Cooling and trapping molecules, since the first experimental realization of MOT [48], is a dynamically expanding field which allows to study fascinating subjects such as ultracold atom collisions [49, 50, 51], photoassociation [52, 53, 54] and creation of Bose Einstein Condensate [55].

In this Chapter the possible photoassociation with the use of fs laser pulses is discussed. This novel approach combines techniques of laser cooling with ultrafast lasers. The results of the pump-probe experiment performed with an 802 nm centered bandwidth pump pulse and a 496 nm centered probe pulse are presented and discussed.